

Fluorescent nanosensors for metal ions

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Abstract

The precise detection of pH levels and metal ions is essential in fields ranging from environmental monitoring to biomedical diagnostics, as both pH variations and metal ion concentrations are crucial indicators of chemical and biological processes. Carbon dots (CDs), known for their photostability, low toxicity, and tuneable fluorescence, have emerged as promising sensors for pH and metal ion detection. This thesis explores the development of CD-based sensors for these applications, integrating machine learning techniques to enhance the sensors' sensitivity, selectivity, and analytical robustness.

The research began with fructose and p-phenylenediamine-derived CDs for pH monitoring across a broad range (pH 3 to 10). Detailed investigations revealed the CDs' fluorescence response to pH changes, influenced by quantum confinement and surface state effects. To refine the precision of pH sensing, Gaussian process regression (GPR) was used to predict pH levels from spectral data accurately, while linear discriminant analysis (LDA) provided precise pH classification. These combined approaches bolstered the CDs' sensitivity to pH, enhancing the reliability of CD-based pH sensors.

The metal-sensing capabilities of CDs were initially explored with biomass-derived CDs. Jacaranda leaf-based CDs demonstrated significant potential as ratiometric pH sensors and selective Cu(II) sensors, while berry-based CDs showed promise for Fe(III) detection. Inspired by these findings, fructose-based CDs were further developed for selective sensing of Cu(II) and Fe(III). To investigate the cross-reactive sensing responses of fructose-based CDs with varying N-doping levels, a four-CD-based sensor array was developed under optimised hydrothermal conditions. Using support vector machine (SVM) and LDA methods, this array successfully classified fourteen metal ions, including complex differentiation among Cu(II), Co(II), and Ni(II) in mixed-ion environments. Rigorous testing in real-world samples, such as environmental water and human urine, confirmed the array's efficacy, offering a practical, sensitive, and cost-effective solution with enhanced accuracy across variable pH conditions.

This research highlights the significant advances in chemical sensing that are made possible through the integration of machine learning with carbon dot technology. Establishing a foundation for robust, versatile, and precise CD-based pH and metal ion sensors, these findings underscore the potential of CDs for applications in environmental, biomedical, and industrial settings, addressing critical needs in selective detection, cost efficiency, and sensor stability.

Statement of originality

I hereby declare that the work presented in this thesis has not been submitted to any other institution for a degree or as part of the requirements for a degree. This thesis has been written and prepared solely by the author, who also designed and analysed all the projects and studies contained within. The same applies to the manuscript submitted RSC Sensor & Diagnosis under review, which was included as part of this thesis.

I confirm that the work in this thesis is original, and all external sources of information and literature used have been appropriately acknowledged and referenced.

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Permission to include the published material has been granted by the corresponding author.

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As supervisor for the candidature upon which this thesis is based, I can confirm that the author attribution statements above are correct.

Hala Zreiqat
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List of abbreviations

a.u.	Arbitrary units
AAS	Atomic absorption spectroscopy
Acc	Accuracy
AES	Atomic emission spectroscopy
AIE	Aggregation-Induced Emission
ATP	Adenosine-5'-triphosphate
ATR	Attenuated total reflectance
B-CD	Biomass based carbon dots
BBB	Blood-brain barrier
BE	Binding energy
Boc	Tert-Butyloxycarbonyl
BODIPY	4,4-Difluoro-4-bora-3a,4a-diaza-s-indacene
CD	Carbon dot
CHEQ	Chelation-Enhanced Quenching
CL	Chemiluminescent
CNS	Central nervous system
DFT	Density functional theory
EDTA	Ethylenediaminetetraacetic acid
EL	Electrochemical
F-CD	Fructose based carbon dots
F1	F1 score
FN-CDs	Fluorine and nitrogen co-doped carbon dots
FRET	Förster resonance energy transfer
FWHM	Full width at half maximum
GCD	Gold carbon dots
GPR	Gaussian process regression
GQDs	Graphene quantum dots
HAD	Heteroscedastic discriminant analysis
HCA	Hierarchical cluster analysis
HCD	Hollow luminescent carbon dots
ICP-MS	Plasma mass spectrometry
ICT	Intramolecular Charge Transfer
LMCT	Ligand-to-Metal Charge Transfer
LOD	Limit of detection
MAE	Mean absolute errors
ML	Machine learning
MOF	Metal-organic frameworks
MSE	Mean squared errors
MWCNTs	Multi-walled carbon nanotubes
OPD	o-phenylenediamine
OvO	One-vs-one

OvR	One-vs-rest
PA	Photoacoustic
PCA	Principal component analysis
PET	Photoinduced electron transfer
PL	Photoluminescent
PLS-DA	Partial least squares discriminant analysis
PN-CDs	Phosphorus and nitrogen co-doped
PPD	p-phenylenediamine
Prec	Precision
QDA	Quadratic discriminant analysis
QY	Quantum yield
R²	Coefficients of determination
RDA	Regularised discriminant analysis
Rec	Recall
Ref.	Reference
ROS	Reactive oxygen species
SVM	Support vector machines
SVM	Linear discriminant analysis
TNP	2,4,6-trinitrophenol
UCPL	Up-conversion photoluminescence
v/v	Volume per volume
VSA	Virtual sensor array
XPS	X-ray photoelectron spectroscopic
λ_{em}	Lamda max (wavelength corresponding to the maximum intensity in fluorescence emission spectrum)
λ_{ex}	Lamda max (wavelength corresponding to the maximum intensity in fluorescence excitation spectrum)
λ_{max}	Lamda max (wavelength corresponding to the maximum result in a spectrum)
σ_f	Signal variance of GPR model's kernel function
σ_l	Length scale of GPR model's kernel function

1 Introduction

1.1 Chemical analytes in health and disease

The impact of diseases on human health is profound, affecting both quality of life and longevity. Understanding the underlying causes of diseases and their progression requires insights into the molecular and chemical changes occurring within the body. Chemical analysis plays a crucial role in this regard, offering solutions that help detect, monitor, and manage health conditions.¹⁻³ By analysing chemical analytes in biological samples such as blood, breath, or urine, researchers can identify biomarkers that signal disease onset, progression, or response to treatment.⁴ For instance, chronic kidney diseases of multi-factorial origin, such as those observed in Sri Lanka, require a comprehensive chemical analysis of water quality and other contributing factors to provide effective solutions.⁵ Furthermore, the analysis of the "blood exposome"—the complete set of chemicals present in the blood—provides key insights into potential environmental and biological factors contributing to diseases, driving further research into their causal links.⁶ Chemical analysis has also been developed as an advanced intervention tool for monitoring toxins and safeguarding human health.^{7, 8} For instance, the assessment of heavy metals and their impact on human health through chemical analysis has led to the implementation of crucial interventions, such as worker safety programs and medical initiatives, aimed at minimising exposure and mitigating health risks.⁹

Many of the critical questions underpinning biomedical research today revolve around chemistry—how small molecules, macromolecules, and chemical environments influence cellular health, or how they are disrupted in disease states.¹⁰⁻¹² It is therefore essential to have reliable methods to measure these chemical analytes both within biological systems and in the environmental systems with which they interact. From public health assessments to disease diagnostics, chemical analysis offers a window into understanding complex health issues. This thesis focuses specifically on the role of pH levels (Chapter 3) and metal ions (Chapter 4-5), both of which play critical roles in cellular function and are often perturbed in disease, setting the stage for the next sections that explore their detailed analysis in biological contexts.

1.2 The need for metal ion sensing

Metal ions are integral to numerous metabolic processes within the human body, supporting critical functions such as respiration, cell growth, enzymatic reactions, immune responses, and gene regulation.¹³⁻¹⁷ These ions act as essential cofactors that maintain the proper functioning

of various biological pathways. However, deviations from normal concentrations can disturb biological homeostasis, potentially leading to diseases like diabetes, cancer,¹⁸ genetic disorders,¹⁹ and neurodegenerative conditions.²⁰ While genetic predispositions are often cited in neurological disorders, growing evidence suggests that metal-induced neurotoxicity plays a significant role in their development and progression.²¹ The importance of metal ion sensing extends across multiple fields. In biological systems, accurate measurement of metal ions helps clarify their roles in vital processes,²² such as the involvement of Zn(II) in DNA synthesis and immunity.²³⁻²⁵ Additionally, sensing toxic heavy metal ions like lead and mercury is crucial, as exposure to these metals can have severe health and environmental consequences.²⁶⁻²⁸ Analytical chemistry also benefits from the development of highly sensitive and selective chemo sensors for metal ions, which are critical for monitoring both essential and toxic metals.²⁹⁻³¹

Transition metals, although present in trace amounts, play indispensable roles in human biology.^{13, 32} Copper, for instance, is vital for mitochondrial function³³, antioxidant defence³⁴, and the formation of connective tissue³⁵. However, excess Cu(II) can cause oxidative stress,³⁶ contributing to neurological diseases such as Alzheimer's and neuroblastoma.³⁷⁻⁴⁰ Copper deficiency, on the other hand, can result in health issues like anaemia, neutropenia, and impaired immune function.⁴¹⁻⁴⁴ The dual nature of copper's role in the body highlights the delicate balance needed to maintain its physiological levels. Similarly, cobalt is an essential component of vitamin B12 (cobalamin),^{45, 46} involved in DNA synthesis⁴⁷ and the maintenance of the nervous system.⁴⁸ It plays a significant role in red blood cell production⁴⁹ and fat and protein metabolism.⁵⁰⁻⁵² However, elevated levels of Co(II) can induce oxidative stress⁵³⁻⁵⁵ and lead to cytotoxicity,^{56, 57} impairing metabolic functions^{33, 58} and increasing the risk of diseases such as cancer⁵⁹ and neurodegenerative disorders.³⁶ Excessive cobalt exposure, often seen in industrial settings,^{60, 61} can result in adverse effects ranging from respiratory problems⁶² to allergies.^{63, 64} Consequently, managing cobalt intake is critical to human health. In above cases, these metal ions play complex, multifaceted roles in human health, where both deficiencies and excesses can lead to significant health problems. As a result, early detection and precise regulation of metal ion concentrations are paramount in preventing disease and maintaining physiological balance.

Toxic metals like mercury, cadmium, and lead, for which there is no known biological function, pose significant risks even at low exposure levels.²⁷ Mercury is a neurotoxin that causes muscle

weakness⁶⁵ and sensory impairments,⁶⁶ while cadmium and lead, known to mimic or antagonise the biological roles of calcium^{67, 68} and zinc,^{69, 70} can lead to cancer,⁷¹ kidney disorders,⁷² and reduced reproductive health.⁷³⁻⁷⁵ Furthermore, organometallic compounds of these metals, such as organolead, are particularly dangerous due to their ability to access biological membranes of the organism by passive diffusion, exacerbating their toxicity.⁷⁶ Therefore, environmental detection efforts should prioritise metal ions to mitigate the harmful effects of these toxic substances.

1.3 The need for pH sensing

The regulation of intracellular pH is essential for maintaining cellular function and supporting numerous biological processes, such as metabolism, vesicle trafficking, cell volume regulation, membrane potential, muscle contraction, and intracellular signalling.⁷⁷⁻⁷⁹ Maintaining an optimal pH balance within cellular compartments ensures physiological stability.⁸⁰ Disruptions in pH levels can have severe consequences, such as impaired signalling pathways and the onset of diseases.⁸¹ For instance, low intracellular pH can lead to protein denaturation, which affects the functionality of enzymes critical for biochemical reactions.⁸² This loss of enzyme activity disrupts cellular homeostasis and can have cascading effects throughout the cell. Acidic conditions may also impair nerve transmission by trapping neurotransmitters within acidic endosomes, causing synaptic dysfunction.^{83, 84} On the other hand, elevated pH levels have been linked to increased growth signalling, potentially leading to uncontrolled cell proliferation, a key factor in cancer progression.^{85, 86} Thus, real-time monitoring of intracellular pH is essential for understanding cellular health and disease progression.

In the biomedical field, pH sensing plays a pivotal role, particularly in diagnosis and health monitoring. Electrochemical pH sensors have advanced significantly due to their compactness and integration with modern techniques.⁸⁷ Metal oxide-based electrochemical sensors are widely used in biomedical contexts, offering high sensitivity and stability.⁸⁸ Fluorescent pH sensors, which benefit from capability to provide intracellular pH, have also gained attention for biomedical applications, providing efficient and accurate pH measurement.⁸⁹ However, there is still a critical need to develop standardised protocols to benchmark these systems, ensuring their reliability and consistency in clinical use.

pH sensing technologies also have important applications in the food industry, where they are used to monitor food quality and safety.^{90, 91} Changes in pH levels can indicate spoilage or

contamination in packaged food products.⁹² Various materials have been explored for these purposes, including natural colorants,^{90, 93} biopolymers,⁹⁴ and metal oxides.⁸⁸ Colorimetric sensors, such as those using chitosan-based dyes or natural plant dyes like *Bauhinia blakeana* Dunn, change colour in response to pH shifts, providing a visual indicator of food freshness.⁹⁵ Additionally, biopolymer-based pH-sensing films and wraps, made from materials like starch, cellulose, and κ -carrageenan, offer real-time monitoring of food quality.^{96, 97} These developments aim to enhance the environmental sustainability and convenience of food packaging, ensuring safer and more effective food monitoring systems. Overall, the development of pH sensors continues to progress across both biomedical and food industry applications, offering innovative solutions for real-time monitoring and improved health and safety outcomes.

1.4 Current sensing techniques

The ability to accurately detect and monitor metal ions and intracellular pH is essential for maintaining the body's homeostasis as stated above. Traditional analytical methods, such as atomic absorption spectroscopy (AAS),⁹⁸ atomic emission spectroscopy (AES),⁹⁹ and inductively coupled plasma mass spectrometry (ICP-MS),¹⁰⁰ are well-established for their precision and sensitivity in detecting trace amounts of elements. However, these methods are limited to laboratory settings due to their high cost, complex instrumentation, and the time-consuming nature of the analysis process. The limitations of these techniques include the need for expensive equipment, large sample quantities, and immobility, making them unsuitable for rapid or point-of-care diagnostics.

In recent years, fluorescence spectroscopy has gained popularity as a highly effective alternative for detecting various analytes and chemical conditions due to its high sensitivity, ease of use, and ability to provide rapid results.¹⁰¹⁻¹⁰⁴ Fluorescent sensor technologies offer great potential for real-time monitoring, as they can be designed to target specific ions or molecules with high selectivity. Various fluorescent materials have been developed for these applications, such as organic dye,¹⁰¹ and carbon dots.¹⁰⁵ Given these constraints, developing new, highly sensitive, and selective fluorescent sensors and sensor arrays for detecting metal ions like Cu(II), Co(II), and Ni(II), as well as for monitoring pH changes in biological systems, remains a critical area of research. Fluorescent sensor arrays offer significant advantages in terms of spatial and temporal resolution, ease of use, and the potential for non-invasive monitoring, making them a promising tool for future diagnostic and therapeutic applications.

1.5 Carbon dots-based sensors for biosensing

Carbon dots (CDs), a novel class of fluorescent carbon-based nanomaterials, have garnered substantial attention in fields such as biosensing, bioimaging, and therapeutic delivery.^{106, 107} These nanomaterials offer a range of advantageous properties, including exceptional biocompatibility, tuneable photoluminescence, low toxicity, and ease of synthesis. Such properties make CDs highly suitable for diverse biomedical applications. The unique physical and chemical properties of CDs are influenced by their size and structural characteristics. Typically less than 10 nm in diameter, carbon dots demonstrate size-dependent photoluminescence due to quantum confinement effects.¹⁰⁸⁻¹¹⁰ The π -conjugated graphitic core of CDs, alongside the option for functionalisation with various moieties, further expands their application range, allowing customisation for specific uses.^{109, 111, 112} Research has shown that controlling synthesis conditions—such as precursor selection, reaction time, and temperature—enables precise tuning of their optical properties, making CDs highly adaptable materials.¹⁰⁸

CDs exhibit extensive photoluminescence (PL) emission, typically covering wavelengths from the ultraviolet (UV) to the near-infrared (NIR) regions.^{113, 114} This broad emission range significantly enhances their versatility in various sensing and bioimaging applications. Notably, CDs have demonstrated full-range UV–Vis–NIR responsiveness, with emission wavelengths commonly spanning from deep red to the near-infrared region.^{115, 116} Such emissions in the red and NIR ranges are particularly advantageous for biological imaging applications due to their improved tissue penetration depth and reduced autofluorescence interference.

More specifically, the emission spectrum of CDs commonly ranges from 390 nm (blue) to over 600 nm (red).¹¹⁷⁻¹¹⁹ The exact emission wavelength is strongly dependent on various synthesis parameters and environmental factors, including particle size, surface functionalisation, solvent selection, and the degree of surface oxidation.^{113, 120, 121} For instance, certain studies have reported significant shifts in emission wavelength influenced by solvent variation, such as a reported shift from 475 nm to 515 nm upon changing solvents.¹²¹ This sensitivity to synthetic and environmental conditions underscores the tuneable and adaptable nature of CDs for specific sensing and imaging purposes.

CDs exhibit unique characteristics beneficial for sensing applications, such as adjustable fluorescence and chemiluminescence, stability, water solubility, ease of surface modification,

and cost-effectiveness derived from green synthesis methods.¹²² Their capabilities include detecting a diverse range of analytes, including environmental parameters, gases, and biological threats.¹¹⁵ Additionally, CDs offer a significant advantage in monitoring pH variations, particularly within biological systems, due to their responsiveness to electronic structure alterations involving protonation-deprotonation processes and energy-level shifts.¹²³ These properties, coupled with inherent low toxicity and biocompatibility, position CDs as promising candidates in advanced sensing technologies.^{124, 125} Recent developments have further expanded the applications and efficiency of CDs, marking them as highly attractive tools in contemporary research.

Recent advances in doped-carbon dots have enhanced their capabilities, expanding their role in biosensing and bioimaging.^{126, 127} CDs are widely used as fluorescent probes for detecting biomolecules like proteins, enzymes, and nucleic acids due to their tuneable fluorescence and selectivity.¹²⁸ Moreover, their biocompatibility makes them ideal for cellular and in vivo bioimaging, offering valuable insights into biological processes and disease states. The versatility of carbon dots extends to theragnostic applications, where they integrate diagnostic and therapeutic functions.¹²⁹ In this context, CDs can facilitate simultaneous disease detection and targeted drug delivery. Their multifunctional nature positions them as a promising tool for modern biomedical approaches. Beyond biomedical applications, CDs also exhibit significant potential in fields such as catalysis, optoelectronics, and environmental monitoring.¹³⁰⁻¹³²

Carbon dots have been proposed as an alternative to traditional semiconductor quantum dots, which have historically been used for similar purposes.¹³³ CDs, however, offer several advantages, including lower toxicity and simpler synthesis processes. The development of CDs has paralleled advancements in quantum dots, which were recognised with the 2023 Nobel Prize in Chemistry. This growing interest in luminescent nanomaterials, including both quantum dots and carbon dots, underscores their importance in fields ranging from biology to energy technologies.

In sensing applications, carbon dots offer significant benefits over traditional methods, particularly in detecting metal ions and pH changes.¹³⁴ Their fluorescence-based sensing capabilities provide low-cost, productive and highly efficient synthesis, convenient, and highly sensitive detection, addressing the limitations of single-target sensing strategies. CDs can be functionalised to detect a variety of analytes, including heavy metals,¹³⁵ and pH fluctuations,¹³⁶ making them versatile tools for biomedical sample testing, environmental monitoring and

diagnostics. The incorporation of metal ions or functional groups can further enhance the stability and performance of carbon dots, ensuring reliable sensing in challenging environments.^{137, 138} Machine learning techniques have also been integrated with carbon dot-based sensor arrays, enhancing the analysis and interpretation of complex data. By applying algorithms such as support vector machines (SVM), researchers can achieve ultrasensitive detection of analytes, including metal ions, antibiotics, and bacteria. These advancements demonstrate the potential of carbon dots in conjunction with machine learning for high-performance sensing applications, particularly in areas such as food safety and biomedical diagnostics.

CDs exhibit remarkable photoluminescent (PL) properties across a broad range of the visible spectrum.¹¹³ Their fluorescence can span from blue to red.¹³⁹ Green-emitting CDs, for instance, often display strong absorption between 350–450 nm, with intense emission centred around 537 nm.¹⁴⁰ Notably, under two-photon excitation, their fluorescence can exhibit red-shifted emission, extending from approximately 550 nm to longer wavelengths such as 630 nm and 680 nm.¹¹⁸ These tuneable optical characteristics make CDs highly versatile for applications in fluorescence-based sensing, imaging, and optoelectronics.

1.6 Overview of the thesis

This thesis begins with a comprehensive literature review. As carbon dots are a key focus of this research, the first half of the literature review delves into the theoretical foundations of carbon dots, exploring their optical properties, fluorescence mechanisms, and current applications in sensing. This section aims to provide a detailed understanding of how carbon dots function as fluorescent nanomaterials and their versatility in biosensing applications.

The second half of the literature review transitions to the role of machine learning, particularly its algorithms and applications in the design and development of fluorescent sensor arrays. Since machine learning plays a pivotal role in this thesis, this section outlines relevant concepts and highlights how computational techniques can be leveraged to enhance sensor performance and accuracy in complex sensing environments.

Chapters 3 and 4 present the core of the experimental work, focusing on the synthesis and development of novel fluorescent carbon dot sensors for metal ion detection. These chapters report on the comprehensive optimisation of carbon dot synthesis and the development of

sensor arrays, demonstrating how machine learning algorithms can be applied to optimise the sensing capabilities of carbon dots for metal ion detection.

In Chapter 5, the research extends into pH sensing, using a carbon dot-based sensor array to explore how different functional groups on CDs influence their pH responsiveness. This chapter proposes theoretical frameworks and experimental verification to enhance our understanding of pH sensing mechanisms in carbon dots.

Finally, Chapter 6 summarises the research findings and offers recommendations for future work, aiming to advance both the synthesis of carbon dots and the integration of machine learning for broader applications in sensor development.

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2 Carbon dots and machine learning for fluorescent sensing applications

2.1 Carbon dots-based sensor development and application

Carbon dots (CDs), a class of emerging nanomaterials, have garnered significant attention due to their exceptional properties and versatile applications in various scientific disciplines.¹⁻³ CDs are typically defined as carbon-based nanoparticles with sizes around 10 nm, encompassing a broad spectrum of morphologies and compositions that include amorphous carbon, graphitic carbon, and nitrogen-doped carbon structures.⁴ The unique characteristics of CDs stem from their size-dependent optical properties, high photostability, low cytotoxicity, and excellent biocompatibility, which collectively position them as promising candidates for a multitude of technological advancements.⁵⁻⁷

The synthesis of CDs can be achieved through multiple methods, broadly categorized into top-down and bottom-up approaches.^{8, 9} Top-down techniques involve the fragmentation of larger carbonaceous precursors such as graphite or polymers,¹⁰ whereas bottom-up methods synthesize CDs from small organic molecules like citric acid and urea.¹¹ These strategies offer a wide range of tuneable properties, enabling researchers to tailor CDs for specific applications by controlling factors such as size, surface chemistry, and optical properties.⁷ One of the most distinctive features of CDs is their size-dependent fluorescence emission, which is governed by quantum confinement effects and surface states.^{12, 13} This property allows CDs to exhibit excitation-dependent emission, where the colour of the emitted light can be adjusted by altering the excitation wavelength. Additionally, CDs can be functionalised with various groups to enhance their solubility, stability, and reactivity, making them suitable for diverse environments and interactions with biological systems.¹⁴

The potential of CDs as fluorescent probes has been extensively explored, particularly in the realm of sensors. Their ability to interact selectively with analytes, coupled with their robust luminescent response, has facilitated the development of highly sensitive and selective detection platforms for various targets, including heavy metals, biomolecules, and environmental pollutants.¹⁵ Moreover, CDs' compatibility with aqueous media and low toxicity have paved the way for their integration into biomedical applications, such as bioimaging, drug delivery, and disease diagnostics.¹⁶ Despite the remarkable progress in CD research, several challenges remain to be addressed for their full-scale implementation. These include the

optimisation of synthesis protocols for consistent quality and scalability, the enhancement of long-term stability and reproducibility, and the elucidation of the precise mechanisms underlying their optical behaviour¹⁷. Furthermore, the exploration of novel functionalities and the development of multifunctional CD-based materials are anticipated to broaden the scope of their applications in advanced sensing technologies and beyond.¹⁸

In the following sections, the first part of this chapter delves deeper into the synthesis methods, fluorescence properties, sensing applications, and future perspective of CDs, focusing on their role as key components in the design of advanced fluorescent sensors. This chapter highlights recent advancements and discusses the current state of the art in this dynamic field, while also identifying the gaps and opportunities for future research endeavours.

2.1.1 Carbon dots synthesis

The synthesis of carbon dots encompasses a broad spectrum of methodologies, each tailored to achieve specific optical and functional properties desired for various applications. Broadly categorised into top-down and bottom-up approaches, the top-down approach involved breaking down larger carbon-based materials, such as graphite or carbon nanotubes, into smaller carbon dots.^{8, 19} In contrast, the bottom-up approach entailed the assembly of carbon dots from smaller organic precursors, including citric acid, glucose, or biomass-derived materials.¹¹ These methods allow for the customisation of CDs' size, surface chemistry, and optical properties, making them versatile for use in advanced sensing technologies.^{7, 20}

2.1.1.1 Hydrothermal synthesis

Hydrothermal synthesis, a popular bottom-up technique, stands out as one of the most widely adopted methods for fabricating high-quality CDs.^{21, 22} Hydrothermal synthesis involves the heating of precursors bathed with solvent in a sealed vessel at elevated temperatures and pressures, facilitating the formation of CDs through a series of complex reactions. This method is favoured for its simplicity, and cost-effectiveness. By changing the precursor selection and synthetic parameters, hydrothermal synthesis enables the production of CDs with tuneable optical properties and high dispersibility, attributes that are crucial for their performance as fluorescent sensors.^{23, 24} **Table 2.1** summarises CDs reported in the literature that have been synthesised via hydrothermal reaction.

Table 2.1 Synthetic parameters and fluorescent properties of carbon dots synthesised via hydrothermal reaction

N O.	Ref	Synthetic conditions	Precursors	Solvent	Excitation wavelength(s) / nm	Emission wavelength(s) / nm
1	25	6-24 h at 200 °C	cassava pulp (4 g), ethylenediamine (0.4 g for NCDs)	deionised water (200 mL)	360	443
2	26	8 h at 180 °C	L-ascorbic acid (0.50 g), L-arginine (0.75 g)	deionised water (50 mL)	320 (CD), 410 (fluorophores)	430 (CD), 580 (rhodamine)
3	27	4 h at 180 °C	CQD W: whole-meal bread (5 g) CQD WS: whole-meal bread (1 g) + soybean flour (4 g) CQD WSL: whole-meal bread (1 g) + soybean flour (4 g) + lemon juice (30 mL)	Milli-Q water or lemon juice (30 mL)	423	400–500
4	28	24 h at 200 °C	citric acid (0.1 g) and porphyrin diacid (0.04 g)	deionised water (10 mL)	300	406
5	29	10 h at 200 °C	o-aminophenol (0.4368 g), ethylenediamine (20 mL)	ultrapure water (30 mL)	430	510
6	30	12 h at 200 °C	ammonium molybdate (0.61 g), Ascorbic acid (0.70 g)	ultrapure water (20 mL)	371	469
7	31	8 h at 180 °C	fresh crocus cancellatus (1 g)	ultrapure water (40 mL)	330	415
8	32	10 h at 220 °C	Peric./pium citri reticulatae powder (1.5 g), Urea (0.6 g)	deionised water (50 mL)	332	407
9	33	1-20 h at 100/150/200/250 °C	L-Cysteine or D-Cysteine (0.1g)	water (10 mL)	350	430
10	34	12 h at 180 °C	p-Phenylenediamine (0.11 g), Ethylenediamine (535 µL)	anhydrous ethanol (3 mL)	488	525
11	30	4 h at 180 °C	Cellobiose (0.2 g), NH ₄ HCO ₃ (1.5 g)	deionised water (10 mL)	230 , 320	420 (single emission peak at both excitation wavelengths)
12	7	8 h at 180 °C	L-Ascorbic acid (1 g), L-arginine (1.5 g)	deionised water (25 mL)	350 (single-photon), 750 (two-photon)	425 (CD), 525 (fluorescein amine)
13	35	6 h at 180 °C	Trimethylolpropane tris[poly(propylene glycol), amine terminated] ether (PEA) and formaldehyde	Water	360	440
14	36	10 h at 200 °C	guava leaves (1 g)	deionised water (60 mL)	370	450
15	37	10 h at 180 °C	red pitaya (5 g)	ultrapure water (10 mL)	350	450
16	38	8 h at 180 °C	ascorbic acid (1 g), arginine (0.5/1/1.5/2 g)	water/ ethanol/ acetonitrile (60 mL)	348-368	442-444
17	39	1-8 h at 180-190 °C	ascorbic acid (2 g)	deionised water (20 mL)	267–280	433
18	40	12 h at 180 °C	acri flavine (10 mg)	ultrapure water (10 mL)	410	505
19	41	10 h at 180 °C	CuCl ₂ ·2H ₂ O (0.1 g), ethanediamine (1 mL)	ultrapure water (29 mL)	320	380
20	42	8 h at 220 °C	phthalocyanine (15 mg)	deionised water (80 mL) and	W-CDs (310), E-CDs (340)	W-CDs (455), E-CDs (507)

N O.	Ref	Synthetic conditions	Precursors	Solvent	Excitation wavelength(s) / nm	Emission wavelength(s) / nm
				anhydrous ethanol (80 mL)		
21	43	12 h at 180 °C	p-Phenylenediamine (PPD) and aspartic acid (ASP)	deionised water	410	535
22	44	2 h at 200 °C	L-tryptophan (0.204 g), L-phenylalanine (0.165 g)	hydrochloric acid solution (0.1 M, 10 mL)	320–380	435
23	45	12 h at 200 °C	candle soot (25 mg)	sodium hydroxide (NaOH) aqueous solution (40 mL of 25 M NaOH)	310	450

2.1.1.2 Microwave-assisted synthesis

Microwave-assisted synthesis, on the other hand, offers a faster and energy-efficient alternative, achieving similar results in minutes rather than hours, making it an attractive choice for industrial scale-up, compared with conventional hydrothermal methods.⁴⁶ By utilising microwaves to induce localised heating, this technique accelerates the reaction kinetics, resulting in shorter synthesis times and potentially higher yields. Microwave-assisted synthesis also allows for better control over the size and morphology of CDs, which can be critical for optimising their sensitivity and selectivity in sensing applications.⁴⁷ Literature examples of CDs synthesised through microwave-assisted procedure are listed in **Table 2.2**.

Table 2.2 Synthetic parameters and fluorescent properties of carbon dots synthesised via microwave assisted synthesis procedure

NO.	Ref	Synthetic Conditions	Precursors	Solvent	Excitation wavelength(s) / nm	Emission wavelength(s) / nm
1	48	2450 MHz for 15 min	urea (2 g), glucose (1 g)	double-distilled water (20 mL)	340	435
2	49	450 W for 5 min	glucose (0.1 g)	DES (7 mL)	320–390 (depending on the solvent used)	380–478 (depending on the solvent used)
3	50	650 W for 6 min	citric acid (6.3 g), tris base (3.6 g)	distilled water (50 mL)	380	440
4	51	420 W for 10 min	psyllium husk (1 g)	acetic acid (30 mL) and deionised water (10 mL)	350	450
5	52	500 W for 20 min	bilimbi fruit extract (30 mL)	distilled water (30 mL)	430	531
6	53	700 W for 3 min	citric acid (0.5 mmol) and urea (0.5 mmol)	water (10 mL)	360 (in water) and 370 (in organic solvents)	450 (in water) and 460 (in organic solvents)
7	54	700 W for 30 min	sewage sludge (2.0 g)	water (50 mL)	322	412
8	55	550 W for 5 min	citric acid (1 g), glutathione or thiourea (1 g)	deionised water (10 mL)	358	436 (N,S-CDs with thiourea), 422 (N,S-CDs with glutathione)
9	56	650 W for 3 min			356	445 (fluorescence),

			triethanolamine (3 mL), phosphoric acid (9 mL)	ultrapure water (8 mL)		518 (phosphorescence)
10	57	200°C for 1.5 h	kelp (5 g), ethylenediamine (0.4 mL)	deionised water (30 mL)	370	450
11	58	600 W for 5 min	citric acid monohydrate (210 mg) and thiourea (210 mg)	water (10 mL)	360	450
12	59	180°C for 5 min	glutathione (0.1 M)	formamide (20 mL)	405 (blue fluorescence) and 600 (red fluorescence)	650 and 680 (red fluorescence) with blue fluorescence component
13	60	200 W, 200°C for 10 min	xylan (0.5 g), ammonium hydroxide (100 µL, 25–28 wt%)	purified water (10 mL)	360	442
14	61	180 W for 90 min	urea (2.5 g), polyethylene glycol (PEG, 2.5 g)	Milli-Q water (50 mL)	320	380
15	62	180°C for 3 min	chitin nanofibers (3 g)	saturated CaCl ₂ in ethanol (20 mL)	370	480
16	63	800 W for 10–15 min	sesame seeds (1 g)	water	365	440
17	64	750 W for 2 min and 30 s for carbon dots, followed by 225 W for 25 min for Zn(OH) ₂ composites	citric acid (5 g), disodium hydrogen phosphate (1 g)	ultrapure water (10 mL)	260	295, 390
18	65	750 W for 2 min and 30 s	citric acid (5 g), dipotassium hydrogen phosphate (K ₂ HPO ₄) (1 g)	ultrapure water (10 mL)	355	440 (for CDs), 450 (for CDs@MIPs)
19	66	200 W for various durations (few min)	sucrose (1.0 g)	phosphoric acid solution (20 mL, concentration varied)	380–490	450–550
20	67	700 W for 5 min	glucose (2.5 g), tryptophan (300 mg)	water (15 mL)	370	457

2.1.1.3 Electrochemical synthesis

Electrochemical synthesis, another bottom-up method, utilises electrical current to drive the conversion of precursors into CDs through the electrochemical carbonisation.⁶⁸ For instance, Deng *et al.* developed a one-pot electrochemical method to synthesise high-quality fluorescent carbon dots from alcohols, where the size of the CDs could be controlled by varying the applied potential.⁶⁹ They found that the diameters of CDs synthesised at different voltages ranged from 2.1 to 4.3 nm, with the QY reaching 15.9%. This method highlights the efficiency of using alcohols as carbon sources, simplifying the process and eliminating the need for complex purification and passivation steps. Similarly, Niu *et al.* proposed a bottom-up electrochemical process using nitriles and ionic liquids to prepare solid-state carbon dots.⁷⁰ In their study, carbon-free electrodes were used, and the positive charges of the ionic liquid facilitated the precipitation of CDs on the cathode. The resulting CDs exhibited a size of around 3 nm, and

they demonstrated excellent photoluminescence and dispersibility in water, making them suitable for bioimaging and metal ion detection.

In another approach, Zhou *et al.* synthesised CDs from multi-walled carbon nanotubes (MWCNTs) via electrochemical ablation.⁷¹ The MWCNTs served as the working electrode, and the cleavage of these large carbon structures resulted in the formation of CDs. The use of acetonitrile and tetrabutylammonium perchlorate as the electrolyte contributed to the efficient production of CDs with a QY of 6.4%. Also, electrochemical approach was employed to synthesize CDs from o-phenylenediamine (OPD).⁷² This method allowed the production of CDs with a large Stokes shift (309 nm), ideal for sensing applications such as the detection of Fe(III) and ascorbic acid. The CDs exhibited strong emission at 570 nm (with excitation at 261 nm) and offered high selectivity and sensitivity in various sensing applications. Electrochemical carbonisation offers several advantages, such as lower costs, tuneable CD sizes, and the ability to incorporate functional groups through the choice of electrolytes.

2.1.1.4 Laser ablation

Laser ablation is a top-down method for the synthesis of CDs. This involves using a pulsed laser to irradiate a carbon target, which leads to the formation of carbon dots. Nguyen *et al.* explored a double-pulse femtosecond laser ablation technique, which was used to generate ultrasmall CDs with an average size of around 1 nm.⁷³ The method employed graphite powder dispersed in ethanol and focused laser beams to ablate the carbon materials, creating smaller particles than those typically produced by single-pulse ablation. The second laser pulse, timed precisely, suppressed the rarefaction wave caused by the first pulse, reducing particle size and enhancing surface functionalisation. Another example of laser ablation-based CDs synthesis was given by Xu *et al.*⁷⁴ They synthesised nitrogen-doped CDs using a femtosecond laser ablation method, wherein graphite powder was ablated in aminotoluene solution at room temperature. The resulting CDs had an average diameter of 2.87 nm and exhibited excitation-independent photoluminescence across the visible spectrum. This property was attributed to surface defects involving C-O and C-N functional groups. The CDs emitted at a peak of 543 nm under 380 nm excitation, with a full width at half maximum (FWHM) of 172 nm. Their photoluminescence was stable across various excitation wavelengths, achieving a quantum yield of 6.5% in aqueous solution. Additionally, the laser power during synthesis was found to influence both fluorescence intensity and emission wavelength. These CDs were subsequently applied in a ratiometric pH sensing platform, leveraging their distinct pH-dependent

luminescence, driven by different functional groups, to enable effective pH sensing. This approach presents a straightforward and efficient method for producing CDs with desirable optical properties for applications in environmental and biological sensing. Laser ablation synthesis resulted in CDs with abundant functional groups, particularly useful for catalytic and sensing applications. While the laser-assisted method can be used to successfully synthesise carbon dots, it has some limitations in terms of size control, uniformity, and complexity of the process.

Each synthesis method offers unique advantages and limitations, with the choice of method determined by the specific application requirements. For example, hydrothermal synthesis is valued for its simplicity and the ability to produce CDs with tuneable fluorescence, while microwave-assisted synthesis is appreciated for its speed and energy efficiency. The range of synthesis approaches, from bottom-up to top-down, provides researchers with flexibility in customising the optical and functional properties of CDs. Following the synthesis of these CDs, their fluorescence properties must be rigorously analysed. The next section will explore the underlying fluorescence mechanisms in detail.

2.1.2 Fluorescence properties of carbon dots

2.1.2.1 Mechanistic insights into fluorescence in carbon dots

The intricate mechanisms underlying the fluorescence of carbon dots are pivotal to their success as sensitive and selective sensors. This section delves into the foundational principles and recent discoveries that elucidate how CDs exhibit their distinctive luminescence properties. Understanding these mechanisms is crucial for the rational design and optimisation of CD-based fluorescent sensors.

Quantum confinement effect

At the core of CDs' optical behaviour lies the quantum confinement effect, a phenomenon that governs the size-dependent fluorescence emission.¹³ CDs, due to their nanoscale size (typically less than 10 nm), exhibit this effect when the particle size becomes smaller than the exciton Bohr radius, leading to discrete energy levels instead of continuous energy bands.^{75, 76} This explains why smaller CDs emit blue light, while larger ones tend to shift towards the red region of the spectrum.⁷ The quantum confinement effect has been widely studied across various semiconductor materials, including graphene quantum dots (GQDs), which represent a novel zero-dimensional quantum-confined system.²³ Studies have demonstrated the size-dependent

absorption and fluorescence spectra of GQDs, where the red-shifted emission is attributed to the increased particle size resulting from the quantum confinement effect. Similar behaviour has been observed in carbon dots with different particle size distributions but similar chemical compositions, suggesting that the quantum confinement effect plays a role in altering their fluorescence emissions. Further evidence points to the persistence of the size-dependent fluorescence in carbon dots even after removing surface oxygen, reinforcing the idea that quantum confinement is a significant contributor to their optical properties.⁷⁷ However, in CDs, quantum confinement alone does not fully explain the observed fluorescent properties, as their fluorescence is influenced by multiple factors, including surface chemistry and functional groups.

Surface defects and functional groups

Surface states, arising from defects and functional groups on the CD surface, significantly influence their optical properties.¹² These surface states can act as trap sites for excited electrons and holes, affecting the emission spectra and the fluorescence lifetime.⁷⁸ The surface of a CD is typically rich in functional groups such as hydroxyl (-OH), carboxyl (-COOH), amine (-NH₂), and others, depending on the synthesis method and precursors used. These functional groups introduce surface states or defect states, which are crucial for fluorescence in CDs.¹² These defects act as trap states for excited electrons, which can transition to lower energy levels and emit light.⁷⁹ Surface defects create localised energy states within the bandgap, leading to multiple emission centres. As a result, CDs often exhibit broad and tuneable emission spectra. The fluorescence in CDs often results from energy transfer between the core (which may exhibit quantum confinement) and the surface defect states. This interplay between the core and surface energy levels gives CDs their characteristic emission properties.

In conclusion, the mechanisms governing the fluorescence of carbon dots are multifaceted and complex, encompassing quantum confinement, surface states, doping, and functionalisation. A deep understanding of these mechanisms is essential for the design and optimisation of CD-based fluorescent sensors, which hold great promise for addressing complex analytical challenges in various fields, from environmental monitoring to biomedicine and beyond.

2.1.2.2 Fluorescence quenching and enhancement mechanisms in carbon dots

The dynamic interplay between quenching and enhancement mechanisms in carbon dots is a fundamental aspect of their fluorescence behaviour, significantly impacting the performance

of CD-based fluorescent sensors. Understanding these mechanisms is crucial for optimising sensor design and achieving high sensitivity and selectivity in various applications. This section delves into the intricacies of fluorescence quenching and enhancement, exploring the underlying principles and their implications for sensor development.

Fluorescence quenching mechanisms

Fluorescence quenching in CDs can occur through several mechanisms, each with distinct implications for sensor design.⁸⁰ Static quenching, a common phenomenon, arises from the formation of a ground-state complex between the fluorophore and the quencher, leading to a decrease in fluorescence intensity.⁸¹ This process is reversible and can be exploited for the detection of analytes that bind to CDs (forming non-fluorescent complex), causing a measurable decrease in fluorescence.⁸² For instance, CDs functionalised with chelating agents can selectively bind to metal ions, resulting in static quenching that is directly proportional to the ion concentration.⁸³ Dynamic quenching, on the other hand, involves a collisional process between the excited-state fluorophore and the quencher molecule, leading to a non-radiative deactivation of the excited state.⁸² This mechanism is concentration-dependent and can be influenced by factors such as temperature, viscosity, and solvent polarity. Dynamic quenching is often utilised in sensors where changes in the environment, such as the presence of a target analyte, can alter the quencher concentration or the physical properties of the medium, resulting in a detectable change in fluorescence intensity.⁸⁴ Another mechanism contributing to fluorescence quenching is photoinduced electron transfer (PET).⁸⁰ In PET quenching, the excited electron of the fluorophore transfers to a nearby acceptor molecule, leading to a decrease in fluorescence. This process can be sensitive to the spatial proximity and relative orientation between the fluorophore and the acceptor, making it a valuable mechanism for the development of sensors that rely on molecular interactions or dynamic changes in the local environment of the sensing system.

Fluorescence enhancement mechanisms

In contrast to quenching, fluorescence enhancement mechanisms in carbon dots (CDs) can result in increased fluorescence intensity, which is equally important for sensor design. One such mechanism is Förster resonance energy transfer (FRET), a non-radiative process in which energy is transferred from an excited donor to an acceptor molecule through dipole–dipole interactions, provided the donor and acceptor are in close proximity (typically 10–100 Å).^{80, 85}

FRET is a distance-dependent process where the donor loses fluorescence as it transfers energy to the acceptor, which in turn gains fluorescence. During this process, a photon excites the donor system, and the resulting energy is transferred to the acceptor molecule, promoting an electron in the acceptor to various higher vibrational levels of its singlet excited state. For efficient FRET, the acceptor's absorption spectrum must overlap with the donor's fluorescence emission spectrum. FRET systems involving CDs are often constructed by pairing CDs with other fluorescent molecules, typically requiring two systems with distinct emission wavelengths. In such systems, the donor CDs transfer energy to an acceptor molecule, enabling enhanced fluorescence for sensor applications.

Plasmonic coupling is another enhancement mechanism that exploits the interaction between CDs and metallic nanoparticles, such as gold or silver.⁸⁶ The localised surface plasmon resonance (LSPR) of the metal nanoparticles can enhance the electric field near the surface, leading to an amplification of the fluorescence signal from nearby CDs.⁸⁷ This effect can be particularly useful in sensors designed to detect low concentrations of analytes, as it provides a significant boost in sensitivity.

Other interaction mechanisms

Beyond energy and charge transfer, other mechanisms influence the interaction between carbon dots (CDs) and analytes. In some cases, the analyte induces conformational changes in the CDs or their functional groups, leading to alterations in fluorescence. These mechanisms are particularly important in sensors designed to detect biomolecules, where analyte-induced structural changes can be directly correlated with the concentration of the target molecule. Interactions such as hydrogen bonding, van der Waals forces, and π - π stacking also contribute to the sensitivity and selectivity of CD-based sensors.⁸⁸ Though weaker, these interactions can stabilise the binding of analytes to the CDs, thereby influencing the fluorescence response in a way that can be leveraged for analyte detection. For instance, π - π stacking interactions between aromatic analytes and CDs may result in either fluorescence quenching or enhancement, depending on the analyte's electronic structure.⁸⁹

In conclusion, the mechanisms of fluorescence quenching and enhancement in carbon dots are integral to the performance of CD-based fluorescent sensors. By understanding and leveraging these mechanisms, researchers can design sensors with improved sensitivity, selectivity, and stability, paving the way for their application in a wide range of fields, from environmental

monitoring to biomedicine and pharmaceutical analysis. As the field advances, the development of more sophisticated sensors will undoubtedly benefit from a deeper insight into the complexities of fluorescence modulation in CDs.

2.1.2.3 Role of surface passivation and functional groups

Surface passivation and functionalisation are crucial strategies for tailoring the optical properties of carbon dots, particularly in enhancing their fluorescence efficiency and stability.⁹⁰ Surface passivation primarily reduces non-radiative decay pathways, which negatively affect the quantum yield (QY) of CDs. Non-radiative processes, such as internal conversion and multiphoton relaxation, can dissipate excited-state energy without photon emission, reducing overall fluorescence efficiency.

Passivating agents, including polyethylene glycol (PEG), polyvinylpyrrolidone (PVP), and amino acids, can coat the surface of CDs, forming a protective barrier that minimises interaction between surface defects and the surrounding environment.^{91, 92} This barrier decreases the likelihood of non-radiative transitions, thereby increasing the QY of CDs. Additionally, surface passivation enhances photostability by protecting CDs from photo-oxidation and degradation caused by prolonged light exposure.⁹³

Functional groups attached to the CD surface not only contribute to passivation but also play a vital role in determining the solubility, reactivity, and selectivity of CDs. The nature of the functional group dictates whether CDs are hydrophilic or hydrophobic, influencing their dispersion in solvents and interactions with analytes.⁸⁸ For example, CDs functionalised with carboxyl groups exhibit high solubility in water, making them ideal for applications in aqueous environments, whereas those with alkyl chains are better suited to organic solvents.⁹⁴

Functional groups also serve as recognition elements, enabling the development of selective sensors. For instance, CDs functionalised with ethylenediaminetetraacetic acid (EDTA) selectively bind to metal ions, resulting in fluorescence changes proportional to the ion concentration.⁹⁵ Similarly, CDs conjugated with aptamers—short single-stranded DNA or RNA molecules—can recognise specific biomolecules, facilitating the creation of highly specific biosensors.^{96, 97}

Moreover, functional groups influence the photophysical properties of CDs. Certain groups act as energy acceptors or donors, leading to FRET, which can result in fluorescence quenching or

enhancement depending on the proximity and orientation of the donor and acceptor molecules. FRET-based sensors leverage this distance-dependent quenching or fluorescence recovery to detect changes in the microenvironment or analyte binding, offering a sensitive detection method.

In addition to improving optical properties and selectivity, functional groups can provide additional functionalities, such as targeting specific cells for applications in imaging and drug delivery. CDs functionalised with peptides or antibodies can selectively target cancer cells, making them useful for both imaging and therapeutic applications. Furthermore, certain functional groups exhibit pH-responsive behaviour, which can be harnessed for controlled drug release or pH detection in biological systems.

The strategic choice of surface passivation and functionalisation is essential for designing CD-based sensors that operate under specific conditions and respond to particular analytes. Optimising these surface properties can lead to sensors with enhanced sensitivity, selectivity, and stability, which are critical for applications in environmental monitoring, food safety, biomedicine, and pharmaceutical analysis.

As the field of CD-based fluorescent sensors evolves, the exploration of novel functional groups and passivation strategies remains a dynamic area of research. Developing multifunctional CDs with enhanced optical properties and reactivity holds significant promise for tackling complex analytical challenges and expanding their application across various scientific disciplines.

2.1.2.4 Tuning of carbon dots' fluorescence characteristics

The tunability of CDs' fluorescence is further enhanced by the effects of doping and surface functionalisation. Doping, the intentional introduction of heteroatoms such as nitrogen, sulfur, and boron into the CD structure, significantly impacts the optical properties.⁹⁸ Doped CDs have been shown to possess higher quantum yields and improved photostability compared to their undoped counterparts, leading to changes in their absorption and emission spectra.⁹⁹ Surface functionalisation, on the other hand, allows for the modification of CDs with various functional groups, enhancing their solubility, stability, and reactivity.¹⁰⁰ These functional groups can also serve as recognition probes for specific analytes, facilitating the development of highly sensitive and selective sensors.

Doping effects on optical properties

Doping, the intentional introduction of heteroatoms such as nitrogen, sulfur, and boron into the carbon dot structure, significantly enhances the optical properties of CDs.⁹⁸ For example, N,S co-doped lignin-derived CDs, as reported by Hou *et al.*, showed exceptional selectivity and sensitivity in detecting metal ions such as Fe(III), Ag(I), Cu(II), and Al(III) using fluorometry.¹⁰¹ This enhanced sensitivity results from the unique interactions between the dopant atoms and the metal ions, which affect fluorescence intensity or lifetime, correlating directly with analyte concentration.

Surface functionalities and their impact

Surface functionalities are another important factor in determining the optical properties of CDs. Functional groups such as hydroxyl (-OH), carboxyl (-COOH), and amine (-NH₂) influence not only the solubility and stability of CDs but also their fluorescence. Hou *et al.* demonstrated that lignin-derived CDs with specific surface groups (-OCH₃, -OH, -COOH) display distinct multicolour emissions, highlighting the significant role of surface chemistry in tuning emission colours.¹⁰¹ Such tuneable emissions are particularly beneficial in ratiometric sensors, where the ratio of two emission peaks allows for precise quantification of analyte concentrations.

In summary, the tuneable fluorescence properties of CDs are governed by a complex interplay of size, surface functionalities, and doping effects. These factors can be strategically manipulated to engineer CDs with desired optical properties for diverse sensing applications. Understanding and controlling these parameters is crucial for developing CDs with optimised performance in specific sensing contexts. Tailoring CDs for high sensitivity and selectivity in response to target analytes necessitates careful consideration of their size, surface chemistry, and doping levels. Future research should focus on the systematic investigation of these parameters to unlock the full potential of CDs as advanced fluorescent sensors. As the field progresses, the ability to precisely control these factors will be essential for developing more sophisticated and reliable sensors that can address complex analytical challenges in environmental monitoring, food safety, biomedicine, and pharmaceutical analysis.

2.1.3 Applications of carbon dot-based fluorescent sensors

2.1.3.1 Biomedical applications

Carbon dots have revolutionised biomedical applications, serving as versatile tools for biosensing, drug delivery, imaging, and theragnostic. Their biocompatibility, low cytotoxicity, and tuneable optical properties make them ideal for interfacing with biological systems, offering unprecedented opportunities in real-time monitoring of biological processes.

Biosensing

CDs have proven to be effective probes for the sensitive and selective detection of biomolecules, including nucleic acids, proteins, and enzymes. The ability of CDs to undergo significant changes in fluorescence upon interaction with specific biomolecules enables the development of biosensors with high sensitivity and selectivity. In photoluminescent (PL) biosensors, CDs can be doped with metal impurities to enhance their optical and sensing capabilities.¹⁰² For instance, cobalt(II)-CD composites with 1-(2-pyridylazo)-2-naphthol (PAN) and cobalt chloride precursors demonstrated high selectivity and sensitivity to Cr(VI) ions, leveraging surface group interactions to weaken surface-trap emissions. CDs have also been employed to detect various ions, where target ions quench fluorescence through electron or energy transfer mechanisms. For example, CDs conjugated with organic dyes function as ratiometric sensors for H₂S in biological fluids and living cells.¹⁰³ Furthermore, CDs modified with amine moieties exhibited stable photoluminescence and colloidal properties, and their interaction with DNA influenced both photoluminescence and absorption due to changes in DNA electrophoretic mobility.¹⁰⁴

In chemiluminescent (CL) biosensors, carbon dots have been widely applied due to their biocompatibility and non-toxic properties. However, their broader use is often limited by challenges such as complex operational procedures, and unstable CL signals. To overcome these issues, CDs have been utilised as signal amplification labels. For example, in a sandwich-structure biosensor, a carbon dots dotted nanoporous gold nanoparticle, labeled signal DNA was captured on the DNA biosensor, enhancing the overall detection sensitivity.¹⁰⁵ Additionally, CDs have been employed in CL systems utilising transition metal ion-catalysed decomposition of hydrogen peroxide (H₂O₂), a Fenton-like reaction, for detecting Fe(II) with minimal interference from Fe(III) and other cations. This strategy improves the selectivity and

sensitivity of the biosensor, making it more effective for detecting metal ions in complex environments.¹⁰⁶

In electrochemical (EL) biosensors, CDs serve as immobilisation platforms for biomolecules such as aptamers, enzymes, and antibodies on electrode surfaces. For example, CDs embedded in bimetallic ZrHf-MOFs exhibited strong aptamer anchoring for early diagnosis of HER2-overexpressed cancer cells.¹⁰⁷ CDs have also been used in combination with other materials, such as graphene oxide and multiwalled carbon nanotubes, to develop ultrasensitive detection systems.¹⁰⁸ Additionally, CDs have been integrated with nanomaterials like gold nanoparticles and Pd-Au nanocomposites to enhance the performance of biosensors, such as those designed for DNA and glucose detection.¹⁰⁹

Drug delivery

Carbon dots have garnered attention as promising carriers for drug delivery, owing to their small size, biocompatibility, and ability to encapsulate therapeutic agents.¹¹⁰ Their surface can be easily functionalised, enabling targeted delivery to specific cells or tissues, thus improving treatment efficacy and reducing side effects. For instance, CDs conjugated with cancer-specific antibodies have been successfully used for targeted delivery of chemotherapy drugs to tumour cells, offering enhanced therapeutic outcomes. Additionally, CDs' inherent fluorescence enables real-time tracking of drug delivery, providing a dual therapeutic and diagnostic (theragnostic) function.

Several studies illustrate the versatility of CDs in drug delivery. Hollow luminescent carbon dots (HCDs), for example, have been used to load doxorubicin (DOX) via π - π stacking and hydrophobic interactions.¹¹¹ This system exhibits pH-controlled drug release, with minimal release at physiological pH and significant release under acidic conditions, mimicking the intracellular environment. Such controlled release enhances the drug's efficacy while minimising its impact on healthy cells. Furthermore, CDs have demonstrated potential in crossing the blood-brain barrier (BBB), making them a promising vehicle for delivering drugs to the central nervous system (CNS), as shown in studies utilising transferrin-functionalised CDs.¹¹²

In addition to their ability to deliver chemotherapy agents, DNA-conjugated CDs (DNA-CDs) have shown promise in drug delivery applications by forming stable complexes with drugs like rhodamine 6G and doxorubicin.¹¹³ These complexes are stable under neutral pH but release the

drugs in more acidic conditions, allowing for controlled release in tumour environments. Overall, CDs offer several advantages, including high drug-loading capacity, long-term stability, and controlled drug release. Their multifunctional capabilities position CDs as a versatile platform for addressing complex therapeutic challenges, particularly in cancer treatment and neurological applications.

Imaging

In the realm of cellular and molecular imaging, CDs have demonstrated remarkable potential.¹¹⁴ Their biocompatibility and low cytotoxicity permit their safe use in live cell imaging, enabling researchers to study cellular dynamics in real time. The tuneable emission properties of CDs can be exploited to visualise multiple cellular components simultaneously, offering a comprehensive view of cellular activities. This capability is crucial for understanding complex biological systems and for the development of targeted therapeutics. Many types of CDs exhibit blue or green fluorescence when excited by UV light, making them well-suited for cellular imaging under fluorescence microscopes.¹¹⁵ Additionally, certain specialised CDs can be excited by long-wavelength light or emit up-conversion photoluminescence (PL), making them suitable for in vivo imaging applications. The nanoscale structure of CDs aids their internalisation into cells, primarily through caveolae-mediated endocytosis, allowing for their incorporation into cytoplasmic regions, and in some cases, labelling of the cell nucleus. Polymer dots (PDs), a subclass of CDs, have also proven effective in imaging living cells and tissues, including MCF-7 cells, at depths of 60-120 μm .¹¹⁶

Traditional CDs, which absorb UV light and emit blue or green fluorescence, face challenges with light penetration in biological tissues. To overcome this, researchers are developing CDs with red or near-infrared emission properties to enhance tissue penetration and reduce photodamage.¹¹⁷ Additionally, CDs with up-conversion photoluminescence (UCPL), which utilise near-infrared photons for imaging, provide a valuable solution by avoiding tissue damage and autofluorescence interference typically associated with UV excitation.¹¹⁸ Multiphoton photoluminescence-emitting CDs have also been explored for cell imaging (such as two-photon), enabling effective in vitro and in vivo bioimaging applications.^{119, 120} CDs are also emerging as powerful contrast agents for photoacoustic (PA) imaging, a technique that combines light excitation with ultrasound detection to achieve deep tissue penetration with high spatial resolution.¹²¹ CDs with high absorption coefficients in the red or near-infrared range can generate detectable sound waves upon laser irradiation, making them effective PA imaging

agents. In addition, afterglow imaging with CDs offers another promising bioimaging modality. By eliminating autofluorescence interference, afterglow imaging allows for clearer visualisation of tissues.¹²² However, the preparation of liquid-phase phosphorescent CDs remains a challenge, which limits broader application of this technique. For example, afterglow imaging has been successfully demonstrated by injecting a CDs@silica complex into mice for subcutaneous imaging.¹²²

Real-time monitoring of biological processes

Carbon dots have proven to be highly effective tools for real-time monitoring of biological processes at both cellular and subcellular levels. Their application in live-cell imaging has allowed for the tracking of dynamic cellular events such as protein localisation, gene expression, and metabolic pathways. This real-time capability is essential for understanding the underlying mechanisms of diseases and for the development of targeted therapies. Several studies have demonstrated the unique abilities of different types of CDs in real-time monitoring. For instance, fluorine and nitrogen co-doped carbon dots (FN-CDs) have been used to track the cell division process in living cells.¹²³ These CDs possess a positively charged surface that binds to DNA and RNA, enabling the visualisation of nucleoli and chromosomes during various stages of cell division. Their long-wavelength excitation and anti-photobleaching properties make them suitable for real-time imaging of cell division from interphase to telophase. Similarly, phosphorus and nitrogen co-doped CDs (PN-CDs) have been utilised as fluorescent probes for real-time detection of reactive oxygen and nitrogen species (ROS and RNS) in macrophages.¹²⁴ These CDs, synthesised from adenosine-5'-triphosphate (ATP), exhibit high quantum yields and selectively detect ROS and RNS, such as ClO, ONOO, and NO. They provide a new platform for monitoring ROS/RNS signaling in live cells, allowing for the study of cellular responses to lipopolysaccharide (LPS)-induced oxidative stress in real-time. Moreover, gold carbon dots (GCDs) have been developed for the real-time monitoring of intracellular biothiol dynamics and ferric ion release during ferroptosis.¹²⁵ GCDs exhibit excellent photobleaching resistance and fast reaction kinetics, allowing for high-resolution, long-term tracking of biothiols like glutathione and cysteine. These GCDs enable precise monitoring of redox changes and ferric metabolism in cells, making them valuable tools for studying redox processes and ferroptosis.

In conclusion, carbon dots have become indispensable tools in the biomedical field, offering a multitude of applications ranging from biosensing and drug delivery to imaging and

theragnostic. Their unique properties and versatility continue to drive innovation in biomedical research, paving the way for new diagnostic and therapeutic strategies. As the field advances, the integration of CDs with other nanomaterials and advanced imaging techniques will likely lead to the development of even more sophisticated and effective biomedical applications, contributing to improved healthcare outcomes.

2.1.3.2 Environmental monitoring

Carbon dots have the potential to significantly enhance environmental monitoring by providing sensitive and selective detection of pollutants in water, air, and soil.^{126, 127} Their unique optical properties, combined with their high stability and green synthesis, make CDs ideal for developing fluorescent sensors that function under various environmental conditions. CDs have been applied as sensors for detecting heavy metal ions, organic pollutants, and other contaminants. For example, CDs have been used to create sensors for the detection of environmental pollutants such as 2,4,6-trinitrophenol (TNP), antibiotics like tetracycline hydrochloride, and pesticides including carbaryl and glyphosate.¹²⁸ Additionally, iron-doped CDs have demonstrated high sensitivity in detecting organophosphorus pesticides.¹²⁸

The advantages of CDs in environmental sensing extend beyond pollutant detection. Their large surface area and diverse surface functional groups allow CDs to act as effective adsorbents, removing pollutants from water.¹²⁹ Furthermore, CDs exhibit photocatalytic properties, enabling the degradation of organic pollutants such as dyes and chemicals under light irradiation.¹³⁰ Their ability to convert solar energy into thermal energy also makes them useful in solar-to-steam generation for water purification.¹³¹ Moreover, CDs have demonstrated antibacterial and antifungal activities, which can be leveraged for wastewater treatment.

Despite these promising applications, challenges remain, particularly in understanding the environmental impact of CDs themselves and improving their performance in environmental remediation. CDs have been incorporated into composite materials to enhance their pollutant degradation capabilities, but further research is needed to establish standardised synthesis protocols and assess their long-term effects. Overall, CDs present a versatile platform for environmental monitoring, pollutant removal, and remediation, offering an efficient and environmentally friendly solution to complex environmental challenges.

2.1.3.3 Food safety and quality control

Ensuring food safety and quality control is paramount in safeguarding public health and maintaining consumer confidence in the food industry. Carbon dot-based fluorescent sensors have emerged as powerful tools for the rapid and sensitive detection of food contaminants, including pesticides, pathogens, and toxins.¹³² Their unique optical properties, coupled with high sensitivity and selectivity, make CDs ideal for developing real-time monitoring systems that can screen for a wide range of harmful substances in food products.

Pesticide detection

The presence of pesticides in food can pose serious health risks, necessitating the development of sensitive and reliable detection methods.¹³³ CDs have been utilised to create sensors capable of detecting trace amounts of pesticides in food samples. For instance, CDs functionalised with specific receptors have been shown to selectively bind to organophosphate and carbamate pesticides, leading to a significant change in fluorescence intensity that can be correlated with pesticide concentration.¹³⁴ These sensors offer quick response times and can be integrated into portable devices for on-site food safety inspections.

Pathogen detection

Foodborne pathogens, such as *Salmonella*, *E. coli*, and *Listeria*, are major contributors to foodborne illnesses. CDs-based sensors have been developed for the detection of these pathogens in food matrices.¹³⁵ By functionalising CDs with aptamers or antibodies specific to the pathogen of interest, researchers have created sensors that can rapidly identify and quantify pathogens at low concentrations.¹³⁶ The use of CDs in these sensors ensures high sensitivity and selectivity, enabling early detection and prevention of foodborne outbreaks.

2.1.4 Future Perspectives

A key trend in CDs synthesis is the development of advanced techniques that allow for greater control over the optical properties of the CDs. Methods such as hydrothermal carbonisation, microwave-assisted synthesis, and laser ablation have been refined to produce CDs with tuneable emission wavelengths and improved photostability. Recent innovations include the use of machine learning algorithms to predict optimal synthesis conditions for achieving desired optical properties. By integrating computational models with experimental data,

researchers can now design CDs with specific emission spectra that are well-suited for specific sensing applications, such as the detection of environmental pollutants or biomarkers in complex matrices.

Surface functionalisation of CDs is another area that has seen significant progress. The strategic attachment of functional groups onto the surface of CDs can significantly enhance their sensitivity and selectivity towards target analytes. This is achieved by creating specific binding sites that facilitate interactions between the CDs and the analyte of interest. Advanced functionalisation strategies, such as the use of bioconjugation techniques, have allowed for the integration of biomolecules such as antibodies, aptamers, and enzymes onto the CD surface. These modifications not only improve the sensitivity of CDs-based sensors but also enable the creation of highly selective sensors capable of distinguishing between closely related molecules, a crucial feature for applications in biomedicine and environmental monitoring.

Environmental sustainability is becoming an increasingly important consideration in the synthesis of CDs. Traditional synthesis methods often involve the use of toxic chemicals and high-energy processes, which can have adverse environmental impacts. To address this, there is a growing trend towards green synthesis routes that employ eco-friendly precursors and mild reaction conditions. For example, the use of plant extracts, food waste, and biopolymers as precursors not only reduces the environmental footprint of CD synthesis but also opens up new possibilities for functionalisation and sensor design. These green synthesis routes align with the principles of green chemistry and contribute to the development of sustainable sensing technologies.

Machine learning algorithms are revolutionising the analysis and interpretation of data generated by CD-based sensors. These algorithms can process large datasets with high-dimensional complexity, identifying patterns and correlations that are not immediately apparent to conventional statistical methods. In the context of sensor development, machine learning can be used to optimise sensor design by predicting the most effective functionalisation strategies or identifying the ideal synthesis conditions for achieving desired optical properties. Moreover, in sensor operation, machine learning can enhance signal processing, enabling the differentiation of true signals from noise and improving the discrimination power of sensor arrays.

2.1.5 Conclusion

The development of CDs-based fluorescent sensors has been marked by remarkable progress, with innovations in synthesis, functionalisation, and integration leading to enhanced performance capabilities. As the field advances, addressing remaining challenges and capitalising on emerging trends will be pivotal in realising the full potential of CDs in the realm of sensing technologies. With their unique optical properties and functional versatility, CDs hold substantial promise for transformative applications across a broad spectrum of fields, spanning from environmental conservation to healthcare improvements and technological advancements. The continued innovation in CD synthesis, functionalisation, and integration with advanced technologies is expected to unlock even greater potential, transforming the way we approach challenges in these critical areas. As interdisciplinary research and collaboration advance, the future of carbon dots in applied sciences is poised to be marked by groundbreaking developments that enhance our quality of life and contribute to global sustainability. The future outlook for carbon dots-based fluorescent sensor research is one of boundless potential. As the technology matures, it will undoubtedly reshape the landscape of sensing technologies. The future of CDs-based sensor technology will be profoundly shaped by interdisciplinary collaboration. Convergence between material science, chemistry, biology, engineering, and data science will foster a fertile ground for innovation, enabling the creation of sensors that transcend traditional boundaries. This collaborative approach will not only accelerate technological advancements but also ensure that the benefits of CDs-based sensors are realised in a socially responsible and sustainable manner.

2.2 Machine learning based sensors array development and application

Fluorescent sensor arrays are an important technology that have a wide range of applications. They consist of an array of fluorescent sensors that can detect and differentiate various analytes, such as metal ions, amino acids, cations and anions, and saccharides.¹³⁷⁻¹⁴² These sensor arrays offer several advantages over traditional single-sensor approaches. They can provide more comprehensive and reliable detection, as the array can cross-react with multiple analytes and generate a unique "fingerprint" response.^{137, 138, 143} This allows for better discrimination and quantification of the target analytes. Fluorescent sensor arrays are particularly useful for the detection of metal ions, as they can differentiate between various metal ions in a sample.^{138, 143} This is important for applications such as environmental monitoring, water quality analysis, and medical diagnostics.¹³⁷ Additionally, fluorescent sensor arrays can be used for the detection of amino acids and saccharides, which are important biomolecules with various applications in fields like biochemistry, food science, and pharmaceutical development.^{142, 144} The rational design of these sensor arrays, including the selection of appropriate fluorescent sensors and the optimisation of the array composition, is crucial for their performance and versatility.^{141, 145} Researchers are continuously working to develop more advanced and efficient fluorescent sensor arrays to address a wide range of analytical challenges.^{137, 146}

There are a number of challenges in the process of developing and optimising a sensor array, specifically around identifying and quantifying similar variants of target analytes, designing the sensing elements, determining optimal parameters, and reducing the complexity of the array. These challenges can be addressed through machine learning (ML). Machine learning algorithms can be used to analyse the complex data from the sensor array, enabling more accurate identification and quantification of target analytes, even for similar variants.¹⁴⁷ ML models can be trained to optimise the design and parameters of the fluorescent sensor array, improving its sensitivity, selectivity, and reusability. Advanced data analysis techniques, such as pattern recognition and feature extraction, can be applied to the sensor array data to extract meaningful insights and improve the overall performance of the system. This chapter will introduce the fluorescent sensor array assisted by ML and provide hands on MATLAB based protocols for those seeking to apply ML models for their projects.

2.2.1 Fundamentals of fluorescent sensor arrays

Fluorescence is the emission of light by a substance that has absorbed light or other electromagnetic radiation.¹⁴⁸ This phenomenon occurs when electrons in the substance are excited to a higher energy level and then return to their ground state, releasing the excess energy as photons. Fluorescent molecules, termed fluorophores, provide a sensitive and selective method to detect and quantify various analytes, and are therefore key to construction of sensor arrays, as they can provide high-dimensional data from the colour or fluorescence changes in response to different analytes. Sensor arrays, also known as "electronic noses" or "optoelectronic noses," are composed of multiple sensing elements that can detect and respond to a wide range of chemical and biological species.¹⁴⁹

A sensor array is defined as a collection of multiple sensing elements, each designed to detect specific analytes or properties, integrated into a single platform. Fluorescent sensor arrays are constructed using a variety of advanced materials, each chosen for its unique properties and suitability for specific sensing applications. Organic dyes, including fluorophores and conjugated polymers, are commonly used for their high sensitivity and tuneable fluorescence.^{150, 151} Nanomaterials like quantum dots, carbon dots and metal nanoparticles offer exceptional brightness, photostability, and size-dependent emission properties, making them ideal for precise sensing.¹⁵² Additionally, metal-organic frameworks (MOFs) and supramolecular assemblies provide customisable frameworks that can enhance selectivity through host-guest interactions.¹⁵³

The application of fluorescent sensor arrays relies on several key principles: fluorescence-based detection, sensor array design, imaging techniques, and data analysis, along with the inherent versatility and sensitivity of fluorescent sensors. Fluorescent sensor elements exhibit changes in their emission properties—such as intensity, wavelength, or fluorescence lifetime—when they interact with target analytes, allowing for highly sensitive and selective detection. Sensor arrays are designed with multiple sensing elements, each containing distinct recognition components that interact with various analytes. This cross-reactive response generates unique "fingerprints" for each analyte, facilitating the detection of complex mixtures. Fluorescence-based sensor arrays can be integrated with imaging techniques, such as fibre bundles, to capture the entire array's fluorescence response simultaneously. The high-dimensional data produced can then be analysed using pattern recognition algorithms to identify and quantify target analytes. These fluorescent sensor arrays have demonstrated remarkable versatility and

sensitivity, capable of detecting a wide range of analytes, including metal ions, phosphates, and other chemical species. Their adaptability and precision make them valuable tools for diverse applications, including environmental monitoring, medical diagnostics, and chemical sensing.

The primary design goals for sensor arrays focus on minimising their number of sensor elements and difficulty of preparation, thereby enhancing practicality and efficiency. One key application of sensor arrays is in cross-reactive chemical sensing, where they detect a wide range of chemical species by utilising multiple sensing elements with varied recognition capabilities. Sensor arrays have also proven highly versatile in analysing complex samples, particularly in biological contexts. These systems offer the potential to overcome the limitations of single-selective sensors, demonstrating robustness and stability when tested in biological sample.^{154, 155} In the food industry, sensor arrays based on metalloporphyrin have been successfully employed to analyse red wine, a product known for its complex composition, showcasing the adaptability of these systems across diverse fields.¹⁵⁶ Achieving these requirements and optimising the sensor array design to its full potential necessitates substantial computational power. By leveraging programming and advanced machine learning models, we can significantly reduce labour costs and greatly enhance work efficiency.

2.2.2 Overview of machine learning algorithms

The performance and contribution of machine learning algorithms to the development of fluorescent sensor arrays are substantial and multifaceted. These algorithms improve the sensitivity, selectivity, and accuracy of detecting and quantifying various analytes. Machine learning-assisted sensor arrays have been applied across multiple fields, achieving significant success in areas such as antibiotic sensing, pesticide monitoring, metal ion detection, and biomolecule sensing.^{157, 158}

2.2.2.1 Supervised Learning Algorithms

Linear discriminant analysis (LDA): LDA is a widely used algorithm for analysing fluorescent sensor array data and identifying patterns or discriminating between different analytes or samples. It effectively extracts relevant features from high-dimensional fluorescent sensor data and builds classification models for analyte identification.¹⁵⁹

Support vector machines (SVMs): SVMs are powerful supervised learning algorithms that handle complex, nonlinear relationships in fluorescent sensor array data. They can be used for

both classification and regression tasks, making them versatile for various fluorescent sensor array applications.¹⁶⁰

Partial least squares discriminant analysis (PLS-DA): PLS-DA is a variant of PLS regression particularly useful for classification tasks with fluorescent sensor array data. It handles multicollinearity in the sensor data and builds robust predictive models for the quantitative and qualitative analysis of analytes.¹⁶¹

Random forests: Random forests are ensemble learning methods combining multiple decision trees to improve the accuracy and robustness of fluorescent sensor array analysis. They handle high-dimensional sensor data, identify important features, and provide reliable predictions for analyte identification and quantification.¹⁶²

Machine learning algorithms significantly enhance the performance of fluorescent sensor arrays by improving sensitivity, selectivity, and accuracy in detecting and quantifying target analytes such as antibiotics, pesticides, metal ions, and biomolecules.¹⁶³ These algorithms excel in efficiently extracting relevant features and reducing the dimensionality of complex, high-dimensional data, resulting in more robust and interpretable models. Their ability to handle nonlinear relationships, multicollinearity, and heterogeneous sensor data makes them versatile across a wide range of applications. Additionally, machine learning facilitates rapid screening and optimisation of sensor array designs, accelerating the development of more effective platforms. Their integration also enables real-time, on-site, and portable analysis, which is particularly valuable for environmental monitoring, food safety, and biomedical diagnostics.

Unsupervised and supervised machine learning algorithms differ significantly in their approach and applications, especially within the context of fluorescent sensor arrays. The key distinction lies in data labelling: supervised learning relies on labelled data, where each input is paired with a corresponding output, enabling the model to predict specific outcomes, such as the type or concentration of analytes. In contrast, unsupervised learning does not require labelled data and focuses on identifying patterns or structures in sensor responses without prior knowledge of the outcomes.

The objectives of these approaches also vary. Applications of unsupervised learning in fluorescent sensor arrays include clustering to group similar sensor responses, dimensionality reduction to handle high-dimensional data, and anomaly detection to identify unusual sensor responses. Pattern discovery can also reveal hidden correlations, offering new insights or

identifying previously unknown analytes. Overall, unsupervised learning is invaluable for data exploration and discovery, especially when labelled data is limited or unavailable. Examples of unsupervised techniques commonly used for sensor arrays are principal component analysis (PCA) and hierarchical cluster analysis (HCA).

Supervised learning approaches, on the other hand aim to map inputs to known outputs, making them ideal for classification and regression tasks in fluorescent sensor arrays, such as identifying different analytes or predicting their concentrations. Unsupervised learning, on the other hand, seeks to uncover the underlying structure of data, using techniques like clustering, dimensionality reduction, and anomaly detection. This is particularly useful for grouping similar sensor responses, simplifying complex data, and discovering novel patterns or outliers. The following section provides more detail about two supervised techniques – LDA and SVM – and their application to fluorescent sensor arrays.

2.2.3 Machine Learning in Fluorescent Sensor Arrays

2.2.3.1 Linear Discriminant Analysis

Linear discriminant analysis (LDA) is a supervised machine learning technique used for dimensionality reduction and classification.¹⁶⁴ It seeks to project data onto a lower-dimensional space while maximising the separation between classes by optimising the ratio of between-class variance to within-class variance. LDA assumes that the data for each class is normally distributed, with identical covariances and linear separability. It is particularly useful in scenarios where these assumptions hold, and it can be used both as a classifier and for dimensionality reduction before applying other classification algorithms. However, it may not perform well in complex, non-linear problems.

LDA was developed by the British statistician Ronald A. Fisher in the 1930s and first introduced in his seminal 1936 paper, "The Use of Multiple Measurements in Taxonomic Problems."¹⁶⁵ Fisher, often regarded as the "father of modern statistics", originally created LDA to classify biological specimens into species based on measured characteristics. The technique involves finding a linear combination of features that maximises the separation between different classes by optimising the ratio of between-class variance to within-class variance. Over time, LDA has become a fundamental method in machine learning, pattern recognition, and data analysis, with applications ranging from text mining to bioinformatics.

Analogy explanation of LDA mechanism

Imagine having a set of fruit trees, including apple and orange trees, with the goal of distinguishing between them based on various characteristics such as colour, size, shape, and brightness. With so much information, it might be challenging to know where to start. LDA assists by finding the "most effective line" that allows for easy differentiation between the two types of trees.

The core idea behind LDA is to identify a linear combination of features—essentially a line—that maximises the separation between different categories (apples and oranges) while minimising the variance within the same category. LDA begins by calculating the within-class variance (differences among apples or oranges) and the between-class variance (differences between apples and oranges). It then seeks a direction that maximises the between-class variance while minimising the within-class variance, effectively finding the optimal line that separates the categories. Once this optimal line is found, all data is projected onto it, creating a new dimension where the categories are as distinct as possible. A threshold is then set on this line to classify new data points—if a new tree falls on one side, it's an apple tree; if on the other, it's an orange tree.

LDA simplifies the classification process by reducing the complexity of the data, improving classification accuracy by maximising the separation between categories, and effectively handling both binary and multi-class classification problems. In essence, LDA helps find the simplest and most effective way to distinguish between categories, making it easier to classify complex data accurately.

Characteristics of LDA

The following is a list of key characteristics of an LDA-based machine learning classifier for array sensing:

Supervised Learning: LDA is a supervised learning algorithm that requires labelled data to learn the underlying structure and identify features that effectively discriminate between classes.

Dimensionality Reduction: LDA functions as a dimensionality reduction technique by projecting high-dimensional feature spaces onto lower-dimensional subspaces, optimising the separation between classes.

Class Separation: The principal objective of LDA is to determine a linear combination of features that maximises the separation between classes by maximising between-class variance while minimising within-class variance.

Gaussian Assumption: LDA operates under the assumption that the data within each class follows a Gaussian distribution, characterised by a class-specific mean and a common covariance matrix.

Optimal Classification: Under the Gaussian assumption, LDA provides an optimal linear classifier by minimising the likelihood of misclassification.

Generative Modelling: LDA models the underlying probability distributions of the classes, enabling not only the prediction of class labels but also the estimation of class probabilities.

Feature Selection: LDA can be employed for feature selection, as it identifies the most discriminative features that significantly contribute to class separation.

Robustness: LDA exhibits robustness to minor deviations from the Gaussian assumption, making it a viable choice for many real-world classification scenarios.

Regularisation: In situations where the covariance matrix is ill-conditioned or when the number of features exceeds the number of samples, regularisation techniques can be applied to LDA to enhance its performance and stability.

Extensions and Variations: Several extensions of LDA, including quadratic discriminant analysis (QDA), regularised discriminant analysis (RDA), and heteroscedastic discriminant analysis (HDA), address specific assumptions and challenges associated with different datasets.

These characteristics make LDA a powerful and widely adopted method for supervised dimensionality reduction and classification, particularly in domains such as pattern recognition, image processing, and bioinformatics.

LDA applications

Linear discriminant analysis has been instrumental in the development and optimisation of fluorescent sensor arrays across multiple research fields, significantly enhancing their ability to detect and differentiate complex analytes with high accuracy (Table 2.3). For example, researchers have applied LDA to analyse data from a fluorescence sensor array designed for metal ion detection.¹³⁹ This array, consisting of nine cross-reactive sensing elements, achieved over 96% accuracy in identifying metal cations such as Ca(II), Mg(II), and Zn(II) in environmental samples, while quantitative analysis exceeded 90% accuracy. LDA played a critical role in maximising the array's discriminatory power, enabling precise classification across a wide range of concentrations and pH levels.

In security and environmental monitoring, Woodka *et al.* (2010) developed a fluorescent polymer sensor array for detecting explosives dissolved in water.¹⁶⁶ Using LDA to analyse the fluorescence response patterns, the array distinguished various explosive compounds with high accuracy, even at low concentrations. This application of LDA not only improved detection accuracy but also facilitated the rapid identification of hazardous materials in environmental samples.

LDA has also proven valuable in medical diagnostics. Mitchell *et al.* (2021) applied LDA in a fluorescent sensor array to detect platinum-based anticancer drugs in biological fluids.¹⁵⁵ The array accurately distinguished between drugs like cisplatin and oxaliplatin and quantified their concentrations in complex biological matrices. LDA was crucial in separating fluorescence response patterns, enabling precise identification and quantification, which is essential for effective chemotherapy monitoring. The integration of LDA into the design of fluorescent sensor arrays has greatly enhanced their accuracy, reliability, and versatility across fields such as environmental monitoring, medical diagnostics, and security. LDA's ability to analyse and classify complex data with precision has made sensor arrays more effective in real-world applications, enabling accurate detection and differentiation of target analytes. A MATLAB based LDA classification protocol and code were presented in Appendix I 7.1.1.

Table 2.3 LDA based fluorescent sensor array application

NO	Ref	Sensor Details	Sensing Targets	Performance	Role of LDA
1	139	Nine small molecule sensors based on organic fluorophores	Metal cations including, Ca ²⁺ , Mg ²⁺ , Cd ²⁺ , Hg ²⁺ , Co ²⁺ , Zn ²⁺ , Cu ²⁺ , Ni ²⁺ , Al ³⁺ , and Ga ³⁺	96% qualitative accuracy and over 90% quantitative accuracy across concentration ranges.	Analysing multidimensional data, effectively clustering patterns and maximising class discrimination.

NO	Ref	Sensor Details	Sensing Targets	Performance	Role of LDA
2	168	Four Conjugated Polyelectrolytes, PPE1, PPE2, PPE-IDA, and PPE-HPA	Metal cations including, Pb^{2+} , Ni^{2+} , Cr^{3+} , Cu^{2+} , Hg^{2+} , and Fe^{3+}	High discrimination of explosives, allowing detection at low concentrations.	Analysed fluorescence response patterns to distinguish between different types of explosives.
3	167	Two complementary cucurbit[n]uril-type receptors	Amino acids (ornithine, lysine, arginine, histidine) and corresponding amines (putrescine, cadaverine, histamine, etc.)	100% correct classification of 10 analytes and control using cross-validation; fluorescence recovery proportional to analyte binding; apparent association constants vary with amino acids and amines, reflecting probe-analyte affinity differences.	Defined a response space with 95% variance, sufficient for clear discrimination of analytes based on fluorescence recovery data from the sensor array.
4	168	Four anionic conjugated polyelectrolytes (CPEs) with a poly(p-phenylene ethynylene) backbone and varying ionic side chains.	Heavy metal ions (Pb^{2+} , Hg^{2+} , Fe^{3+} , Cr^{3+} , Cu^{2+} , Mn^{2+} , Ni^{2+} , Co^{2+})	Clear differentiation of 8 metal ions using quenching effects on the CPEs. LDA transformed quenching response into canonical scores; accurate differentiation of ions in 2D and 3D plots. Detection sensitivity: 100 nM for most ions.	Transformation of the quenching responses of the four-PPE sensor array into canonical scores, achieving clear separation and clustering of eight metal ions in both 2D and 3D discrimination plots, with 95.8% variance explained.
5	169	Four types of carbon nanodots (CDs) synthesised from different amino acids (isoleucine, leucine, histidine, tyrosine).	Eight antibiotics (TC, OTC, CTC, SM, NEO, CHL, SPM, ERY)	100% correct discrimination of 8 antibiotics using LDA, with unique fluorescence patterns ("fingerprints"). Binary and ternary antibiotic mixtures were also effectively identified, with high accuracy even in real samples like human urine.	Enabled the discrimination of antibiotics based on fluorescence changes in a sensor array. Antibiotics were well-separated into distinct clusters, demonstrating clear differentiation. Used for both individual antibiotics and mixtures.
6	170	Colour-coded sensor array based on Ln-MOFs (Ln = Eu, Tb, Gd)	10 nitro-antibiotics (e.g., RNZ, NFZ, DNZ, MNZ, FD)	Detection limit as low as ppb level (e.g., TNZ 22.81 ppb, FD 40.01 ppb). Effective discrimination of both individual and mixtures of antibiotics in water samples with recovery rates from 90.8% to 110.8%.	Used for reducing data dimensions and visualising separability of antibiotic classes, achieving 100% correct classification of the 10 antibiotics. LDA also allowed for the construction of concentration-dependent response trends for quantification.
7	171	Bovine serum albumin-encapsulated gold nanoclusters (BSA-AuNCs) were used as fluorescence probes and three different dopamine (DA) concentrations as nonspecific receptors.	Eight heavy metal ions (Zn^{2+} , Ca^{2+} , Mn^{2+} , Fe^{3+} , Ni^{2+} , Cu^{2+} , Co^{2+} , and Sr^{2+})	Achieved 100% accuracy in discriminating 8 heavy metal ions using LDA. The sensor array showed good sensitivity with a LOD for Co^{2+} at 46.32 nM. Mixtures of metal ions were also discriminated with high accuracy. Fluorescence recovery response was used for analysis.	Used to transform fluorescence recovery data into canonical scores. This allowed for the successful discrimination of all metal ions, achieving 100% correct classification. The LDA also provided a linear quantitative relationship for some ions.

2.2.3.2 Support Vector Machines

Support vector machines (SVMs) are a prominent class of supervised machine learning algorithms, utilised primarily for classification tasks, though they can also be adapted for regression purposes.¹⁷² At the core of SVM lies the objective of identifying the optimal hyperplane that maximally separates different classes of data within a high-dimensional feature space. The SVM algorithm seeks to maximise the margin between these classes, where the margin is defined as the distance between the hyperplane and the nearest data points from each class, known as support vectors.

The development of SVMs began in the 1960s with Vladimir Vapnik's introduction of the maximum-margin hyperplane concept, which formed the foundation of the algorithm.¹⁷³ In the 1990s, Vapnik, alongside Bernhard Schölkopf and Corinna Cortes, advanced SVMs by formalising the algorithm and introducing key innovations such as kernel functions for non-linear classification and slack variables to handle non-separable data. These contributions, documented in several influential papers, established SVMs as a robust and versatile machine¹⁷⁴ learning technique, gaining widespread adoption in both research and industry for their strong theoretical underpinnings and practical efficacy.

Analogy explanation of SVM mechanism

Imagine a plot of land with apple trees and orange trees. The task is to draw a line that separates the apple trees from the orange trees, so that in the future, anyone who sees this line will know whether apples or oranges are planted on that side of the land. The goal of SVM is to find the "optimal" boundary line that perfectly separates the apples and oranges, ensuring that this line is as far away as possible from the closest points in each category. These closest points are called "support vectors." SVM will start by trying to draw many different lines between the apple trees and orange trees. These lines might differ in direction and position. SVM doesn't just choose any line that separates the two categories; it selects the line that is the farthest from the nearest apple and orange trees (the support vectors). The aim here is to ensure that, no matter where a new tree appears on this plot, as long as it falls on a particular side of the line, it can be confidently identified whether it is an apple or an orange tree. Sometimes, the apples and oranges might be mixed in such a way that it is difficult to separate them with a straight line. SVM can use a technique called the 'kernel trick' to map the data into a higher-dimensional space. In this new space, it becomes easier to find a line that separates the two

categories. Although this process cannot be directly visualised in two dimensions, the computer is capable of executing it in higher-dimensional space. Once SVM identifies the optimal boundary, a new data point—such as a previously unseen fruit—can be classified by determining on which side of the boundary it lies, thereby distinguishing whether it is an apple or an orange.

Characteristics of SVMs

The following is a list of key characteristics of an SVM-based machine learning method for array sensing:

Data Representation and Kernel Functions: SVMs often involve transforming the input data into a higher-dimensional space using a kernel function. This transformation enables the algorithm to capture complex, non-linear relationships that may not be apparent in the original feature space. Various kernel functions, such as linear, polynomial, and radial basis function (RBF) kernels, can be employed depending on the nature of the data and the specific problem at hand.

Optimal Hyperplane Determination: The primary goal of the SVM algorithm is to find the hyperplane that best separates the data into distinct classes while maximising the margin. The support vectors, which are the data points closest to the hyperplane, play a crucial role in determining its position. These points are the most informative instances in the dataset, as they directly influence the decision boundary.

Regularisation and Overfitting Prevention: SVMs incorporate a regularisation parameter, denoted as C , which balances the trade-off between maximising the margin and minimising the classification errors on the training set. This regularisation helps to prevent overfitting, particularly in cases where the data may contain noise or when the feature space is of high dimensionality.

Handling Non-Separable Data: In scenarios where the data is not linearly separable, SVMs introduce slack variables that allow for some data points to be misclassified, effectively permitting them to be on the wrong side of the margin. This flexibility enables SVMs to manage more complex datasets while still striving for an optimal classification boundary.

Multiclass Classification: Although SVMs are inherently binary classifiers, they can be extended to handle multiclass classification problems through strategies such as one-vs-rest (OvR) or one-vs-one (OvO). These techniques involve decomposing the multiclass problem into multiple binary classification tasks, which are then aggregated to produce the final classification outcome.

SVM applications

In the early 2000s, researchers began exploring the application of support vector machines (SVMs) for analysing data from fluorescent sensor arrays, often referred to as "electronic noses" or "electronic tongues". These arrays, composed of multiple fluorescent dyes or receptors, generate complex fluorescence patterns in response to various analytes. The need for accurate and robust classification algorithms led to the adoption of SVMs, which proved particularly effective due to their ability to handle high-dimensional and non-linear data. SVMs demonstrated strong performance in classifying fluorescence patterns and identifying analytes, even in noisy environments. The kernel-based approach of SVMs further enhanced classification accuracy by incorporating prior knowledge about sensor arrays and analytes, facilitating applications in environmental monitoring, food and beverage analysis, and medical diagnostics.

The integration of SVMs into sensor array design has advanced the accuracy and applicability of these technologies across various fields (Table 2.4). For example, Chang Liu *et al.* (2018) employed SVM regression to model the complex relationship between sensor signals and herbicide concentrations in a supramolecular fluorescent sensor array.¹⁷⁵ This system demonstrated high effectiveness in detecting paraquat and diquat, even in the presence of interfering substances, underscoring its utility for environmental safety monitoring. Similarly, Yanchao Zhu *et al.* (2021) developed a fluorescent sensor array-based electronic tongue, using SVM to achieve 100% accurate classification of nine Chinese tea samples, highlighting its potential in food authentication.¹⁷⁶

Further advancements include Zijun Xu *et al.*'s (2020) development of a dual-channel fluorescence sensor array for detecting tetracycline antibiotics in river water and milk.¹⁷⁷ SVM was pivotal in analysing fluorescence patterns, enabling precise classification and identification of various tetracyclines. Min Li *et al.* (2023) used SVM in a two-element fluorescence sensor array to differentiate between flavanols, complex compounds found in

Kaempferia galangal L. and honey, demonstrating SVM's role in enhancing sensitivity and specificity in food analysis.¹⁷⁸

In summary, SVM has made a significant impact on the development of sensor arrays, enabling researchers to handle complex, nonlinear datasets with greater accuracy and reliability. Its versatility in managing high-dimensional data and improving sensor array performance across fields such as environmental monitoring, food safety, and biomedical diagnostics makes it an invaluable tool in advancing sensor technologies. A MATLAB based SVM classification protocol and code are presented in Appendix I 7.1.2.

Table 2.4 SVM based fluorescent sensor array application

No	Ref	Sensor Details	Excitation Wavelength	Sensing Targets	Performance	Role of SVM
1	176	Six soluble conjugated polymeric nanoparticles (SCPNS) embedded in waterborne polyurethane.	365 nm	Nine Chinese tea samples	The sensor array achieved 100% accuracy in tea discrimination through linear SVM classification with leave-one-out cross-validation.	To predict and classify the tea samples based on the fluorescence quenching response data.
2	179	Three sensing elements. A virtual sensor array (VSA) based on a single metal-organic framework (MOF) recognition unit (MOF-74 (Zn)) with the excitation wavelength as external regulatory factors (three region of EEM profiles).	200–400 nm	Four aromatic pollutants (phenylamine, chlorobenzene, nitrobenzene, and phenol) in both single and binary mixtures.	Achieved classification accuracy of 100% for single analytes and 96% for mixtures using a PCA-SVM model with 10-fold cross-validation. Quantitative predictions for concentration showed acceptable error ranges except for chlorobenzene.	Qualitative classification and quantitative predictions of the pollutants, enhancing classification accuracy and efficiency by combining with principal component analysis (PCA).
3	180	Four different fluorescent compounds (AFGlu 1–4)	365 nm	Glucose concentrations in bio-fluids, specifically artificial tears for monitoring diabetes.	The random forest model achieved 96.7% classification accuracy with a mean squared error (MSE) of 0.060 mM. The SVM regression model yielded an MSE of 0.026 mM for glucose concentration prediction.	To estimate glucose concentrations with finer granularity, providing continuous glucose monitoring with high accuracy.
4	177	Two quantum dots, QR-CDs (Quinaldine Red) and CPC-CDs (Cetylpyridinium Chloride).	320 nm (QR-CDs), 370 nm (CPC-CDs).	Four tetracycline antibiotics: tetracycline (TC), oxytetracycline (OTC), doxycycline (DOX), metacycline (MTC).	The array successfully distinguished all four tetracyclines at concentrations between 1.0 μ M and 150 μ M, achieving 100% accuracy for 72 unknown samples using SVM and LDA. The system showed excellent selectivity and anti-interference ability in complex media, such as river water and milk samples.	Data processing and classification, processing both linear and non-linear classification problems.

5	181	Six sensors. AC-CQDs (CQDs) functionalised with five different amino acids (glutamine, histidine, arginine, lysine, and proline).	450 nm	Eleven metal ions (Mo^{5+} , Fe^{3+} , Cr^{3+} , Er^{3+} , Yb^{3+} , La^{3+} , Ni^{2+} , Cu^{2+} , Co^{2+} , Pb^{2+} , Mn^{2+}).	Achieved 100% classification accuracy for the metal ions using LDA and SVM models. The SVM also predicted the concentration of individual metal ions in mixtures with good accuracy.	Quantitative regression analysis for estimating the concentrations of metal ions in mixtures. SVM contributed to accurate classification and prediction of metal ion concentrations.
6	182	Six sensing elements. AMS (ams-OSRAM AG, Austria) AS7263 sensor unit with six spectral bands (3×2 photo diode array)	610, 680, 730, 760, 810, 860 nm	Maize canopies responding to increased temperatures and water deficit, specifically focusing on leaf rolling (LR) as a drought stress indicator.	The SVM model achieved over 80% accuracy in stratified cross-validation, with comparable performance in other validation procedures.	Classification of multispectral sensor data for detecting leaf rolling in maize, yielding high accuracy and precision for drought stress detection in maize canopies.
7	183	Four fluorescent polymer thin films (ADS518PT, ADS2008P, PTMA, PURET)	440 nm	Explosive compounds including TNT (2,4,6-trinitrotoluene), DNT (2,4-dinitrotoluene), ammonium nitrate, and nitrotoluene.	The sensor array showed a recognition rate of 100% within 5 seconds using the SVM neural network. The system demonstrated high robustness to additive noise and could detect explosives at the sub parts per billion (ppb) level.	Pattern recognition method for classifying the fluorescent quenching data, achieving a 100% recognition rate after 5 seconds.

2.2.3.3 Conclusion

This literature review explores the roles of linear discriminant analysis, and support vector machines in data analysis, focusing on their complementary strengths in dimensionality reduction and classification. LDA, while also used for dimensionality reduction, primarily focuses on maximising class separability by identifying the linear combination of features that best distinguishes between classes. This makes LDA particularly useful for supervised classification tasks where clear separation of classes is needed.

SVM is recognised for its robust classification capabilities, especially in scenarios where data is not linearly separable. By finding the optimal hyperplane that maximises the margin between classes, SVM effectively handles both linear and non-linear classification problems, thanks to its use of kernel functions. The review underscores the synergistic use of these techniques, particularly how LDA can preprocess data to enhance SVM's performance by reducing noise and focusing on the most discriminative features. Together, LDA, and SVM form a powerful trio in machine learning, with LDA simplifying data and SVM providing strong classification capabilities. Their combined use is particularly effective in complex, high-dimensional datasets, ensuring both computational efficiency and high accuracy in predictive models. These methods remain essential in various applications, from pattern recognition to bioinformatics, due to their robust theoretical foundations and adaptability to evolving data challenges.

The integration of machine learning into the development of fluorescent sensor arrays offers significant advantages, particularly in handling complex, high-dimensional data. Techniques like SVM, and LDA enable the extraction of meaningful patterns from noisy data, leading to the creation of highly sensitive and specific sensor arrays. ML's flexibility allows researchers to explore non-linear relationships, resulting in innovative sensor designs that traditional methods might overlook. However, there is a need to balance model complexity with interpretability, ensuring that the underlying mechanisms of sensor arrays are understood and can be practically implemented. For future research, it is recommended to explore advanced ML techniques such as deep learning and ensemble methods to enhance the accuracy and robustness of sensor arrays. Hybrid models that combine ML with domain-specific knowledge could lead to more guided and efficient sensor development. Additionally, focusing on interpretable ML models is crucial for ensuring that the outputs of these sensors are not only accurate but also understandable to end-users, particularly in critical applications like healthcare and environmental monitoring.

Further research directions include leveraging data augmentation and simulation to overcome the challenges of limited experimental datasets, fostering cross-disciplinary collaboration to enhance ML integration, and conducting real-world application testing to validate the performance of ML-designed sensor arrays. These steps will help bridge the gap between theoretical models and practical applications, ensuring that fluorescent sensor arrays can perform reliably in diverse and complex environments

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3 Carbon dots-based pH sensor array

This chapter is currently under consideration for publication in *Sensors and Diagnostics* (second round of revisions). It is presented here as submitted, with supplementary figures included in the main text for enhanced readability.

This chapter will present a sensor array for precise pH monitoring, based on carbon dots (CDs) synthesised from fructose and p-phenylenediamine through a one-step hydrothermal method. The CDs exhibited significant photostability and different fluorescence emissions in different pH conditions, making them suitable for real-time pH sensing across a wide pH range of 3 to 10. Our mechanistic studies revealed how surface functionalisation affects the pH response. For statistical analysis of our array data, this thesis employed Gaussian process regression to create a predictive model for determining pH levels of test samples based on the array spectral data, and linear discriminant analysis for high-precision classification of pH values. This combined approach enabled a comprehensive analysis of the CDs' pH-sensitive fluorescence, demonstrating a significant methodological advancement in pH sensor technology. Our research advances the understanding of the fluorescence mechanisms of CDs in response to pH variations. Furthermore, our study demonstrates the power of integrating machine learning techniques to improve the performance and application scope of fluorescent nanomaterials. These findings have important implications for chemical sensing across a range of fields, including environmental monitoring and biomedical diagnostics

3.1 Introduction

Determination of pH is a crucial analytical challenge across a range of industrial challenges, from food spoilage¹ to environmental monitoring² to a broad range of biological questions. For example, pH sensing is crucial for biomedical applications due to its significant role in drug delivery, disease diagnosis, and physiological condition monitoring.³⁻⁷ The development of pH-responsive amphiphilic polymer nano-assemblies enhances drug delivery efficiency while reducing side effects.⁵ Additionally, flexible potentiometric pH sensors for wearable technologies highlight the importance of real-time health monitoring.⁸ These sensors are particularly valuable for continuous pH monitoring in bodily fluids, facilitating the early detection of health condition changes or abnormalities.^{9, 10} Moreover, pH determination is vital in clinically relevant diagnoses, where sensors are utilised in drug control-released carriers and other diagnostic tools.¹¹

A broad range of analytical tools have been developed to measure pH within complex environments, including H^+ permeable microelectrodes,¹² advanced nuclear magnetic resonance (NMR) techniques,¹³ and fluorescent sensors.¹⁴ The latter are advantageous as they do not require physical breaching of cell membranes, and they can be coupled to inexpensive and readily available instruments for quantifying and imaging fluorescence. A broad range of pH-responsive fluorescent sensors have been reported to date, including those based on fluorescent proteins,¹⁵ organic fluorophores,¹⁶ and fluorescent nanomaterials.¹⁷ The protonatable groups typically have a working range within ± 1 pH unit of the pK_a , therefore limiting the working range of the final assay to less than 2 pH units.

One approach to address this limitation is to simultaneously use multiple sensors with different pK_a values, and therefore, different pH working ranges. This can be achieved through the use of a sensor array, built by combining multiple sensors, each with specific properties, into a single platform.¹⁸ Array-based sensing offers several advantages, including increased selectivity and sensitivity, and reduced interference from extraneous substances.^{6, 19, 20} This versatility enables the detection of a wide range of pH values and the ability to tailor the sensor array for specific applications or environments. The use of array sensing technologies in pH sensing applications can lead to enhanced accuracy compared to traditional methods. This improvement in accuracy is crucial for various fields where precise pH measurements are essential, such as environmental monitoring and biomedical applications. This approach has been elegantly demonstrated by Kim and co-workers, who used a library of 30 fluorescent sensors to achieve pH determination within 0.2 pH units.²¹

This thesis was interested in developing an array with fewer sensor elements, in order to increase efficiency of synthesis and array application. This thesis therefore aimed to develop a new sensor array based on carbon dots (CDs) due to their previously reported pH-responsive properties.²²⁻²⁴ Traditional pH indicators, such as dyes and organic fluorophores, face limitations, including reduced stability, susceptibility to photobleaching, and potential cytotoxicity.²⁵ Conversely, CDs have emerged as a superior alternative, renowned for their robust fluorescence, exceptional photostability, low toxicity, and facile synthesis.²⁶⁻²⁸ As small carbon nanoparticles, CDs display unique optical properties due to quantum confinement effects, surface modifications, and diverse carbonaceous compositions.²⁹⁻³¹ These features render CDs ideal for pH sensing applications, as they can significantly alter their emission wavelength and intensity in response to environmental pH changes, thus serving as effective

fluorescent probes for pH differentiation across a spectrum of applications.^{22, 32} Recent research has focused on exploring the fluorescence mechanisms of CDs.³³⁻³⁵ Understanding these mechanisms is crucial for several reasons: it paves the way for enhancing their fluorescent properties, facilitates the creation of novel CDs with tailored features, and aids in developing standardised methodologies for their characterisation. This chapter focused on a mechanistic study of the surface functionality impact on CDs pH response.

Here we synthesised CDs from fructose and p-phenylenediamine precursors. After determining the surface groups contributing to pH sensing, this thesis meticulously analysed the pH sensing capabilities exhibited by the CDs array using two advanced statistical methodologies: Gaussian process regression (GPR) and linear discriminant analysis (LDA).

3.2 Results and discussion

3.2.1 Synthesis of and characterisation of carbon dots

3.2.1.1 Carbon dots synthesis procedure and morphology

A series of CDs were synthesised via a one-pot hydrothermal method. Two carbon sources were selected as precursors: fructose, which can also introduce hydroxyl groups; and p-phenylenediamine (PPD), which can also introduce amino groups. The heating time and temperature were selected based on our previous work.³⁶ This study focused on altering the ratio of precursors to further investigate its effect on the surface functionality of the synthesised CDs. In total, five different fructose: PPD ratios were used (100:0; 92:8; 69:31; 43:57; and 25:75) to give five CDs, named CD100, CD92, CD69, CD43 and CD25, respectively. The hydrothermal reactions were carried out in an oven preheated to 180 °C for 8 h to ensure consistent synthetic conditions. The synthesised CDs were purified by filtration through a 0.22 µm cellulose acetate filter to remove large and aggregated particles (TEM figures of the synthesised CDs were illustrated in Figure 3.1). The average sizes of the CDs were ranged from 8 to 15 nm as listed in Table 3.1.

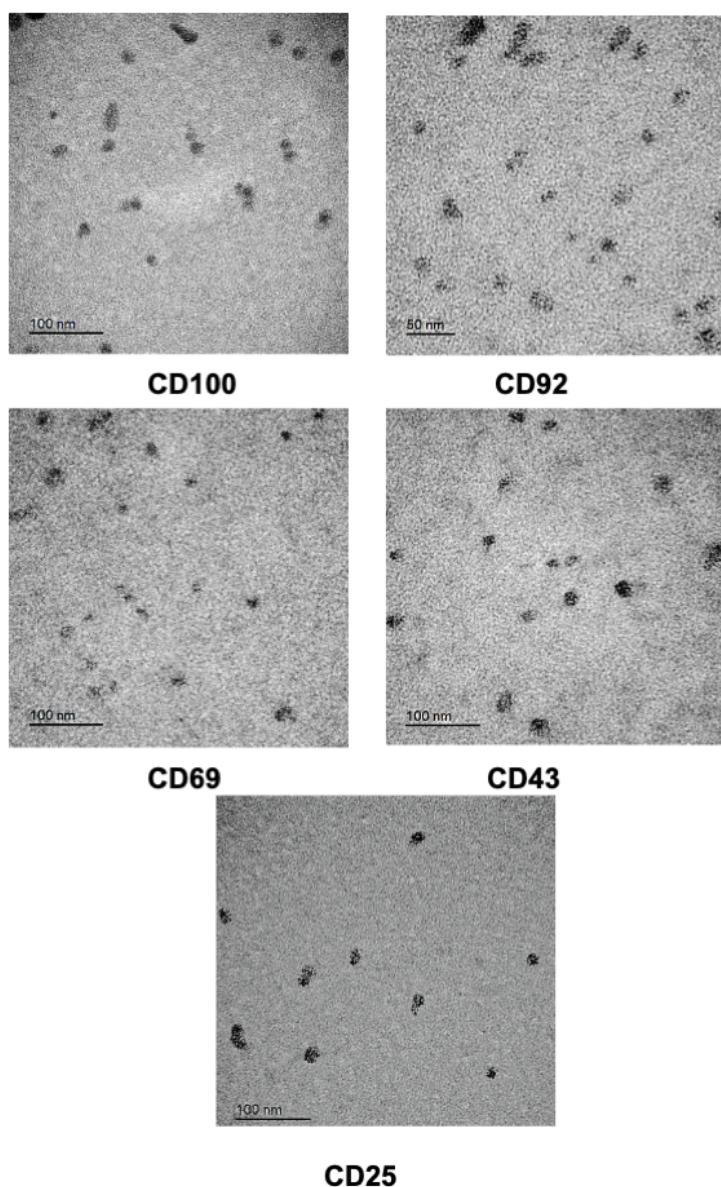


Figure 3.1 TEM figures of synthesised carbon dots

Table 3.1 Size analysis of synthesised carbon dots

Carbon dots	average size/nm	standard deviation/nm
CD100	10.72	6.38
CD92	7.95	3.14
CD69	10.71	4.19
CD43	11.06	4.94
CD25	15.45	5.35

In this study, CDs such as CD43 and CD25 exhibited hydrodynamic diameters slightly exceeding 10 nm (as summarized in Table 3.1), which may be attributed to surface passivation, partial aggregation, or limitations in filtration during post-synthesis purification. Despite their

size, these CDs displayed strong excitation-dependent fluorescence and optical properties consistent with quantum confinement effects typically seen in smaller carbon nanomaterials.

Regarding photostability, representative CDs (e.g., CD43) retained consistent fluorescence intensity and pH-responsive behaviour after being stored for three months at room temperature in sealed vials. This indicates that the CDs remained stable under ambient conditions, and their optical functionality was not compromised by thermal or oxidative degradation over time (see Section 3.2.8 for details on long-term photostability).

3.2.1.2 Attenuated total reflectance spectroscopy

The attenuated total reflectance (ATR) spectroscopy provided structural insights into the five synthesised CDs. CD100, synthesised from fructose alone, displayed bands at 3300, 2970, 1660, 1405, and 1050 cm^{-1} , corresponding to the stretching vibrations of O-H, C-H, C=O, COO-, and C-O respectively (Figure 3.2). Introduction of some nitrogen doping, in CD92, and CD69, gave rise to an additional peak at approximately 1500 cm^{-1} , corresponding to N-H. CD43 and CD25 had the highest N-doping levels, and therefore exhibited strong ATR peaks located at N-H (1510 cm^{-1}), as well as peaks corresponding to C-N/C-O (1390, 1260, and 1070 cm^{-1}).

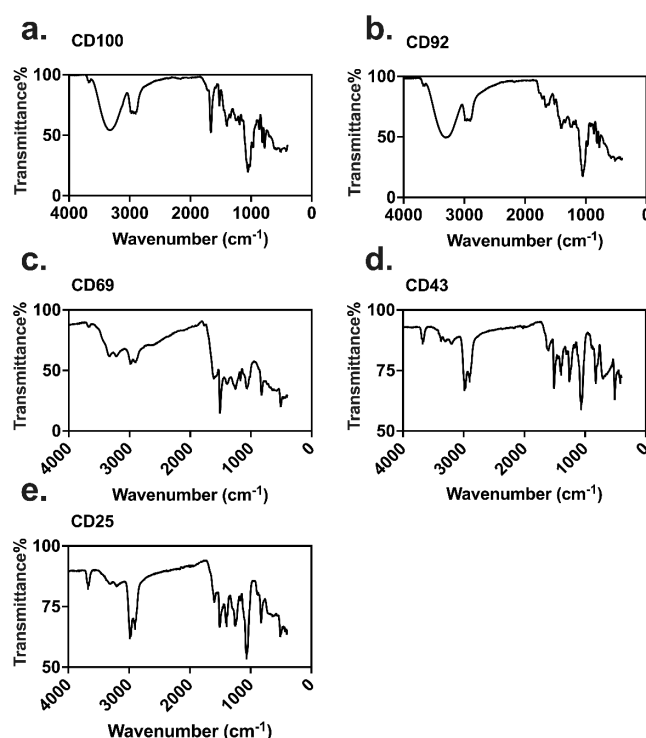


Figure 3.2 Attenuated total reflectance (ATR) spectra of the five carbon dots (a. CD100, b. CD92, c. CD69, d. CD43, and e. CD25).

3.2.1.3 X-ray photoelectron spectroscopy

X-ray photoelectron spectroscopic (XPS) analysis of CDs (Figure 3.4, Figure 3.5 and Table 3.2-5) gave insight into the surface functionality of the particles. CD100 showed 67.5% carbon (C–C/C=C, C–O, and O–C=O) and 32.5% oxygen (–OH and =O), in agreement with the ATR results (Figure 3.2). As the degree of N-doping increased from CD92 to CD25, the nitrogen surface content determined by XPS increased from 5.5% to 17.1%, with a concomitant decrease in the oxygen atomic percentage (from 26.3% to 5.8%). This corresponds to an increase in the proportion of –OH groups and a corresponding increase in the proportion of amino groups.

Table 3.2 Summary of X-ray photoelectron spectroscopy (XPS data) for carbon dots.

Carbon dot	Orbital	Peak BE ^a / eV	FWHM ^b / eV	Area (P) / CPS.eV	Atomic %
CD100	C1s	285.89	3.46	1101342.77	67.5
	O1s	532.97	2.73	1281038.55	32.5
CD92	C1s	286.12	2.9	1070425.44	68.2
	N1s	401.78	3.77	133969.56	5.5
	O1s	533.28	3.24	996332.28	26.3
CD69	C1s	285.93	2.74	1078439.21	70.6
	N1s	400.77	3.93	220901.22	9.3
	O1s	532.9	3.2	740176.29	20.1
CD43	C1s	285.87	3.41	1104498.22	75.3
	N1s	400.13	3.21	311061.83	13.7
	O1s	532.74	3.59	393557.56	11.1
CD25	C1s	284.84	2.85	2140792.94	77.1
	N1s	399.03	2.78	738685.36	17.1
	O1s	531.36	3.23	389977.55	5.8

- a. BE = binding energy
- b. FWHM = full width at half maximum

Table 3.3 Summary of XPS data of the carbon region (C 1s) for carbon dots

Carbon dot	Functional group	Peak BE / eV	FWHM / eV	Area (P) / CPS.eV	Atomic %
CD100	C-C/C=C	284.8	1.3	61871.39	43.9
	C-O	286.33	1.37	67489.98	47.9
	O-C=O	287.89	1.2	11562.66	8.2
CD92	C-C/C=C	284.8	1.53	100684.8	71.9
	C-O / C-N	286.65	1.65	18782.78	13.4
	O-C=O	288.82	1.53	20591.59	14.7
CD69	C-C/C=C	284.8	1.45	76479.39	53.6
	C-O / C-N	285.46	2.8	50598.77	35.5
	O-C=O	288.72	1.85	15583.86	10.9
CD43	C-C/C=C	284.8	1.53	23897.45	16.3
	C-O / C-N	285.94	2.65	114934.7	78.5
	O-C=O	289.36	3.37	7574.72	5.2
CD25	C-C/C=C	284.8	0.92	36832.34	14.1
	C-O / C-N	285.53	2.03	217490.85	83.5
	O-C=O	288.16	1.4	6229.01	2.4

Table 3. 4 Summary of XPS data of the oxygen region (O 1s) for carbon dots

Carbon dot	Functional group	Peak BE / eV	FWHM / eV	Area (P) / CPS.eV	Atomic %
CD100	-OH	532.77	1.63	138287.57	85.0
	=O	533.26	2.07	24319.89	15.0
CD92	-OH	531.61	1.45	19714.04	14.6
	=O	532.51	2.51	114910.67	85.4
CD69	-OH	531.36	1.41	19107.95	18.5
	=O	532.37	2.54	84049.2	81.5
CD43	-OH	532.65	3.08	52160.32	100
CD25	-OH	531.52	1.41	11596.08	23.4
	=O	532.53	2.76	37861.02	76.6

Table 3.5 Summary of XPS data of the nitrogen region (N 1s) for carbon dots

Carbon dot	Functional group	Peak BE / eV	FWHM / eV	Area (P) / CPS.eV	Atomic %
CD92	Amino N	400.78	2.13	8525.03	50.7
	Graphitic N	402.64	1.82	8270.06	49.3
CD69	Amino N	400.13	1.71	16827.02	60.1
	Graphitic N	402.16	1.93	11138.72	39.9
CD43	Amino N	399.79	2.67	41330.05	100
CD25	Amino N	398.92	1.63	105106.06	100

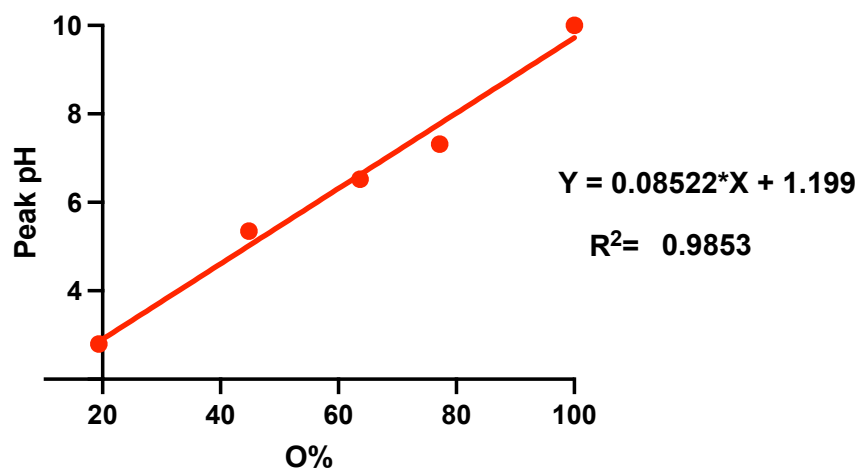


Figure 3.3 Linear relationship between the atomic percentage of oxygen in hydroxyl group (vs. nitrogen in amino group) in CDs and the pH of maximum fluorescence intensity (characterised by XPS; $R^2 = 0.9853$).

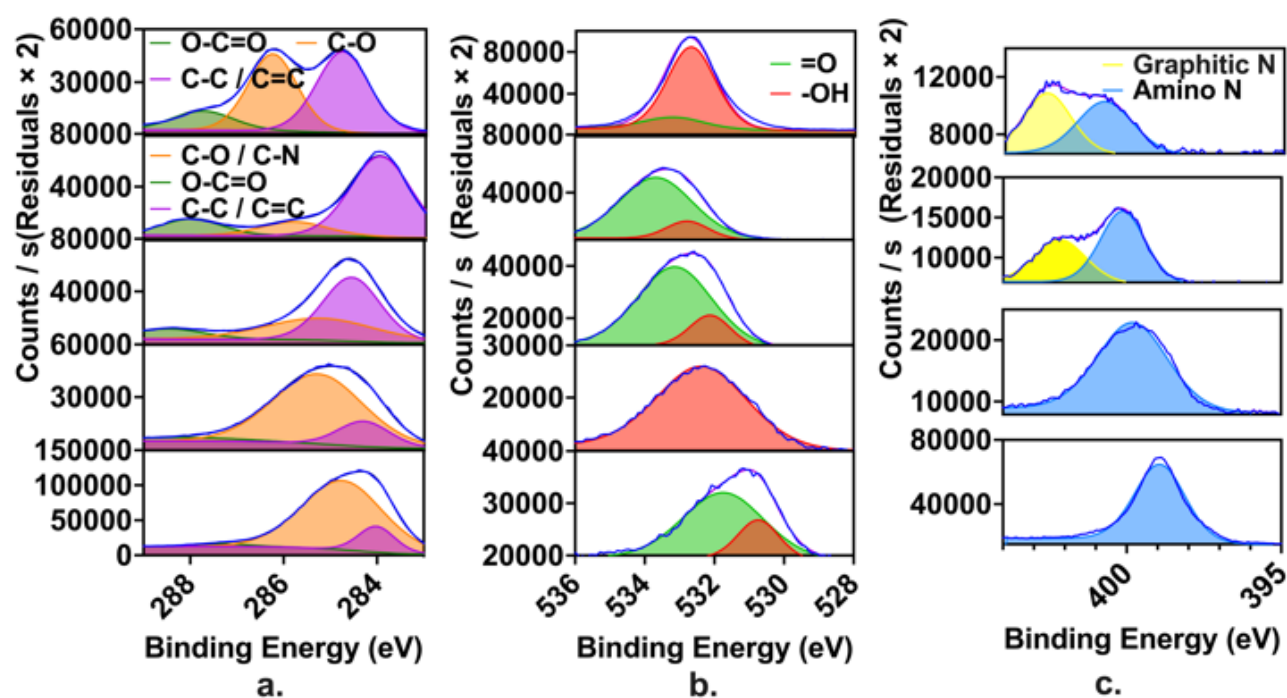


Figure 3.4 High resolution XPS spectra of the five CDs, listed vertically in the order of CD100, CD92, CD69, CD43 and CD25, showing (a) carbon region (C 1s), (b) oxygen region (O 1s) and (c) nitrogen region (N 1s).

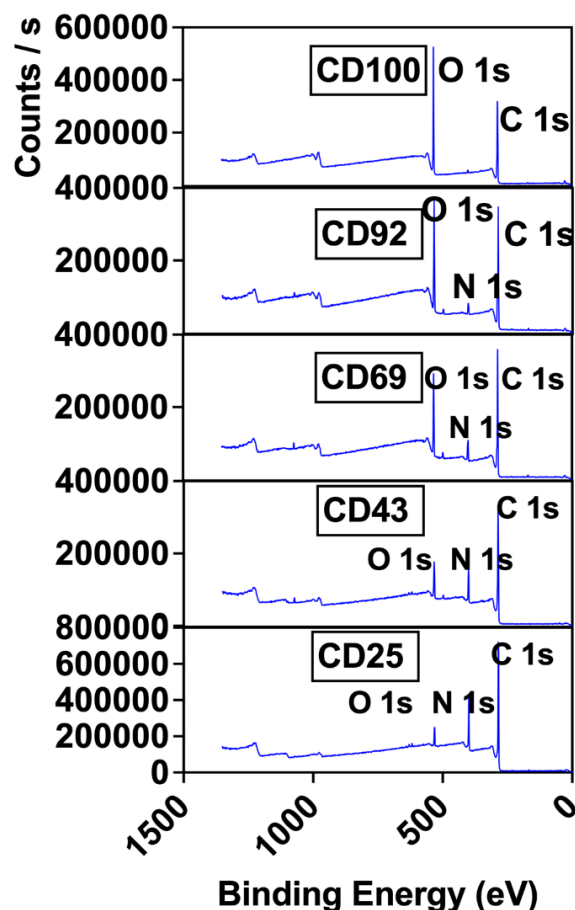


Figure 3.5 XPS spectra of the five CDs, in order of CD100, CD92, CD69, CD43 and CD25 from top to bottom.

3.2.1.4 Absorption

UV-visible spectroscopy gave further insight into the composition of the CDs (Figure 3.6, the absorption spectrums of five CDs under different pH buffers were presented in Figure S3.1). CD100 shows a predominance of oxygen-containing functional groups, such as hydroxyl and carboxyl groups, evidenced by strong absorption between 290-310 nm, corresponding to aromatic or graphitic π - π^* transitions,^{37,38} and a secondary peak between 320-350 nm, corresponding to n - π^* transitions of C=O groups,³⁹ suggesting significant oxidation.⁴⁰ With the N-doping level increased, CD92 to CD25 showed an absorption increase around 350-400 nm, which was related to the π - π^* transitions of the aromatic C=C bonds and n - π^* transitions of C=N heterocycles. This shift was evidenced by a lower intensity in the 290-310 nm range and diminished n - π^* transitions in the 320-350 nm range.

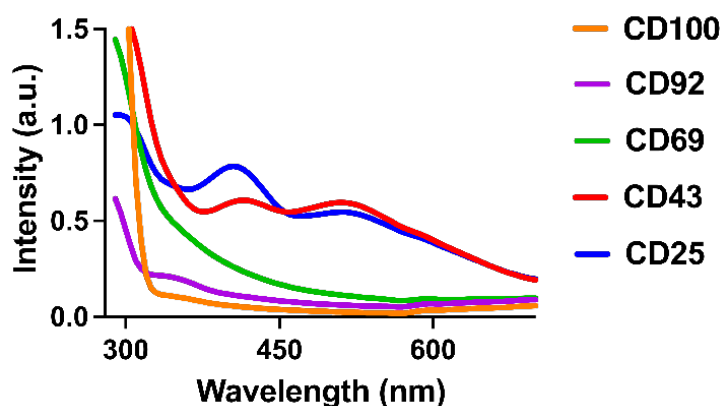


Figure 3.6 UV-vis absorption of five CDs under pH 7.00 buffer (a. CD100, b. CD92, c. CD69, d. CD43, and e. CD25, f. summary of CDs, at 0.25 mg/mL respectively.)

3.2.2 Fluorescence characterisation and pH response of carbon dots

The fluorescence excitation and emission spectra of the synthesised CDs are shown in Figure 3.8, the full range fluorescence excitation and emission spectra (with λ_{ex} 330-450 nm, and λ_{em} 470-590 nm) was presented in Figure 3.7. The excitation peaks for these CDs range from 354 nm to 393 nm, and the emission peaks span from 510 nm to 545 nm (listed in Table 3.6). CD100 exhibits a Stokes shift of 150 nm, whereas the introduction of nitrogen source precursors for N-doping lead to an initial decrease to 117 nm (CD92), then a steady increase to 186 nm (CD25). This ascending trend in Stokes shift, concomitant with an increase in amino groups on the surface of CDs, is consistent with an augmented separation between the electronic energy levels, leading to enhanced red emission.^{41, 42} Furthermore, the increase in nitrogen doping influenced the solubility of CDs, as the proliferation of $-\text{NH}_2$ groups led to the formation of hydrophobic CDs. These hydrophobic CDs were prone to aggregation, resulting in variations in fluorescence.^{42, 43} Additionally, PPD, characterised by its conjugated structure, contributed to the enlargement of sp^2 domains, which was advantageous for the red shift in the emission wavelength of CDs.^{42, 44}

Table 3.6 Summary of the fluorescence features of the synthesised CDs

CD	Excitation maximum/nm	Emission maximum/nm	Stokes shift/nm
CD100	360	510	150
CD92	393	510	117
CD69	369	530	161
CD43	354	525	171
CD25	359	545	186

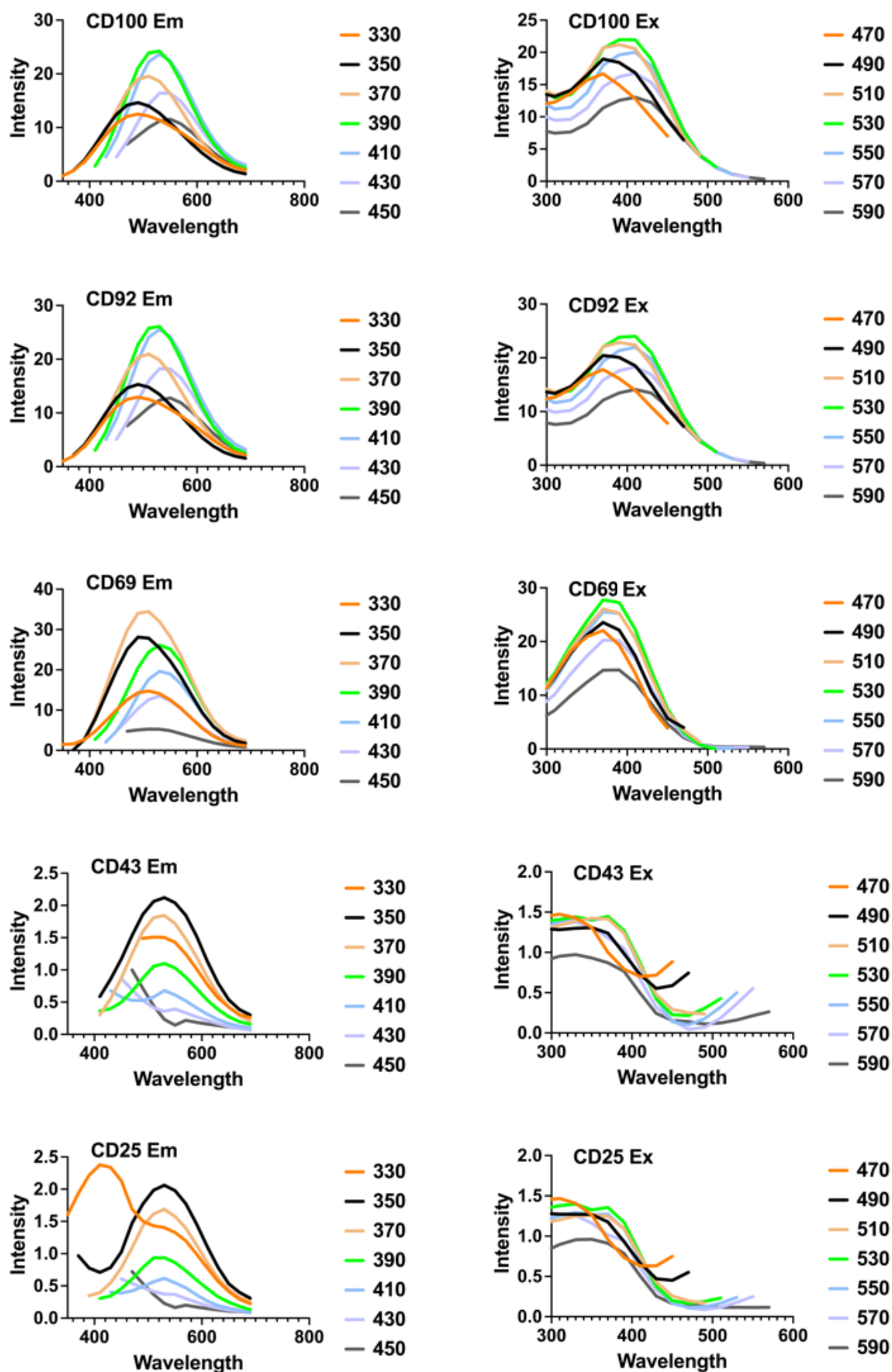


Figure 3.7 Fluorescence excitation and emission spectra of synthesized CDs (with λ_{ex} 330-450 nm, and λ_{em} 470-590 nm.)

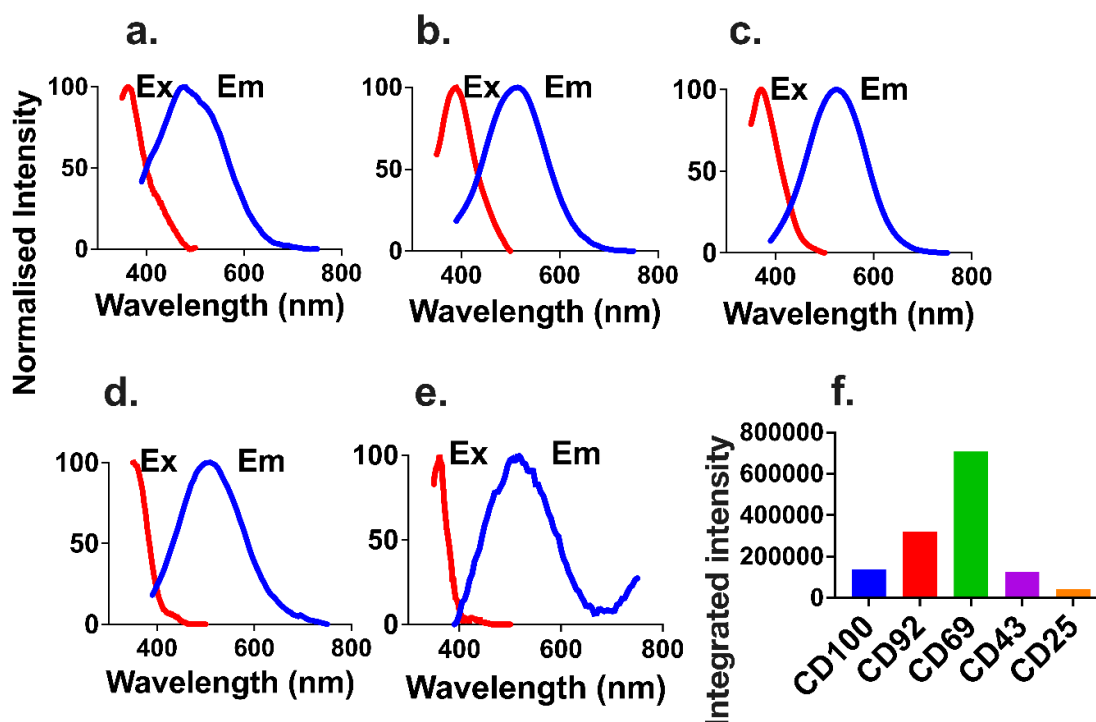


Figure 3.8 Fluorescence spectra of the synthesised CDs: (a-e) show excitation (red) and emission (blue) spectra for CD100, CD92, CD69, CD43, and CD25, respectively; (f) summarises the fluorescence intensity variations among the CD series (1 mg/mL CDs).

In addition to variation in fluorescence wavelengths, total emission intensity also varies across the series of CDs (Figure 3.8 (f)). As the proportion of the nitrogen source PPD increased during hydrothermal synthesis, the fluorescence intensity of the CDs initially rose and then declined, with CD69 exhibiting the highest fluorescence intensity. Thus, the effect of nitrogen doping on the fluorescence intensity of CDs was not straightforwardly linear. It involved a complex interplay of factors that could either enhance or quench fluorescence, depending on the specific levels of doping and the environmental conditions.⁴⁵

The absorption and emission spectra of the CDs (Figure 3.9 and Table 3.7) demonstrate a distinct dependency on pH levels, with clear shifts in peak positions and variations in intensity. For instance, CDs with a higher oxygen content, such as CD100, consistently exhibited emission peaks around 500 nm at low pH, accompanied by Stokes shifts ranging from 85 to 160 nm. On the other hand, CDs with a higher nitrogen content, like CD25, presented higher emission peaks at lower pH levels, with Stokes shifts ranging from 130 to 190 nm. These differences implied that the fluorescence properties of CDs were significantly affected by their oxygen and nitrogen content, which in turn influenced their sensitivity and stability across various pH levels. Specifically, higher nitrogen content appears to stabilise both the emission

and excitation wavelengths over a broader pH range. Conversely, CDs with a higher oxygen content displayed more pronounced shifts, particularly in their emission peaks. These findings highlight the potential use of CDs in pH sensing applications. They demonstrate how variations in oxygen and nitrogen content could be leveraged to tune the fluorescence properties of CDs for specific sensing needs.

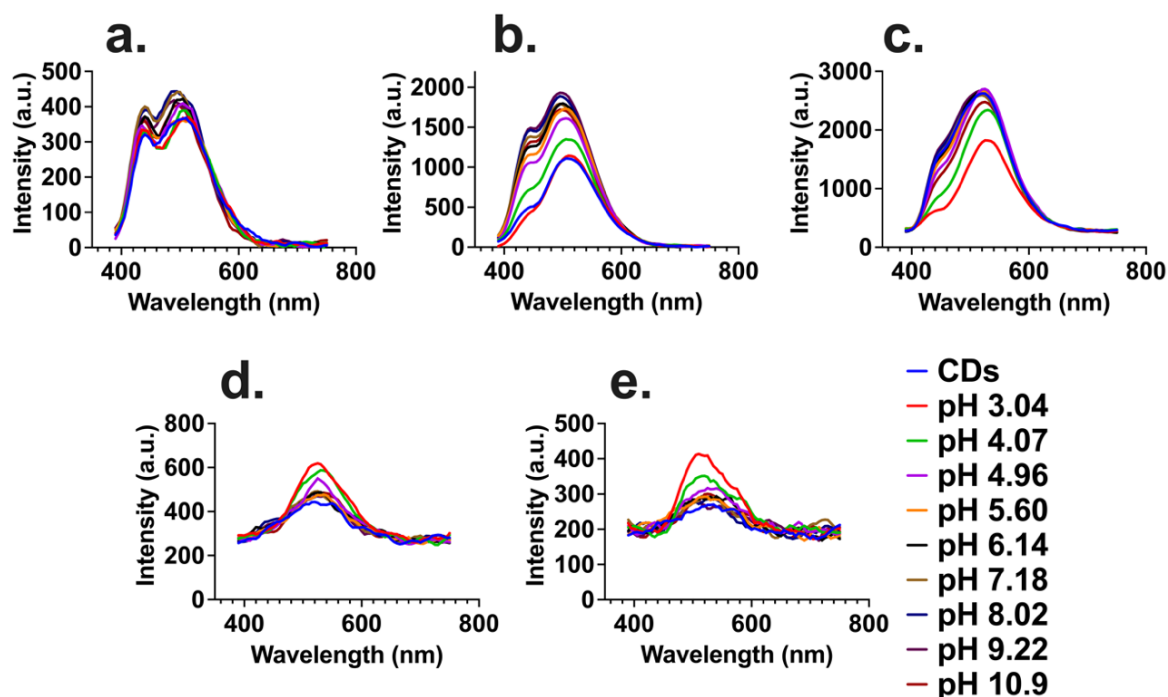


Figure 3.9 Fluorescence spectra of aqueous solutions of carbon dots (1 mg/mL) in different pH conditions. (a) CD100, (b) CD92, (c) CD69, (d) CD43, and (e) CD25.

The emission spectra presented in Figure 3.9 appear more structured and exhibit additional peaks in comparison to those in Figure 3.8. This variation arises from differences in the measurement instrumentation and spectral resolution. Figure 3.8 was recorded using a high sensitivity fluorometer with broader wavelength intervals, resulting in smoother emission profiles. In contrast, Figure 3.9 was measured using a microplate reader equipped with narrower wavelength steps and a different optical geometry, which can introduce additional spectral features or noise artefacts due to increased resolution and differences in detection configuration.

We found a clear transition from pH turn-on to pH turn-off response with the N-doping levels increased (Figure 3.10). The fluorescence spectra of the synthesised CDs with full pH range were presented in Figure 3.9 and the naked eye observation of CDs under different pH were presented in Figure S3.2. CD100 exhibits turn-on behaviour with increasing pH (Figure 3.10(a)). This thesis hypothesise that this pH response is dominated by surface $-OH$ groups. In

comparison, CD25, which is rich in amino groups, presents a turn-off fluorescence response with increasing pH (Figure 3.10(e)). The intermediate CDs, CD92, CD69 and CD43, show a turn-on pH response in acidic environments and a turn-off response in basic environments (Figure 3.10(b-d)). This can be explained by the competition between hydroxyl groups, which dominate in acidic environments, and amino groups, which take over control in basic environments.

Table 3.7 Photophysical properties of aqueous solutions of carbon dots (1 mg/mL) in different pH conditions.

Carbon dot	Items	CDs only	pH 3.04	pH 4.07	pH 4.96	pH 5.60	pH 6.14	pH 7.18	pH 8.02	pH 9.22	pH 10.9
CD100	Emission maximum / nm	500	495	520	505	510	490	495	485	480	505
	Excitation maximum / nm	355	360	360	365	375	370	390	395	395	400
	Stokes shift / nm	145	135	160	140	135	120	105	90	85	105
CD92	Emission maximum / nm	510	515	505	505	500	500	490	500	500	500
	Excitation maximum / nm	395	370	385	370	370	385	370	375	370	370
	Stokes shift / nm	115	145	120	135	130	115	120	125	130	130
CD69	Emission maximum / nm	525	530	525	515	525	525	515	525	510	525
	Excitation maximum / nm	370	375	370	370	370	370	365	370	365	370
	Stokes shift / nm	155	155	155	145	155	155	150	155	145	155
CD43	Emission maximum / nm	535	520	530	525	535	525	515	535	530	525
	Excitation maximum / nm	365	380	370	370	375	370	370	365	350	350
	Stokes shift / nm	170	140	160	155	160	155	145	170	180	175
CD25	Emission maximum / nm	525	520	520	520	555	560	505	540	540	510
	Excitation maximum / nm	370	380	370	370	370	370	375	350	370	365
	Stokes shift / nm	155	140	150	150	185	190	130	190	170	145

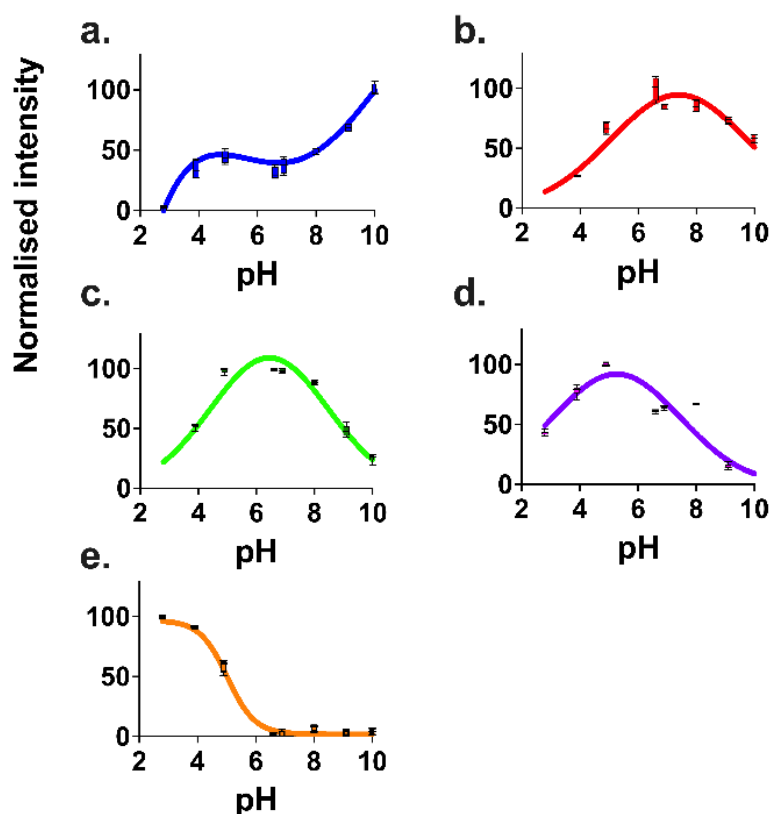


Figure 3.10 Synthesised CDs with variable pH response patterns (1 mg/mL CDs; $\lambda_{ex} = 370$ nm, λ_{em} integrated across 390–750 nm) in buffered Milli-Q water (25 mM buffers, pH 3–10): (a-e) pH responses of individual CDs: CD100, CD92, CD69, CD43, and CD25, respectively.

3.2.3 Carbon dots-based pH sensors' fluorescence recovery

To evaluate the applicability of carbon dot-based probes for pH monitoring, a fluorescence recovery test was conducted using CD100 and CD25. CD100, a pH turn-on sensor, exhibited fluorescence activation under basic conditions, while CD25, a pH turn-off sensor, responded to acidic environments. For each probe, a 10 mL solution (1 mg/mL) was prepared, and pH adjustments were made using 5 M HCl and NaOH. Fluorescence integral intensities were recorded during alternating pH cycles.

CD100 displayed clear pH-dependent fluorescence (as presented in Figure 3.11(a)), increasing in basic conditions with an initial integral intensity of 16, and decreasing to 8 under acidic conditions. Subsequent cycles showed partial fluorescence recovery in basic environments (13), followed by suppression in acidic conditions (7.5), confirming its reliable response as a basic pH-responsive probe. In contrast, CD25 exhibited stronger fluorescence suppression under acidic conditions (as illustrated in Figure 3.11(b)). Its fluorescence integral intensity dropped from 75 in basic conditions to 32 after acidification. During pH cycling, CD25 showed

significant variations, with integral intensities peaking at 244 in basic conditions and falling to 40 in acidic conditions, highlighting its sensitivity to pH decreases. These tests confirmed the distinct pH responsiveness of both probes. CD100 consistently responded to basic pH, making it suitable for applications involving pH increases, while CD25 demonstrated a dynamic response to acidic environments, making it highly effective for detecting pH decreases. The rapid and reversible pH sensing capabilities of these CDs underscore their potential for real-world applications where pH fluctuations are frequent.

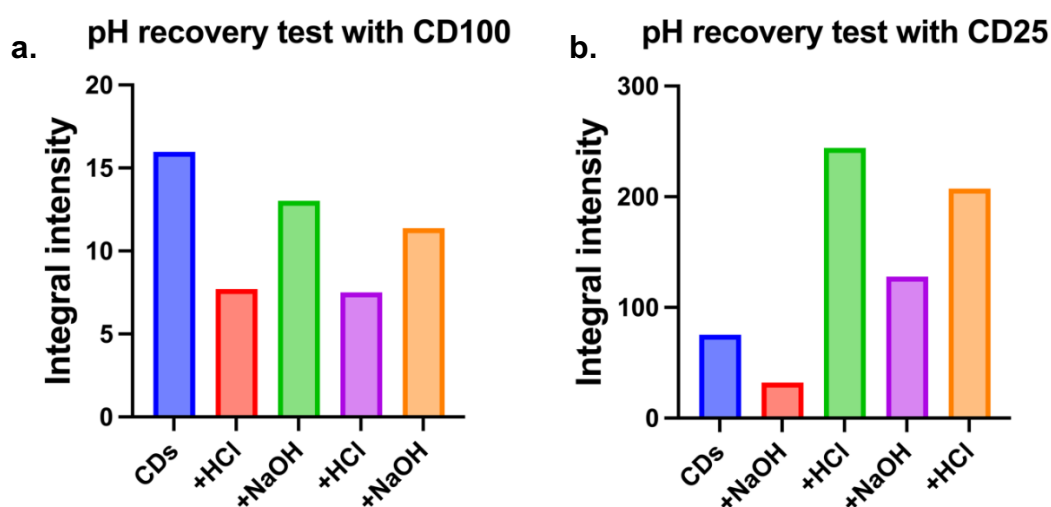


Figure 3.11 pH recovery test with a. CD100 and b. CD25 (10 mL 1 mg/mL CDs solution, pH adjusted by 5 M NaOH and HCl).

3.2.4 Mechanistic study of carbon dot pH response

From our observations of the pH sensitivity of fluorescence, this thesis can hypothesise the effects of hydroxyl and amino groups on the pH response of CDs. The presence of hydroxyl groups promotes the transformation of CDs into pH-sensitive 'turn-on' sensors with increasing pH, while amino groups drive CDs to function as pH-sensitive 'turn-off' sensors.

The oxygen content of the CD (determined both by precursor ratio and by XPS characterisation) is well-correlated with the pH value at which the highest fluorescence intensity, with R^2 of 0.9853, was observed (Figure 3. 3). This is consistent with our hypothesis that the surface functional groups influence the pH responsiveness of CDs.

To validate our hypothesis, the hydroxyl groups were masked on CD100 and CD69 using TBS protection, and the amino groups on CD69 and CD25 using Boc protection (Figure 3.12 (a-b)). This approach enabled us to investigate the specific impact of hydroxyl and amino groups on

the fluorescence and pH response of the CDs. For CD100, masking the –OH groups using TBS protection resulted in significant fluorescence quenching, very little pH sensitivity (Figure 3.12(c)). This result supports our hypothesis that the pH turn-on response of CD100 can largely be attributed to the presence of –OH groups.

For CD25, masking the –NH₂ groups led to a significant enhancement in fluorescence, but a dampening of the pH-responsive behaviour (Figure 3.12(d)). The fluorescence enhancement may be due to the fact that amino group themselves are fluorescence quenchers through surface passivation, and through non-radiative pathways for energy dissipation.^{46, 47} The turn-off performance of CD25 decreased by 55%, confirming our hypothesis that –NH₂ groups were responsible for the CDs' pH turn-off response and limited the CDs' fluorescence.

CD69 exhibited a pH response pattern characterised by fluorescence turn-on in the pH range of 3-7 and turn-off in the pH range of 7-10, as displayed in Figure 3.10(c). This behaviour was due to its surface functionalisation, which contained considerable amounts of both –OH and –NH₂ groups. For CD69, the original sample had a peak fluorescence value at pH 6.525, as determined from plot fitting results. After masking the –OH groups, the peak shifted to the acidic range at pH 4.816, and the fluorescence was clearly quenched compared to the original CD69. Conversely, after masking the –NH₂ groups, the peak shifted to the basic range at pH 8.163, and the fluorescence was enhanced.

These results confirmed that hydroxyl groups promoted the transformation of CDs into pH-sensitive 'turn-on' sensors, while amino groups drove CDs to function as pH-sensitive 'turn-off' sensors. The masking experiments validated our hypothesis and provided a deeper understanding of the mechanisms underlying the pH responsiveness of CDs.

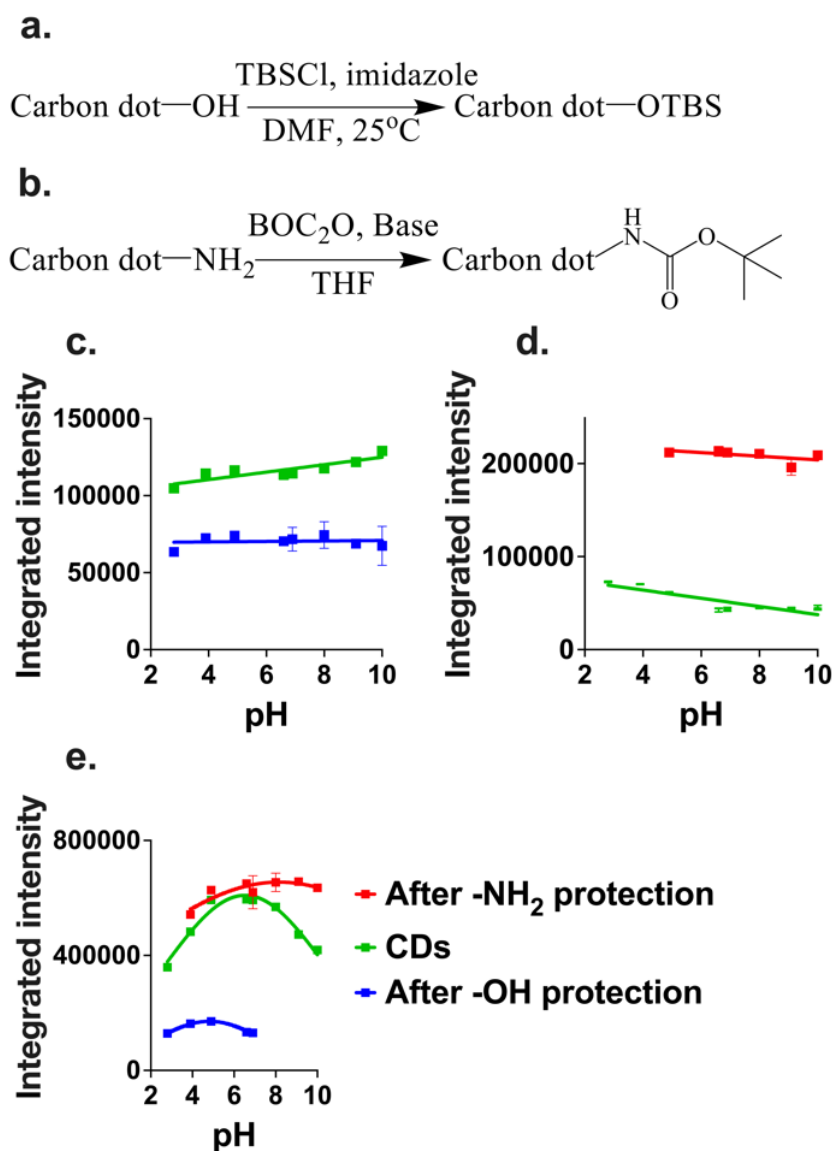


Figure 3.12 (a) Reaction scheme for CD TBS protection; (b) reaction scheme for CD BOC protection; (c-e) The pH response of CDs, including CDs without surface modification (green lines), CDs with $-\text{OH}$ groups masked (blue lines), and CDs with $-\text{NH}_2$ groups masked (red lines): (c) CD100, (d) CD25, and (e) CD69. 1 mg/mL CDs ($\lambda_{\text{ex}} = 370 \text{ nm}$, λ_{em} integrated across 390–750 nm) in buffered Milli-Q water (25 mM buffers, pH 3–10).

3.2.5 Application of CDs to a pH sensor array

With the original 5 CDs and the 4 masked CDs (CD100—OH protected; CD25—NH₂ protected; CD69—OH protected; and CD69—NH₂ protected) to hand, their potential was investigated by using as an array to distinguish and determine different pH values. The fluorescence emission spectra and calculated integrated emission intensity were collected for each CD at each pH value, in quadruplicate. These data were then subjected to principal component analysis (PCA) and linear discriminant analysis (LDA) using SPSS (IBM, 29.0.1.0 (171)).

PCA was utilised to analyse feature sets, such as those obtained by adding sensors to a standard configuration, and to understand how the different features compared and contributed to the overall sensor array performance. From Figure 3.13, PCA revealed that the combined features of the 9 CDs contributed significantly to the overall variance, indicating their collective importance in accurately sensing pH levels. This comprehensive approach leverages the strengths of each individual CD, leading to an enhanced and reliable pH sensing capability. The classification accuracy of LDA classification by using this 9 CDs array was confirmed to be 100% for both classification accuracy and cross-validation accuracy, as presented in Figure S3.3 and Table S3.1.

We had therefore demonstrated that these 9 CDs were able to correctly classify these pH values, over a greater than 7 pH range. However, the application of 9 elements to a sensor array is still cumbersome, and so we sought to reduce the number of sensor elements required to still give 100% correct classification. The process of reducing the number of array elements can be achieved by an iterative trial-and-error approach,⁴⁸ or by examination of the correlation statistics derived from PCA,⁴⁹ as we have previously described. However, both techniques are cumbersome, and do not enable investigation of all permutations of the full sensor set. We therefore sought to develop an algorithm that would let us rapidly and comprehensively perform array reduction.

To address this challenge, a MATLAB-based classifier was developed, implanted with LDA and support vector machine (SVM) to carry out array-based classification and array optimisation for CDs based pH array development. A 4-fold cross-validation setup was implemented for robust performance evaluation. This program automatically tested all potential combinations driven by the 9 CD sensor features: a total of 511 combinations. Both SVM and LDA classifiers were trained and evaluated for each combination, individually. The performance metrics, including accuracy, precision, recall, and F1 score, were calculated and averaged over the cross-validation folds (Table S3.3). The F1 score represented the mean of precision and recall, giving equal weight to both metrics. An F1 score ranges from 0 to 1, with 1 indicating a perfect model where all predictions are correct, and 0 representing the worst possible performance. In general, combinations containing our protected CDs were less accurate than those containing the original 5 CDs, consistent with the fact that masking groups led to less pH sensitivity.

The best-performing feature sets for both SVM and LDA were identified based on the highest accuracy. The SVM model used a linear kernel with a one-vs-one approach for multi-class classification, while the LDA model employed a pseudo-linear discriminant type. This program provided a comprehensive evaluation of the sensor features, identifying the most effective combinations for pH level classification, and ensuring reliable and robust performance through meticulous data normalisation and cross-validation. Just using CD43 alone was found to achieve 100% classification accuracies for both SVM and LDA. However, since we sought to use a sensor array with at least two elements for subsequent pH quantification, we then looked for two sensor combinations that contained CD43 (Table S3.4). 100% classification accuracies for both SVM and LDA classifiers were achieved by using combinations of CD43 and CD69, and CD43 and CD25. These results were verified by SPSS-based LDA, and 100% classification accuracies for both original classification and cross validation were achieved for both arrays. The LDA classification results are shown in Figure 3.14, Table S3.8-10 (CD43 and CD69), and Table S3.11-13(CD43 and CD25).

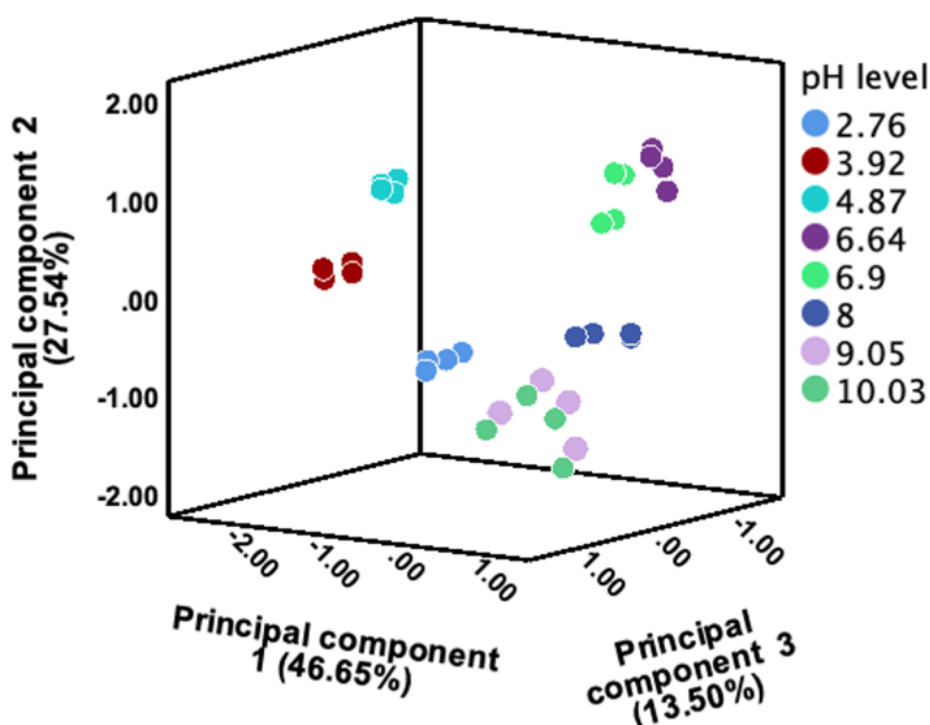


Figure 3.13 PCA plot by using 9 CDs to classify pH ranging from 3 to 10 (Each data point represents the mean value obtained from four independent replicates ($n = 4$), demonstrating the pH sensitivity of each carbon dot formulation under consistent measurement conditions.)

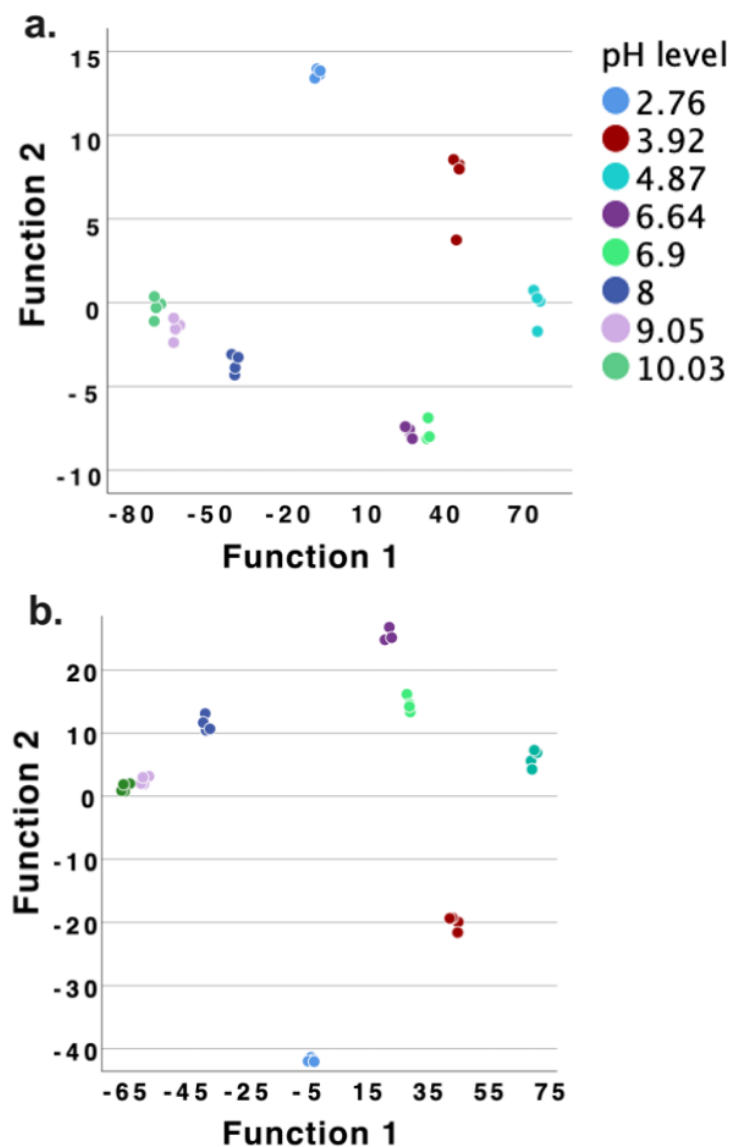


Figure 3.14 The LDA classification results by using two CDs based array (top: CD43 and CD69; bottom: CD43 and CD25) as pH sensor array to discriminate 8 different pH environments.

3.2.6 Gaussian process regression-based pH prediction.

With these two robust two-sensor arrays in hand, we sought to use a machine learning approach to accurately quantify pH. Gaussian process regression (GPR) is a non-parametric, Bayesian machine learning method used for regression problems, based on the Gaussian process — a collection of random variables where any finite subset follows a multivariate Gaussian distribution.⁵⁰ GPR is particularly effective for modelling complex and non-linear relationships between input features and target variables.⁵¹ In our study, we developed and rigorously evaluated a MATLAB-based GPR model for predicting pH values based on a comprehensive CDs based fluorescence responses, treated as features, described in the supplementary

information. Using our model, we could explore all possible combinations of CDs, but here we report the investigation of only the one-sensor CD43 and two-sensor CD43 and CD69, and CD43 and CD25, which were found to be most promising by LDA and SVM analysis. In the preliminary GPR model, we took out the pH 6.9 sample from the dataset and added it as an unknown to test the GPR. Our GPR model demonstrated superior performance, as evidenced by high coefficients of determination (R^2), low mean squared errors (MSE), and minimal mean absolute errors (MAE) across all sets of arrays (Table S3.14).

Of the three arrays tested, the array built using CD43 and CD69 achieved the best prediction for the “unknown” pH 6.9 sample. For the kernel parameters, the length scale (σ_l) was calculated to be 427.7, indicating a significant influence of input features on predictions. The signal variance (σ_f) was 0.316, representing the variation of function values from their mean. The standard deviation of predicted values ($n=4$) was 0.110, indicating consistent predictions close to the average value. This demonstrates that the GPR model can predict pH values close to those measured by a pH meter.

The R^2 was 1, indicating that the model explains approximately 100% of the variance in the data. The MSE was 0, suggesting a good model fit. The MAE was 0.009, indicating that the model's predictions were accurate.

To further validate the performance of the pH sensor array constructed using CD43 and CD69, GPR tests were conducted at pH 4.07 and 9.05, (as presented in Figure 3.15 and Table S19). At pH 4.07, the standard deviation of the predicted values ($n=4$) was 0.106, with an average predicted pH of 4.040 ± 0.106 . The prediction range was 3.934 to 4.145, and the R^2 was 0.987. MSE was 0.055, and the MAE was 0.165. For pH 9.05, the standard deviation of the predicted values was 0.0359, with an average predicted pH of 8.970 ± 0.0359 . The prediction range was 8.934 to 9.006, and the R^2 value was 0.996. The MSE was 0.019, and the MAE was 0.102. These results demonstrate that the pH sensor array provided accurate and precise pH predictions, with very high R^2 values and low error metrics for both pH levels tested in acidic and basic conditions.

Overall, the CDs-based pH sensor array performed well in predicting pH values (from acidic to alkaline) based on the fluorescence response of the three sensors using the GPR model. The high R^2 value, low MSE, and low MAE indicated a good fit and accurate predictions. The standard deviation of predicted values was relatively small, indicating

consistency in predictions. The model's predictions for the test pH value were reasonably close to the actual value, confirming its effectiveness.

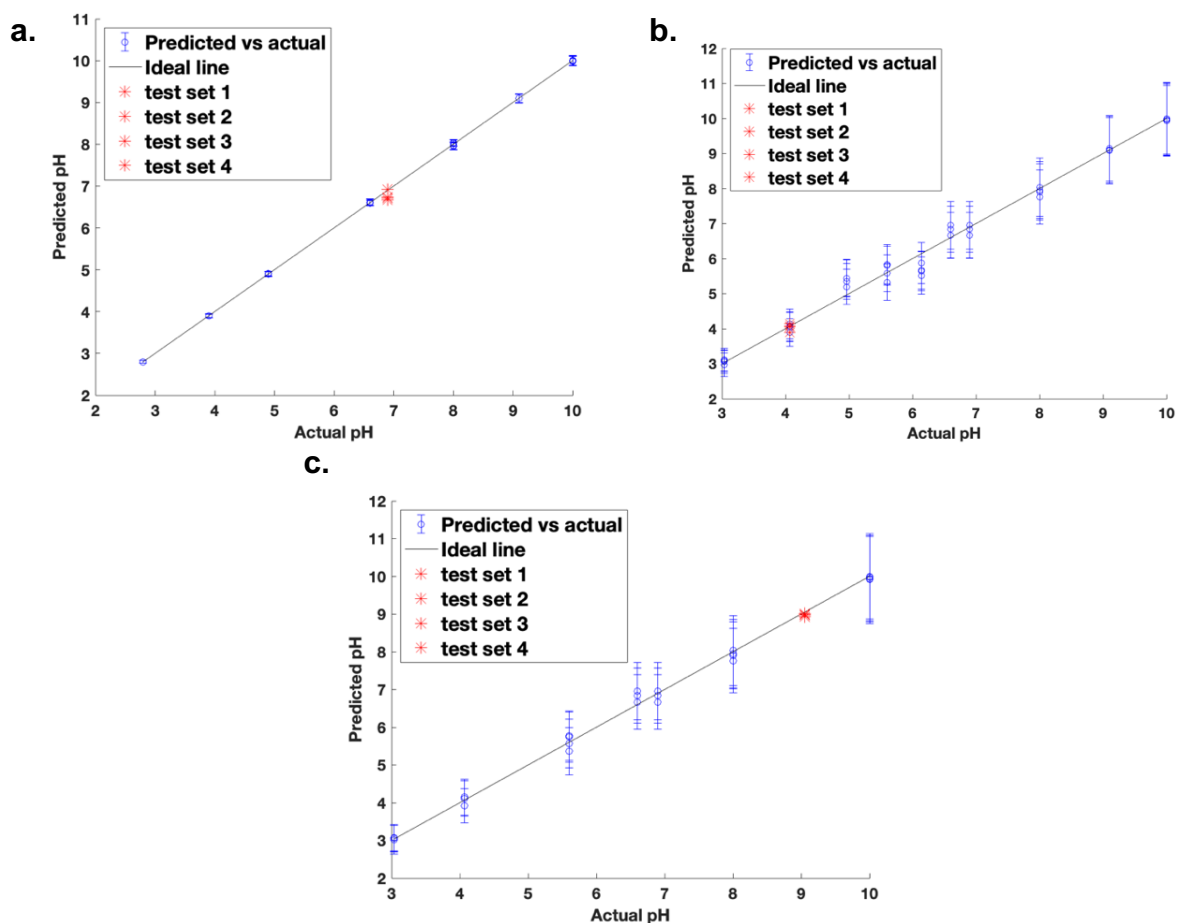


Figure 3.15 GPR prediction by using 2 CDs array built by CD43 and CD69 (from top to bottom the test pH values were 6.9, 4.07, and 9.5).

3.2.7 GPR-based pH prediction application with optimised CDs sensor array for milk's pH sensing

To further investigate the application of our pH sensor array for monitoring pH in food, we applied our optimised sensor array (CD69 and CD43) to determine the pH values of both fresh and expired A2 full-cream milk. The results were benchmarked against standard pH meter readings, where the fresh milk (tested 10 days before its best before date) exhibited a pH of 6.74, and the expired milk (tested 20 days after its best before date) recorded a pH of 5.20.

The performance of the GPR model, using CD69 and CD43 as features, demonstrated a high degree of accuracy in predicting the pH of fresh milk (Figure 3.16(a)). The model predicted an average pH value of 6.89 ± 0.18 , with a range of predictions between 6.70 and 7.07. This closely aligned with the actual pH of 6.74, reflecting the reliability of the sensing array in

accurately detecting minor pH variations in fresh milk. The model exhibited a high R^2 value of 0.971, indicating a strong correlation between the predicted and observed pH values. Furthermore, the MSE (0.084) and MAE (0.234) were relatively low, supporting the model's capacity to provide precise pH predictions.

For the expired milk (Figure 3.16(b)), the GPR model demonstrated strong predictive accuracy. The model predicted an average pH of 5.62 ± 0.48 , with a prediction range of 5.14 to 6.09, closely approximating the actual pH value of 5.20. The high R^2 (0.9772) reflected a strong correlation between predicted and observed values, with low error rates—an MSE of 0.066 and an MAE of 0.179—indicating precise prediction. The relatively small standard deviation (0.48) confirmed the model's robustness. These results suggest that the sensor array retained high accuracy even in expired milk, despite the potential influence of factors like microbial spoilage and chemical decomposition. The naked eye observation of pH sensor array applied for spoiled milk was presented in Figure S3.4.

Our CD-based sensor array exhibited considerable promise as a rapid and reliable method for assessing milk freshness. The high level of accuracy observed in milk pH predictions underscored its potential application in food quality monitoring systems, where real-time and non-invasive pH sensing is of significant commercial interest.

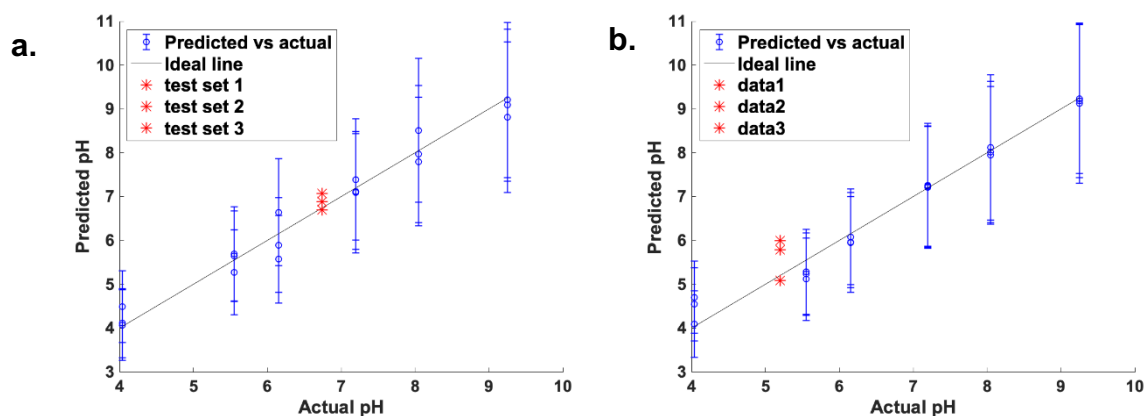


Figure 3.16 GPR prediction by using 2 CDs array built by CD43 and CD69 (left: fresh milk and right: spoiled milk).

3.2.8 Carbon dots-based pH sensors' stability at room temperature

To evaluate the fluorescence stability of CDs as pH sensors, CD solutions were prepared and stored in sealed transparent vials, left on a laboratory bench for three months at room temperature without sunlight protection. After this period, the CDs retained their pH responsiveness, indicating that they remained functional and stable despite environmental exposure. Although the overall fluorescence intensity decreased, the surface functional groups responsible for pH sensitivity, including -OH, -COOH, and -NH₂ groups, remained active and continued to mediate effective pH sensing.

The pH response of each CD was presented in Figure S3.5. CD100, which contains only -OH and -COOH groups, maintained a strong fluorescence response in basic conditions, functioning as a pH turn-on sensor. CD92, which also contains -OH and -COOH groups, with a small presence of -NH₂ groups, displayed a more balanced pH response across a wider range. The presence of amino groups enhanced CD92's sensitivity in slightly acidic conditions due to the protonation of -NH₂. CD25, rich in amino groups, exhibited high fluorescence at lower pH values, demonstrating its heightened sensitivity in acidic environments. Although CD25's fluorescence intensity was reduced after aging, it continued to show detectable pH-dependent fluorescence. Both CD69 and CD43, which initially acted as turn-on-then-turn-off sensors, transitioned to turn-off sensors after aging. The presence of -NH₂ groups played a crucial role in these CDs' pH response, particularly in acidic conditions, where protonation enhanced fluorescence.

After three months of ambient storage, CD69 and CD43 transitioned from turn-on-then-turn-off pH sensors to turn-off sensors, likely due to surface chemical changes caused by aging. Initially, their non-monotonic fluorescence behaviour arose from the combined effects of amino, hydroxyl, and carboxyl groups, with protonation of amino groups enhancing fluorescence at mid pH. Aging in unshielded, transparent vials likely led to partial oxidation or degradation of these surface groups, particularly amino functionalities, reducing their capacity for protonation-induced fluorescence enhancement.^{52, 53} Consequently, fluorescence steadily decreased with rising pH, simplifying their behaviour to a turn-off response. Although the sensors retained pH responsiveness, the altered surface chemistry and reduced signal intensity contributed to diminished sensing precision.

The GPR model results for the three-month-aged CD sensor array (CD69 and CD43) provided additional insights into the stability and predictive accuracy of the aged sensors. For the test at pH 7.2, the GPR model, using a squared exponential kernel, predicted an average pH of 7.721 ± 0.041 . Despite aging, the sensor array demonstrated a reasonable degree of accuracy in estimating pH, with an R^2 value of 0.641, indicating moderate predictive strength. However, the mean absolute error (MAE) of 1.211 and mean squared error (MSE) of 2.138 highlighted reduced precision compared to the fresh samples. The degradation of surface-active sites responsible for proton and hydroxide ion interactions likely contributed to this decrease in accuracy.

In conclusion, the carbon dots exhibited significant stability over three months of aging, underscoring their potential for long-term pH sensing applications. While fluorescence intensities diminished, the CDs' fundamental pH sensing capabilities remained intact, driven by their functional surface groups. CDs with higher amino group content continued to respond strongly in acidic environments, although the optimal fluorescence response shifted towards more acidic pH ranges as the sensors aged. Although the predictive accuracy of the GPR model decreased for the aged CDs compared to fresh samples, the results confirmed that the CDs retained sufficient pH responsiveness to remain functional as a sensing array. Their prolonged stability under environmental exposure further supports their suitability for long-term sensor applications.

3.3 Conclusions

The research presented herein successfully demonstrated the use of CDs synthesised from fructose and p-phenylenediamine for effective pH monitoring across a broad range. This chapter conducted a comprehensive spectroscopic analysis to assess changes in CDs' fluorescence response to pH variation. The results gleaned from this study offer deep insights into the roles hydroxyl group and amino group play in the pH sensitivity of CDs. We verified the functional group impacts by masking the functional groups respectively and established an exhaustive understanding of the pH-sensing capabilities of CDs.

This thesis was able to use this set of CDs to distinguish between pH values over a greater than 7 pH unit range, and in fact we have shown that just two carbon dots can be used to construct an array that gives 100% accuracy of classification, as well as a very high accuracy in quantification of pH. Employing Gaussian process regression and linear discriminant analysis,

a sophisticated analytical framework was developed that not only predicts pH levels with remarkable accuracy but also classifies them with high precision. Importantly, we have shown that these methods can be used to rapidly and efficiently scan a full set of array data to identify the most promising set with the best performances.

In this study, GPR was employed to construct a comprehensive multivariate regression model aimed at predicting pH levels in diverse test sensing environments. By analysing variations in the spectral features of the CDs' fluorescent emission in response to pH changes, GPR facilitated a nuanced understanding of the relationship between spectral attributes and pH levels, thereby enabling accurate and reliable pH estimation. The inherent flexibility of GPR, coupled with its ability to provide confidence intervals, significantly bolstered its applicability in sensor technology and analytical chemistry.

Concurrently, LDA was utilised as a powerful statistical tool for dimensionality reduction and classification within the pattern recognition domain.^{54, 55} By maximising the ratio of between-class variance to within-class variance, LDA ensured optimal separation among different classes.^{56, 57} In our CDs-based array, LDA harnessed the unique fluorescence-based pH responses of the CDs to create distinct fingerprints for varying pH levels. This enabled the effective reduction of data dimensionality while retaining critical distinguishing features, thereby streamlining the classification process.

This study contributes to the fields of nanotechnology and sensor development by providing detailed mechanistic insights into the fluorescence behaviour of CDs under different pH level. This study not only contributes to the fundamental understanding of CDs but also offers practical guidelines for the future design and optimisation of pH-responsive CDs, expanding their potential in various scientific and industrial applications. It also showcased the integration of nanomaterials with machine learning to create more sensitive, selective, and versatile sensing technologies. Our optimised two-CD sensor array was utilised for pH sensing in milk. During the sensor stability and fluorescence recovery tests, the CD sensors exhibited a dynamic fluorescent response to pH fluctuations, demonstrating their effectiveness in detecting pH variations. Future research should aim to refine the synthesis and functionalisation of CDs to further enhance their sensitivity and selectivity for specific ions or molecules, thereby expanding their utility in a wider range of applications such as environmental monitoring, industrial processes, and biomedical diagnostics.

3.4 Experimental methods

3.4.1 Materials and instrumentation

All chemical reagents and solvents employed in this study were of laboratory or analytical grade, acquired from Sigma-Aldrich, Merck, or Combi-Blocks. These chemicals were used as procured, without further purification. A2 full-cream milk was purchased from a local Woolworths store and was left at room temperature to allow it to spoil for the experiment.

X-ray photoelectron spectroscopy (XPS) measurements were collected using a Kratos Axis Nova spectrometer (Kratos Analytical, UK), equipped with a monochromatised aluminium X-ray source (Al K α , 1486.6 eV), operating at 10 mA and 15 kV (150 kW). Both survey and high-resolution spectra were recorded at detector pass energies of 160 and 20 eV, respectively. The obtained XPS data were processed and analysed using Thermo Advantage software (version 5.9902). Photophysical evaluations were conducted using a PerkinElmer EnSpire Multimode Plate Reader, with experiments carried out in 300 μ L, 96-well polypropylene microplates (Item No.: 655209, Greiner Bio-One), ensuring standardised and reproducible measurement conditions.

3.4.2 Preparation of carbon dots

A one-step hydrothermal/solvothermal synthesis route was used to prepare CDs using fructose and p-phenylenediamine (PPD) as precursors. The precursors were mixed in 50 mL of Milli-Q water, then processed by ultrasonication to achieve a homogeneous mixture. The solution was then transferred into a 100 mL Teflon-lined stainless-steel autoclave chamber with a 50% filling ratio and heated at 180 °C for 8 h. The initial purification was performed by passing the solution through a 0.22 μ m cellulose acetate Millipore filter (Millipore, USA) to remove large or agglomerated particles. The sample was then concentrated by rotary evaporation and dried using a freeze dryer. A dark brown powder was obtained, which was subsequently used to produce a 1 mg/mL CDs solution by adding Milli-Q water. The synthesis process was universal and applicable for all CDs.

To prepare CDs of different varieties with varying proportions of –OH and –NH₂ groups, the weight ratio of precursors and synthesis time were adjusted for each CDs species, as detailed in Table 3.8. This table illustrated the variations in precursor ratios and synthesis times used to

achieve the different functional group compositions in the CDs, which influenced their fluorescence and pH responsiveness.

Table 3.8 Summary of synthesis conditions for fructose and p-phenylenediamine-based CDs

CD	fructose / g	p-phenylenediamine / g	synthesis time / h	synthesis / °C	temperature
CD100	0.5	0	8	180	
CD92	0.43	0.07	8	180	
CD69	0.278	0.222	8	180	
CD43	0.147	0.353	8	180	
CD25	0.078	0.422	8	180	

3.4.3 pH buffer preparation

A series of standard pH buffers was prepared using different buffering agents to cover a range of pH values from 3 to 10: sodium citrate dihydrate (50 mM) for pH 3 – 5; potassium phosphate dibasic (50 mM) for pH 6 – 8; and sodium bicarbonate (50 mM) for pH 9-10. Each buffer solution was meticulously adjusted using either NaOH or HCl to achieve the desired pH level. The resultant pH values were accurately determined using a SevenCompact pH meter. The measured pH values for each buffer are detailed in Table 3.9.

Table 3.9 pH buffer preparation details

Expected pH	actual pH	chemical 1	chemical 2	buffer conc. / mM
3	2.76	sodium citrate dihydrate	citric acid	50
4	3.92	sodium citrate dihydrate	citric acid	50
5	4.87	sodium citrate dihydrate	citric acid	50
6	6.9	potassium phosphate dibasic	potassium phosphate monobasic	50
7	6.64	potassium phosphate dibasic	potassium phosphate monobasic	50
8	8	potassium phosphate dibasic	potassium phosphate monobasic	50
9	9.05	sodium bicarbonate	sodium carbonate (anhydrous)	50
10	10.03	sodium bicarbonate	sodium carbonate (anhydrous)	50

3.4.4 TBS protection of carbon dots

CDs (50 mg) were dissolved in N,N-dimethylformamide (DMF) (10 mL). Subsequently, imidazole (18.5 μ L) and tert-butyldimethylsilyl chloride (25 mg) were added to the solution at a controlled temperature of 0 °C. The reaction mixture was gradually allowed to attain room temperature and was stirred continuously for 24 h to ensure complete reaction. To terminate the reaction, an adequate amount of water was introduced to the mixture. The resulting mixture was then concentrated under reduced pressure, leading to the formation of a solid CDs product with hydroxyl groups masked.

The obtained solid was redissolved in Milli-Q water and transferred to a dialysis bag (molecular weight cutoff: 2 kDa) for purification purposes. The dialysis process was conducted over 24 h to remove any unreacted reagents and by-products.

3.4.5 Boc protection of carbon dots

For amino group protection, 50 mg of CDs were dissolved in N,N-dimethylformamide (10 mL), followed by the addition of di-tert-butyl dicarbonate (50 mg) and triethylamine (25 μ L). The reaction mixture was stirred at room temperature for 24 h. After the reaction, the mixture was concentrated under reduced pressure to obtain the solid product. The resulting solid was then purified using a similar dialysis procedure as described for hydroxyl group protection. The dialysis process, conducted over a span of 24 h with a molecular weight cutoff of 2 kDa, effectively removed any unreacted reagents and by-products.

3.4.6 Fluorescence spectroscopy

To prepare the CDs solution, the synthesised CDs were first dispersed in Milli-Q water then mixed with pH buffer, achieving a final concentration of 1 mg/mL of CDs solution and 25 mM of pH buffer (50% v/v). For fluorescence measurements, precisely 200 μ L of this CDs solution was pipetted into the wells of 96-well plates. Using an EnSpire Multimode Plate Reader (Revvity), the fluorescence spectra were then recorded. The settings for excitation and emission wavelengths were carefully selected based on the unique properties of the CDs under investigation. Emission spectra were diligently collected over a wavelength range of 400-700 nm. To ensure the reliability and accuracy of the measurements, each sample underwent triplicate testing. This rigorous approach not only confirmed the reproducibility of the results but also enhanced the overall precision of the fluorescence data obtained.

3.4.7 Statistical analysis of array data

The fluorescence data obtained from the CDs array were subjected to sophisticated statistical analyses. Specifically, Gaussian process regression was used to construct predictive models for estimating pH levels. The spectral data served as inputs to the GPR model, which provided predictions accompanied by confidence intervals to indicate the reliability of these predictions. Furthermore, linear discriminant analysis was employed to categorise the fluorescence responses into distinct pH levels. The LDA model was designed to maximise the separation between different pH classes, thereby enhancing the accuracy of classification.

GPR and LDA analyses were conducted using MATLAB (MathWorks, USA). Custom scripts were meticulously developed to handle the fluorescence data, train the models, and evaluate their performance comprehensively. To ensure the robustness and generalisability of the models, cross-validation techniques were employed to validate the results from both GPR and LDA.

3.4.8 Carbon dot sensors' fluorescence recovery test

CD100 and CD25 were synthesised and prepared as 10 mL solutions at a concentration of 1 mg/mL in Milli-Q water. For the fluorescence recovery test of CD100, an initial 200 μ L aliquot of the solution was loaded in quadruplicate into a 96-well fluorescent sensing plate to serve as the blank control (represented by the blue columns in Figure S3). To create an acidic environment and quench the fluorescence of CD100, 200 μ L of 5 M HCl was added to the solution, which was then mixed thoroughly using sonication. After mixing, four 200 μ L aliquots were collected and transferred into the sensing plate. Next, 400 μ L of 5 M NaOH was added to neutralise the acidic environment and shift the solution to a basic pH, allowing CD100's fluorescence to be activated ("turned on"). Four additional 200 μ L replicates were taken and loaded into the plate. The cycle was repeated by adding 400 μ L of 5 M HCl, followed by sonication, and four replicate samples were collected. Finally, 400 μ L of 5 M NaOH was added to return the solution to basic conditions, and four more replicates were transferred into the plate. Fluorescence measurements were conducted using a plate reader with an excitation wavelength of 370 nm, and emission spectra were recorded from 390 to 700 nm.

For the fluorescence recovery test of CD25, a similar procedure was followed, with the order of HCl and NaOH additions reversed. In this case, 200 μ L of 5 M NaOH was added first, followed by 400 μ L of 5 M HCl, then 400 μ L of NaOH, and finally 400 μ L of HCl. After each

addition, the solution was mixed thoroughly, and four 200 μL aliquots were collected. Fluorescence measurements were performed under the same conditions as for CD100.

3.4.9 Carbon dots-based pH sensor array for milk pH sensing

Fresh A2 full-cream milk was purchased from a local Woolworths store, and a spoiled sample was prepared by leaving the milk at room temperature. A carbon dots sensor array, consisting of CD43 and CD69, was utilised for sensing the pH of both fresh and spoiled milk samples. The pH values of the milk samples were measured using a pH meter as a reference. The synthesised CDs were first dispersed in Milli-Q water and then mixed with the milk samples and pH buffer. For the test group, Milli-Q water was used in place of the pH buffer. The final solution contained 1 mg/mL CDs, 25 mM pH buffer, and the milk sample, with each component contributing 33.33% (v/v) to a total volume of 200 μL per well. The procedure was consistent with the previously described fluorescence spectroscopy and statistical analysis of array data.

3.5 Reference

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4 Carbon dots-based metal ion sensor

Metal ion sensing has emerged as a crucial field of study, given the significant impact that metal ions, particularly heavy metals, can have on human health and the environment, as discussed in Chapter One. Heavy metals like lead, cadmium, and mercury, which are often discharged into natural water sources, can enter the food chain, leading to toxic effects on human health.¹⁻³ These metals have been linked to various health issues, including neurological disorders, kidney damage, and developmental problems. Consequently, accurate detection and monitoring of metal ion concentrations at trace levels are essential for ensuring public health and environmental safety. Advanced chemical analysis techniques play a pivotal role in monitoring these metals and understanding their interactions with biological systems.^{4, 5} These techniques not only help in identifying the presence of hazardous metal ions but also contribute to developing intervention strategies.

The biological significance of metal such as iron and copper further underscore the importance of metal ion detection. Iron is essential for various physiological processes, including oxygen transport and energy production,^{6, 7} but its excess can lead to cytotoxicity and oxidative stress.⁸⁻¹⁰ Copper, another vital trace element, is crucial for bone health,¹¹ immune function,¹² and neurological development,¹³ but elevated levels can cause toxicity and are associated with diseases like Alzheimer's.^{14, 15} Detecting and quantifying these metal ions in biological and environmental systems is therefore critical for understanding their roles in health and disease. Sensors capable of distinguishing between metal ions such as Cu and Fe, and with sensitivity to oxidation state, are particularly valuable in clinical diagnostics, environmental monitoring, and biological research, where precise measurements are needed for treatment and intervention.

However, current metal ion sensing methods face several limitations (as discussed in Section 1.4), including the high cost of instrumentation, lack of portability, limited detection sensitivity, and interference between analytes and other ions, making the detection of low concentrations particularly challenging. In this context, carbon dots have emerged as an effective and versatile class of fluorescent sensors for metal ion detection. CDs offer several advantages, including ease of synthesis, cost-effectiveness, and tuneable optical properties, making them ideal candidates for developing highly sensitive and selective metal ion sensors.¹⁶⁻¹⁹ Research has shown that CDs can detect a variety of metal ions, including Fe(III), Cu(II), Hg(II), and Pb(II),

with remarkable accuracy, even in complex samples such as environmental water or biological fluids.^{18, 20, 21}

This chapter will focus on the use of carbon dots as selective metal ion sensors, with particular emphasis on the detection of Fe(III) and Cu(II) ions. These metal ions are not only biologically significant but also serve as valuable models for exploring the broader capabilities of CDs in metal ion sensing. The chapter will begin by introducing selective fluorescence sensors for Fe(III) and Cu(II), followed by a detailed examination of their sensing selectivity. Initially, natural ingredients such as berries will be used to synthesise CDs, and their metal sensing properties will be evaluated. Inspired by the successful use of nature-derived precursors, the focus will then shift to chemical ingredients, with fructose being used as the primary carbon source for CD synthesis. Fructose-based CDs will be applied as selective sensors for Fe(III), while nitrogen-doped fructose-based CDs will be developed for the selective detection of Cu(II) ions.

4.1 Selective fluorescent sensor for Fe(III) and Cu(II) sensing

4.1.1 Selective fluorescent sensing

Fluorescent sensors have become a powerful tool in analytical chemistry due to their high sensitivity and versatility, which are derived from their ability to undergo distinct changes in fluorescence response upon interacting with analytes, as presented in Figure 4.1.²² These changes can manifest in different ways, such as "turn-on," "turn-off," or ratiometric responses, each providing a unique method for detecting and quantifying target metal ions. For instance, turn-on sensors experience an increase in fluorescence intensity upon binding with an analyte (Figure 4.1(c)), making them advantageous for their simplicity in detection. Turn-off sensors, on the other hand, decrease in fluorescence intensity when exposed to the target ion (Figure 4.1(b)). Ratiometric sensors, however, offer more robust measurements by using a change in the fluorescence emission at two different wavelengths, providing a built-in standard for enhanced accuracy.

The selectivity of fluorescent probes is critical, especially when applied to complex biological or environmental samples where multiple analytes coexist. Effective probes must target specific metal ions without interference from other substances, avoiding false positives or inaccurate readings (Figure 4.1(d-e)). Selectivity is particularly challenging when sensing metal ions with similar chemical properties, such as Zn(II) and Cd(II) or Cu(II) and Ni(II).^{23, 24}

While selective probes are typically preferred, there are instances where cross-reactive sensors (Figure 4.1(f)), which respond to multiple analytes, can also be useful in creating a broader sensing platform.

The working mechanism of selective fluorescent metal ion sensors relies on several key design strategies. These sensors are engineered to selectively bind metal ions using ligands or receptors tailored with specific functional groups or donor atoms that form stable complexes with the target ions.^{25, 26} Selectivity is further refined through the use of chelation effects,^{27, 28} steric hindrance,²⁹ and fluorophore selection,³⁰ ensuring that the sensor binds preferentially to the target ion over others. By integrating these strategies, fluorescent sensors achieve selective detection of metal ions like Fe(III), Cu(II), and others, making them invaluable tools in biological, environmental, and clinical applications. The development of these sensors continues to evolve, focusing on improving selectivity, sensitivity, and compatibility with real-world samples, ensuring their relevance in addressing key challenges in metal ion detection.

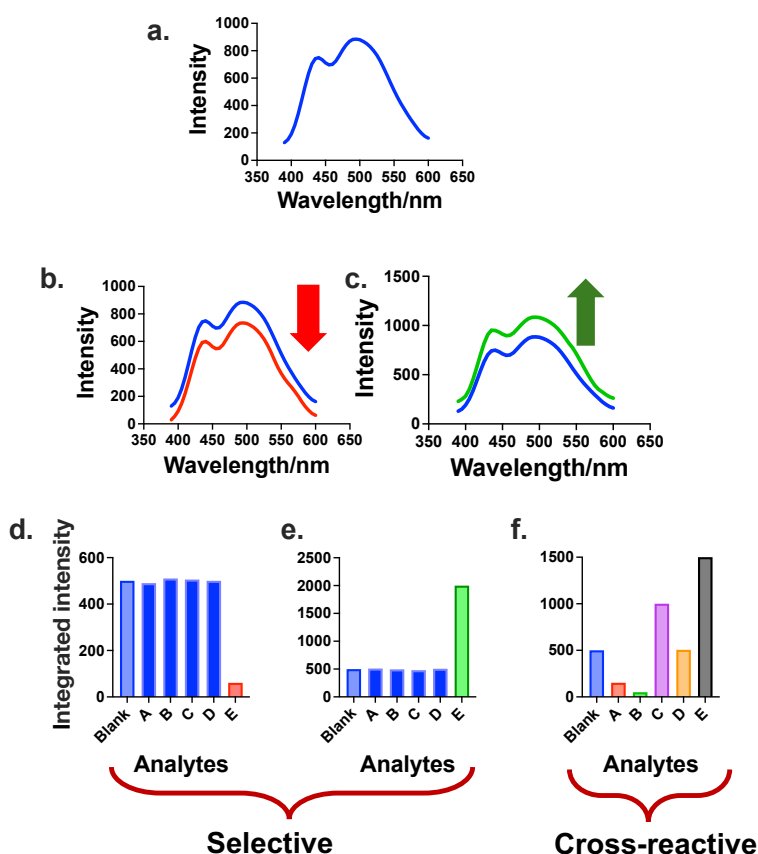


Figure 4.1 Types of fluorescence responses and fluorescent sensors. (a) emission spectrum of fluorescent sensors before interaction with analytes (blank); (b-c) fluorescence response after interaction with analytes: (b) turn-off response, (c) turn-on response; (d-f) representative responses

of fluorescent sensors: (d) selective turn-off sensor, (e) selective turn-on sensor, (f) cross-reactive sensor.

4.1.2 Sensing of Fe(III)

Iron in its ferric state (Fe(III)) exhibits unique chemical properties that make it a key element in coordination chemistry and material science.³¹ Fe(III) forms stable complexes with various ligands, particularly nitrogen-containing ligands and hydroxypyridinone-based chelating agents, which display strong binding affinity and stability.^{32, 33} These properties are crucial in designing Fe(III)-selective sensors. The following sections outline general design principles for different classes of Fe(III) sensors, highlighting key examples.

4.1.2.1 Turn-off sensors for Fe(III)

Turn-off fluorescent sensors are widely used for Fe(III) detection due to Fe(III)'s tendency to quench fluorescence (listed in Table 4.1). In these systems, fluorescence intensity decreases upon Fe(III) binding, providing a simple yet effective method for detection. For example, pyrimidine- and imidazole-based sensors exhibit fluorescence quenching when coordinated with Fe(III), making them useful in bioimaging and environmental monitoring applications.³⁴⁻³⁷ A pyrimidine Schiff-base probe has been successfully used to image Fe(III) in human prostate carcinoma cells, shedding light on Fe(III)'s involvement in cancer biology.³⁴

4.1.2.2 Turn-on sensors for Fe(III)

In contrast to turn-off sensors, turn-on fluorescent sensors exhibit enhanced fluorescence upon Fe(III) binding. These sensors often involve inhibition of photoinduced electron transfer (PET),^{38, 39} which results in a significant increase in fluorescence intensity. For instance, Chen *et al.* reported a turn-on chemodosimeter-based probe for Fe(III) detection that operates through PET suppression, making it ideal for bioimaging applications.⁴⁰ Turn-on sensors offer the advantage of larger fold changes in fluorescence, which enhances sensitivity, especially in trace-level Fe(III) detection. This approach allows for a clearer signal, particularly useful in aqueous and biological environments. Researchers have reported a group of chemosensors probes that rely on the hydrolytic cleavage of imine linkages to generate a turn-on fluorescence response in the presence of Fe(III).⁴¹⁻⁴³ These probes are particularly useful in one-time-use detection scenarios, where continuous monitoring is unnecessary. Their ability to detect Fe(III) in live cells demonstrates their practical applications in biological systems, where sensitivity and specificity are essential.

4.1.2.3 Ratiometric sensors for Fe(III)

Ratiometric sensors represent another important class of Fe(III) detection tools. These sensors monitor fluorescence intensity at two different wavelengths, creating an internal standard that improves measurement accuracy and reliability. By comparing the ratio of intensities at two wavelengths, ratiometric sensors can reduce the influence of environmental factors, leading to more precise quantification of Fe(III), even in complex matrices. For example, Wang *et al.* demonstrated a 2-pyridylthiazole-based probe with ratiometric turn-on responses, offering high selectivity for Fe(III) in aqueous systems.⁴⁴ Similarly, Kuwar *et al.* introduced a Schiff-base receptor that provided a dual-response mechanism, featuring a ratiometric signal for Fe(III) and a turn-off response for Cu(II), thereby enhancing the sensor's ability to distinguish between metal ions in more complex environments.⁴⁵

4.1.2.4 Advanced detection mechanisms and sensors

Advanced Fe(III)-selective sensors have also been developed using mechanisms such as Förster resonance energy transfer and chelation-enhanced fluorescence.⁴⁶⁻⁴⁸ These methods allow for higher sensitivity, enabling the detection of Fe(III) at ultra-low concentrations. The strategic inclusion of donor atoms such as nitrogen and oxygen in the sensor's ligand design enhances selectivity, forming stable complexes with Fe(III).^{49, 50} This approach has been effectively applied in sensors based on BODIPY, which demonstrate high specificity and efficiency in detecting Fe(III).^{51, 52}

Additionally, metal-organic frameworks (MOFs) have been integrated into sensor designs due to their porous structures and high affinity for metal ions. For example, Eu(III)-based MOFs can selectively detect Fe(III) via cation-exchange mechanisms, acting as turn-off luminescent sensors with high sensitivity.⁵³ Flow-through fluorescent sensors, which offer real-time monitoring of Fe(III) and total inorganic iron, provide another advanced approach to Fe(III) detection, particularly beneficial for environmental applications like water quality monitoring.⁵⁴

The development of Fe(III)-selective fluorescent sensors has yielded a broad spectrum of detection tools, each with unique strengths. From turn-off and turn-on mechanisms to ratiometric and chemodosimeter-based designs, these sensors are applicable across a range of fields, including environmental monitoring, industrial processes, and biological systems. As

sensor technologies continue to evolve, combining novel signalling mechanisms with advanced materials promises even greater precision and applicability for Fe(III) detection.

Table 4.1 Fe(III) selective fluorescent sensors

Ref	Fluorophore	λ_{ex} / nm	Fluorescence Response	Solvent	LOD	Application
55	Anthracene.	350	Turn-off	DMSO-H ₂ O (1:1 v/v)	5.09 μM	The sensor is used for detecting Fe(III) ions in water, identifying benzaldehyde as a toxic pollutant, and detecting antibiotics (particularly those with nitro groups such as Nitrofurazone, Metronidazole, and Furazolidone) in environmental samples.
56	5-Nitro isophthalic acid derivative	340	Turn-off	DMSO-MeOH (1:9 v/v)	9.18 μM	A simple, portable method for detecting and differentiating Fe(III) and Fe(II) in real-life samples, suitable for field applications in environmental and industrial monitoring.
57	Carbon quantum dots (CQDs).	350	Turn-off	Water	Fe ²⁺ : 6.5 μM ; Fe ³⁺ : 2.5 μM .	The CQDs serve as a selective and sensitive fluorescent probe for detecting Fe ²⁺ and Fe ³⁺ ions in water, which can be applied for water quality monitoring and medical diagnostics.
58	Flavonol derivative (HPCB).	365	Turn-off	DMSO/H ₂ O (1:9 v/v)	0.265 μM	Detection of Fe(III) in real water samples. EC-HPCB-loaded filter paper for Fe(III) detection by fluorescence quenching, allowing detection by the naked eye.
59	Triphenylamine (TPA).	300	Turn-off	H ₂ O /THF (6:4 v/v).	0.14 ppm	Detection of Fe(III) in environmental and biological samples. Sequential detection of deferasirox (DFX), an oral iron chelator, in real samples (such as water samples).
60	Zn-based luminescent metal-organic framework (MOF).	363	Turn-off	Water	1.05 μM	This MOF sensor can be used for the detection of Fe(III) ions and trinitrophenol (TNP) in environmental and water samples. It has good recyclability and anti-interference properties, making it a practical sensor for real-world applications.
61	Redox-active macromolecular ratiometric sensor (MRS3).	360	Turn-off	DMSO	0.486 ppb	The sensor is applicable for detecting Fe(III), Cu(II), and Hg(II) ions in environmental samples, with potential uses in water quality monitoring and environmental protection due to its high sensitivity and selectivity. It also provides visible colorimetric changes, allowing for naked-eye detection.
62	Carbazole-based fluorescent probe	375	Turn-off	DMSO	Fe(III): 1.38 nM. Fe(II): 98.3 nM.	The sensor can be applied for detecting Fe(II) and Fe(III) ions in environmental water samples, with successful detection in real-world water samples such as tap, lake, and river water.
63	Coumarin derivative (AIE-active).	325	Turn-on	DMSO/H ₂ O (1:9, v/v, pH = 7.2)	0.488 μM	The sensor can be applied for detecting Fe(III) in water samples, with potential use as a test kit for environmental monitoring. It also showed reversible behaviour in logic gate applications with EDTA, enhancing its practical utility.

Ref	Fluorophore	λ_{ex} / nm	Fluorescence Response	Solvent	LOD	Application
64	Rhodamine B.	490	Turn-on	Water and acetonitrile (1:1 v/v)	N/A	pH-sensitive fluorescent biosensor for bioimaging in living cells, particularly in acidic environments like lysosomes. It detects Fe(III) and Cu(II) ions, with Cu(II) detection leading to near-infrared fluorescence, useful for deep tissue imaging.
65	Naphthyl (donor) and Rhodamine (acceptor).	320	Turn-on	EtOH/H ₂ O (1:1 v/v)	83 nM	The sensor can be applied for Fe(III) detection in environmental and biological systems due to its high selectivity and sensitivity.
66	BODIPY-based compounds.	378 (QBPH and PBPH); 353 (QBP and PBP)	Ratiometric	DMF-buffer mixed solution (1:1, v/v)	2.59 - 6.27 μ M	The probes are applied for the detection of Fe(III) in living cells, particularly for near-infrared imaging and sensing of Fe(III) in A549 cells (lung carcinoma cells). The probes exhibited good sensitivity and selectivity for Fe(III) ions and demonstrated successful application in bioimaging.
67	N, S-doped carbon dots (N/S-CDs).	344	Ratiometric	Water	83 nM	This ratiometric fluorescent probe is used for the selective detection of Fe(III) ions in environmental water samples, offering high precision with the coupling of fluorescence and SOS.

4.1.3 Sensors for Cu(II)

Copper in its divalent state, Cu(II), exhibits unique chemical properties that make it both an essential trace element and a versatile target for sensing applications.^{68, 69} Cu(II) plays critical roles in biological systems, including participation in enzymatic catalysis, electron transport, and redox reactions.⁷⁰ However, elevated levels of Cu(II) can be toxic to living organisms, catalysing the formation of reactive oxygen species (ROS) through Fenton-type reactions, which can lead to oxidative stress and cellular damage.⁷¹ As a borderline Lewis acid, Cu(II) prefers to bind with hard ligands, such as nitrogen and oxygen, a characteristic that makes it an ideal target for selective detection by fluorescent chemosensors.⁷² Building on these chemical properties, researchers have developed a wide range of fluorescent chemosensors to detect Cu(II) ions with high sensitivity and selectivity (listed in Table 4.2). General design principles and specific examples of each class of sensor are discussed below.

4.1.3.1 Turn-off sensors for Cu(II)

Cu(II) exhibits the heavy atom effect, which enhances spin-orbit coupling and leads to fluorescence quenching.^{73, 74} This property, coupled with its redox activity, makes Cu(II) an

ideal target for developing turn-off fluorescent sensors, which have proven highly effective in both environmental and biological monitoring applications.

Turn-off fluorescent sensors, such as those developed by Chen *et al.*, exhibit selective quenching of fluorescence in the presence of Cu(II) ions. These sensors produce distinct visual colour changes, allowing for effective detection of Cu(II) in biological systems, including HeLa cells.⁷⁵ Similarly, He *et al.* reported a naphthalimide-based sensor with a remarkable 30-fold quenching response and a detection limit as low as 64 ppb. This sensor was successfully applied in both solution and cellular media, including MCF-7 cells, without observable toxicity.⁷⁶ These turn-off sensors are particularly valuable for detecting Cu(II) in real-world samples such as biological fluids and environmental water, where high sensitivity is required to identify low concentrations of Cu(II). Zheng *et al.* synthesized BODIPY-based chemosensors demonstrated strong fluorescence quenching in the presence of Cu(II), making them excellent tools for environmental monitoring.⁷⁷

4.1.3.2 Turn-on sensors for Cu(II)

In addition to turn-off sensors, turn-on fluorescent sensors have been developed to detect Cu(II) through mechanisms such as Cu(II)-promoted hydrolysis.⁷⁸ One example is the semicarbazide-based fluorescent sensor introduced by Hu and his research group, which undergoes Cu(II)-induced hydrolysis, leading to a turn-on fluorescence response.⁷⁹ Turn-on sensors generally function by disrupting non-radiative decay pathways, which results in fluorescence enhancement. For instance, Shi *et al.* reported indoleamine-based chemosensors exhibit high binding affinities for Cu(II), resulting in a marked increase in fluorescence upon interaction with the ion.⁸⁰ These sensors are highly advantageous for real-time monitoring of Cu(II) concentrations, particularly in biological samples where turn-on sensors are especially valuable, as they enable the detection of small fluctuations in Cu(II) concentration, facilitating the monitoring of dynamic processes in living systems with enhanced accuracy and sensitivity.

4.1.3.3 Ratiometric sensors for Cu(II)

Ratiometric fluorescent sensors provide a highly effective approach for Cu(II) detection by monitoring fluorescence intensity changes at two distinct wavelengths, thus offering an internal standard that reduces the influence of environmental variables. This dual-wavelength method significantly improves the accuracy and reliability of measurements. For example, Yang *et al.* developed BMPUN, a naphthalene derivative with two urea groups, as a ratiometric fluorescent

sensor for Cu(II).⁸¹ Additionally, a novel hemicyanine-based ratiometric fluorescent probe was designed for the selective and sensitive detection of Cu(II) in fully aqueous solutions.⁸² This probe exhibits a large red shift (55 nm) in emission and up to a 22-fold change in emission intensity ratio upon binding to Cu(II), driven by a Cu(II)-triggered hydrolysis reaction of the picolinate moiety. These probes demonstrate high sensitivity and selectivity for Cu(II) over other metal ions. Moreover, Yu *et al.* developed another ratiometric sensor incorporating rhodamine B and naphthalimide units linked via a CN bond, which detects Cu(II) through an intramolecular FRET mechanism with a detection limit of 0.25 μM .⁸³ This sensor has proven successful for real-time monitoring of Cu(II) concentrations in cellular media, emphasizing its utility in biological research, where precision and accuracy are paramount. By utilizing the internal standard provided by dual-wavelength measurements, these sensors generate more consistent and reproducible data, enhancing their applicability across various environmental and biological contexts.

Recent advancements in Cu(II) sensors include the development of multifunctional probes capable of detecting multiple analytes. Huang *et al.* designed a novel 4-amino-1,8-naphthalimide-based fluorescence bifunctional sensor consisting of the morpholine moiety as a receptor site for Cu(II) binding.⁸⁴ Their sensor exhibited an efficient fluorescence turn-on sensing response for the Hg(II) ion and a fluorescence turn-off response for Cu(II) in aqueous solution. Similarly, Mu *et al.* reported a novel two-channel fluorescence sensor is a dual-channel fluorescence sensor that can selectively detect both Cu(II) and Hg(II), with visual colour changes and a low detection limit of 3×10^{-7} M for Cu(II).⁸⁵ These multifunctional sensors represent an innovative approach to metal ion detection, enabling simultaneous monitoring of multiple ions in complex environments.

Table 4.2 Cu(II) selective fluorescent sensors

Ref	Fluorophore	λ_{ex} / nm	Fluorescence Response	Solvent	LOD	Application
86	Quinoline-based compound.	410	Turn-off	DMSO/HEPES buffer (20 mM, pH 7.22, 9:1 v/v)	8.68 μM	This sensor can be applied for the detection of Cu(II) in biological and environmental samples, with further application in detecting sulfide ions (S^{2-}) by fluorescence recovery. It can also be used as a reversible sensor for Cu^{2+} and S^{2-} detection with colorimetric changes visible to the naked eye.

Ref	Fluorophore	λ_{ex} / nm	Fluorescence Response	Solvent	LOD	Application
87	Aminonaphthol.	320	Turn-off	Water	N/A	The sensor is designed for the detection of Cu(II) ions in aqueous solutions, with potential applications in environmental monitoring and analysis due to its high selectivity for copper ions in a broad pH range.
88	BODIPY	550	Turn-off	HEPES–DMSO (9:1, v/v) (20 mM HEPES, buffer pH 7)	N/A	The sensor was successfully used for imaging intracellular Cu(II) in human liver cancer (BEL 7402) cells, demonstrating its potential for biological applications in detecting copper ions within living cells.
89	Benzimidazole.	243	Turn-off	HEPES-buffered solution (100% aqueous, 20 mM, pH 7.4)	2.77 μ M	The sensor was successfully applied for detecting Cu(II) and S^{2-} in biological systems, including imaging in human esophageal cancer cells (Ec-9706). It is highly selective for Cu(II) in environmental and physiological systems and can also be used as a recyclable sensor for S^{2-} .
90	Fluorescein dye (attached to single-stranded DNA), with FRET transfer to ATTO 590 dye.	470	Turn-off	MOPS buffer (10 mM, pH 7) with NaCl (50 mM)	20 nM	The sensor is designed for selective detection of Cu(II) in biological samples with a tuneable emission wavelength (using different DNA-linked acceptor dyes), making it suitable for applications in biological systems where autofluorescence below 550 nm is an issue. It can be used for monitoring Cu^{2+} levels in biological environments, such as blood serum and brain tissues related to diseases like Alzheimer's.
91	1,8-naphthalimide moiety.	444	Turn-off	Tris-HCl buffer (pH = 7.2) in DMF (1:1, v/v).	0.32 μ M	The sensor was successfully applied for detecting Cu(II) in water samples, including tap water and Yellow River water, showing good recovery rates, making it suitable for environmental monitoring.

Ref	Fluorophore	λ_{ex} / nm	Fluorescence Response	Solvent	LOD	Application
92	Benzimidazole moiety.	256	Turn-off	Water	N/A	The fluorescent sensor is used for detecting transition metal ions, especially for bioimaging and analytical chemistry. The mechanisms for Zn(II), Cd(II), and Cu(II) were elucidated, with potential applications in environmental and biological sample analysis, where detecting these metal ions is critical for understanding their roles in health and disease.
93	BINOL (1,1'-Bi-2-naphthol).	291	Turn-off	HEPES buffer, pH 7.4	4 μM	The sensor was used for detecting Cu^{2+} and sulfide ions (S^{2-}) in water, showing potential for environmental monitoring, especially for water pollution detection. The dual-response mechanism allows for selective detection of both Cu^{2+} and sulfide with high sensitivity.
94	Benzimidazo[2,1-a]benz[de]isoquinoline-7-one.	370	Turn-on	Acetonitrile–water (9:1, HEPES buffer, pH 7.4)	13 nM	The sensor can be applied for detecting Cu(II) and Fe(III) ions in environmental samples, with high selectivity and sensitivity. It was successfully used for the quantitative detection of Cu(II) and Fe(III) in tap water samples.
95	Coumarin.	460	Turn-on	80% aqueous MeCN (acetonitrile) in phosphate buffer solution (pH = 7.2)	N/A	The sensor was applied for Cu(II) detection in living systems, including LLC-MK2 cells (kidney cells from a monkey) and in vivo imaging in ICR mice. It was shown to selectively detect Cu(II) accumulation in liver and kidney tissues, demonstrating potential applications in biological imaging and environmental monitoring.
96	Rhodamine-6G derivative.	480	Turn-on	HEPES buffer (20 mM, pH 7.0) with 50% water/ CH_3CN (1:1, v/v)	Below 2 ppb	The sensor was successfully applied for imaging Cu(II) in living cells, particularly in SPC-A-1 (lung cancer) cells, demonstrating its potential

Ref	Fluorophore	λ_{ex} / nm	Fluorescence Response	Solvent	LOD	Application
						for use in biological and environmental monitoring.
97	Naphthalimide-based fluorophore.	451	Ratiometric	Ethanol-water solution (40:60, v/v) with 50 mM HEPES buffer (pH 7.2)	10 nM	This sensor was designed for highly selective ratiometric detection of Cu(II) in aqueous solutions, making it useful for environmental monitoring and biological applications. The visible emission shift also allows for colorimetric detection of Cu(II).
81	Naphthalene-based derivative (BMPUN).	310	Ratiometric	CH ₃ CN/DMSO (9:1 v/v)	N/A	The sensor can be applied for detecting Cu(II) ions with high selectivity in environmental and biological samples, showing potential utility in environmental monitoring and chemical analysis.

4.1.4 Overview

This section introduced the various types of fluorescent sensors, including turn-off, turn-on, and ratiometric sensors, each utilising distinct mechanisms for detecting metal ions. The discussion then shifted to the selective fluorescent sensors for Fe(III) and Cu(II), emphasizing their unique chemical properties and the range of approaches used in sensor design. These sensors have proven to be highly effective in both biological and environmental applications, offering high sensitivity, selectivity, and practical utility for detecting and quantifying Fe(III) and Cu(II).

As the focus moves towards carbon dots-based metal ion fluorescent sensors in the following section, there is significant potential for further advancements in Fe(III) and Cu(II) sensing. Carbon dots, celebrated for their exceptional optical properties, tuneable photoluminescence, and ease of surface modification, present a promising platform for developing highly sensitive and selective sensors. Doping carbon dots with elements such as nitrogen and sulfur enhances their performance in detecting Fe(III) and Cu(II) ions, especially in complex environments like biological systems. These innovations in CDs-based sensors are expected to offer more efficient, reliable, and practical detection methods for Fe(III) and Cu(II) ions, with wide-reaching applications in environmental monitoring, medical diagnostics, and industrial processes.

4.2 Biomass-based carbon dots for metal ions sensing

Carbon dots are a novel carbon-based zero-dimensional nanomaterial (introduced in Chapter 2 2.1). CDs derived from natural ingredients, or biomass-based sources, represent an exciting and eco-friendly approach to synthesising nanomaterials with broad applications.⁹⁸⁻¹⁰⁰ Unlike conventional methods that often rely on non-renewable and potentially toxic chemical sources, biomass-based carbon dots offer a sustainable and environmentally conscious alternative.

Numerous natural materials have been successfully utilised as carbon sources for the synthesis of green CDs, including apple,^{101, 102} orange,^{103, 104} bamboo leaf,^{105, 106} garlic,^{107, 108} and tea leave.^{109, 110} These biomass-derived carbon dots (B-CDs) retain the excellent optical properties of traditional CDs, such as tuneable fluorescence, high photostability, and good water solubility, while offering enhanced biocompatibility and lower toxicity.¹¹¹ The process of converting low-value biomass waste into high-value functional materials not only contributes to waste reduction but also provides a cost-effective solution for large-scale production.

The synthesis of biomass-based carbon dots typically employs methods such as hydrothermal treatment, microwave-assisted treatment, or electrochemical processes (covered in Chapter 2 2.1.1). These techniques enable the straightforward and scalable production of CDs, often with minimal need for complex post-synthesis modifications. Due to their environmentally friendly production and ease of surface functionalisation, biomass-based CDs have gained significant attention for applications in biological imaging, sensing, catalysis, and drug delivery.¹¹²⁻¹¹⁴ Among these applications, biomass-derived CDs demonstrate exceptional potential for use in highly sensitive and selective fluorescent sensors for metal ion detection, particularly in both environmental and biological systems. The types of biomass-based CDs employed for metal ion sensing are outlined in Table 4.3. Inspired by the successful application of natural biomass sources, this chapter focuses on synthesizing carbon dots using berries as the carbon source and evaluating their use in selective metal ion sensing. Specifically, carbon dots were synthesised via a hydrothermal reaction. Additionally, nitrogen-doped biomass-based CDs (N-BCDs) were prepared to assess how doping affects the properties of CDs, as discussed in the previous chapter. The following sections will explore the potential of these CDs as fluorescent sensors for Fe(III) and Cu(II), further advancing their application in metal ion sensing.

Table 4.3 Biomass-based carbon dots' metal ions sensing

NO.	Source	Method	Excitation wavelength/nm	Sensing application	Ref
1	Broccoli	Hydrothermal	355	Ag(I)	115
2	Peach gum	Hydrothermal	370	Au(III)	116
3	Bamboo leaves	Hydrothermal	365	Cu(II)	117
4	Pipe tobacco	Hydrothermal	360	Cu(II)	118
5	Banana juice	Hydrothermal	330	Cu(II)	119
6	Coconut water	Microwave hydrothermal	350	Cu(II)	120
7	Coriander Leaves	Hydrothermal	320	Fe(III)	121
8	Papaya	Hydrothermal	370	Fe(III)	122
9	Potato	Hydrothermal	323	Fe(III)	123
10	Black tea	Hydrothermal	320	Fe(III)	124
11	Egg white	Hydrothermal	315	Fe(III)	125
12	Honey	Hydrothermal	338	Fe(III)	126
13	Goose feathers	Microwave hydrothermal	340	Fe(III)	127
14	Pomelo peel	Hydrothermal	360	Hg(II)	128
15	Yam	Hydrothermal	320	Hg(II)	129
16	Cucumber Juice	Hydrothermal	514	Hg(II)	130
17	Lemon peel	Hydrothermal	441	Hg(II)	131
18	Prosopis juliflora leaves	Hydrothermal	325	Hg(II)	132
19	Flour	Microwave	365	Hg(II)	133
20	Serum albumin	Microwave	345	Pb(II)	134

4.2.1 Carbon dots synthesis based on berries and leaves

Building on the successful application of biomass-based CDs (B-CDs) in metal ion sensing, this thesis also begins with careful selection of biomass as initial precursors to explore the use of CDs as metal sensors. Specifically, jacaranda leaves and berries were chosen as the carbon sources, and B-CDs were synthesised through a hydrothermal reaction. Jacaranda trees, known for their iconic purple blooms, hold a special place in Sydney's heart, particularly at the University of Sydney. A historically significant *Jacaranda mimosifolia* once graced the south-eastern corner of the main quadrangle, a legacy continued by its clone, which now stands in the same spot, symbolising a blend of beauty and academic resilience. The choice of biomass precursors leverages their natural composition, which is rich in organic compounds suitable for carbon dot formation, providing an environmentally friendly and sustainable approach for

metal ion sensing applications. These naturally-derived CDs were then evaluated for their potential as selective metal sensors.

4.2.2 Jacaranda leaves-based carbon dots for cation sensing and metal ions sensing

Three types of Jacaranda leaf-based CDs were prepared using hydrothermal reactions under different loading conditions: Jacaranda_CD (synthesised from 5 g Jacaranda leaves with 50 mL MilliQ water), Jacaranda_(NaOH)_CD (synthesised from 5 g Jacaranda leaves with 50 mL 0.2 M NaOH). The N-doping impacts on CDs' fluorescence properties has been introduced in Chapter 2 2.1.2.4, and discussed in Chapter 3 3.2.4. The properties of CD can be fine-tuned by varying the N-doping level. So that we also prepared Jacaranda_(N-doped)_CD (synthesised from 5 g Jacaranda leaves and 0.5 g p-phenylenediamine (PPD) with 50 mL MilliQ water).

4.2.2.1 ATR characterisation of jacaranda-based carbon dots

The ATR-FTIR spectra revealed several key functional groups on the surface of the Jacaranda-based CDs (Figure 4.2). A broad peak around 3200–3500 cm^{-1} , corresponding to O-H and N-H stretching, was observed in all spectra, indicating the presence of hydroxyl or amine groups. These groups contribute to the hydrophilicity of the CDs and their solubility in water. Additionally, a peak in 1700–1750 cm^{-1} associated with C=O stretching vibrations was noted, confirming the presence of carbonyl or carboxyl groups (-COOH), which are formed during the carbonisation process. The C-H stretching peaks at 2800–3000 cm^{-1} suggest the presence of alkyl groups, while peaks between 1000–1300 cm^{-1} reflect C-O or C-N stretching, confirming the presence of oxygen- and nitrogen-containing functional groups.

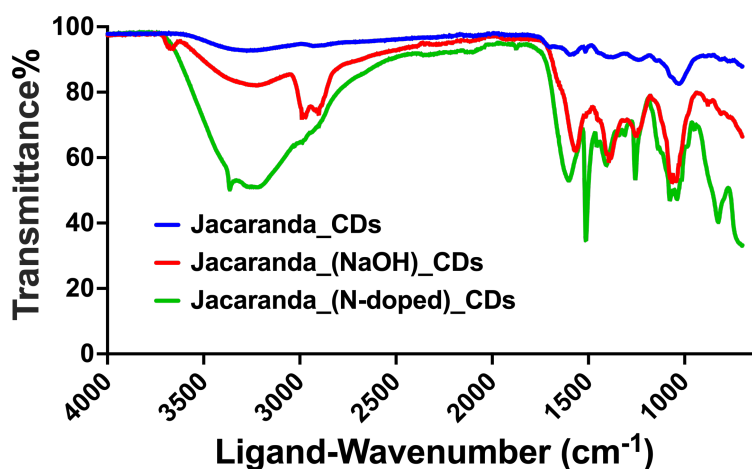


Figure 4.2 ATR of jacaranda leave-based carbon dots

4.2.2.2 Fluorescent properties of jacaranda-based carbon dots

All Jacaranda-based carbon dots (CDs) exhibited the typical excitation-dependent emission behaviour common to carbon dots. This property is attributed to the range of surface functional groups present, which caused shifts in emission peaks as the excitation wavelength increased. The fluorescence spectra for each type of carbon dot were scanned over an excitation range from 300 nm to 500 nm.

Jacaranda_CD

The Jacaranda-derived carbon dots (Jacaranda_CD) demonstrated strong blue fluorescence, with an emission peak observed at 430 nm when excited at 300 nm (Figure 4.3(a)). As the excitation wavelength increased, the emission peak shifted to longer wavelengths, reaching 650 nm at 500 nm excitation. This red shift was mainly attributed to the diverse surface functional groups, particularly oxygen-containing ones, which influenced the emission characteristics. The broad emission bands observed across all excitation wavelengths suggested a range of emissive states, likely due to surface functionalisation. The strong blue fluorescence at shorter excitation wavelengths indicated that Jacaranda_CD was suitable for applications requiring high-intensity blue emission, such as fluorescence-based sensors.

Jacaranda_(NaOH)_CD

The Jacaranda_(NaOH)_CD sample exhibited a distinctive dual-emission behaviour, setting it apart from the other carbon dots (Figure 4.3(b)). When excited at 300 nm, the primary emission peak was at 430 nm, indicating blue fluorescence. However, at 400 nm excitation, a second peak appeared at 655 nm, representing a significant red shift. This dual-peak behaviour was linked to the NaOH-assisted synthesis, which likely introduced additional oxygen-containing functional groups, such as hydroxyl and carboxyl groups. These functional groups influenced the electronic transitions, resulting in emissive centres that responded to different excitation energies. The ability to exhibit both blue and red fluorescence suggested that Jacaranda_(NaOH)_CD had strong potential for ratiometric sensing applications, where the ratio of two different fluorescence signals could improve measurement accuracy.

Jacaranda_(N-doped)_CD

The Jacaranda_(N-doped)_CD sample displayed enhanced fluorescence in the blue-green region, attributed to nitrogen doping, which introduced additional emissive centres (Figure

4.3(c)). At 300 nm excitation, the emission peak appeared at 430-440 nm, producing strong blue fluorescence. As the excitation wavelength increased to 320 nm, the peak shifted to 450-460 nm, and at longer wavelengths such as 400 nm, the emission shifted further to the green region around 480-600 nm. The presence of nitrogen functional groups contributed to the increased fluorescence intensity, especially at shorter excitation wavelengths, making Jacaranda_(N-doped)_CD well-suited for applications requiring high-intensity blue-green emission. The broad emission bands indicated that nitrogen doping enhanced the versatility of these carbon dots for sensing applications, where sensitivity to environmental changes is crucial.

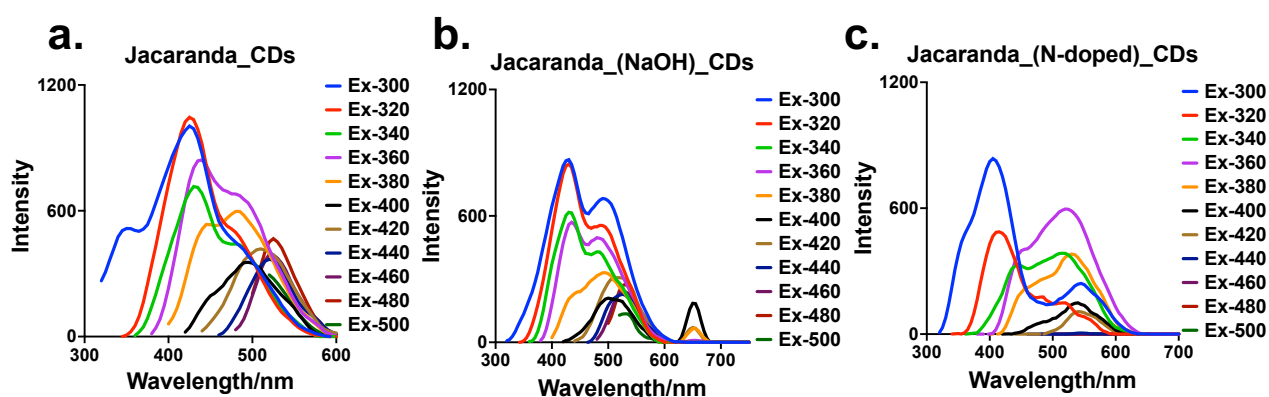


Figure 4.3 fluorescent spectrums of jacaranda-based carbon dots (1 mg/mL CDs, under λ_{ex} ranged from 300 to 500 nm); a: Jacaranda_CDs; b: Jacaranda_(NaOH)_CDs; c: Jacaranda_(N-doped)_CDs

4.2.2.3 pH sensing potential of jacaranda-based carbon dots

The pH response analysis of the jacaranda-based CDs was conducted, revealing distinct behaviours across the different samples. Jacaranda_CD acted as a pH turn-on sensor, consistent with the findings from the CDs-based pH sensing study in Chapter 3 Carbon dots and machine learning for fluorescent sensing applications. The effect of N-doping was reconfirmed, as it shifted the sensor's response from turn-on to turn-off, as observed in Jacaranda_(N-doped)_CD. Both of these pH sensing responses were based on the emission peak at 425-430 nm under an excitation wavelength of 300 nm.

In Jacaranda_(NaOH)_CD, the same emission peak also functioned as a pH turn-on sensor. However, this sample exhibited an additional significant fluorescence peak at 650 nm under an excitation wavelength of 400 nm. This second peak was found to operate as a full-range pH turn-on sensor, effective across pH values from 3 to 10.

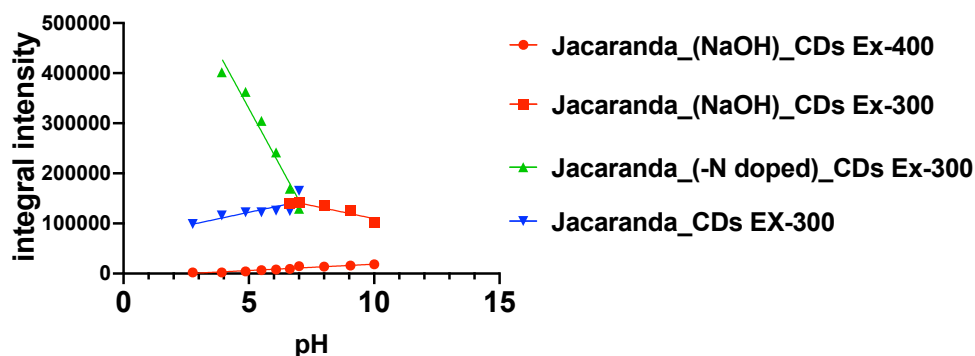


Figure 4.4 Jacaranda based carbon dots' pH response (1 mg/mL CDs, under λ_{ex} at 300 and 400 nm)

The potential for developing a ratiometric pH sensor was inspired by combining the N-doping effect with the second fluorescence peak induced by the basic synthesis conditions. It was anticipated that by synthesising Jacaranda_(NaOH)_(N-doped)_CD, the first peak (under 300 nm excitation) could function as a pH turn-off signal, while the second peak (under 400 nm excitation) could serve as a pH turn-on signal.

Following this hypothesis, Jacaranda_(NaOH)_(N-doped)_CD was synthesised, and its pH response was examined, confirming the initial prediction. The first peak under excitation at 300 nm acted as a pH turn-off signal (inherited from Jacaranda_(N-doped)_CD), while the second peak under excitation at 400 nm continued to function as a pH turn-on signal (inherited from Jacaranda_(NaOH)_CD). This dual fluorescence response allows Jacaranda_(NaOH)_(N-doped)_CD to be employed as a ratiometric pH sensor, offering complementary and reliable signals for accurate pH detection across various conditions.

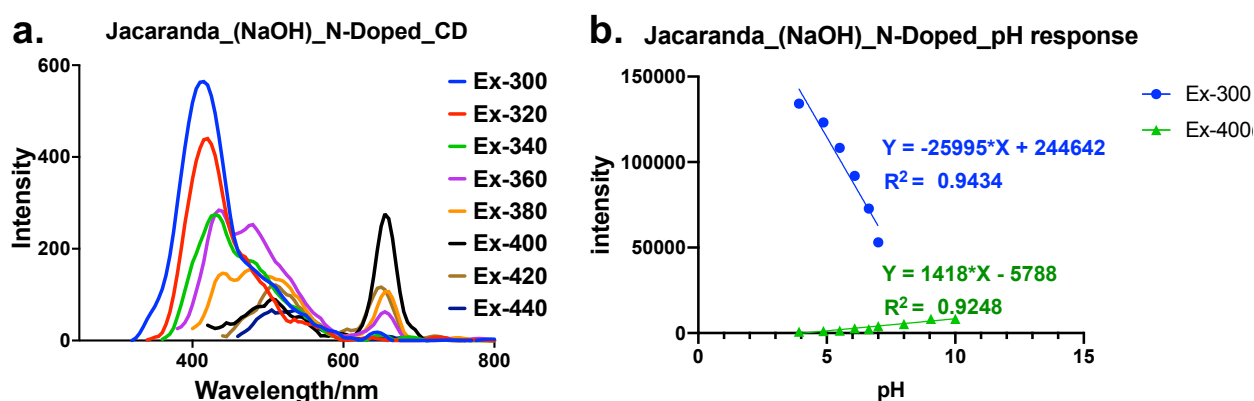


Figure 4.5 Fluorescence emission spectra of jacaranda_(NaOH)_(N-doped)_CD (1 mg/mL CDs, under λ_{ex} ranged from 300 to 500 nm) and linear fit of jacaranda_(N-doped)_CD at different pH levels (1 mg/mL CDs, under λ_{ex} at 300 and 400 nm)

4.2.2.4 Potential of jacaranda based-carbon dots for metal ions sensing

The fluorescence response of Jacaranda_(N-doped)_CDs under different excitation wavelengths demonstrated a distinct shift in behaviour, highlighting the tunability of these carbon dots for metal ion sensing. When excited at 320 nm (left panel), the CDs acted as a selective Cu(II) turn-off sensor, with significant quenching of fluorescence in the presence of Cu(II). Other metal ions such as Al(III), Ag(I), Ca(II), Co(II), and Zn(II) showed minimal impact on fluorescence, indicating limited interaction with the CDs at this excitation wavelength. This selective response suggested that Jacaranda_(N-doped)_CDs could effectively distinguish Cu(II) from other metal ions under these conditions, but there is not a very strong dynamic range.

In contrast, at 420 nm excitation (right panel), the CDs exhibited a cross-reactive sensing behaviour, where multiple metal ions caused noticeable changes in fluorescence. Both Co(II) and Cu(II) led to quenching, while Ni(II) triggered a fluorescence turn-on response, increasing the intensity compared to the blank. This shift from selective to cross-reactive sensing illustrated the CDs' adaptability, enabling them to detect a broader range of metal ions when excited at different wavelengths. The ability to respond differently to metal ions at varying excitation wavelengths demonstrated the versatility of these carbon dots in metal sensing applications.

The change in fluorescence response at different wavelengths was attributed to the complex surface functionalisation of the carbon dots, which resulted from their natural precursor. Different fluorescent emissive sites on the surface were activated at different excitation wavelengths, leading to variable interactions with metal ions. At 320 nm, emissive sites responsible for Cu(II) quenching were dominant, producing selective sensing. At 420 nm, a wider array of emissive sites became active, allowing the CDs to exhibit cross-reactivity towards several metal ions. This tuneable response offers a flexible approach to designing carbon dot-based sensors, allowing researchers to control selectivity and cross-reactivity by adjusting the excitation wavelength.

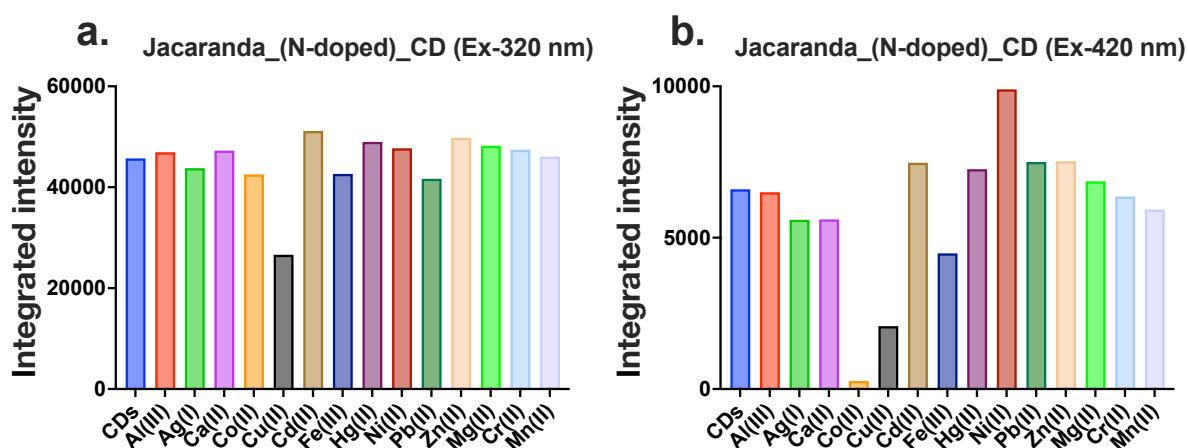


Figure 4.6 Fluorescence response of Jacaranda_(N-doped)_CDs to various metal ions under different excitation wavelengths (a: $\lambda_{ex}=320$ nm; b: $\lambda_{ex}=420$ nm)(1 mg/mL CDs; 100 μ M of metal ions).

4.2.3 Potential of berries based-carbon dots for Fe(III) sensing

The fluorescence properties of carbon dots synthesized from berry-based precursors, namely blueberry, raspberry, and strawberry, offer significant potential for metal ion sensing applications. The characterization of these carbon dots, presented in Figure and Table 4.4, reveals their distinct optical properties, including their excitation and emission spectra, Stokes shifts, and selective interactions with metal ions. These features make berry-based carbon dots particularly promising for developing sensitive and selective fluorescent sensors.

The fluorescence properties of berry-based carbon dots demonstrate distinct excitation and emission characteristics. Blueberry-based carbon dots exhibit an excitation peak at 400 nm and an emission peak at 477 nm, with a Stokes shift of 77 nm, indicating stable fluorescence and selective responses to various metal ions. Raspberry-based carbon dots, with an excitation peak at 356 nm and an emission peak at 458.9 nm, show a larger Stokes shift of 102.9 nm, enhancing their sensitivity for detecting low concentrations of metal ions. Strawberry-based carbon dots, with an excitation peak at 373.1 nm and an emission peak at 448 nm, have a Stokes shift of 74.9 nm and demonstrate selective interactions with specific metal ions, reinforcing their potential use in metal ion sensing applications.

The fluorescence spectra and bar graphs for all three berry-based carbon dots highlight their selective interactions with metal ions. Notably, Fe(III) ions significantly quench the fluorescence of all these carbon dots, indicating their suitability as turn-off sensors for Fe(III) detection. In summary, biomass-based carbon dots exhibit promising optical properties and

selective responses to metal ions, particularly Fe(III), suggesting their use as effective turn-off sensors. These findings lay the groundwork for further exploration of carbon dots in selective metal ion sensing, which will be discussed in the following section. However, while these initial findings demonstrate the feasibility of using berry-derived carbon dots for Fe(III) sensing, further investigation is necessary to confirm their practical applicability. Future steps could include refining the synthesis process to enhance the sensitivity and selectivity of these carbon dots for Fe(III). For instance, optimising surface functionalisation through targeted doping or adjusting synthesis conditions might improve their response to Fe(III) over competing ions. Additionally, testing these carbon dots under varied environmental conditions, such as different pH levels and ionic strengths, would help evaluate their robustness and reliability as Fe(III) sensors. This future work would ultimately provide a deeper understanding of their practical effectiveness, guiding their development into reliable, real-world Fe(III) sensing platforms.

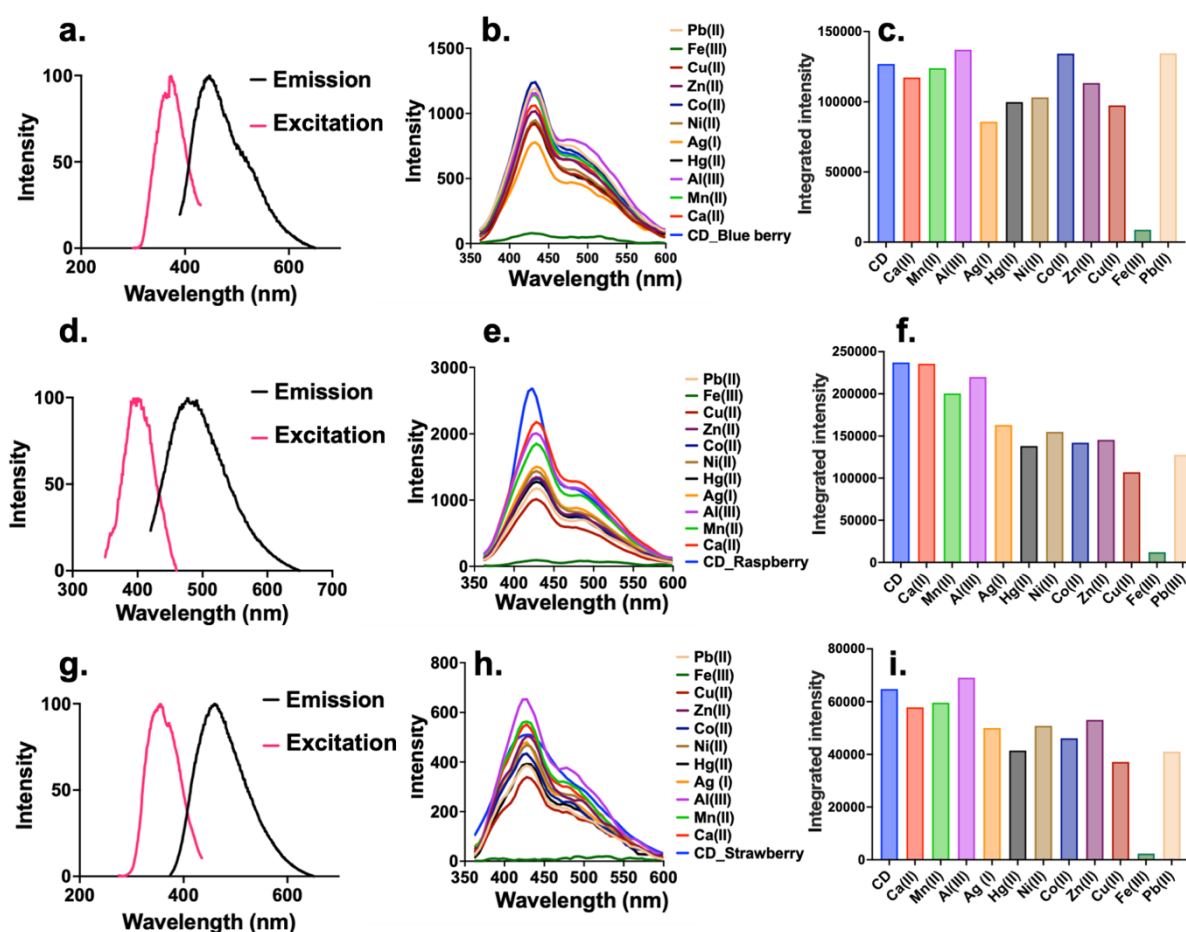


Figure 4.7 Fluorescence emission and excitation spectra (a,d,g), full spectrum of carbon dots with metal ion interactions (1 mg/mL CDs; 100 μ M metal ions, λ_{ex} = 340 nm, λ_{em} ranging from 360–600

nm for figure b, e, and h), and selectivity response (1 mg/mL CDs; 100 μ M metal ions, λ_{ex} = 340 nm, λ_{em} integrated across 360–600 nm, for figure c, f, and i) of carbon dots derived from blueberries, raspberries, and strawberries. Figures a–c represent blueberry-based carbon dots (in figure a, λ_{ex} = 400 nm, λ_{em} = 475 nm for excitation and emission spectra respectively), d–f represent raspberry-based carbon dots (in figure d, λ_{ex} = 355 nm, λ_{em} = 460 nm), and g–i represent strawberry-based carbon dots (in figure g, λ_{ex} = 370 nm, λ_{em} = 450 nm). The bar graphs illustrate the fluorescence selectivity responses towards various metal ions at a concentration of 100 μ M.

Table 4.4 fluorescence properties of berry driven carbon dots

Precursor	Excitation wavelength/nm	Emission wavelength/nm	Stockes shift/nm
Blue berry	400	477	77
Raspberry	356	458.9	102.9
Strawberry	373.1	448	74.9

4.3 Fructose based carbon dots-based metal ion sensing

Despite the advantages of using natural precursors, such as biocompatibility, the synthesis process can result in structurally complex and non-homogeneous CDs, limiting their practical applications. The variability in size distribution and the challenges in controlling the CDs' fluorescence properties, such as emission wavelength, further restrict the consistency of naturally derived CDs. To address these limitations, this section shifts the focus to the use of pure chemical precursors, aiming for more controlled and predictable synthesis outcomes. This approach will allow for better control over the CDs' properties, enhancing their potential for metal ion sensing applications.

Building on the success of carbon dots as metal ion sensors, this section explores the synthesis of fructose-based carbon dots (F-CDs) and nitrogen-doped F-CDs (N-doped F-CDs) using *p*-phenylenediamine (PPD) as the nitrogen source. Researchers have noted that Jacaranda leaves contain free nectar sugars,¹³⁵ including fructose, and berries are also rich in fructose. This inspired the selection of fructose as a simplified and uniform carbon source for CDs synthesis, aiming to produce more homogeneous structures post-synthesis. N-doping, achieved using PPD, was previously demonstrated to influence the properties of carbon dots, including their selectivity for Cu(II) ions, as seen in the case of Jacaranda leaves-based carbon dots.

4.3.1.1 Synthesis of and characterisation of fructose-based carbon dots and nitrogen-doped F-CDs

Fructose-based carbon dots (F-CDs) were synthesised via a hydrothermal reaction, using 0.5 g of pure fructose as the precursor. For the N-doped F-CDs, the precursor was a mixture of 0.227 g fructose and 0.273 g *p*-phenylenediamine (PPD), totalling 0.5 g. Both types of CDs were

produced under identical conditions, with the reaction carried out at 180°C for 6 hours. The resulting solutions displayed an orange hue, and under UV light, the F-CDs emitted a light blue fluorescence, while the N-doped F-CDs showed a light green-yellow fluorescence (Figure 4.8). After synthesis, the solutions were freeze-dried to obtain the powdered form of these CDs for further analysis.

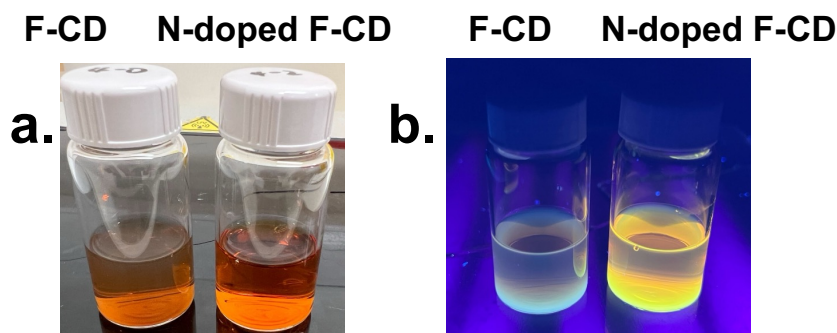


Figure 4.8 Carbon dots solutions; *a: under day light; b: under UV lamp*

4.3.1.2 ATR

The ATR-FTIR spectra for both fructose-based carbon dots (F-CDs) and nitrogen-doped fructose-based carbon dots (N-doped F-CDs) reveal key differences in surface functional groups resulting from nitrogen doping. Both spectra show a broad peak in the 3200–3500 cm^{-1} region, which can be attributed to O-H stretching from hydroxyl groups or N-H stretching from amine groups. This peak is more pronounced in the N-doped F-CDs, indicating an increase in amine groups due to nitrogen doping, while in F-CDs, it primarily reflects the presence of hydroxyl groups.

A clear C=O stretching peak appears around 1700–1750 cm^{-1} in both spectra, indicating the presence of carbonyl or carboxyl groups. However, the peak is slightly more pronounced in F-CDs, while in N-doped F-CDs, it may be slightly shifted due to interactions with nitrogen-containing groups like amides or imines. Additionally, the 2800–3000 cm^{-1} region shows C-H stretching peaks, reflecting the presence of alkyl groups, though these are relatively less prominent in both types of CDs. The 1000–1300 cm^{-1} region, associated with C-O or C-N stretching, shows stronger peaks in N-doped F-CDs, confirming the incorporation of nitrogen-containing functional groups.

The nitrogen doping effect is further supported by the appearance of amide peaks in the 1500–1600 cm^{-1} region in the N-doped F-CD spectrum, indicating N-H bending or C-N stretching

from amides or imines. These results confirm that nitrogen doping successfully introduced nitrogen-containing groups like amines and amides, while the basic structure, including hydroxyl and carbonyl groups, was retained. The modified surface chemistry of N-doped F-CDs contributes to their distinct fluorescence properties and enhanced potential for sensing applications.

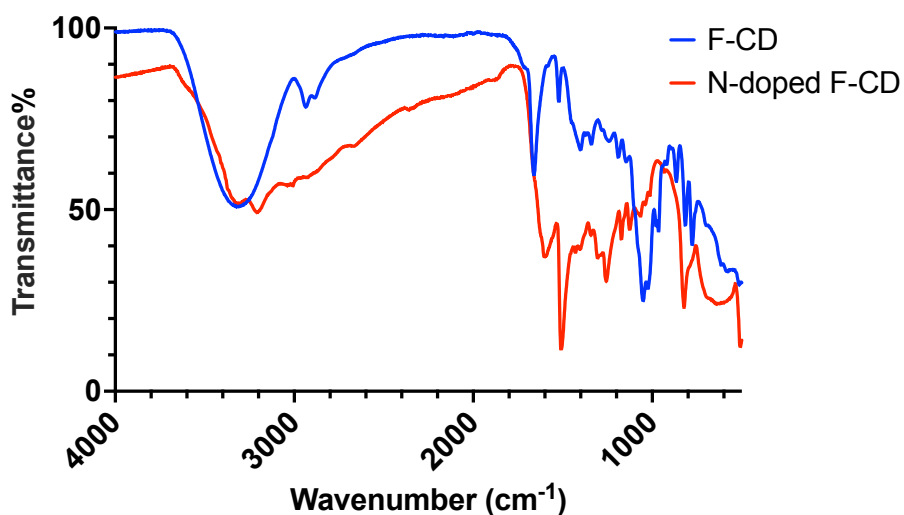


Figure 4.9 ATR of F-CD (blue line) and N-doped F-CD (red line)

4.3.1.3 Absorption study

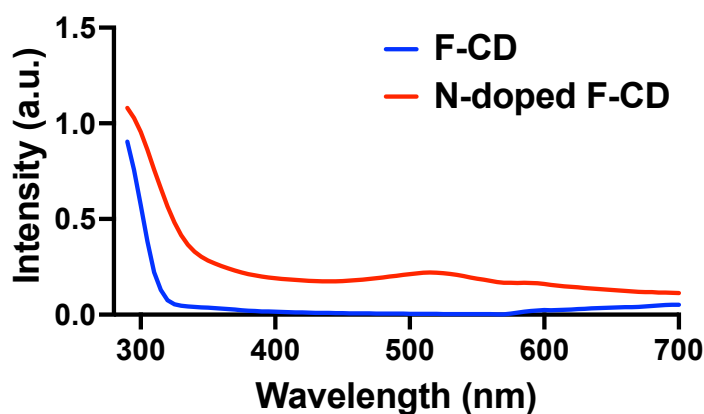


Figure 4.10 Absorption spectrum of F-CD (blue line), and N-doped F-CD (red line)

The absorption spectra of both F-CD and N-doped F-CD provided insight into their optical properties and electronic transitions. Both types of carbon dots exhibited strong absorption in the UV region (200–300 nm), characteristic of carbon dots, primarily due to π – π^* transitions associated with the conjugated carbon core (C=C bonds). This absorption diminished significantly beyond 300 nm, with a slight tailing into the visible region, likely influenced by surface states or functional groups.

When comparing the two, F-CD showed a strong absorption peak between 220–280 nm, sharply decreasing as it approached 300 nm. In contrast, N-doped F-CD exhibited not only stronger absorption in this region but also a broader tail into the visible spectrum. This suggests that nitrogen doping enhanced the light absorption properties, introducing $n-\pi^*$ transitions attributed to nitrogen-containing groups, such as amines or amides. The broader absorption of the N-doped F-CD, particularly in the visible region, indicates the formation of additional surface states, making them more versatile for applications like sensing or bioimaging.

In conclusion, the nitrogen doping in N-doped F-CD enhanced their absorption capacity, broadening the spectral range into the visible region. This improvement, driven by additional $n-\pi^*$ transitions, makes the N-doped F-CDs more suitable for applications requiring strong light absorption and visible light activity, such as photocatalysis and bioimaging.

4.3.1.4 Fluorescence study

In the fluorescence analysis, the F-CD (Figure 4.13(a)) exhibited an excitation peak around 320 nm, with an emission peak between 450–460 nm, characteristic of typical blue fluorescence. This excitation-emission gap corresponds to electronic transitions likely arising from $\pi-\pi^*$ transitions within the carbon core structure. In contrast, the N-doped F-CD (Figure 4.16(a)) displayed a similar excitation peak at 320 nm, but the emission was red-shifted to around 480 nm. This shift is attributed to nitrogen doping, which introduces new surface functional groups and electronic states, modifying the fluorescence behaviour. The nitrogen doping not only shifted the emission wavelength but also broadened the emission spectrum, enhancing the tunability of the optical properties. As a result, the N-doped F-CD exhibited a broader and red-shifted emission compared to the F-CD, reflecting the impact of doping on their fluorescence characteristics.

4.3.2 Cu(II) sensing using N-doped fructose-based carbon dots

4.3.2.1 Optimisation of carbon dot concentration

The fluorescence response of N-doped F-CDs to varying concentrations of Cu(II) provided valuable insights into optimising F-CD density for effective Cu(II) detection (Figure 4.11). At lower Cu(II) concentrations (0–20 μM), fluorescence intensity showed minimal quenching, with a slight "turn-on" effect observed at lower N-doped F-CDs densities. For example, at 10 μM Cu(II) and 0.05 mg/mL F-CDs, a slight increase in fluorescence was observed, while at 1

mg/mL F-CDs, fluorescence remained stable, offering a reliable baseline for sensing applications.

As Cu(II) concentrations increased (40–100 μM), a more pronounced fluorescence quenching effect was observed, particularly at higher N-doped F-CDs densities. For instance, at 100 μM Cu(II) and 1 mg/mL N-doped F-CDs, fluorescence intensity dropped significantly, indicating strong quenching. This demonstrated that higher N-doped F-CDs concentrations, particularly around 1 mg/mL, provided good sensitivity for Cu(II) detection at moderate levels.

At higher Cu(II) concentrations (200–500 μM), fluorescence quenching became more consistent across all N-doped F-CDs densities, with a more pronounced effect at 1 mg/mL N-doped F-CDs. Even at 500 μM Cu(II), significant quenching was evident, highlighting the efficiency of higher N-doped F-CDs concentrations in Cu(II) sensing. The results suggested that 1 mg/mL of N-doped F-CDs was the optimal concentration for reliable and sensitive Cu(II) detection, exhibiting stronger and more consistent quenching effects across moderate to high Cu(II) concentrations. Lower N-doped F-CDs concentrations demonstrated weaker quenching, making higher densities, particularly 1 mg/mL, more suitable for Cu(II) sensing applications.

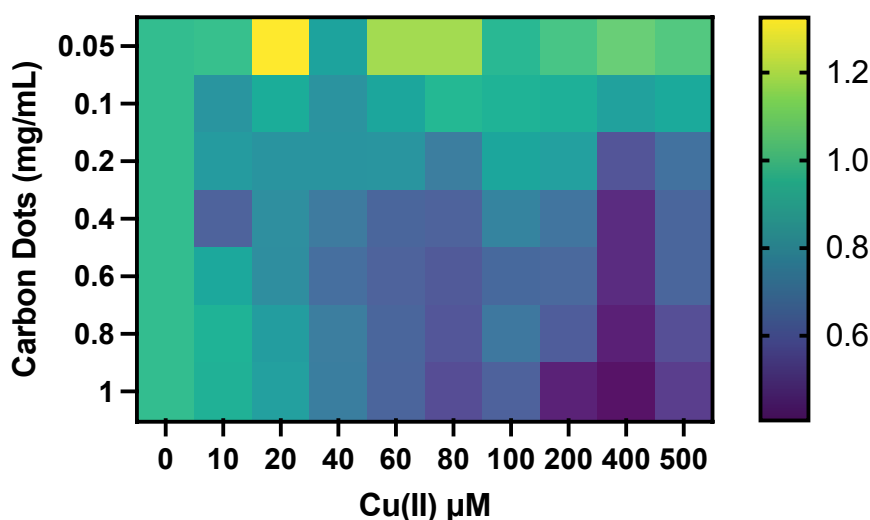


Figure 4.11 Fluorescence quenching of N-doped fructose-based carbon dots at varying Cu(II) concentrations and F-CD densities ($\lambda_{\text{ex}} = 370 \text{ nm}$)

Figure 4.12 visually demonstrates that 1 mg/mL N-doped F-CDs achieves the highest fluorescence intensity and effective quenching when exposed to 100 μM Cu(II). The green bar, representing this concentration, shows the strongest fluorescence compared to other F-CD concentrations, confirming that 1 mg/mL maintains high initial fluorescence prior to quenching. This concentration also exhibits the most significant fluorescence "turn-off" effect, indicating

optimal performance for sensing applications. In contrast, lower concentrations of F-CDs (e.g., 0.1 mg/mL, 0.4 mg/mL) show weaker fluorescence and less efficient quenching, suggesting reduced interaction with Cu(II). The consistent use of 100 μ M Cu(II) across all conditions highlights that the observed quenching is primarily due to F-CD density. Overall, the figure reinforces that 1 mg/mL N-doped F-CDs is the optimal concentration for Cu(II) detection, providing robust fluorescence and efficient quenching, while lower densities are less suitable for such applications.

The observed concentration-dependent fluorescence quenching of the CDs may arise from a combination of mechanisms, including aggregation-induced quenching (AIQ), aggregation-enhanced emission (AEE), and inner filter effects (IFE).^{136, 137} At low concentrations, a slight fluorescence increase suggests potential AEE behaviour, possibly due to Cu(II)-induced formation of emissive aggregates. However, at higher concentrations, the predominant quenching is likely due to AIQ or IFE, where increased particle proximity facilitates non-radiative decay or optical attenuation. Further clarification could be achieved through dynamic light scattering or absorbance-corrected fluorescence assays to distinguish genuine quenching from inner filter artefacts.

N-doped F-CDs for Cu(II) sensing under different CD concentration

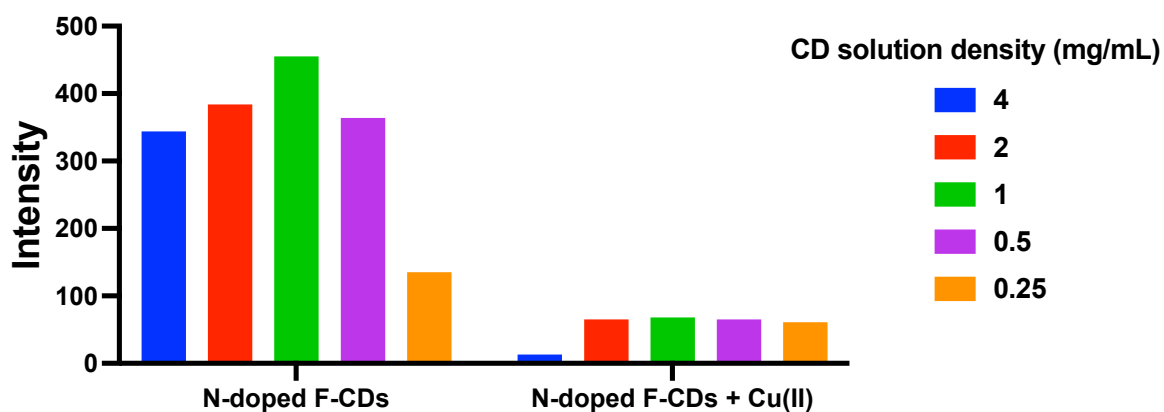


Figure 4.12 Fluorescence response of N-doped fructose-based carbon dots at varying concentrations, CDs alone (left) and CDs with 100 μ M Cu(II) (right) (λ_{ex} = 370 nm)

4.3.2.2 Cu(II) selectivity

N-doped F-CDs were investigated for their ability to detect Cu(II) (Figure 4.13(b-d)). In the absence of Cu(II), the N-doped F-CD exhibited a quenched fluorescent peak at 525-530 nm. Upon the addition of Cu(II), there was a noticeable decrease in fluorescence intensity, indicating that Cu(II) effectively quenched the fluorescence of the N-doped F-CDs. To assess

sensitivity, various concentrations of Cu(II) ranging from 0 to 80 μM were tested. The inset in Figure 4.13(d) shows the relationship between $(A_0-A)/A_0$ and Cu(II) concentration, where A_0 and A represent the integrated fluorescence intensities from 400-700 nm in the absence and presence of Cu(II), respectively. The detection limit was estimated to be 36 μM , with a linear range from 5 to 80 μM and an R^2 value of 0.984.

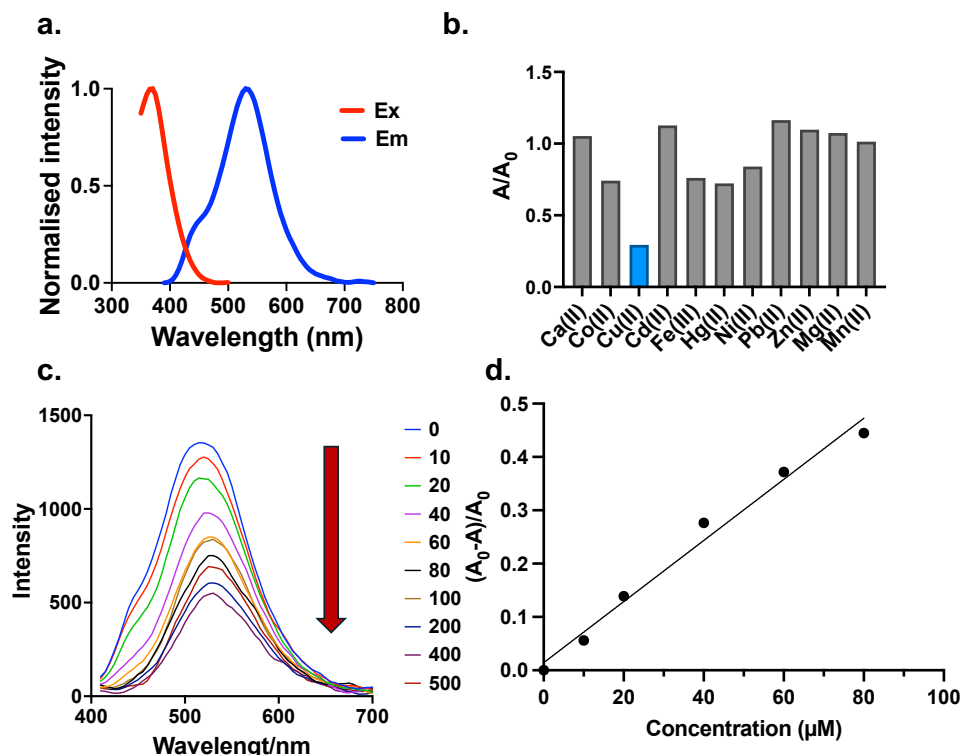


Figure 4.13 (a) Fluorescence spectrum of N-doped F-CDs; (b) Integrated FL intensity differences of F-CD dispersion between the blank (A_0) and solutions containing various metal ions (A) ($\lambda_{\text{ex}} = 370$ nm; 100 μM metal ions; 1 mg/mL CDs); (c) Fluorescence spectra of N-doped F-CDs in the presence of different Cu(II) concentrations (from top to bottom: 0 to 500 μM); (d) Dependence of $(A_0-A)/A_0$ on Cu(II) concentrations within the range of 0–80 μM (1 mg/mL CDs; $\lambda_{\text{ex}} = 370$ nm, with emission integrated from 400 nm to 700 nm). In (b) and (d) “A” represented the integrated area under the fluorescence spectrum.

4.3.2.3 Fluorescence stability

In order to investigate the stability of the CDs, a time-course study was implemented. Figure 4.14 illustrates the fluorescence intensity of N-doped fructose-based carbon dots (CDs) over time, demonstrating their stability when exposed to varying concentrations of Cu(II). The blue line, representing CDs without Cu(II), shows a gradual fluorescence decay from an initial intensity of around 800 a.u., indicating good intrinsic stability over 120 minutes with only minor loss in intensity. In contrast, the red and green lines, corresponding to CDs exposed to 50 μM and 100 μM Cu(II), respectively, exhibit pronounced fluorescence quenching, with

initial intensities of approximately 400 a.u. and 250 a.u. The quenching effect is clearly concentration-dependent, as the higher Cu(II) concentration induces greater quenching. Despite the initial reduction in fluorescence, both Cu(II) conditions show stable intensities over time, indicating that the quenching is persistent without further degradation. These observations highlight the potential of N-doped fructose-based CDs as effective Cu(II) sensors, where their stable fluorescence and reliable quenching behaviour enable consistent sensing performance.

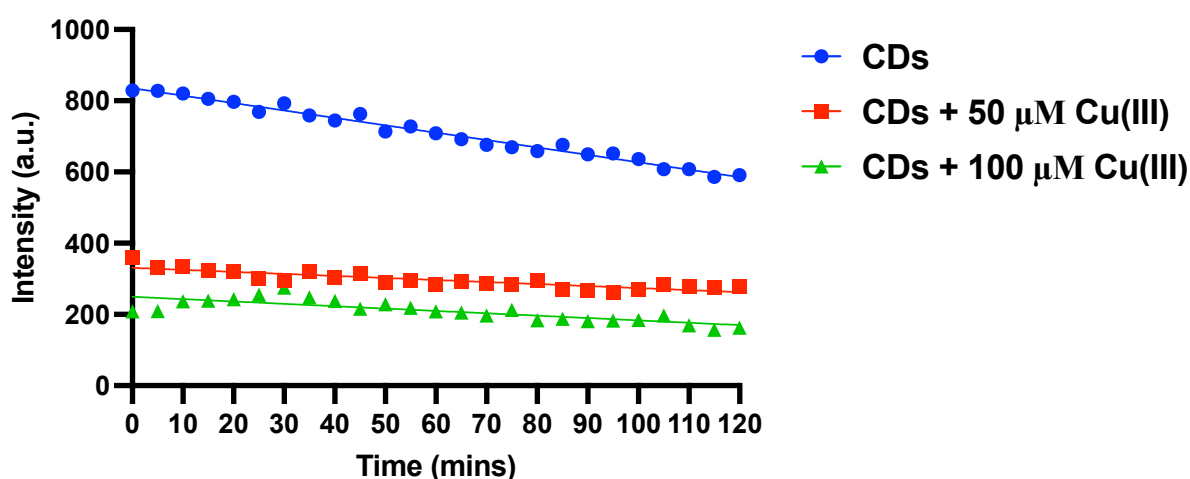


Figure 4.14 Time-dependent fluorescence stability of N-doped fructose-based carbon dots in the presence of Cu(II) at different concentrations (1 mg/mL CDs, $\lambda_{\text{ex}} = 370$ nm, highest intensity collected): CDs alone (blue), CDs with 50 μM Cu(II) (red), and CDs with 100 μM Cu(II) (green)

4.3.2.4 Fluorescence response of N-doped F-CD to Cu(II) with presence of other metal ions

The fluorescence study of N-doped fructose-based CDs reveals distinct responses to Cu(II) and other metal ions, highlighting the selective quenching effect of Cu(II). In the experiment, the blue bars represent the fluorescence intensity ratio (I/I_0) of the CDs in the presence of a mixture of Cu(II) and another metal ion, while the red bars show the fluorescence intensity when CDs are exposed to individual metal ions without Cu(II). Key observations demonstrate that Cu(II) dominates fluorescence quenching, as seen by the significantly lower blue bars across all metal ions, especially for Ca(II), Zn(II), Cd(II), Mg(II), and Al(III). These metal ions show a noticeable fluorescence "turn-on" effect without Cu(II), indicated by the red bars exceeding 1.0. Zn(II), Al(III), and Ca(II) show particularly strong fluorescence enhancement in the absence of Cu(II), while Cd(II) and Mg(II) exhibit slight enhancement. Conversely, Pb(II) and Mn(II) show minimal fluorescence changes, suggesting limited interaction with the CDs.

In contrast, the presence of Cu(II) in combination with these ions results in a notable decrease in fluorescence, with the blue bars consistently lower, reinforcing the dominant quenching effect of Cu(II). This indicates that the N-doped Fructose CDs are highly effective for selective Cu(II) detection, as they suppress fluorescence even in the presence of metal ions that would typically enhance it. These findings underscore the potential of these CDs in selective Cu(II) sensing applications, particularly in complex environments where multiple ions are present.

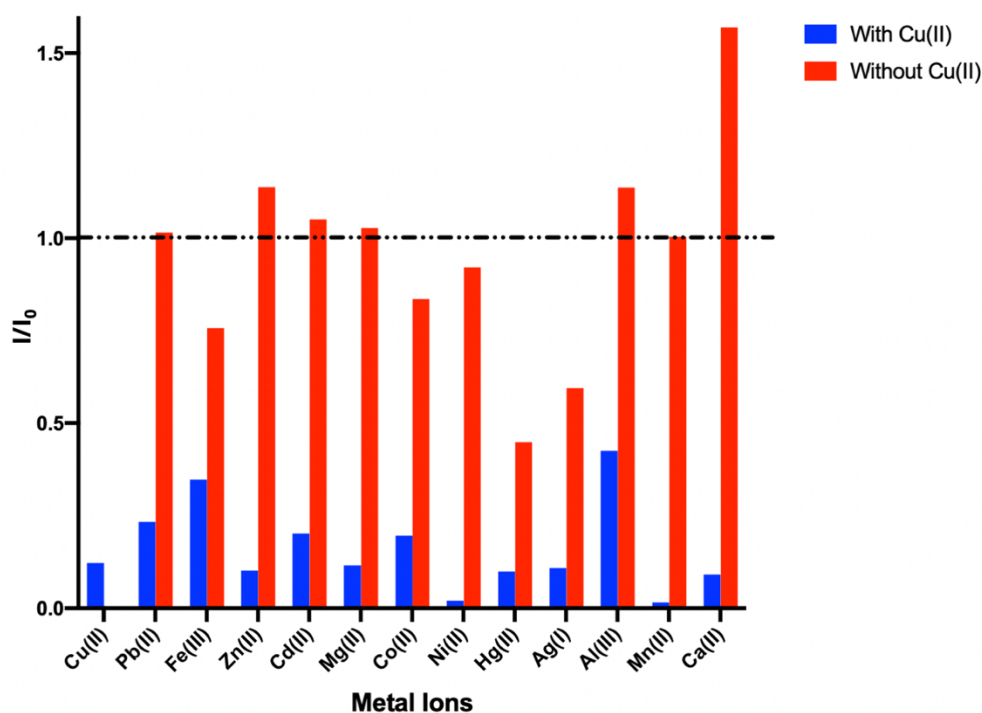


Figure 4.15 Fluorescence response of N-Doped Fructose based CDs to Cu(II) in the presence of various metal ions (1 mg/mL CDs, $\lambda_{ex} = 370$ nm, highest intensity collected). Blue bars represent the fluorescence intensity ratio (I/I_0) of N-doped Fructose CDs in the presence of a mixture of Cu(II) and another metal ion (total metal ion concentration controlled at 100 μ M, with Cu(II) at 40 μ M and the other metal ion at 40 μ M). Red bars represent the fluorescence intensity of CDs in the presence of individual metal ions without Cu(II) (metal ion concentration at 80 μ M).

4.3.3 Fe(III) sensing by using fructose-based carbon dots

F-CD was investigated for its ability to detect Fe(III). In the absence of Fe(III), the F-CD exhibited a quenched fluorescent peak at 480-530 nm. Upon the addition of Fe(III), there was a noticeable decrease in fluorescence intensity, indicating that Fe(III) effectively quenched the fluorescence of the F-CD. To assess sensitivity, various concentrations of Fe(III) ranging from 0 to 80 μ M were tested. The inset in Figure 4.16 b shows the relationship between $(A_0 - A)/A_0$ and Fe(III) concentration, where A_0 and A represent the integrated fluorescence intensities from 400-700 nm in the absence and presence of Cu(II), respectively. The detection limit was estimated to be 44 μ M, with a linear range from 5 to 80 μ M and an R^2 value of 0.977. Future

work could aim to enhance the detection limit and sensitivity range of F-CDs for Fe(III) by optimising synthesis parameters, such as adjusting precursor ratios or incorporating additional functional groups known to interact with Fe(III). Additionally, exploring the performance of F-CDs in varied pH environments or in the presence of competing metal ions would help establish their selectivity and stability across different conditions. By refining these aspects, F-CDs could be further developed into practical, high-performance sensors for Fe(III) applicable in environmental and biomedical settings.

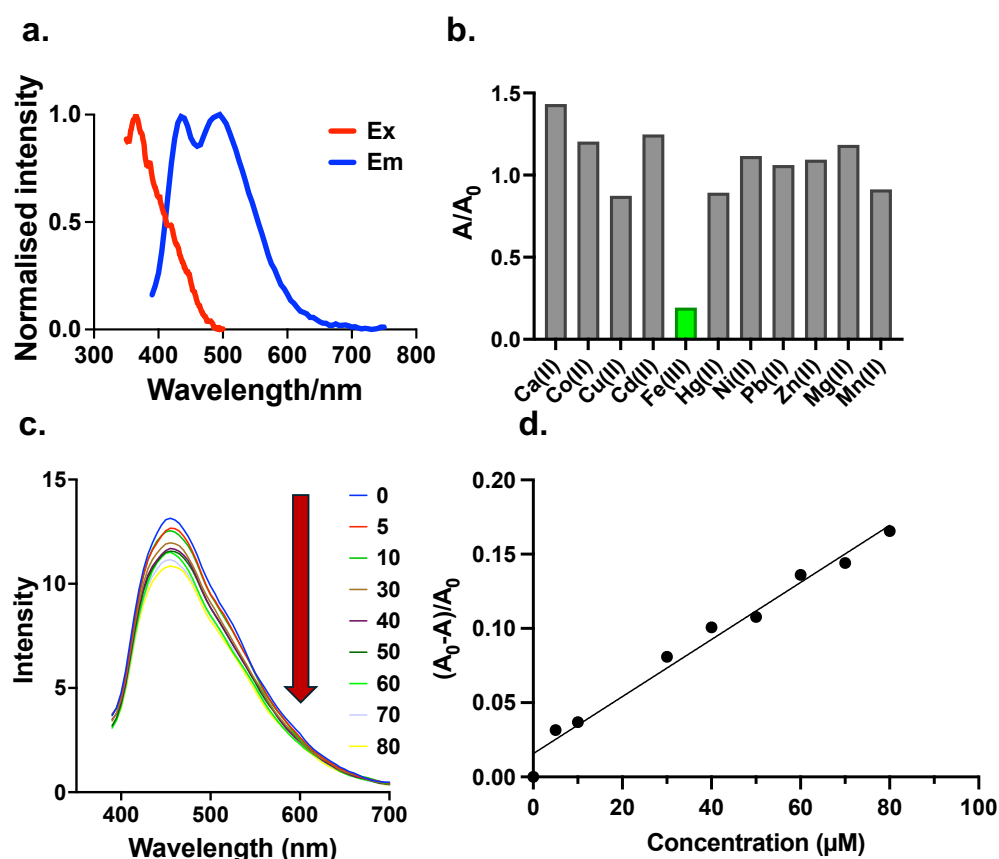


Figure 4.16 (a) Fluorescence spectrum of F-CDs; (b) Integrated FL intensity differences of F-CD dispersion between the blank (A_0) and solutions containing various metal ions (A) ($\lambda_{\text{ex}} = 370 \text{ nm}$; $100 \mu\text{M}$ metal ions; 1 mg/mL CDs); (c) Fluorescence spectra of F-CDs in the presence of different Fe(III) concentrations (from top to bottom: 0 to $80 \mu\text{M}$); (d) Dependence of $(A_0 - A)/A_0$ on Fe(III) concentrations within the range of $0\text{--}80 \mu\text{M}$ (1 mg/mL CDs; $\lambda_{\text{ex}} = 370 \text{ nm}$, with emission integrated from 400 nm to 700 nm). In (b) and (d), “A” represented the integrated area under the fluorescence spectrum.

4.4 Conclusion

In conclusion, this chapter introduced fluorescent sensors for Cu(II) and Fe(III) detection, investigating both biomass-based and chemically synthesised carbon dots (CDs). The study demonstrated the synthesis and application of Jacaranda-derived and fructose-based CDs, with

a particular focus on their capabilities for Fe(III) and Cu(II) sensing. Synthesised via hydrothermal methods, both pristine and nitrogen-doped CDs were examined, revealing how nitrogen doping significantly influenced their optical properties and enhanced their sensing capabilities. Excitation-dependent emission properties were prominent in Jacaranda-derived CDs, where nitrogen doping further intensified fluorescence and shifted emission spectra. These changes underscored the impact of surface functionalisation in modulating the CDs' response to metal ions.

For Cu(II) detection, nitrogen-doped fructose-based CDs demonstrated high selectivity, with effective quenching and a linear fluorescence response to Cu(II) concentration, achieving a detection limit of 36 μM . Fructose-based CDs also exhibited quenching behaviour in the presence of Fe(III), with a detection limit of 44 μM and a consistent linear response range. This selective reactivity underscores the potential of these CDs for applications in environmental monitoring, industrial processing, and biochemical assays, where distinct responses to Cu(II) and Fe(III) are required.

The potential of Jacaranda-derived CDs was also evaluated for pH sensing. Specifically, Jacaranda_(NaOH)_CDs exhibited dual fluorescence peaks—at 430 nm and 650 nm—with the 650 nm peak demonstrating full-range pH sensitivity, marking Jacaranda_(NaOH)_CDs as promising pH sensors. By combining nitrogen doping with basic synthesis conditions, a Jacaranda_(NaOH)_(N-doped)_CD was developed, effectively functioning as a ratiometric pH sensor. This configuration offered a pH turn-off signal at 430 nm and a turn-on signal at 650 nm, establishing a robust and reliable method for detecting pH fluctuations across various environments.

Future directions for research were highlighted during the synthesis of fructose-based CDs, where adjustments to N-doping levels resulted in varied responses to multiple metal ions. This cross-reactive response suggests strong potential for developing a comprehensive metal sensor array capable of detecting a broad range of ions with high sensitivity and selectivity. Further exploration of this concept will be detailed in Chapter 5.

This research underscores the versatility of carbon dots, particularly when customised through doping and tailored synthesis approaches. Surface functionalisation was shown to be instrumental in defining the optical properties of CDs and their metal ion interactions. The success in developing selective and sensitive CDs for Cu(II) and Fe(III) detection, alongside

the novel ratiometric pH sensing capabilities, paves the way for practical applications in environmental monitoring, industrial sensing, and biomedical diagnostics. Future research may expand on these findings, examining additional doping strategies to optimise sensitivity, enhancing selectivity, and broadening the applicability of CDs across diverse metal ions and complex environmental matrices.

4.5 Experimental section

4.5.1 Materials

All chemical reagents and solvents were of laboratory or analytical grade, sourced from Sigma-Aldrich, Merck, or Combi-Blocks. The precursors used for carbon dot synthesis were fructose and p-phenylenediamine (PPD). A pH 7 buffer solution (50 mM) was prepared using sodium phosphate dibasic heptahydrate and sodium phosphate monobasic monohydrate. Metal salts included aluminium(III) nitrate, silver(I) nitrate, calcium(II) chloride, cobalt(II) nitrate hexahydrate, copper(II) nitrate trihydrate, cadmium(II) nitrate tetrahydrate, iron(III) nitrate nonahydrate, mercury(II) nitrate monohydrate, nickel(II) nitrate hexahydrate, lead(II) nitrate, zinc(II) nitrate hexahydrate, magnesium(II) nitrate, chromium(III) chloride hexahydrate, and manganese(II) chloride. All chemicals were used as received, without additional purification.

X-ray photoelectron spectroscopy (XPS) measurements were conducted on a Kratos Axis Nova spectrometer (Kratos Analytical, UK) equipped with a monochromatised aluminium X-ray source (Al K α , 1486.6 eV) operating at 10 mA and 15 kV (150 W). Survey and high-resolution spectra were collected at detector pass energies of 160 and 20 eV, respectively, and were processed using Thermo Advantage software (v5.9902). Photophysical studies were performed on a PerkinElmer EnSpire Multimode Plate Reader in 300 μ L, 96-well polypropylene microplates (Greiner Bio-One, Item No.: 655209). Berries for CD synthesis were purchased from a local fruit store, and the Jacaranda leaves were collected from the researcher's garden.

4.5.2 Synthesis of biomass-based and fructose-based carbon quantum dots

Carbon dots were synthesised via a hydrothermal method, with specific adjustments made based on the precursor and target doping conditions. For biomass-based CDs, either 5 g of frozen berries or 5 g of fresh Jacaranda leaves were blended with 10 mL of Milli-Q water to produce a juice, which was then transferred to a 50 mL Teflon-lined stainless-steel autoclave and diluted with an additional 40 mL of Milli-Q water, reaching a total volume of 50 mL. For

Jacaranda_(NaOH)_CD and Jacaranda_(NaOH)_(N-doped)_CD, the Milli-Q water was replaced with 0.2 M NaOH solution; additionally, for Jacaranda_(N-doped)_CD and Jacaranda_(NaOH)_(N-doped)_CD, 0.5 g of p-phenylenediamine (PPD) was added to the mixture. In the fructose-based CDs synthesis, 0.5 g of fructose was dissolved in 50 mL of Milli-Q water. For nitrogen-doped fructose-based CDs, a mixture of 0.227 g of fructose and 0.273 g of PPD was dissolved in 50 mL of Milli-Q water and transferred to the autoclave.

The hydrothermal reaction for all CDs was conducted in an oven preheated to 180 °C and maintained for 6 hours. After cooling to room temperature, the solutions were centrifuged for 10 minutes to remove any solid residues, and the supernatants were filtered through a 0.22 µm cellulose acetate filter to remove any larger or aggregated particles. The purified solutions were then concentrated by rotary evaporation and freeze-dried to yield a dark brown powder, which was subsequently reconstituted in Milli-Q water to prepare a 1 mg/mL CD solution for further analysis.

4.5.3 Metal ions detection

To prepare arrays for distinguishing various metal ions, freshly prepared aqueous metal salt stock solutions (100 mM) were combined with aqueous carbon dot sensor stock solutions (1 mg/mL) and Milli-Q water in 300 µL 96-well polypropylene microplates to achieve the desired concentrations. Each well was adjusted to a final volume of 200 µL using a 20 mM phosphate buffer at pH 7.0. Fluorescence emission spectra for each well were recorded from 390 to 750 nm at 5 nm increments upon excitation at 370 nm. Data were collected using a PerkinElmer EnSpire Multimode Plate Reader, with the microplate agitated for 30 seconds prior to measurement.

Detection of Cu(II): To assess the detection capabilities of N-doped F-CDs for Cu(II), a series of Cu(II) solutions were prepared with final concentrations of 0, 10, 20, 40, 60, 80, 100, 200, 400, and 500 µM in Milli-Q water. These solutions were added to the N-doped F-CD solution (200 µL, 1 mg/mL), mixed, and incubated for 10 minutes at room temperature. The fluorescence emission spectra were recorded at an excitation wavelength of 370 nm. Selectivity studies were also conducted by introducing potentially interfering metal ions, including Pb(II), Fe(III), Zn(II), Cd(II), Mg(II), Co(II), Ni(II), Hg(II), Ag(I), Al(III), Mn(II), and Ca(II), under the same experimental conditions.

Detection of Fe(III): For Fe(III) detection, a series of Fe(III) solutions were prepared with final concentrations of 0, 5, 10, 30, 40, 50, 60, 70, and 80 μM in Milli-Q water. These solutions were added to the F-CD solution (200 μL , 1 mg/mL), mixed, and incubated for 10 minutes at room temperature. The fluorescence emission spectra were then recorded at an excitation wavelength of 370 nm.

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5 Carbon dots-based metal ions sensor array

5.1 Introduction of carbon dots-based sensor array for metal ions sensing

Metal ions play a key role in both biological and environmental applications.¹⁻³ In biological systems, metal ions are essential for life, underpinning numerous metabolic processes in every living cell.⁴⁻⁶ Indeed, almost half of all enzymes are dependent on a metal ion for function.⁷ Beyond their vital role in biology, metal ions are integral to various industrial applications, particularly in addressing environmental challenges.^{8, 9} For example, iron and aluminium ions are used in coagulation and flocculation processes to remove suspended particles and organic matter from water, making it clearer and safer for consumption,¹⁰ while iron fertilisation is being explored as a method to stimulate the growth of phytoplankton in the oceans, which can absorb carbon dioxide from the atmosphere and potentially mitigate climate change.¹¹

It is important to note that while metal ions have beneficial roles, they can also be detrimental to both biological and environmental systems. Under specific biological conditions, disruption in the regulation of essential metals leading to imbalanced concentrations can cause cellular damage and contribute to the development of diseases.¹² This dysregulation is associated with many diseases including Alzheimer's and Parkinson's.^{13, 14} Furthermore, heavy metals, which were not encountered by humans during evolution, play no positive physiological role and are associated with significant toxicity.

The complexity of studying and monitoring metals in various matrices, especially when they exist in complex mixtures, is a significant analytical challenge.¹⁵ These mixtures often contain multiple metal species, each with its own unique chemical properties, making detection, analysis, and quantification particularly challenging. Environmental samples, like mining waste,¹⁶ feature metals in diverse chemical states and concentrations, necessitating advanced analytical techniques. In human health, particularly in body fluids such as cerebrospinal fluid,¹⁷ the interaction of metals with biological molecules complicates their analysis due to their influence on bioavailability. These studies highlighted the need for sophisticated methodologies to accurately assess metals in such varied contexts. This intricate nature of metal analysis underscores the importance of developing sensitive, selective, and cost-efficient techniques for metal ion screening to gauge their impact on human health and environmental systems, as well as to mitigate potential adverse effects.

It is therefore crucial to access highly sensitive, selective, and cost-efficient techniques for screening metal ions. This understanding is vital to gauge their impact on human health and environmental systems and to mitigate potential negative consequences. A range of techniques have been developed for metal ion sensing and screening, including colorimetric,¹⁸ electrochemical,¹⁹ and fluorescent sensors.^{20, 21} Electrochemical methods, while widespread, often exhibit lower sensitivity and require extensive sample preparation. Fluorescent methods, in contrast, depend on specific reactions between target metal cations and fluorescent materials. However, traditional fluorescent detection, which uses one sensor per analyte, is hampered by interference from similarly valence states or electron-configured metal ions, and requires as many selective sensors as there are analytes. The capability to simultaneously detect multiple metals in complex solutions is paramount in both environmental and biological contexts. As mentioned above, in environmental samples like mining waste,¹⁶ identifying, and quantifying various metal species concurrently is essential to understand their impact, including toxicity and bioavailability. Similarly, in biological systems, especially in complex fluids such as cerebrospinal fluid,¹⁷ simultaneous metal detection enables a more accurate assessment of their synergistic or antagonistic interactions and their potential health implications. Therefore, advancing analytical methods for concurrent metal detection is crucial, as it enhances the efficiency of analysis and provides deeper insights into the roles and impacts of metals in diverse matrices.²² This advancement is particularly significant for clinical diagnostics and environmental monitoring, where understanding the complex interplay of metals is vital.

Array-based sensing offers an alternative to traditional selective sensing modes for metal ion detection.²³⁻²⁵ Sensor arrays operate by using a combination of cross-reactive, non-selective probes that can differentiate metal ions based on their unique chemical properties.²⁶ Each sensor in the array has a specific chemical moiety that can selectively bind to a specific metal ion, resulting in a unique fluorescent response. The combination of responses from each sensor in the array can provide a unique fingerprint for each metal ion, allowing for the differentiation of metal ions.²⁷ Array-based sensing offers several advantages, including increased selectivity and sensitivity, reduced interference from interfering substances, and the ability to detect multiple metal ions simultaneously.^{25, 26, 28-32} For example, distinguishing between metal ions like Cu(II), Co(II), and Ni(II) is challenging with traditional fluorescent sensors. These ions exhibit similar binding affinities and fluorescence responses due to their comparable coordination chemistry, electronic configurations, and redox potentials.³² Furthermore, interference from other metal ions, organic

compounds, and biological matrices in samples can compromise the accuracy and reliability of sensing results.

One challenge in the development of arrays is the requirement to prepare a relatively large set of sensor elements, which can be synthetically challenging. We and others have demonstrated the use of other methods to introduce diversity of sensor elements, such as by using a single probe in different pH values or different solvents.^{29, 32} We have also focussed on the development of facile reactions that can generate a range of alternate products or different solvents.^{29, 32} We have also focussed on the development of facile reactions that can generate a range of alternate products,³¹ and this brought to our attention the potential of nanomaterials as sensor elements. In particular, we identified that the photophysical and sensing properties of carbon dots (CDs) could be readily tuned by small changes in the synthetic procedure.^{33, 34}

Over the past decade, there has been a remarkable increase in the exploration of carbon dots (CDs) as highly promising nanoprobcs for biosensing and bioimaging applications.³⁵ The surge in interest is primarily driven by the unique optical properties exhibited by CDs, their ease of surface functionalisation, chemical inertness, rapid cellular uptake, low toxicity, and excellent biocompatibility.³⁶ Notably, CDs have demonstrated the ability to rapidly detect targeted molecules with detection limits ranging from the micro- to nanomolar range.³⁷ These features have paved the way for diverse applications of CDs, including biosensors, gene transmission, drug carriers,³⁸ and bioimaging probes.³⁹ The exceptional fluorescence properties of CDs have further contributed to their popularity in analytical chemistry, particularly in the fields of environmental and biological sensing and imaging.⁴⁰

CDs have demonstrated rapid response and superior sensitivity and selectivity towards metal ions in both biological and environmental environment settings, making them a promising alternative to traditional methods.⁴¹⁻⁴³ In previous studies, we developed nitrogen-doped CDs with size-dependent optical properties.³³ We observed a red shift in excitation and emission wavelength as the size of the CDs increased.³⁹ We further enhanced these CDs by conjugating them with fluorescein isothiocyanate, leading to a two-photon dual-emissive sensor with high selectivity for Fe(III) ions.³⁹ The utilisation of these CD-based fluorescent multifunctional probes proved to be a promising approach for achieving precise imaging in deep tissues.³⁹ Over the past decade, there has been a remarkable increase in the exploration of carbon dots (CDs) as highly promising nanoprobcs for biosensing and bioimaging applications.³⁵ The surge in interest is primarily driven

by the unique optical properties exhibited by CDs, their ease of surface functionalisation, chemical inertness, rapid cellular uptake, low toxicity, and excellent biocompatibility.³⁶ Notably, CDs have demonstrated the ability to rapidly detect targeted molecules with detection limits ranging from the micro- to nanomolar range.³⁷ These features have paved the way for diverse applications of CDs, including biosensors, gene transmission, drug carriers,³⁸ and bioimaging probes.³⁹ The exceptional fluorescence properties of CDs have further contributed to their popularity in analytical chemistry, particularly in the fields of environmental and biological sensing and imaging.⁴⁰

To design a highly efficient CD-based metal sensing array, this chapter synthesised a set of novel fluorescent CDs using fructose and p-phenylenediamine (PPD). The synthetic conditions and precursor ratios were varied to yield 22 distinct CDs. This chapter then collected and analysed the fluorescence responses of each CD to 14 metal ions through advanced statistical analysis. Facing the challenge of reducing the sensor elements from an overwhelming pool of over 4 million combinations, we leveraged these statistical methods within a MATLAB-based machine learning framework. This approach automated the exploration of all potential sensor combinations, culminating in the identification of an optimal subset of four sensors achieving 100% classification accuracy. This refined array was subsequently applied in aquatic environmental samples, demonstrating exceptional performance and accuracy.

5.2 Results and discussion

To contextualise the performance differences observed among the CDs investigated in this chapter, it is essential to provide a brief overview of their distinct synthetic origins. The CD library employed for the metal ion sensing array was deliberately developed using a diverse range of carbon sources and synthetic parameters to promote structural and surface functional variation. In contrast to the more targeted approaches described in previous chapters, this study systematically varied the ratios of carbon- and nitrogen-rich precursors, such as fructose and PPD, alongside controlled adjustments to reaction time and temperature. This strategy yielded CDs with tailored emission profiles and a broad distribution of surface functionalities, including $-\text{COOH}$, $-\text{OH}$, and $-\text{NH}_2$ groups, as characterised by FTIR and XPS analyses (see Section 5.2.9). Notably, certain CDs were enriched with amino groups, while others exhibited predominantly hydroxyl or carboxyl functionalisation, directly impacting their interaction with specific metal ions. The synthesis design thereby enabled the generation of a cross-reactive sensor array, capable of distinguishing

between fourteen metal ions based on their differential fluorescence quenching or enhancement behaviours. Further details regarding the synthesis protocols for each CD are provided in Section 5.4.2, but this overview serves to clarify the basis for the sensor performance comparisons that follow.

5.2.1 CD library development

The selection of hydrothermal synthesis parameters, including heating time, temperature, and precursor ratios, is known to significantly impact the ultimate photophysical properties of CDs.³³ As presented in Figure 5.1, we utilised the fluorescence intensity of a series of CDs synthesised at temperatures ranging from 120 °C to 200 °C to determine the optimal synthesis temperature. The CDs exhibited the highest fluorescence intensity at 180 °C; therefore, this temperature was chosen for synthesising the series of CDs. A critical aspect of the synthesis involved adjusting the amount of PPD in the precursor mixture, which comprised fructose and PPD as the major carbon and nitrogen sources, respectively. This adjustment allowed us to tailor the type and ratio of functional groups on the CDs' surface, notably increasing the presence of amino groups when the PPD quantity was raised. We envisaged that these changes would not only affect the photophysical properties of the CDs but also their selectivity towards different metal ions, a key factor in their effectiveness as a sensing array to produce distinct fingerprints for each analyte. Consequently, a series of 22 different CDs were synthesised, each using varied precursor ratios and subjected to either a 6 or 8 h reaction time at 180 °C. After synthesis, these four CDs appear as orange-red solutions and exhibit bright green fluorescence under a UV lamp. After freeze-drying for 48 h, a dark brown dried powder of CDs was collected.

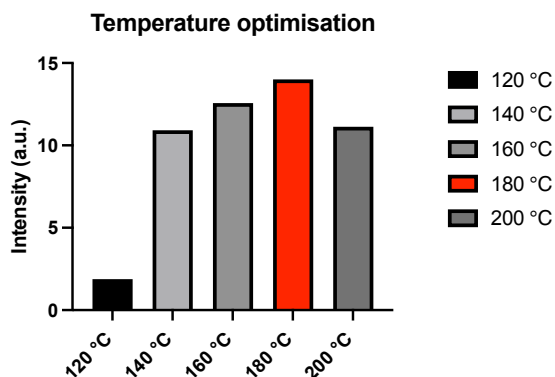


Figure 5.1 Synthesis temperature optimisation of carbon dots' hydrothermal reaction

In terms of synthetic yield and product characteristics, the hydrothermal reaction generally achieved a yield of 85%. While altering the PPD ratio did not significantly change the yield, it did have a significant impact on the CDs' properties. An increase in PPD ratio rendered the CDs more hydrophobic compared to those synthesised solely with fructose, which exhibited greater hydrophilicity. Moreover, CDs synthesised with higher levels of PPD participation were found to be more amenable to freeze-drying, facilitating the production of a dry product. On the other hand, CDs produced exclusively from fructose exhibited a tendency to absorb moisture from the environment during storage. The diverse characteristics of these CDs, influenced by their distinct synthesis conditions, underpin their potential as a versatile sensor array for metal ion detection.

5.2.2 Response of CD fluorescence to metal ions

Figure S5.1 presents the summarised fluorescence emission spectra of carbon dots (CDs) under an excitation wavelength of 370 nm. The naked eye observation of the addition of different metal ions to these CDs meeting is shown in Figure S5.2. These CDs displayed emission peaks spanning from 390 nm to 700 nm, with the most emissive peaks observed in the range of 440 nm to 530 nm. For CDs synthesised from identical proportions of fructose and *p*-phenylenediamine (PPD) but subjected to varying synthesis time (6 h or 8 h), the emission spectra exhibited consistent main emission peaks and shoulder peaks. The variation in emission peaks among different CDs can be attributed primarily to differences in the precursor composition. These compositional differences lead to variations in nitrogen (N)-doping levels and the nature of functional groups present on the surface of each CD. Such modifications in the surface functionality directly influence the photoluminescence properties, explaining the observed spectral differences.

CDs serve as potential candidates for metal ion sensing due to their unique fluorescent properties. In this study, we examined the fluorescent responses of 22 different CDs against 14 metal ions. The data were categorised based on whether the fluorescence intensity was enhanced (turn-on) or reduced (turn-off), as shown in Figure 5.2.

From the synthesised CDs library, some of the CDs were characterised by an increase in fluorescence intensity when exposed to certain metal ions, a phenomenon predominantly observed in those classified under the turn-on category. CD_1-0_6 h and CD_3-17_8 h stand out as frontrunners, displaying particularly strong turn-on reactions across multiple metal ions. For instance, they show pronounced responses to Al(III), Ca(II), and Cd(II). Although some CDs, such

as CD_9-1_6 h and CD_4-1_6 h, exhibit slight turn-on behaviour with specific metal ions like Ag(I) and Ni(II).

Several CDs exhibit a decrease in fluorescence intensity upon interaction with specific metal ions. For instance, CD_1-4_6 h demonstrates a strong turn-off response to Co(II), suggesting its potential as a Co(II) sensor. Likewise, CD_4-2_6 h exhibits notable fluorescence reduction when exposed to Hg(II). CDs offer a versatile platform for metal ion detection, with their ability to produce varied fluorescence responses. The distinct characteristics of different CDs underscore their crucial role in the field of fluorescence-based metal ion detection. Owing to their ability to produce varied fluorescence responses, CDs present a versatile platform for metal ion sensing. Importantly, no sensor was selective for a single metal ion, making them ideal for a cross-reactive array approach. This versatility and selectivity variation laid the groundwork for the development of our comprehensive metal sensing array, which employed advanced statistical methods for enhanced accuracy and efficiency.

For all metal ion fluorescence measurements presented in this chapter, blank controls were included in each experimental set to establish baseline emission intensities. These controls comprised CD solutions at a fixed concentration of 1 mg/mL in buffer without the addition of metal ions, thereby ensuring that any observed changes in fluorescence could be directly attributed to metal ion interactions rather than dilution effects or environmental artefacts. To maintain consistency across all test conditions, each fluorescence solution—including both test and control samples—was prepared with identical total volume and solvent composition. Metal ion stock solutions were introduced in small volumes ($\leq 5 \mu\text{L}$), such that the final CD concentration remained unchanged. Unless otherwise specified, all fluorescence response data were collected using CD solutions at 1 mg/mL, with metal ion solutions added to achieve a final concentration of 80 μM . This experimental design ensured that fluorescence variations arose solely from analyte-specific quenching or enhancement, rather than from changes in CD concentration. The reliability of this approach was further confirmed through replicate measurements and comparative analysis of control spectra.

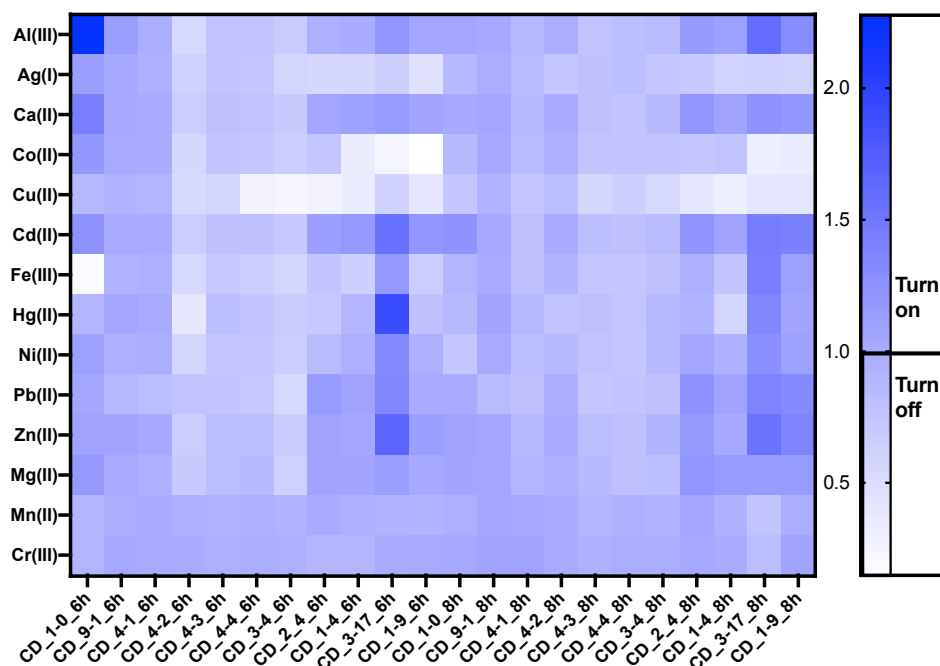


Figure 5.2 Heat map of the fluorescence response of carbon dots during metal ions sensing. The fluorescence response values are derived from the ratio of the integral of the fluorescence spectrum of the carbon dots (CDs) post-interaction with the metal analyte to the integral of the fluorescence spectrum of the CDs in the absence of the analyte. These values were obtained using an excitation wavelength (λ_{ex}) of 370 nm and an emission wavelength range (λ_{em}) of 390–750 nm. A ratio greater than one indicates a 'turn-on' fluorescence response upon encountering the analyte, while a ratio less than one signifies a 'turn-off' response. Values close to one represent no significant fluorescence response to the analyte. (80 μ M metal ion, 1 mg/mL CDs solutions)

5.2.3 LDA classification with whole library carbon dots

In this study, two major statistical techniques were used for classification tasks to improve the array's performance. They are linear discriminant analysis (LDA) and support vector machines (SVM), introduced in Chapter 2 2.2. LDA classification, a supervised classification method, involves finding a linear combination of features that characterise or separate two or more classes of objects. LDA seeks to maximise the distance between classes while minimising the variance within each class. It assumes that the data are normally distributed, and the covariance matrices of the classes are equal. LDA can be used for both binary and multiclass classification problems. SVM classification is also a supervised classification method that involves finding a hyperplane that separates two classes of data points. The hyperplane is found by maximising the margin, which is the distance between the hyperplane and the closest data points of each class. SVM can handle both linearly separable and nonlinearly separable data, by using a kernel function to map the data into a higher-dimensional space where the data may be linearly separable. Given the varied

fluorescent responses of CDs during metal ion sensing, we initially employed SPSS-based LDA to examine the array's performance by using all twenty-two CDs to discriminate 14 metal ions. The workflow is illustrated in Figure 5.3.

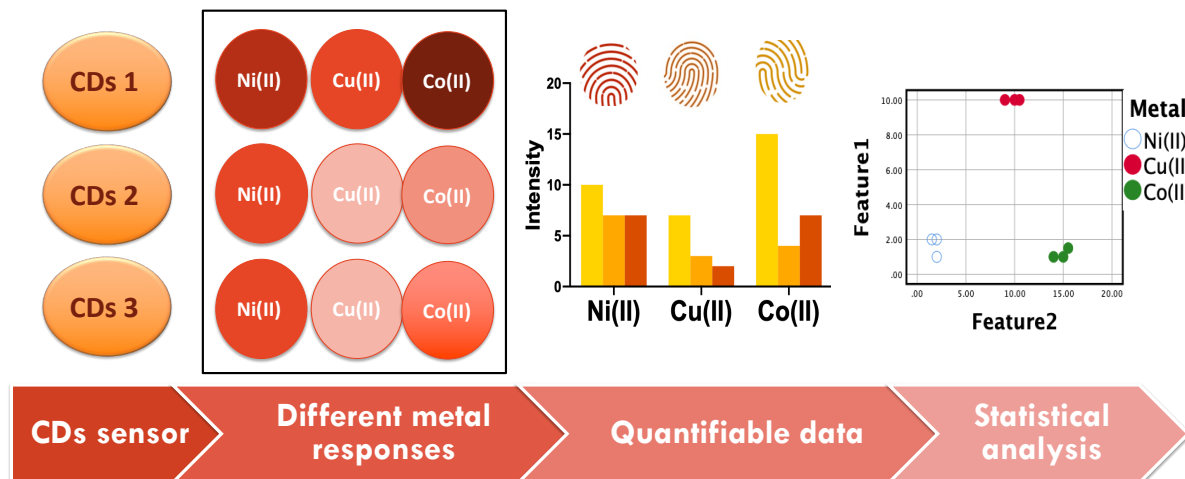


Figure 5.3 A schematic representation of a carbon dots-based sensing array for the discrimination of metal ions. Cu(II), Co(II) and Ni(II) are given as example analytes. Each sensor gives a different and characteristic set of responses to each metal ion. These can be quantified to give the fingerprint response for each metal analyte, which can be analysed by statistical methods to maximise discrimination of each analyte.

For each sensor, the quadruplicate measurements for each metal ion (56 cases in total) were processed and included in the analysis without exclusions. In our analysis involving 14 different metal ion targets, we assigned an equal prior probability to each metal ion. This implies that, under the assumption of our model, each type of metal ion was equally likely to be the analyte in any given test case. This uniform distribution of probabilities aligns with the design of our experiment, where each metal ion type was equally represented. As a result, 100.0% of original grouped cases correctly classified (Figure 5.4). Cross-validation was conducted to evaluate the array's performance. In cross validation, each case was classified by the functions derived from all cases other than that case, resulted as 96.4% of cross-validated grouped cases correctly classified. One case of Mg(II) sensing was miss identified as Ca(II), and one case of Cr(III) was miss identified as Mn(II), as shown in Supporting material Table S5.1.

According to the canonical discriminant functions, eigenvalues show how much variance is captured by each function.⁴⁴ The first function captures 70.0% of the variance. Followed by the second function with 22.0 %. Cumulatively, by the third function (3.3%), 95.3% of the variance in the dataset is accounted for. This means the first three functions are very significant in

distinguishing between groups. The canonical correlations for the first three functions were 1.000, 0.999 and 0.992 for the first three functions. They are close to 1, indicating a strong relationship between the groups and variables.⁴⁵ Wilks' Lambda measures the extent to which the means of the dependent variables differ between groups. Smaller values mean larger differences.⁴⁶ In this analysis, all the functions have very small Wilks' Lambda values (close to 0), suggesting that the group means differ significantly, as listed in Table 5.1. In conclusion, the LDA performed extremely well in classifying the metal ions. The first three discriminant functions were particularly significant in differentiating the groups. Our synthesised CDs have proven to be excellent choices, demonstrating effectiveness in distinguishing between various metal ions.

Table 5.1 Summary of canonical discriminant function of 22 CDs based array for 14 metal ions' LDA classification

Eigenvalues				
Function	Eigenvalue	Variance (%)	Cumulative (%)	Canonical Correlation
1	1407.675 ^a	70	70	1
2	443.514 ^a	22	92	0.999
3	65.419 ^a	3.3	95.3	0.992
4	38.593 ^a	1.9	97.2	0.987
5	29.481 ^a	1.5	98.6	0.983
6	11.278 ^a	0.6	99.2	0.958
7	6.615 ^a	0.3	99.5	0.932
8	5.243 ^a	0.3	99.8	0.916
9	1.780 ^a	0.1	99.9	0.8
10	1.207 ^a	0.1	99.9	0.74
11	.709 ^a	0	100	0.644
12	.456 ^a	0	100	0.56
13	.139 ^a	0	100	0.349
a. First 13 canonical discriminant functions were used in the analysis.				
Wilks' Lambda				
Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1 through 13	0	1252.978	286	<.001
2 through 13	0	984.713	252	<.001
3 through 13	0	759.125	220	<.001
4 through 13	0	603.873	190	<.001

5 through 13	0	467.763	162	<.001
6 through 13	0	341.329	136	<.001
7 through 13	0.001	248.541	112	<.001
8 through 13	0.009	173.426	90	<.001
9 through 13	0.058	105.664	70	0.004
10 through 13	0.16	67.83	52	0.069
11 through 13	0.353	38.534	36	0.356
12 through 13	0.603	18.712	22	0.663
13	0.878	4.813	10	0.903

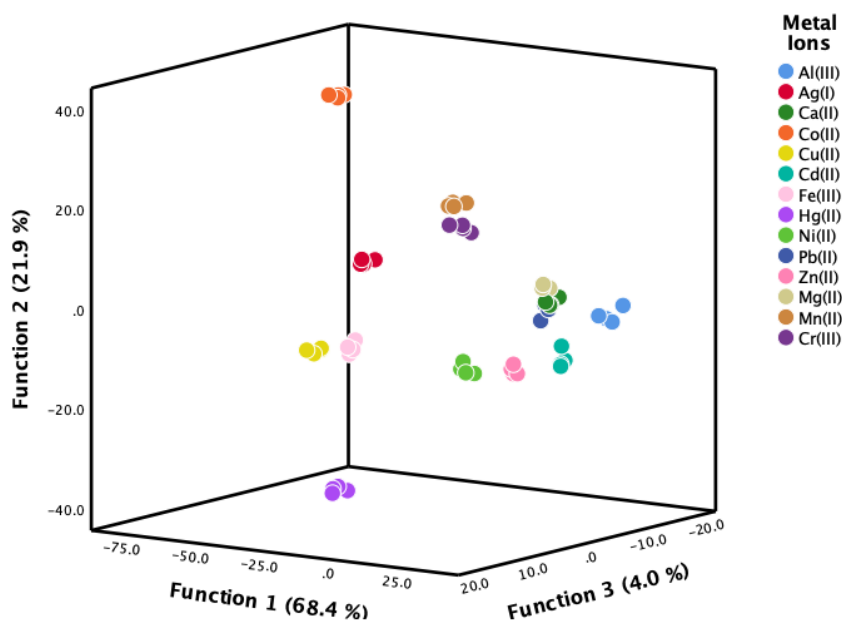


Figure 5.4 LDA classification of 22 CDs based array for 14 metal ions classification with 100% classification accuracy and 96.4% cross validation accuracy. (80 μ M metal ion, 1 mg/mL CDs solutions, with four replicates)

5.2.4 SVM and LDA based array optimising MATLAB program

After confirming the metal ions classification performance with our synthesised CDs, we then shifted our focus to further optimising the array. This optimisation involved implementing a dimensionality reduction to streamline. We wanted to reduce the number of CDs used for our metal sensor array. We aimed to find the optimised array containing fewest number of CDs and capable to provide 100% classification accuracy to classify 14 metal ions. To achieve this, we designed an ML-based MATLAB program. The first section of the program utilised LIBSVM and LIBSVM-FarutoUltimate toolboxes for SVM classification evaluation.⁴⁷⁻⁴⁹ Then, for the second section of the program, we implanted LDA classification to verify the SVM optimisation results. As a result,

we achieved a list of array combinations which could performance 100% classification accuracy for both SVM and LDA.

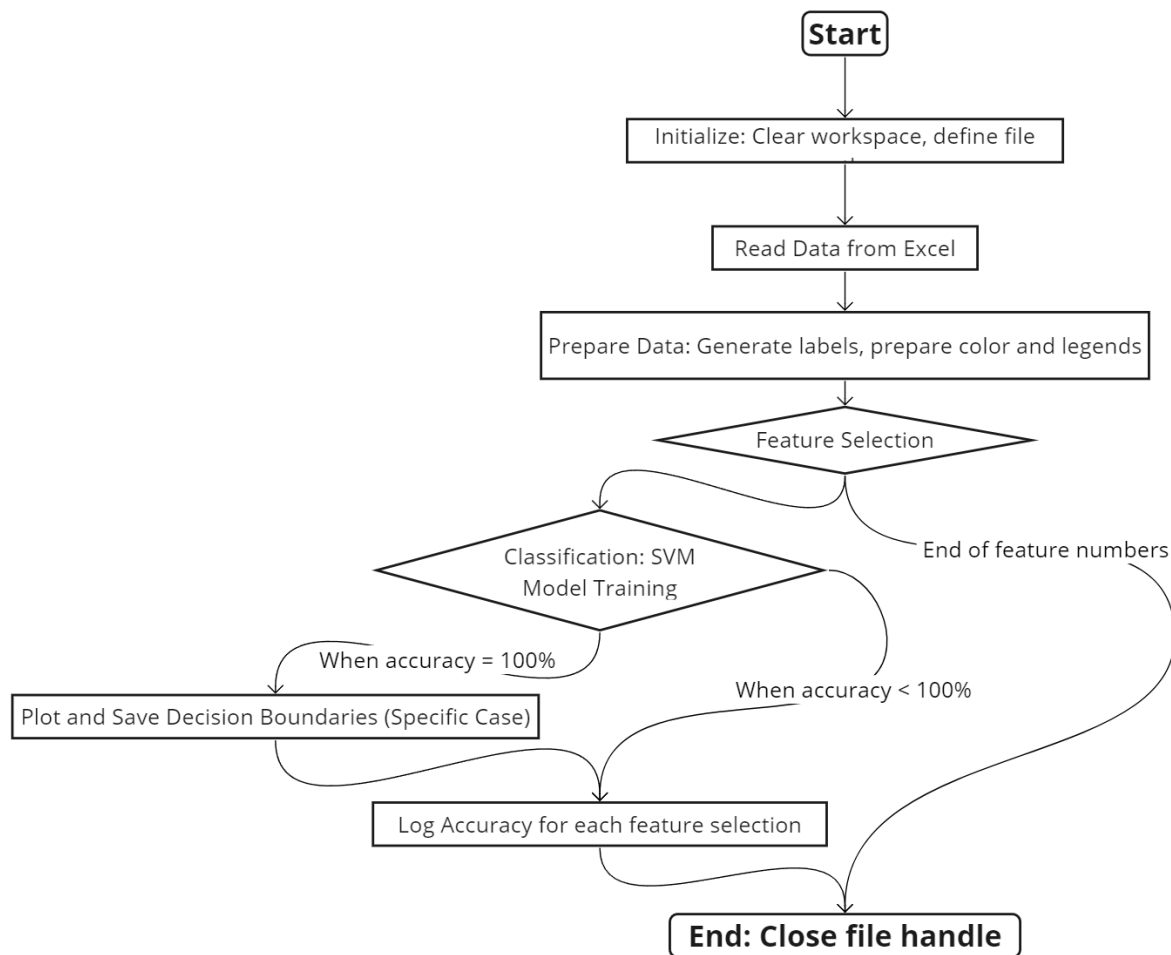
5.2.5 MATLAB based SVM classification for array optimisation

5.2.5.1 Coding development and working mechanism

In the development of practical sensor arrays, minimising the number of sensor elements, while ensuring their efficacy, is paramount. This necessity is driven by the need to reduce costs, simplify complexity, and enhance operational efficiency. Preliminary observations from Figure 5.2, coupled with correlations discerned through statistical analysis, suggested potential redundancies in sensor responses. To empirically ascertain the most efficacious sensor combination, a more rigorous methodology was employed.

Our MATLAB programme was specifically developed to address this challenge, aiming to systematically diminish the sensor count in the array while being guided by their fluorescence responses to the fourteen target metal ions. The programme's algorithm assessed all conceivable combinations of the 22 CD sensors, totalling 4,194,303 potential selections. For each combination, the programme methodically executed a sequence of steps: it computed and contrasted the SVM classification outcomes, entailing the training and testing of the SVM model with our dataset (Scheme 5.1).

This detailed procedure was focused not merely on identifying a combination with a reduced number of sensors but on pinpointing the optimal set. By concentrating on the SVM and LDA classification efficacy of each combination, our programme successfully identified the most effective and efficient subset of sensors. This practical solution to sensor array design is a significant progression from initial statistical correlations, offering a more definitive and application-focused approach in sensor array optimisation. The designed MATLAB programme serves to perform SVM and LDA classification on a given dataset for metal analytes. The dataset is represented as an Excel file, where each row corresponds to a sample (metal analyte) and each column represents a feature (CD sensor). The program incorporates feature selection and hyperparameter optimisation techniques to enhance the SVM model's classification accuracy through cross-validation.



Scheme 5.1 Carbon dots-based array for metal ions classification by applying the MATLAB based SVM classifier.

The code (given in supporting material 7.3.3) begins by reading the Excel file containing the dataset using the *xlsread* function. It then sets up the class labels for the dataset and creates a 4-fold cross-validation object using the *crossvalind* function.

Next, the code initialises some plotting variables and loops over the feature numbers from 1 to 20, indicating the number of features that will be selected for the SVM model. For each feature number, it creates all possible combinations of the given number of features using the *nchoosek* function.

Then, for each combination of features, the code uses the *SVMcgForClass* function to perform hyper-parameter optimisation on the SVM model using the training data, and then trains and evaluates the SVM model using the optimised hyper-parameters and testing data. This process is repeated for each of the 4-folds of the cross-validation.

If the selected feature combination has only two features and it is the first fold, the code creates a 2D plot of the SVM decision boundary using the *gscatter* function and saves it in a specified directory.

Finally, the code outputs the classification accuracy for each feature combination and feature number to a text file named *result.txt*. If any feature combination achieves a 100% classification accuracy, the code logs the combination to the text file.

In terms of the design principles, this code follows the principles of modularity and abstraction by breaking down the classification task into smaller subtasks that are handled by separate functions. It also uses loop structures to iterate over different combinations of features and hyper-parameters, reducing code redundancy and increasing code efficiency. Additionally, the code employs cross-validation techniques to validate the accuracy of the SVM model, which is a standard approach in machine learning.

5.2.5.2 Array simplification results

Based on the SVM classification accuracy results generated by our program (Figure 5.5), it was evident that as the number of CDs in the array increased, both the average SVM classification accuracy and the number of array selections achieving 100% accuracy also increased. The results of array simplification, produced by our MATLAB program, are detailed in the supporting material section 7.3.3.

For arrays consisting of one or two CDs, no combination achieved 100% SVM classification accuracy. However, for arrays based on three CDs, four combinations were identified that provided 100% classification accuracy with SVM. The sensing performances of these combinations were further validated using SPSS-based LDA, as shown in Table 5.2 and Table S5.2-5. Despite the excellent metal sensing performance with three-CD arrays, none of the combinations achieved 100% accuracy for both original classification and cross-validated classification using LDA in SPSS. Consequently, we shifted our focus to arrays using four CDs, aiming to identify an optimal combination that could achieve 100% accuracy in both LDA and SVM classifications. As a result, forty-one four-CD array combinations achieving 100% SVM classification accuracy were identified and will be further validated, analysed, and discussed in the following sections, along with results from the second section of the MATLAB program using LDA classification (see section 5.2.6).

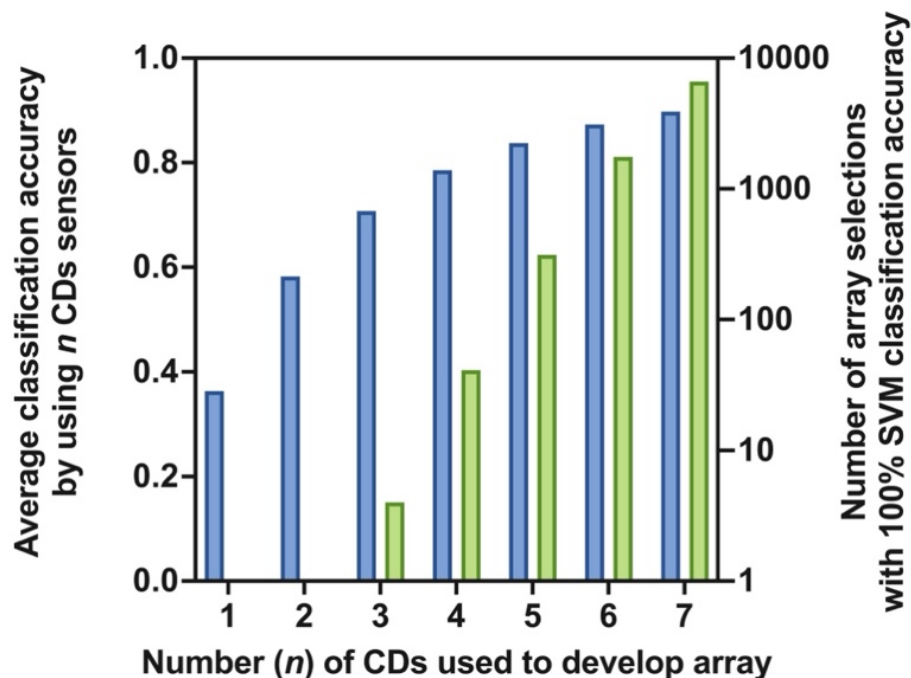


Figure 5.5 Carbon dots-based metal sensing array simplification results, where blue bars (left y axis) represented the average classification accuracy by using n CDs sensing as array to classify 14 metal ions. With the number n increase, the average classification accuracy was increased; The green bars (right y axis) represent the number of array selections which achieved 100% SVM classification accuracy

Table 5.2 SPSS based LDA classification verification of three CDs-based array selections

Array selections	original classification accuracy (%)	cross-validated classification accuracy (%)
CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h	91.1	82.1
CD_2-4_6 h, CD_1-4_6 h, CD_1-9_6 h	98.2	94.6
CD_2-4_6 h, CD_1-9_6 h, CD_9-1_8 h	94.6	89.3
CD_2-4_6 h, CD_1-9_6 h, CD_1-4_8 h	100	96.4

From the above results we learn that in our quest to develop most optimised metal sensor arrays based on CDs, previous analysis shows that arrays with fewer than four sensors cannot achieve 100% classification accuracy, for both original and cross validation classification. Therefore, we focus on a four-CDs configuration, which has shown considerable promise. Our MATLAB-based SVM classification identified 41 four-CD arrays as detailed in

Table 5.3. In the following sections, we will introduce LDA classification based on MATLAB we applied to find the optimal four-CDs based metal sensing array, by introducing parameters that can quantify the classification performance (seen section 5.2.6).

Table 5.3 Summary of the 4 CDs based array with 100% classification accuracy given by MATLAB based SVM classifier

NO.	Carbon dots array combination	NO.	Carbon dots array combination
1	CD_6h_1_0 CD_6h_4_1 CD_6h_2_4 CD_8h_3_17	22	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_1_9
2	CD_6h_1_0 CD_6h_4_1 CD_6h_1_4 CD_6h_3_17	23	CD_6h_1_0 CD_6h_1_9 CD_8h_4_1 CD_8h_1_4
3	CD_6h_1_0 CD_6h_4_1 CD_6h_1_9 CD_8h_3_17	24	CD_6h_1_0 CD_6h_1_9 CD_8h_4_2 CD_8h_1_9
4	CD_6h_1_0 CD_6h_4_3 CD_6h_1_9 CD_8h_1_9	25	CD_6h_1_0 CD_6h_1_9 CD_8h_4_3 CD_8h_1_4
5	CD_6h_1_0 CD_6h_4_4 CD_6h_1_9 CD_8h_4_2	26	CD_6h_1_0 CD_6h_1_9 CD_8h_4_4 CD_8h_1_9
6	CD_6h_1_0 CD_6h_4_4 CD_6h_1_9 CD_8h_1_4	27	CD_6h_1_0 CD_6h_1_9 CD_8h_2_4 CD_8h_3_17
7	CD_6h_1_0 CD_6h_3_4 CD_6h_1_9 CD_8h_9_1	28	CD_6h_1_0 CD_6h_1_9 CD_8h_1_4 CD_8h_3_17
8	CD_6h_1_0 CD_6h_2_4 CD_6h_1_4 CD_6h_1_9	29	CD_6h_1_0 CD_6h_1_9 CD_8h_3_17 CD_8h_1_9
9	CD_6h_1_0 CD_6h_2_4 CD_6h_1_4 CD_8h_3_17	30	CD_6h_4_1 CD_6h_2_4 CD_6h_1_4 CD_8h_1_4
10	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_9_1	31	CD_6h_4_1 CD_6h_2_4 CD_6h_3_17 CD_6h_1_9
11	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_4_2	32	CD_6h_4_1 CD_6h_2_4 CD_6h_1_9 CD_8h_9_1
12	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_2_4	33	CD_6h_4_1 CD_6h_2_4 CD_6h_1_9 CD_8h_1_4
13	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_1_4	34	CD_6h_3_4 CD_6h_2_4 CD_6h_1_4 CD_6h_1_9
14	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_3_17	35	CD_6h_3_4 CD_6h_1_4 CD_6h_3_17 CD_8h_3_17
15	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_1_9	36	CD_6h_3_4 CD_6h_3_17 CD_6h_1_9 CD_8h_3_17
16	CD_6h_1_0 CD_6h_1_4 CD_6h_3_17 CD_8h_1_9	37	CD_6h_2_4 CD_6h_1_4 CD_6h_3_17 CD_6h_1_9
17	CD_6h_1_0 CD_6h_1_4 CD_6h_1_9 CD_8h_3_17	38	CD_6h_2_4 CD_6h_1_9 CD_8h_9_1 CD_8h_4_2
18	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_4_1	39	CD_6h_2_4 CD_6h_1_9 CD_8h_3_17 CD_8h_1_9
19	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_4_4	40	CD_6h_1_4 CD_6h_1_9 CD_8h_3_17 CD_8h_1_9
20	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_1_4	41	CD_6h_3_17 CD_8h_9_1 CD_8h_1_4 CD_8h_3_17
21	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_3_17		

5.2.6 MATLAB based LDA classification for array optimisation and verification

After SVM classification listed the optimal four-CDs based array, MATLAB based LDA programs were designed to explore and verify the metal four-CDs sensing performance of the based metal sensing array combinations.

5.2.6.1 LDA based array optimising results for four-CDs based array

We designed a MATLAB based LDA program (MATLAB based LDA classifier A as shown in the supporting material 3.3.3) to carry out LDA classification to verify the classification performance of the 41 CDs combinations given by the previous SVM program. As a result, 16 out of 41 combinations of the four-CDs based metal sensing array achieved 100% classification

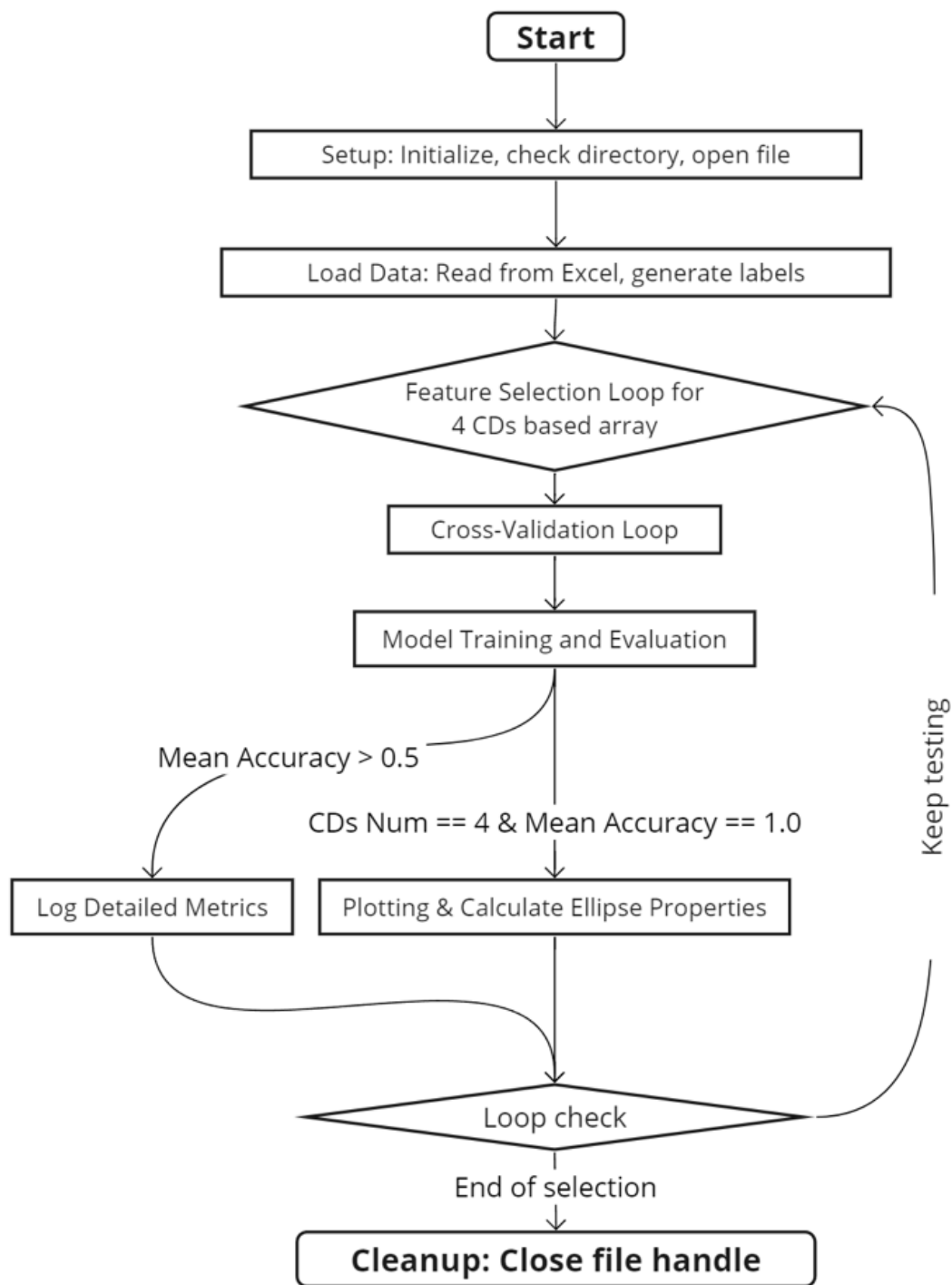
accuracy, which significantly narrowed the scope of the array selections. The working mechanism of LDA classifier A is shown in Scheme 5.2, and the working mechanism of the classifier is explained in supporting material 3.3.3.

5.2.6.2 Single CDs based LDA classification for CDs' sensing performance analysis

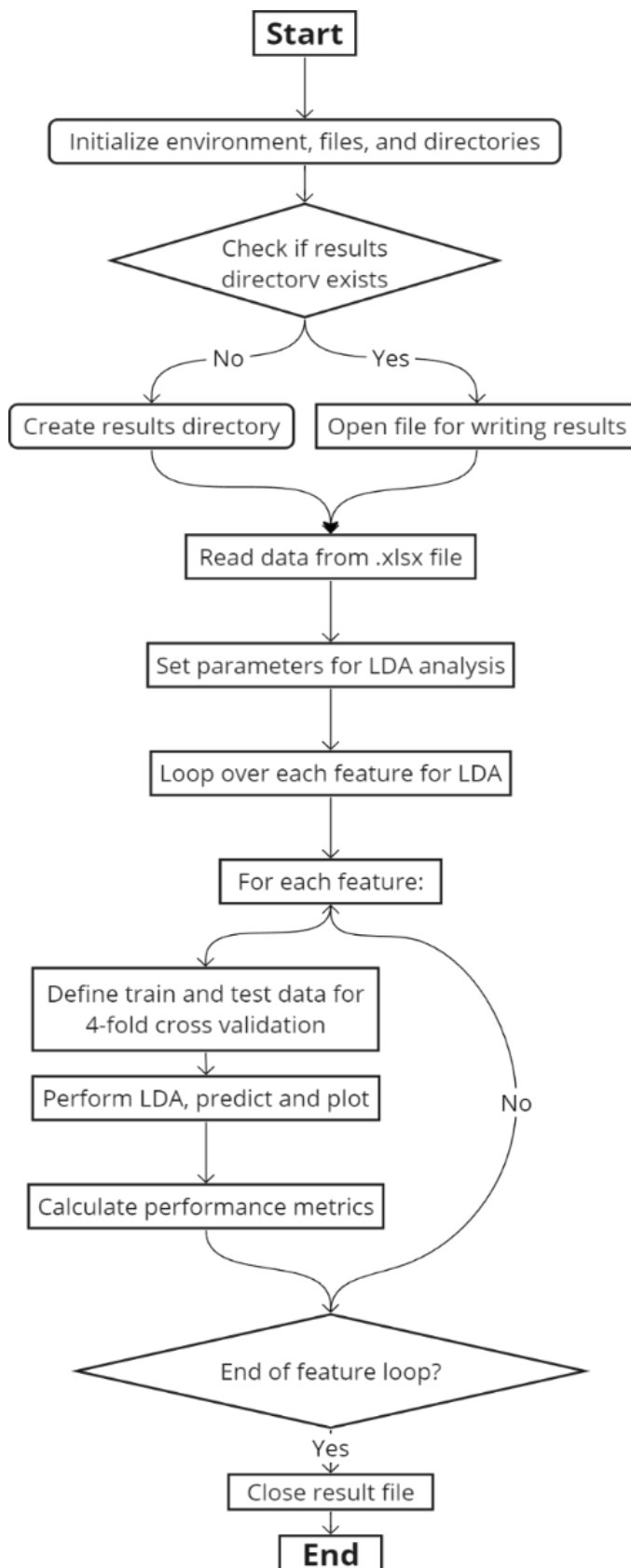
We also developed a MATLAB based program (MATLAB based LDA classifier B as shown in supporting material 3.3.4) implanted with the LDA classifier to investigate the performance of single CDs based metal ions sensing and summarised their classification accuracy in Table 5.4. From the accuracy studies given by LDA classifier B, the mean accuracies of CDs synthesised under 6 h were generally higher than the CDs synthesised under 8 h. The working mechanism of our MATLAB program has been illustrated in Scheme 5.3, the working mechanism of the classifier was explained in supporting material 3.3.4. As listed in Table 5.1, CD_2_4_6 h and CD_1-9_6 h achieved the highest mean accuracy by using them for one CD based LDA classification.

Table 5.4 Single CDs based metal ions classification by MATLAB based LDA classifier

No.	Carbon dots	Mean Accuracy	No.	Carbon dots	Mean Accuracy
1	CD_1-0_6 h	0.428571	12	CD_1-0_8 h	0.214286
2	CD_9-1_6 h	0.196429	13	CD_9-1_8 h	0.232143
3	CD_4-1_6 h	0.160714	14	CD_4-1_8 h	0.196429
4	CD_4-2_6 h	0.482143	15	CD_4-2_8 h	0.25
5	CD_4-3_6 h	0.285714	16	CD_4-3_8 h	0.339286
6	CD_4-4_6 h	0.303571	17	CD_4-4_8 h	0.232143
7	CD_3-4_6 h	0.357143	18	CD_3-4_8 h	0.25
8	CD_2_4_6 h	0.678571	19	CD_2_4_8 h	0.267857
9	CD_1-4_6 h	0.517857	20	CD_1-4_8 h	0.410714
10	CD_3-17_6 h	0.535714	21	CD_3-17_8 h	0.446429
11	CD_1-9_6 h	0.714286	22	CD_1-9_8 h	0.553571



Scheme 5.2 Four CDs based array for metal ions classification by using MATLAB based LDA classifier A.



Scheme 5.3 Single Carbon dot-based metal ions sensing by MATLAB based LDA classifier *B*

5.2.6.3 Optimisation results of MATLAB based LDA classifier

The MATLAB-based LDA classifier analysed all possible combinations of the four CDs in the sensor array, identifying 73 combinations with 100% LDA classification accuracy, as listed in Table 5.5. Furthermore, 16 four-CD sensor array combinations achieved 100% classification accuracy with both SVM and LDA classifiers as based on MATLAB, listed in Table 5.6.

Table 5.5 Summary of the 4 CDs based array with 100% classification accuracy given by MATLAB based LDA classifier

NO.	Carbon dots array combination	NO.	Carbon dots array combination
1	CD_1-0_6 h, CD_9-1_6 h, CD_3-17_6 h, CD_1-9_6 h	38	CD_1-0_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_1-4_8 h
2	CD_1-0_6 h, CD_9-1_6 h, CD_1-9_6 h, CD_9-1_8 h	39	CD_1-0_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_3-17_8 h
3	CD_1-0_6 h, CD_9-1_6 h, CD_1-9_6 h, CD_3-17_8 h	40	CD_1-0_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_4-2_8 h
4	CD_1-0_6 h, CD_4-2_6 h, CD_3-17_6 h, CD_1-9_6 h	41	CD_1-0_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_2-4_8 h
5	CD_1-0_6 h, CD_4-2_6 h, CD_1-9_6 h, CD_9-1_8 h	42	CD_1-0_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_1-4_8 h
6	CD_1-0_6 h, CD_4-2_6 h, CD_1-9_6 h, CD_4-1_8 h	43	CD_1-0_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_3-17_8 h
7	CD_1-0_6 h, CD_4-2_6 h, CD_4-3_6 h, CD_1-9_6 h	44	CD_1-0_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_1-9_8 h
8	CD_1-0_6 h, CD_4-2_6 h, CD_2-4_6 h, CD_1-9_6 h	45	CD_1-0_6 h, CD_1-9_6 h, CD_4-2_8 h, CD_4-3_8 h
9	CD_1-0_6 h, CD_4-3_6 h, CD_1-9_6 h, CD_3-17_8 h	46	CD_1-0_6 h, CD_1-9_6 h, CD_4-2_8 h, CD_1-9_8 h
10	CD_1-0_6 h, CD_4-3_6 h, CD_2-4_6 h, CD_1-9_6 h	47	CD_1-0_6 h, CD_1-9_6 h, CD_4-3_8 h, CD_3-17_8 h
11	CD_1-0_6 h, CD_4-3_6 h, CD_1-4_6 h, CD_1-9_6 h	48	CD_1-0_6 h, CD_1-9_6 h, CD_4-4_8 h, CD_3-17_8 h
12	CD_1-0_6 h, CD_3-4_6 h, CD_3-17_6 h, CD_1-9_6 h	49	CD_1-0_6 h, CD_1-9_6 h, CD_1-4_8 h, CD_1-9_8 h
13	CD_1-0_6 h, CD_2-4_6 h, CD_3-17_6 h, CD_1-9_6 h	50	CD_1-0_6 h, CD_1-9_6 h, CD_3-17_8 h, CD_1-9_8 h
14	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_1-0_8 h	51	CD_4-2_6 h, CD_4-3_6 h, CD_1-9_6 h, CD_4-3_8 h
15	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_9-1_8 h	52	CD_4-2_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_1-4_8 h
16	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_4-1_8 h	53	CD_4-2_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_1-9_8 h
17	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_4-2_8 h	54	CD_4-2_6 h, CD_2-4_6 h, CD_1-4_6 h, CD_1-9_6 h
18	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_3-4_8 h	55	CD_4-2_6 h, CD_1-4_6 h, CD_3-17_6 h, CD_1-4_8 h
19	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_2-4_8 h	56	CD_4-2_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_3-17_8 h
20	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_1-4_8 h	57	CD_4-2_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_1-4_8 h
21	CD_1-0_6 h, CD_2-4_6 h, CD_1-9_6 h, CD_1-9_8 h	58	CD_4-2_6 h, CD_1-9_6 h, CD_1-4_8 h, CD_1-9_8 h
22	CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, CD_1-9_6 h	59	CD_4-2_6 h, CD_1-9_6 h, CD_3-17_8 h, CD_1-9_8 h
23	CD_1-0_6 h, CD_1-4_6 h, CD_3-17_6 h, CD_1-9_6 h	60	CD_4-3_6 h, CD_3-4_6 h, CD_2-4_6 h, CD_1-9_6 h
24	CD_1-0_6 h, CD_1-4_6 h, CD_3-17_6 h, CD_1-4_8 h	61	CD_4-3_6 h, CD_2-4_6 h, CD_3-17_6 h, CD_1-9_6 h
25	CD_1-0_6 h, CD_1-4_6 h, CD_1-9_6 h, CD_9-1_8 h	62	CD_2-4_6 h, CD_1-4_6 h, CD_3-17_6 h, CD_1-9_6 h
26	CD_1-0_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_9-1_8 h	63	CD_2-4_6 h, CD_1-4_6 h, CD_3-17_6 h, CD_1-0_8 h
27	CD_1-0_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_4-1_8 h	64	CD_2-4_6 h, CD_1-4_6 h, CD_3-17_6 h, CD_9-1_8 h
28	CD_1-0_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_4-3_8 h	65	CD_2-4_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_4-3_8 h

NO.	Carbon dots array combination	NO.	Carbon dots array combination
29	CD_1-0_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_1-4_8 h	66	CD_2-4_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_2-4_8 h
30	CD_1-0_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_3-17_8 h	67	CD_2-4_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_3-17_8 h
31	CD_1-0_6 h, CD_3-17_6 h, CD_1-9_6 h, CD_1-9_8 h	68	CD_2-4_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_4-2_8 h
32	CD_1-0_6 h, CD_1-9_6 h, CD_1-0_8 h, CD_9-1_8 h	69	CD_2-4_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_1-4_8 h
33	CD_1-0_6 h, CD_1-9_6 h, CD_1-0_8 h, CD_4-1_8 h	70	CD_2-4_6 h, CD_1-9_6 h, CD_4-4_8 h, CD_3-17_8 h
34	CD_1-0_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_4-1_8 h	71	CD_2-4_6 h, CD_1-9_6 h, CD_3-17_8 h, CD_1-9_8 h
35	CD_1-0_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_4-2_8 h	72	CD_1-4_6 h, CD_3-17_6 h, CD_1-4_8 h, CD_3-17_8 h
36	CD_1-0_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_4-3_8 h	73	CD_1-4_6 h, CD_1-9_6 h, CD_4-1_8 h, CD_3-17_8 h
37	CD_1-0_6 h, CD_1-9_6 h, CD_9-1_8 h, CD_4-4_8 h		

Table 5.6 Summary of the 4 CDs based array with 100% classification accuracy given by both MATLAB based SVM classifier and LDA classifier

NO.	Carbon dots array combination	NO.	Carbon dots array combination
1	CD_6h_1_0 CD_6h_2_4 CD_6h_1_4 CD_6h_1_9	9	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_1_4
2	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_9_1	10	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_3_17
3	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_4_2	11	CD_6h_1_0 CD_6h_1_9 CD_8h_4_1 CD_8h_1_4
4	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_2_4	12	CD_6h_1_0 CD_6h_1_9 CD_8h_4_2 CD_8h_1_9
5	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_1_4	13	CD_6h_1_0 CD_6h_1_9 CD_8h_3_17 CD_8h_1_9
6	CD_6h_1_0 CD_6h_2_4 CD_6h_1_9 CD_8h_1_9	14	CD_6h_2_4 CD_6h_1_4 CD_6h_3_17 CD_6h_1_9
7	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_4_1	15	CD_6h_2_4 CD_6h_1_9 CD_8h_9_1 CD_8h_4_2
8	CD_6h_1_0 CD_6h_1_9 CD_8h_9_1 CD_8h_4_4	16	CD_6h_2_4 CD_6h_1_9 CD_8h_3_17 CD_8h_1_9

The frequency of each carbon dot in the combinations identified by the SVM and LDA classifiers is summarised in Figure 5.6 and Table S5.7 in supporting material. These occurrences reflect the frequency with which each carbon dot was included in the array for metal ion discrimination. Notably, CD_1-9_6h, CD_1-0_6h, and CD_2-4_6h emerged as the top three most frequently selected in all three conditions.

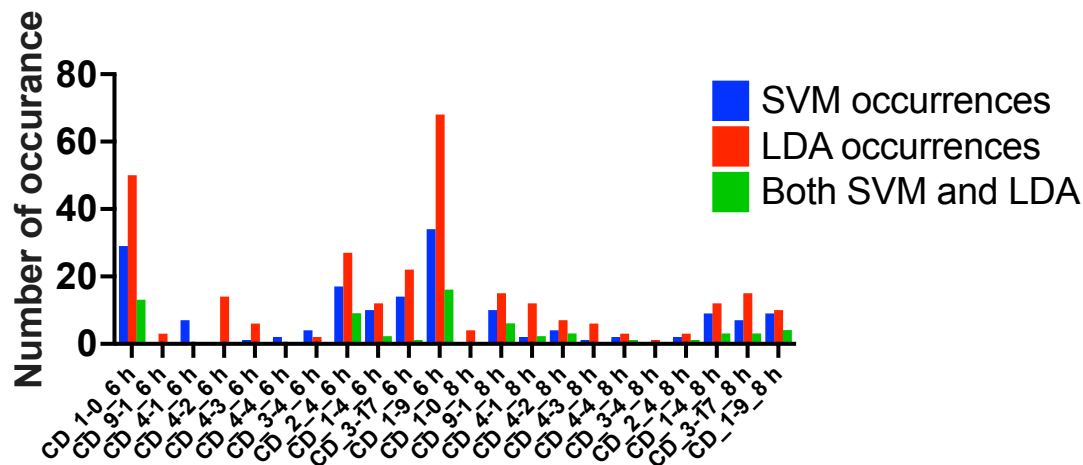


Figure 5.6 The occurrences of each CDs involved in the array combinations with 100% classification accuracy given by both MATLAB based SVM classifier and LDA classifier

Additional parameters provided by the LDA classifier include ellipsoid volumes and ellipsoid distances. Ellipsoid volumes assess the stability and robustness of the array's performance, calculated from the first three functions generated by the LDA classifier. Smaller volumes indicate greater stability and robustness during metal ion classification. Ellipsoid distances measure the separation between the centres of each metal ion's ellipsoid, with larger distances suggesting enhanced performance in metal ion discrimination.

The ellipsoid volumes for all 16 four-CD array combinations are summarised in the supporting material Table S5.8. Notably, the first combination (CD_6h_1_0, CD_6h_2_4, CD_6h_1_4, and CD_6h_1_9) and the fifteenth combination (CD_6h_2_4, CD_6h_1_9, CD_8h_9_1, and CD_8h_4_2) exhibited the smallest ellipsoid volumes. The first combination provided more stable classification across a range of metal ions, including **Ag(I), Ca(II), Co(II), Cu(II), Cd(II), Fe(III), Hg(II), Ni(II), Pb(II), Zn(II), Mg(II), and Mn(II)**, compared to the fifteenth combination. Conversely, the fifteenth combination performed better with the remaining seven metal ions: **Al(III), Ca(II), Cu(II), Fe(III), Pb(II), Mg(II), and Cr(III)**.

The ellipsoid distances for the first and fifteenth combinations are listed in Table 5.7. Generally, in the comparison of the distances between metal ion analytes in the fifteenth combination, the first combination was greater than the second one in all the average metal distance. Therefore, the first combination (CD_6h_1_0, CD_6h_2_4, CD_6h_1_4, and CD_6h_1_9) was selected as the optimised array for discriminating 14 metal ions. The selection of CDs for the final array combination was further validated in the subsequent PCA section 5.2.7.

Table 5.7 The ellipsoid distance matrix of the first combination (CD_6h_1_0, CD_6h_2_4, CD_6h_1_4, CD_6h_1_9) and the fifteenth combination (CD_6h_2_4, CD_6h_1_9, CD_8h_9_1, CD_8h_4_2)

CD_6h_1_0, CD_6h_2_4, CD_6h_1_4, CD_6h_1_9														
Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)
Al(III)	0	3224.3	239.3	4672	3969.2	884.9	2350	1445.5	704.9	279.9	349	268.3	805.4	599
Ag(I)	3224.3	0	3365.2	1455.4	745.9	4070.4	918.4	1812.5	2557.7	3112.5	3477.3	3108.6	2465.6	2685.5
Ca(II)	239.3	3365.2	0	4817.5	4109.7	706.8	2473.2	1562.5	810.3	263.7	120.7	263	906	686
Co(II)	4672	1455.4	4817.5	0	725.1	5523.3	2366.2	3267.1	4011.5	4565.3	4930.1	4561.6	3919	4139.4
Cu(II)	3969.2	745.9	4109.7	725.1	0	4814.4	1651.3	2554	3301.4	3856.7	4221.5	3852.7	3209.1	3428.9
Cd(II)	884.9	4070.4	706.8	5523.3	4814.4	0	3173.8	2263.3	1513.1	959.2	593.2	962.1	1606.8	1386.1
Fe(III)	2350	918.4	2473.2	2366.2	1651.3	3173.8	0	911.5	1663.5	2216	2581.8	2213	1567.8	1788.4
Hg(II)	1445.5	1812.5	1562.5	3267.1	2554	2263.3	911.5	0	752.5	1306.2	1671.2	1302.6	658.1	877.4
Ni(II)	704.9	2557.7	810.3	4011.5	3301.4	1513.1	1663.5	752.5	0	557.2	920.3	552.1	107.2	130.2
Pb(II)	279.9	3112.5	263.7	4565.3	3856.7	959.2	2216	1306.2	557.2	0	366.8	22.3	648.4	430.2
Zn(II)	349	3477.3	120.7	4930.1	4221.5	593.2	2581.8	1671.2	920.3	366.8	0	369.1	1014.2	793.8
Mg(II)	268.3	3108.6	263	4561.6	3852.7	962.1	2213	1302.6	552.1	22.3	369.1	0	645.3	425.6
Mn(II)	805.4	2465.6	906	3919	3209.1	1606.8	1567.8	658.1	107.2	648.4	1014.2	645.3	0	223
Cr(III)	599	2685.5	686	4139.4	3428.9	1386.1	1788.4	877.4	130.2	430.2	793.8	425.6	223	0
Average	1414	2357	1452	3497	2889	2033	1848	1456	1256	1327	1529	1325	1270	1257
CD_6h_2_4, CD_6h_1_9, CD_8h_9_1, CD_8h_4_2														
Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)
Al(III)	0	2668.3	114.4	3626.7	3023.9	753.2	1598.4	1265.8	541.8	67.4	380.3	94.9	508.5	169.4
Ag(I)	2668.3	0	2780.1	966.5	356.8	3421.5	1070.6	1402.9	2126.9	2613.9	3047.8	2578	2162.9	2499
Ca(II)	114.4	2780.1	0	3737.4	3135.9	641.7	1709.8	1378.2	654.6	167	267.7	202.2	618.3	281.7
Co(II)	3626.7	966.5	3737.4	0	625.3	4379.2	2029	2364.4	3086.6	3570.7	4005	3535.3	3119.2	3457.3
Cu(II)	3023.9	356.8	3135.9	625.3	0	3777.1	1426.9	1758.2	2482.3	2969.9	3403.6	2933.9	2519	2854.6
Cd(II)	753.2	3421.5	641.7	4379.2	3777.1	0	2351.4	2019	1294.9	808.5	374.2	843.9	1260	922.5
Fe(III)	1598.4	1070.6	1709.8	2029	1426.9	2351.4	0	337	1057.7	1543.5	1977.5	1507.7	1092.3	1429
Hg(II)	1265.8	1402.9	1378.2	2364.4	1758.2	2019	337	0	724.1	1212.6	1645.7	1176.3	763.3	1096.7
Ni(II)	541.8	2126.9	654.6	3086.6	2482.3	1294.9	1057.7	724.1	0	490.3	921.9	453.5	77.2	373
Pb(II)	67.4	2613.9	167	3570.7	2969.9	808.5	1543.5	1212.6	490.3	0	434.3	37.5	451.5	119.6
Zn(II)	380.3	3047.8	267.7	4005	3403.6	374.2	1977.5	1645.7	921.9	434.3	0	469.9	885.8	549.1
Mg(II)	94.9	2578	202.2	3535.3	2933.9	843.9	1507.7	1176.3	453.5	37.5	469.9	0	416.1	82.2
Mn(II)	508.5	2162.9	618.3	3119.2	2519	1260	1092.3	763.3	77.2	451.5	885.8	416.1	0	339.6
Cr(III)	169.4	2499	281.7	3457.3	2854.6	922.5	1429	1096.7	373	119.6	549.1	82.2	339.6	0
Average	1058	1978	1121	2750	2233	1632	1366	1225	1020	1035	1312	1024	1015	1012

5.2.7 Principal component analysis for twenty-two CDs

In the development of CDs based metal sensing arrays, principal component analysis (PCA) serves as a critical statistical tool to enhance the selection of optimal sensor combinations. Following the successful application of LDA and SVM classification, which pinpointed the optimised four-CDs array (CD_6h_1_0, CD_6h_2_4, CD_6h_1_4, CD_6h_1_9) for classifying 14 metal ions with 100% accuracy, PCA offers additional insights. This method provides a different perspective, helping to confirming the contributions of these four CDs from the most promising array.

PCA is widely recognised for its utility in various scientific and industrial fields, facilitating the analysis and interpretation of complex data sets. By reducing the dimensionality of data while retaining the most significant variance, PCA can visually simplify the understanding of the interactions within carbon dots arrays. This simplification aids in discerning the underlying patterns that govern sensor responses, which is instrumental in refining the design of differential sensing systems. Thus, by applying PCA, we can better interpret the data plots generated from differential sensing studies, leading to more effective discrimination and differentiation of analytes.

5.2.7.1 Dataset Overview

The dataset consists of fluorescent responses from 22 carbon dot sensors to 14 different metal ions. Each CD sensor's response to each metal ion has been recorded in quadruplicate, leading to a total of 56 readings per sensor. The aim of the PCA on this dataset is to investigate the relationships between the carbon dots' responses. This analysis will help in guiding the selection of sensors to develop a metal sensing array, optimising for diversity and sensitivity in detecting various metal ions.

5.2.7.2 KMO test, Bartlett's test and the total variance explained of 22 CDs' PCA

The Kaiser-Meyer-Olkin (KMO) measure of sampling adequacy was employed to examine the partial correlations among variables. A KMO value close to 1 suggests a stronger suitability of the dataset for factor analysis. Specifically, a KMO value greater than 0.9 is considered excellent, above 0.7 is acceptable, and below 0.5 indicates that the data is not suitable for factor analysis. Additionally, Bartlett's test of sphericity confirmed the presence of significant correlations among variables, with an approximate chi-square and a significance level of $p < 0.001$, providing a robust basis for factor analysis. In our study, as provided in Table 5.8, the PCA conducted on 22 carbon

dots (CDs) resulted in a KMO of 0.803, confirming the data's suitability for factor analysis, while Bartlett's test of sphericity was significant ($p < 0.001$), further validating the interdependencies among variables.

Regarding the total variance explained (in **Table 5.9**), eigenvalues greater than 1 generally indicate that the corresponding factors are significant contributors to the dataset. Factors with eigenvalues below 1 are typically discarded as they contribute insufficient information. In this analysis, the cumulative percentage used to assess the cumulative contribution rate of factors showed that the first four components, each with eigenvalues exceeding 1, accounted for 84.534% of the total variance. This substantial cumulative contribution rate suggests that these four factors explain nearly 85% of the variability in the dataset, justifying the extraction of the first four factors.

This analysis provides strong evidence that a sensor array composed of four different CDs may offer an optimal configuration, effectively balancing complexity and explanatory power, thereby guiding the final design of the CD-based sensor array for enhanced performance in applications.⁵⁰

Table 5.8 KMO and Bartlett's Test of twenty-two carbon dots' PCA

KMO and Bartlett's Test		
Kaiser-Meyer-Olkin Measure of Sampling Adequacy.		0.803
Bartlett's Test of Sphericity	Approx. Chi-Square	1764.99
	df	231
	Sig.	<.001

Table 5.9 Total Variance Explained of twenty-two carbon dots' PCA

Total Variance Explained									
Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	10.699	48.632	48.632	10.699	48.632	48.632	6.982	31.736	31.736
2	4.184	19.02	67.652	4.184	19.02	67.652	6.929	31.493	63.23
3	2.219	10.085	77.737	2.219	10.085	77.737	2.538	11.535	74.765
4	1.495	6.797	84.534	1.495	6.797	84.534	2.149	9.769	84.534
5	0.696	3.163	87.697						
6	0.58	2.636	90.333						
7	0.464	2.109	92.442						

Total Variance Explained									
Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
8	0.392	1.78	94.223						
9	0.262	1.19	95.412						
10	0.221	1.005	96.417						
11	0.17	0.773	97.19						
12	0.14	0.635	97.824						
13	0.124	0.564	98.388						
14	0.089	0.404	98.793						
15	0.069	0.313	99.106						
16	0.05	0.226	99.332						
17	0.038	0.172	99.504						
18	0.035	0.16	99.664						
19	0.029	0.132	99.796						
20	0.022	0.1	99.895						
21	0.018	0.082	99.977						
22	0.005	0.023	100						
Extraction Method: Principal Component Analysis.									

5.2.7.3 Correlation matrix analysis

The correlation matrix is a critical component for PCA as it helps to understand the pairwise relationships between variables—in this case, the different CD sensors. Each entry in the matrix provides the correlation coefficient between two sensors, which ranges from -1 to 1, indicating the strength and direction of their linear relationship (as shown in Figure 5.7).

CD_1-0_6 h has a moderately high positive correlation with CD_9-1_6 h (0.544), indicating that these sensors might respond similarly to the presence of certain metal ions.

CD_1-4_6 h and CD_2-4_6 h show a high positive correlation with each other (0.868), suggesting redundancy in their responses.

CD_4-3_6 h and CD_4-4_6 h (0.872), as well as CD_4-3_8 h and CD_4-4_8 h (0.811), have very high positive correlations, which may indicate that they are sensing similar features or aspects of the metal ions.

Some pairs of CDs, such as CD_4-2_6 h and CD_1-0_6 h (-0.053), have a slight negative correlation, indicating that they might be sensitive to different properties of the metal ions.

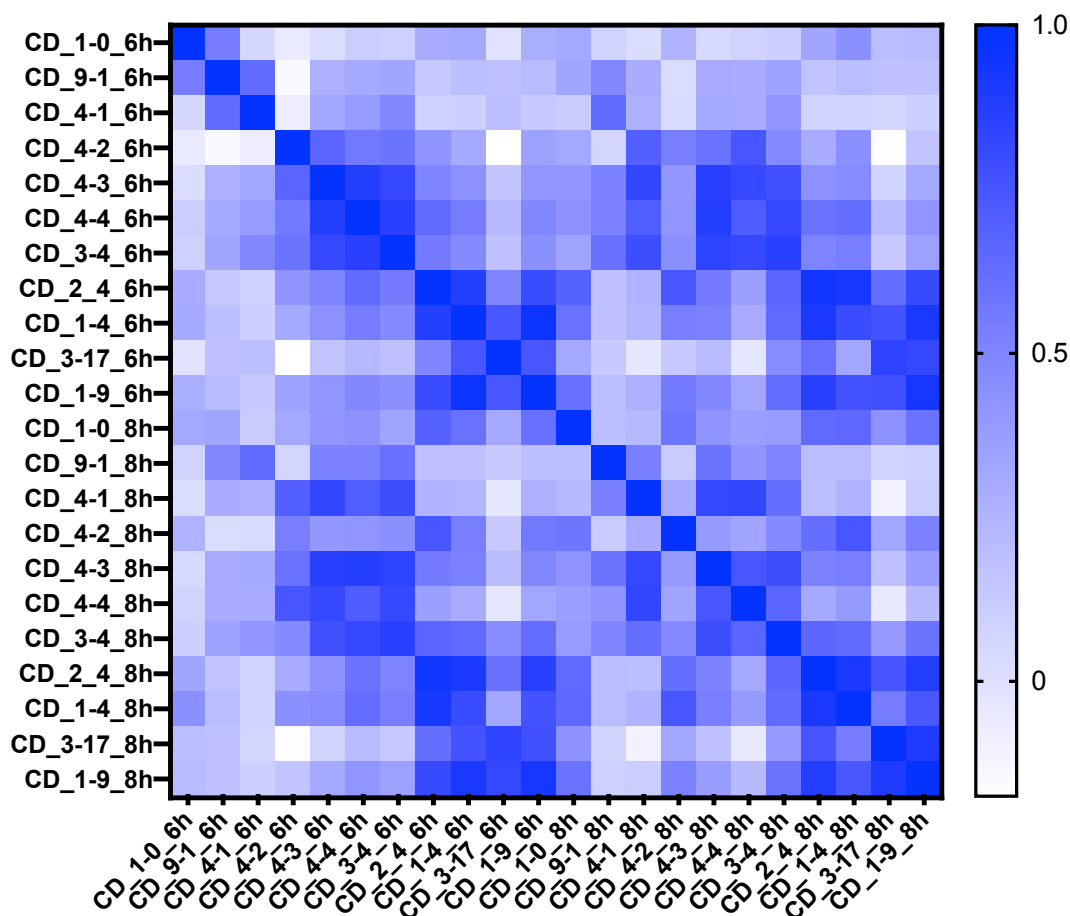


Figure 5.7 Correlation matrix of 22 CDs, (80 μ M metal ion, 1 mg/mL CDs solutions)

5.2.7.4 Component matrix analysis

The component matrix (listed in Table 5.10) presents the loadings of each variable (CD sensors) on the extracted principal components. Loadings are correlations between the variables and the principal components and can range from -1 to 1. From the total variance explained matrix Table 5.9, the first 4 components were extracted.

Table 5.10 Component Matrix of 22 CDs

CDs	Component			
	1 (48.63%)	2 (- 19.02%)	3 (-10.09%)	4 (-6.8%)
CD_1-0_6 h	0.279	-0.211	0.217	0.805

CDs	Component			
	1 (48.63%)	2 (- 19.02%)	3 (-10.09%)	4 (-6.8%)
CD_9-1_6 h	0.366	0.148	0.735	0.417
CD_4-1_6 h	0.332	0.321	0.719	-0.019
CD_4-2_6 h	0.547	0.442	-0.633	0.031
CD_4-3_6 h	0.77	0.511	-0.076	-0.111
CD_4-4_6 h	0.837	0.373	0	-0.072
CD_3-4_6 h	0.809	0.477	0.058	-0.078
CD_2_4_6 h	0.871	-0.319	-0.194	0.097
CD_1-4_6 h	0.849	-0.444	-0.027	-0.081
CD_3-17_6 h	0.514	-0.572	0.331	-0.479
CD_1-9_6 h	0.835	-0.432	-0.017	-0.089
CD_1-0_8 h	0.671	-0.212	-0.056	0.318
CD_9-1_8 h	0.465	0.441	0.535	-0.045
CD_4-1_8 h	0.604	0.68	-0.073	-0.066
CD_4-2_8 h	0.658	-0.131	-0.381	0.295
CD_4-3_8 h	0.811	0.437	-0.015	-0.13
CD_4-4_8 h	0.666	0.593	-0.121	0.035
CD_3-4_8 h	0.872	0.196	0.103	-0.188
CD_2_4_8 h	0.858	-0.426	-0.084	0.035
CD_1-4_8 h	0.843	-0.268	-0.193	0.278
CD_3-17_8 h	0.561	-0.705	0.219	-0.206
CD_1-9_8 h	0.775	-0.573	0.031	-0.161

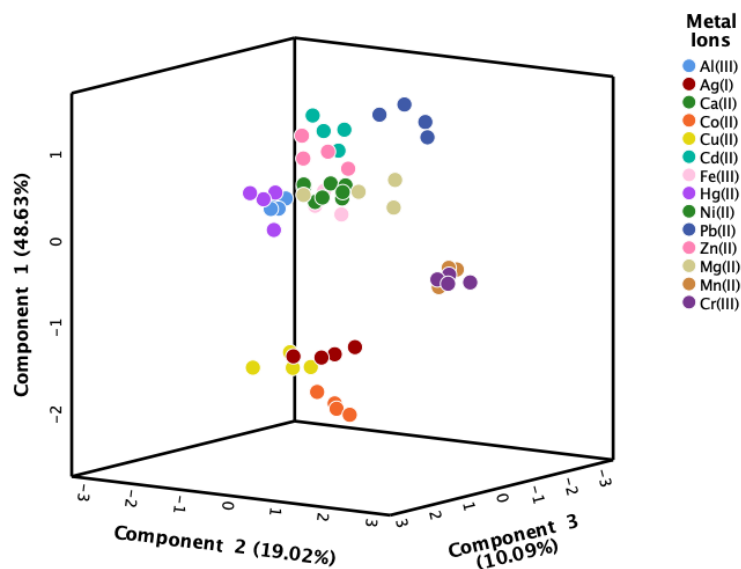


Figure 5.8 PCA plot of cross-reactive of twenty-two carbon dots for fourteen metal ions sensing, (80 μ M metal ion, 1 mg/mL CDs solutions, with four replicates)

5.2.7.5 PCA based CDs selection guidance

PCA is useful in providing guidance on the selection CDs for the final sensor array. Firstly, by including sensors with high loadings on Component 1, such as CD_2-4_6 h, and CD_1-4_6 h, distinction between metal ions can be maximised. CDs like CD_1-0_6 h, with its high loading on Component 4, can be useful for detecting unique metal ion features not captured by other sensors. Sensors with different loadings, such as CD_9-1_6 h (positive loading) and CD_4-2_6 h (negative loading) from Component 3, enable determination of differential responses to ionic properties. The above suggestions can be used as a reference in the process of screening CDs as metal sensing array, but the results of SVM and LDA classification must still be incorporated to determine the final selection of CDs.

PCA was therefore conducted to discern the most reliable and efficient sets of four sensors from the 41 CD combinations identified by the SVM analysis. The PCA was carried for all of the 22 CDs, as presented in the previous sections and the component matrix (listed in Table 5.10) presents the loadings of each variable (CD sensors) on the extracted principal components. Loadings are correlations between the variables and the principal components and can range from -1 to 1. From the PCA, 4 components were extracted.

The analysis results firstly identified two CDs, CD_2-4_6 h and CD_1-4_6 h, as having substantial contributions to the variance of the dataset. These particular CDs exhibited high communalities, with extraction values standing at 0.925 and 0.931 respectively, thereby capturing a considerable proportion of the variance. Furthermore, they showed significant loadings on the first two principal components, with CD_1-4_6 h bearing loadings of 0.871 and -0.319, and CD_2-4_6 h with 0.809 and 0.477. Such robust loadings underpin their critical role in encapsulating a broad spectrum of variance, which is imperative for the development of a responsive sensing array capable of effectively distinguishing between various metal ion properties. In another words, CD_2-4_6 h and CD_1-4_6 h's metal ions sensing performances can be used to represent most of the cross-reactive sensing performances of the set of 22 CDs. Significantly, the most accurate 3 sensor arrays identified by SVM analysis heavily featured these two probes. Then PCA recommended CD_1-0_6 h.

Analysed by the single CD based LDA analysis, the classification performance of the 22 CDs for metal ion identification, as presented in the supporting material 3.4.3, is illustrated through one-

feature-based LDA classification results. The plots effectively demonstrate the CDs' ability to classify metal ions, highlighting a clear distinction between groups of metal ions that can be readily separated and those that cannot. Metal ion groups that exhibit significant separability underscore the efficacy of the selected CDs in differentiating between specific types of metal ions, showcasing their substantial differences in sensing capabilities. Conversely, the plots also reveal the limitations of CDs in distinguishing between metal ion groups with overlapping characteristics, where the distinction is less pronounced. These visual representations provide a valuable assessment of the CDs' specificity and selectivity in metal ion detection, emphasising the importance of feature selection in optimising classification performance. Incorporating the insights from the previous PCA analysis, the LDA classification results can be used to supporting the PCA results. Under PCA analysis, four principal components were identified, with Component 1 representing the typical metal ion sensing characteristics of CDs and Component 4 highlighting the more unique and specific sensing capabilities. The analysis revealed that CDs such as CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h, among others, demonstrated higher coefficients in Component 1, indicating their effectiveness in representing the group's general metal ion sensing properties, particularly towards Cu. These CDs also exhibited high mean classification accuracy, underscoring their stability and accuracy in metal ion detection. Conversely, CDs with higher coefficients in Component 4, like CD_1-0_6 h, were identified as offering more distinct sensing responses to specific metal ions such as Al(III), Zn(II), Hg(II), and Ca(II), with above-average mean classification accuracy. CDs associated with Components 2 and 3, however, were noted for their less satisfactory mean classification accuracy and less distinctive sensing responses, thus providing limited utility for array sensing. Consequently, an array comprising CDs like CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h could potentially yield a robust classification effect for metal ion sensing targets, leveraging both typical and unique sensing capabilities.

5.2.8 The SPSS based LDA analysis of the optimised CDs based metal sensing array

An SPSS-based LDA classification was carried out for the above suggested array combination that included CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h, to verify the sufficiency of the array. According to the verification results, this array can identify and quantify fourteen metal ions, including Al(III), Ag(I), Ca(II), Co(II), Cu(II), Cd(II), Fe(III), Hg(II), Ni(II), Pb(II), Zn(II), Mg(II), Cr(II), and Mn(II), at a concentration of 80 μ M. Array data was collected using four replicates of each condition, as shown in Figure 5.9. LDA performed on the 14 metal responses revealed that

the four CDs sensors could distinguish all 14 metal ions from each other with 100% correct classification (100% cross-validated). This was mainly attributed to the first three discriminant functions, which together explained 99.2% of the variance. Having identified this set of four sensors as being optimal, we sought to further characterise this set of carbon dots.

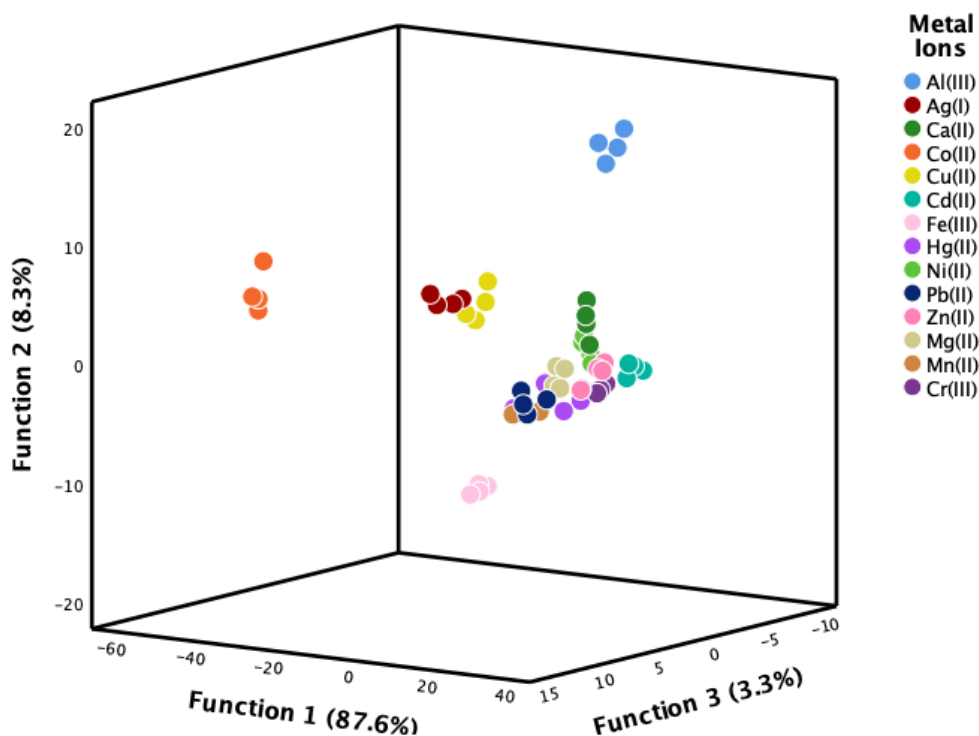


Figure 5.9 3D LDA score plots with the first three functions for the analysis of 14 metal ions (80 μ M metal ion, 1mg/mL CDs ($\lambda_{ex} = 370$ nm, $\lambda_{em} = 390-750$ nm)) in buffered Milli-Q water (20 mM, pH 7.0) by using CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h based array.(with four replicates)

5.2.9 Carbon dots characterisation

5.2.9.1 Carbon dots' morphological and fluorescent characterisation

The characterisation focused on the optimised CDs selections, including CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h. After synthesis, these four CDs appear as orange-red solutions and exhibit bright green fluorescence under a UV lamp. After freeze-drying for 48 hours, a dark brown dried powder of CDs was collected. Morphologies of CDs were observed by TEM (as shown in Figure 5.10).

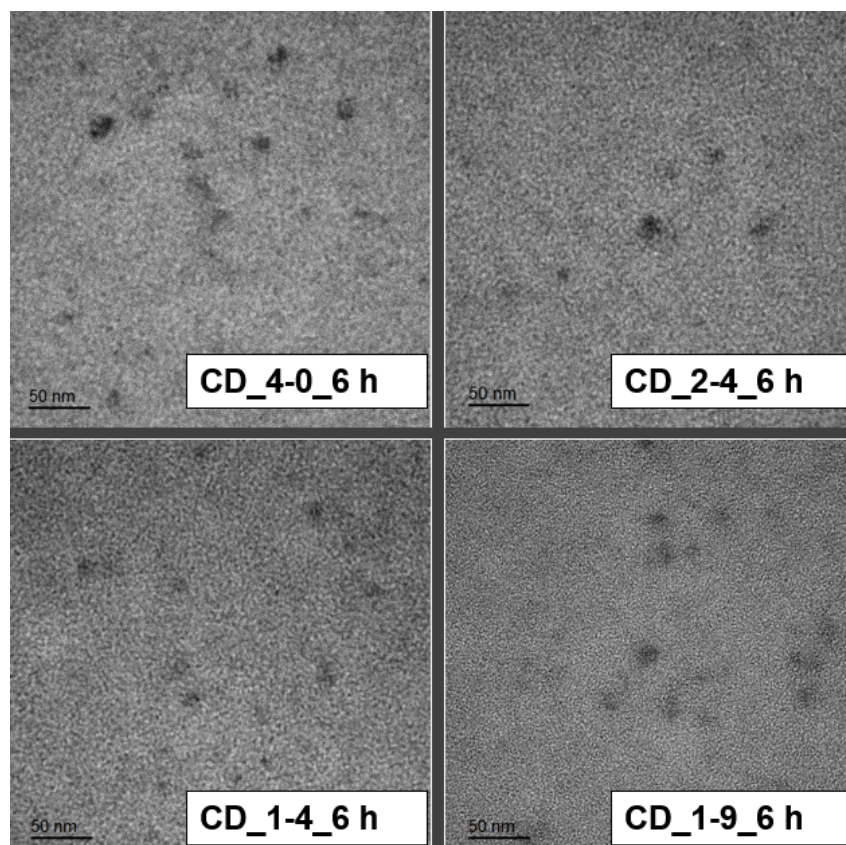


Figure 5.10 TEM images of CDs (CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, CD_1-9_6 h)

CD_1-0_6 h, synthesised from pure fructose without a nitrogen source, exhibits two emission peaks that are excitation-dependent. This means that the emission peak values of CD_1-0_6 h shift with varying excitation wavelengths. The excitation-dependent fluorescence characteristic is frequently observed in carbon dots (CDs). Such variable emission is often ascribed to 'defect states' encompassing a wide energy spectrum, which can be attributed either to the graphitised core of the CDs or the prevalent oxygenated groups, including carbonyl, carboxylic acid, and ether.⁵¹ Specifically, under an excitation wavelength of 370 nm, CD_1-0_6 h exhibits the highest emission intensities at 530 nm. On the other hand, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h were synthesised using two components: fructose as the carbon source and PPD as the nitrogen source. These CDs display emission spectra with a single peak, and their fluorescence is excitation-independent. The emission peak for these CDs is consistently located at 530 nm, with the highest emission peak occurring at an excitation wavelength of 370 nm (as shown in). The variance of the emission intensities of CDs by meeting with different metal ions was presented in Figure 5.12.

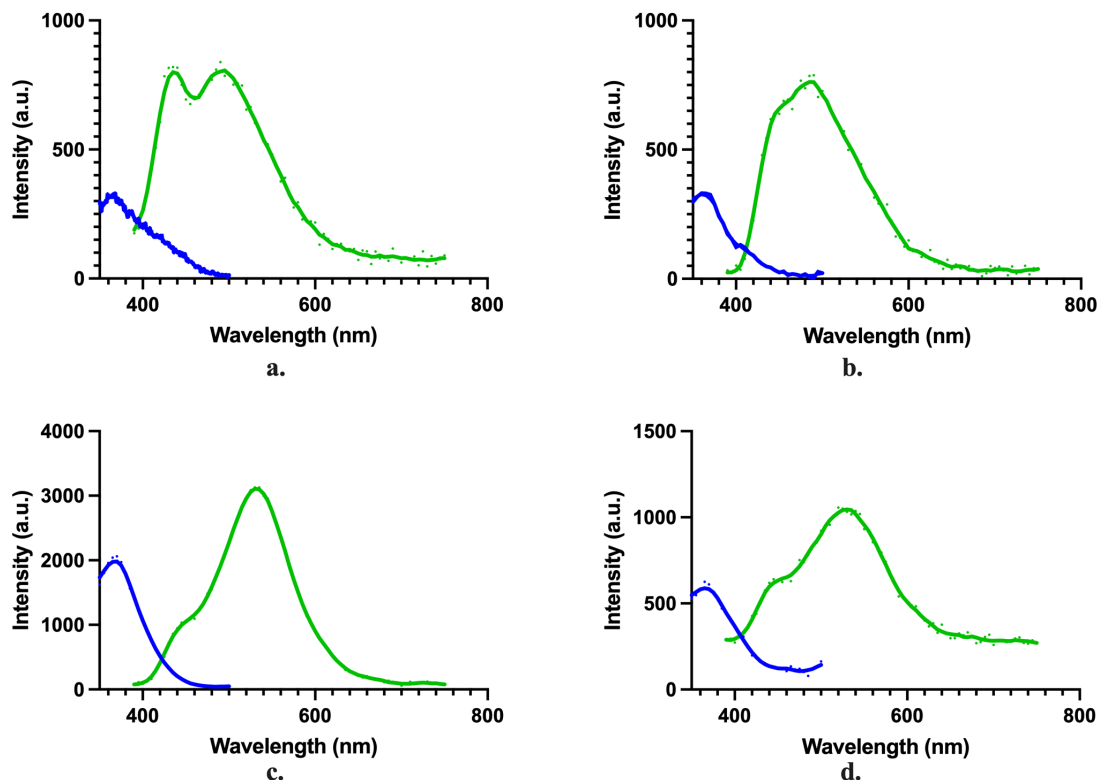


Figure 5.11 The emission (green, $\lambda_{ex} = 370$ nm) and excitation (blue, $\lambda_{em} = 530$ nm) spectra of CDs (a. CD_1-0_6 h, b. CD_2-4_6 h, c. CD_1-4_6 h, d. CD_1-9_6 h at 1g/L) in PBS buffer at pH 7

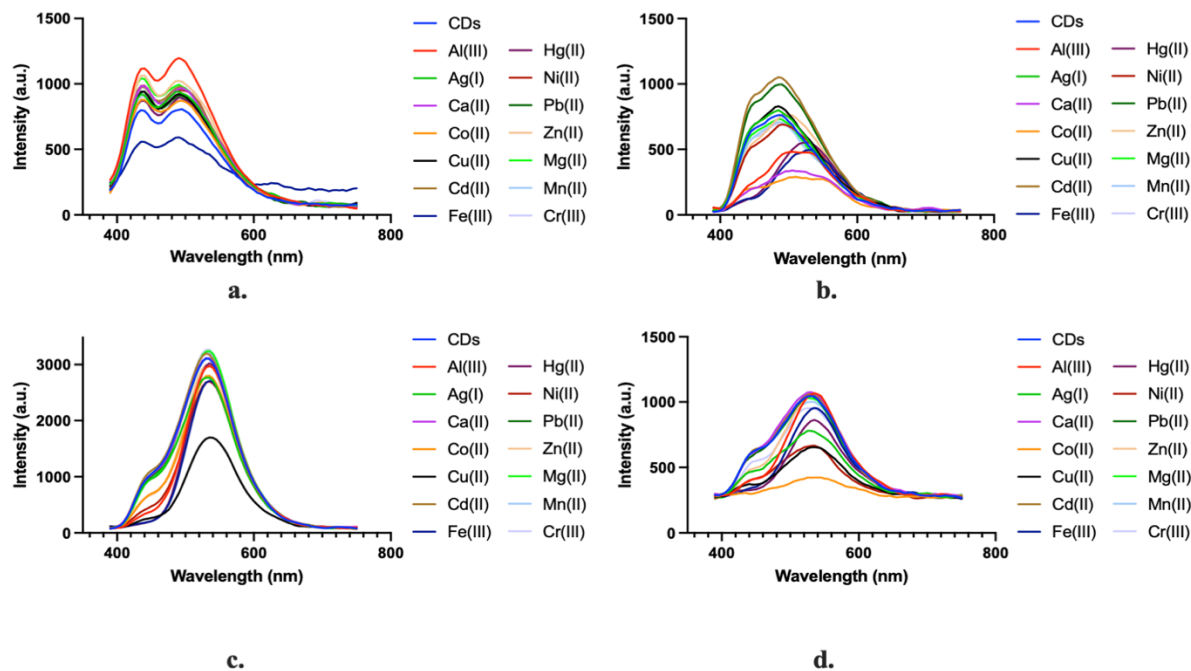


Figure 5.12 The emission spectra of CDs during metal ions sensing (a. CD_1-0_6 h, b. CD_2-4_6 h, c. CD_1-4_6 h, d. CD_1-9_6 h at 1mg/mL, with 80 μ M metal ion, ($\lambda_{ex} = 370$ nm, $\lambda_{em} = 390-750$ nm)) in PBS buffer at pH 7

5.2.9.2 ATR characterisation

ATR spectroscopy was utilised to analyse the surface functional groups of the selected CDs (Figure 5.13). The transmittance spectra revealed several characteristic absorption bands that are indicative of the various functional groups present on the CDs. The broad band centred around $3200\text{--}3600\text{ cm}^{-1}$ was observed across all CD types, which is characteristic of O-H stretching vibrations. The presence of such a broad peak suggests hydrogen-bonded hydroxyl groups, likely due to the presence of alcohols or phenols, which confer hydrophilicity to the CDs. In CD_1-0_6 h, an additional peak near 2920 cm^{-1} was attributed to C-H stretching vibrations from alkane chains, indicating the presence of alkyl functionalities on the surface of the CDs. In the absence of nitrogen-related functional groups for this CD type, the peak around 1580 cm^{-1} , generally associated with C=C stretching in alkenes or aromatic compounds, was not indicative of N-H bending. The broad features observed around $1200\text{--}1000\text{ cm}^{-1}$ were indicative of C-O stretching from alcohols, acids, esters, or ethers. The spectra for the other CDs showed similar O-H stretching vibrations but differed in the relative intensities and positions of peaks associated with other functional groups. CD_2-4_6 h exhibited a less intense O-H stretching peak, suggesting a variation in the hydrogen bonding or hydroxyl group concentration. This CD also showed a less pronounced C-H stretching peak and subdued features in the fingerprint region, indicating a possible difference in the presence or environment of functional groups.

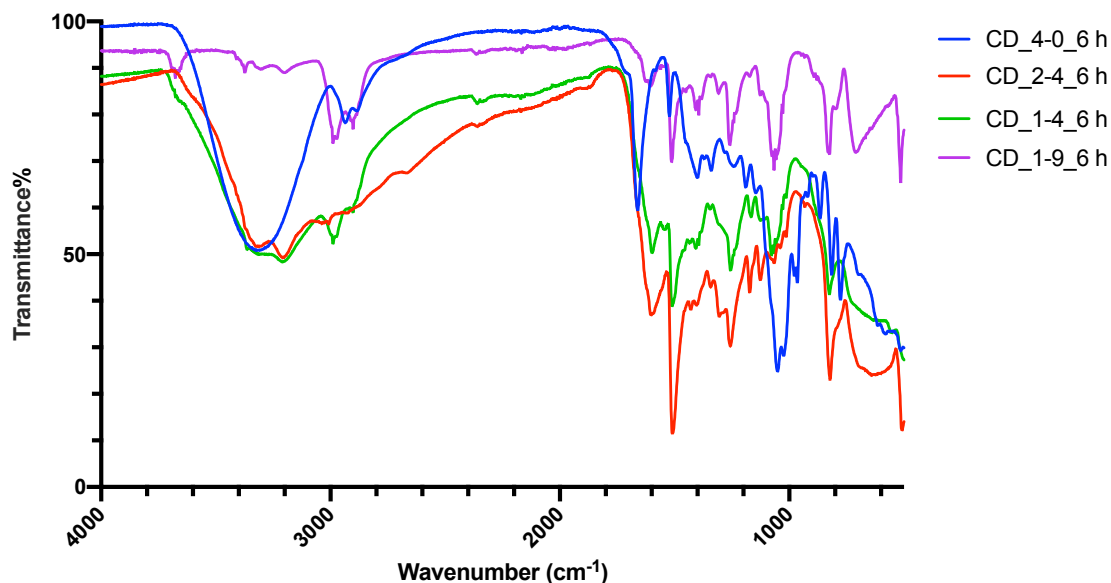


Figure 5.13 ATR spectra of the CDs derived from fructose and p-phenylenediamine

5.2.9.3 XPS characterisation

In alignment with the previously conducted ATR analysis, which suggested the presence of various functional moieties, XPS data further elucidated the precise chemical environment of the CDs. Differences in the atomic percentages of oxygen, carbon, and nitrogen derived from the XPS survey scans indicated variability in the quantity and type of surface functional groups among the CDs, which can be attributed to the distinct precursor materials or synthetic routes employed in their production, as shown in Table 5.11. By delving into the C1s peaks, a more granular picture emerged, showcasing a mix of graphitic or aliphatic carbon structures, along with oxygenated and nitrous carbon atoms. These results not only affirm the multifunctional nature of the CDs' surfaces but also highlight the variation in functional groups, which is particularly pronounced in the CDs synthesised from different precursors.

The XPS spectra exhibit three principal peaks across the samples, correlating with the core levels of C1s, N1s, and O1s. These peaks are situated approximately at binding energies of 284, 400, and 531 eV, respectively, indicative of the carbon, nitrogen, and oxygen elements present on the CDs' surfaces. The XPS C1s binding energies (BE) indicate the presence of various carbon-containing functional groups on the surface of the carbon dots (CDs), as shown in Table 5. 12. These peaks reflect the diversity of the chemical environments of carbon on the surface of the CDs and are indicative of their functionalisation. The relative atomic percentages provide insight into the abundance of these functional groups, which is crucial for the CDs' potential applications in various fields due to their influence on solubility, reactivity, and biological interactions. The C1s region of CD_1-0_6 h displayed peaks at binding energies of 285.87 and 288.14 eV, denoting the presence of graphitic or aliphatic carbon (C-C/C-H) and oxygenated carbon species (C-O), respectively. This CD showed no N1s peak, aligning with the absence of nitrogenous functional groups as confirmed by ATR. The oxygen content, evidenced by the O1s peak, was consistent with the hydroxyl functionalities observed in the ATR spectrum. The three fructose and PPD-derived CDs, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h, showed C1s peaks that could be attributed to both graphitic/aliphatic carbon and nitrogen-containing functionalities, as indicated by the presence of N1s peaks around 400 eV. These findings align with the ATR analysis, which identified N-H bending vibrations and C-N stretching. The C1s region also revealed oxygenated carbon, aligning with the presence of carbonyl and hydroxyl groups detected in the ATR spectra.

Table 5.11 XPS analysis of four CDs (peak binding energy and element content of C, O and N)

CDs	C1s			
	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
CD_1-0_6 h	286.4	3.46	1166466.33	65.45
CD_2-4_6 h	285.28	2.83	1268354.34	80.7
CD_1-4_6 h	285.1	2.81	1192482.36	78.92
CD_1-9_6 h	284.94	3.04	1032979.09	72
CDs	O1s			
	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
CD_1-0_6 h	533.29	2.69	1488346.41	34.55
CD_2-4_6 h	532.49	3.53	319865.42	8.42
CD_1-4_6 h	532.11	3.28	367210.48	10.05
CD_1-9_6 h	531.9	3.25	373754.14	10.78
CDs	N1s			
	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
CD_1-0_6 h				
CD_2-4_6 h	399.86	2.85	265469.91	10.88
CD_1-4_6 h	399.25	2.84	258573.36	11.02
CD_1-9_6 h	399.25	2.86	383490.27	17.22

Table 5. 12 XPS analysis of four CDs (C1s analysis)

CD_1-0_6 h				
Name	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
C-C or C-H	284.77	1.73	82114.21	53.71
C-O	286.14	0.92	33726.38	22.08
C=O	287.03	2.21	36952.33	24.21
CD_2-4_6 h				
Name	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
C-C or C-H	284.78	1.96	132930.9	79.28
C-O or C-N	285.46	3.37	28017.59	16.72
C=O	288.59	3.37	6703.17	4.01
CD_1-4_6 h				
Name	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
C-C or C-H	284.79	1	6447.84	4.47
C-O or C-N	285.79	1.61	107699.8	74.72
C=O	286.15	3.06	29984.41	20.81

CD_1-9_6 h				
Name	Peak BE	FWHM eV	Area (P) CPS.eV	Atomic %
C-C or C-H	284.81	1.03	21648.05	15.8
C-O or C-N	285.63	2.15	101605.9	74.21
C=O	288.59	3.37	13643.23	9.98

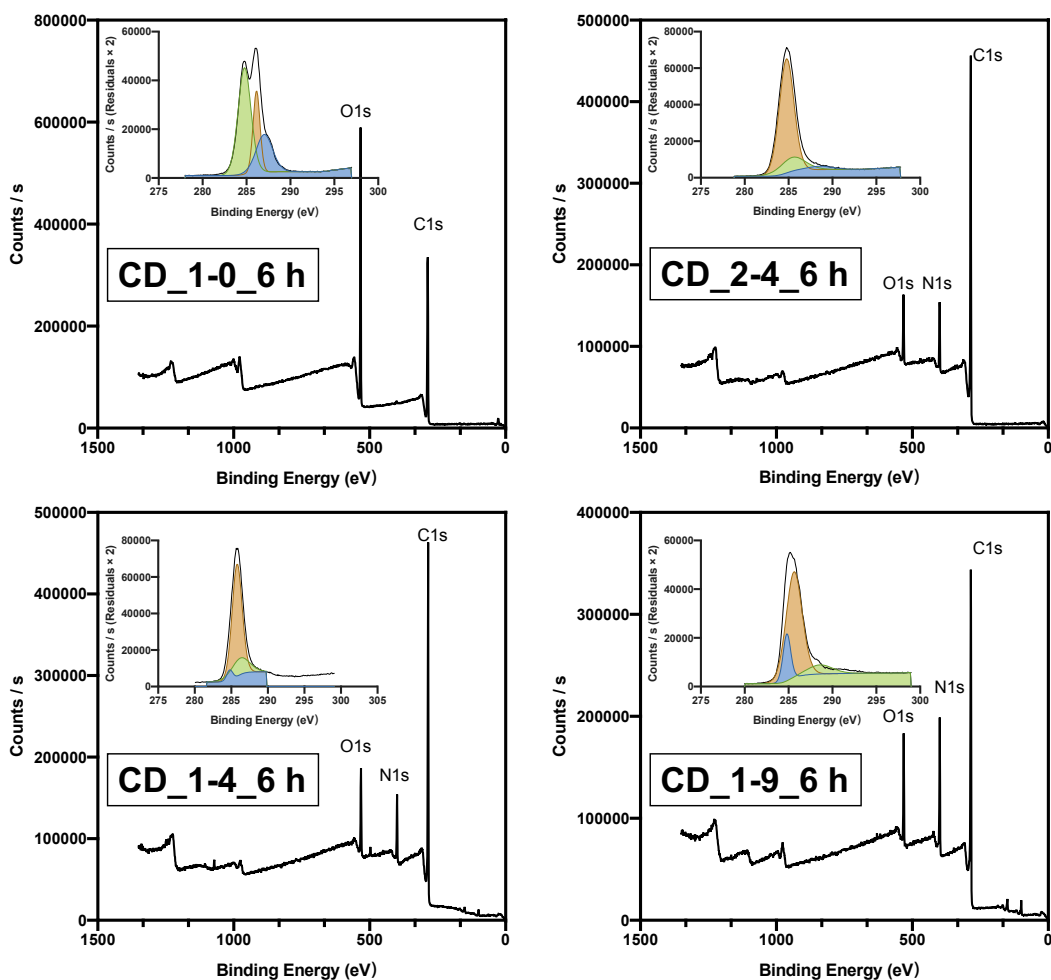


Figure 5.14 XPS analysis of four-CDs-based metal sensing array (*CD_1-0_6 h*, *CD_2-4_6 h*, *CD_1-4_6 h* and *CD_1-9_6 h*)

5.2.9.4 UV-vis characterisation

UV-vis spectroscopy was performed for the 4 CDs (Figure 5.15). *CD_1-0_6 h* exhibits a prominent absorption peak near 257 nm, which could correspond to the π - π^* transitions typically associated with conjugated C=C bonds⁵²⁻⁵⁵. This is supported by the ATR and XPS analyses, which reveal

the presence of alkyl functionalities and graphitic or aliphatic carbon structures, respectively, without evidence of nitrogen-containing groups.

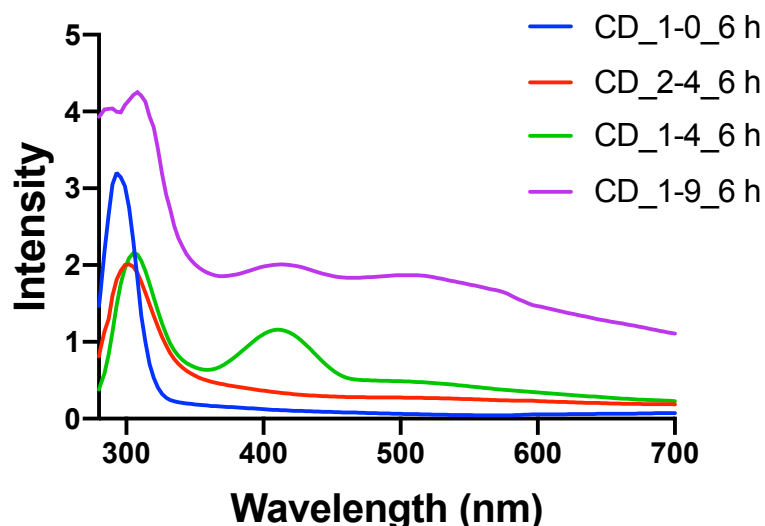


Figure 5.15 UV-vis absorption of four-CDs-based metal sensing array (CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h and CD_1-9_6 h)

CD_2-4_6 h presents a slightly shifted major peak at approximately 254 nm, suggesting a similar π - π^* transition but with possible variations in the electronic environment due to different surface functional groups or degrees of conjugation. The less intense peak might indicate a lower concentration of conjugated domains or a change in the CDs' surface chemistry, which aligns with the ATR findings of subdued O-H stretching and C-H stretching peaks.

CD_1-4_6 h and CD_1-9_6 h both display primary absorption peaks also around 254 nm, with an additional absorption feature around 302 nm, which may indicate n - π^* transitions associated with C=O bonds, respectively. These results are consistent with the presence of carbonyl and hydroxyl groups as detected by ATR spectroscopy and the presence of oxygenated carbon species from the XPS data. The presence of N1s peaks in the XPS spectra for these CDs suggests nitrogen functionalities that could also influence the UV-vis absorption patterns. Therefore, the conclusion to be drawn from these observations is twofold. Firstly, the CDs indeed have sufficiently distinct structures and surface chemistries to explain their different optical responses, as evidenced by the shifts in absorption peaks and the detection of specific functional groups. Secondly, it is remarkable how these nanomaterials exhibit pronounced sensitivity in their optical behaviours, even when the structural differences are subtle. Figure 5.16 illustrates the variations in absorption

of the CDs upon interaction with different metal ions. The changes in the colours of the CDs due to interaction with various metal ions are depicted in the supporting material, Figure S5.2.

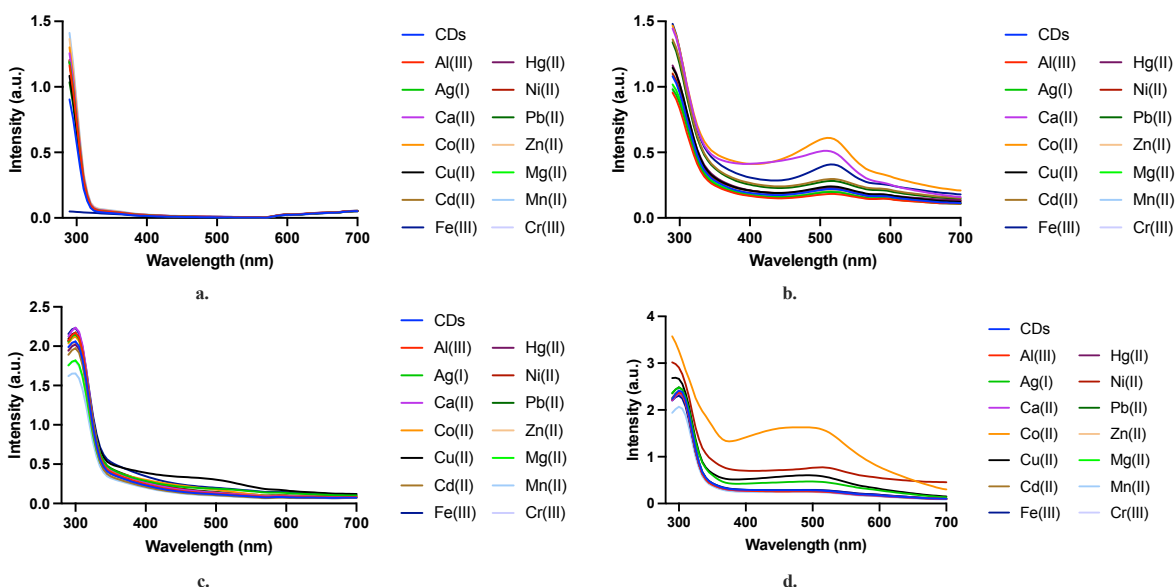


Figure 5.16 UV-vis absorption of four-CDs after meeting with different metal ions (a. CD_1-0_6 h, b. CD_2-4_6 h, c. CD_1-4_6 h and d. CD_1-9_6 h, at 0.25 mg/mL with metal ions at 20 μ M)

Associated with Figure 5.12, CD_1-0_6 h exhibited decreases in both fluorescence and absorption intensities after interaction with Fe(III), which can be attributed to the interaction between the carbon dots and Fe(III) ions. The observed fluorescence quenching may result from energy transfer from the carbon dots to the Fe(III) ions, leading to reduced light emission.^{56, 57} Additionally, the decrease in absorption intensity could arise from complex formation between the Fe(III) ions and the carbon dots. This complex formation might alter the electronic structure of the carbon dots, thereby affecting their absorption properties and resulting in diminished absorption intensity. For CD_2-4_6 h and CD_1-9_6 h, following exposure to Co(II) ions, there was a decrease in fluorescence intensity, while the absorption intensity increased around 530 nm. This reduction in fluorescence can be explained by energy transfer processes between the carbon dots and Co(II) ions. The interaction between the -NH₂ groups on the carbon dots and the Co(II) ions likely results in fluorescence quenching due to energy transfer mechanisms, leading to a reduction in fluorescence intensity, as observed in various studies on fluorescence detection of metal ions such as Co(II). Concerning the increased absorption, the presence of Co(II) ions may induce changes in the electronic structure of the carbon dots, leading to alterations in their absorption properties.⁵⁸

This could manifest as new absorption bands or shifts in existing absorption peaks, as reported in studies on the detection of Co(II) ions using carbon dots.^{58, 59}

The CDs characterised in this study exhibited notable variations in surface functional groups and photophysical properties, which contribute to their differential responses toward metal ions. For example, CDs such as CD100 and CD92, which contain abundant carboxyl (-COOH) and hydroxyl (-OH) groups but minimal amine (-NH₂) content, demonstrated strong fluorescence quenching in response to cations like Cu(II) and Fe(III), likely due to chelation via oxygen donors. In contrast, CDs enriched with amino groups (e.g. CD25 and CD69) exhibited enhanced or less quenched responses under similar conditions, suggesting altered metal binding affinity and charge transfer dynamics. Despite these differences, the CDs shared a comparable core graphitic structure and similar excitation/emission profiles, indicating that their sensing diversity arises primarily from surface state modulation rather than core size or morphology. This chemical diversity underpins the orthogonal fluorescence responses leveraged in the array-based metal ion sensing described in subsequent sections.

5.2.10 Carbon dots-based sensor array application for metal ions sensing

5.2.10.1 Discrimination of metal ion binary and ternary mixtures

To demonstrate the capabilities of the developed four-CDs-based sensing array, we conducted tests to differentiate mixtures of three metal ions: Co(II), Cu(II), and Ni(II). These three ions were selected as model analytes due to the difficulty of distinguishing them using traditional fluorescent dyes^{31, 32, 60}, highlighting the advantages of the CDs-based array.

The concentration of the CDs utilised in these experiments was 1 mg/mL, and the pH of the solutions was controlled using buffered Milli-Q water at a pH of 7.0. Binary mixtures (1:1) of Co(II)/Cu(II), Cu(II)/Ni(II), and Co(II)/Ni(II) were prepared with a total concentration of 80 μ M. Additionally, a ternary mixture (1:1:1) of Co(II)/Cu(II)/Ni(II) was prepared with equal volumes and a total concentration of 80 μ M. The results demonstrated that the four CDs-based sensing array successfully discriminated all the mixture combinations with a classification accuracy of 100% in both the initial analysis and the leave-one-out cross-validation test (Figure 5.17). The first three discriminant functions accounted for 95.1%, 3.7%, and 0.8% of the variance, respectively.

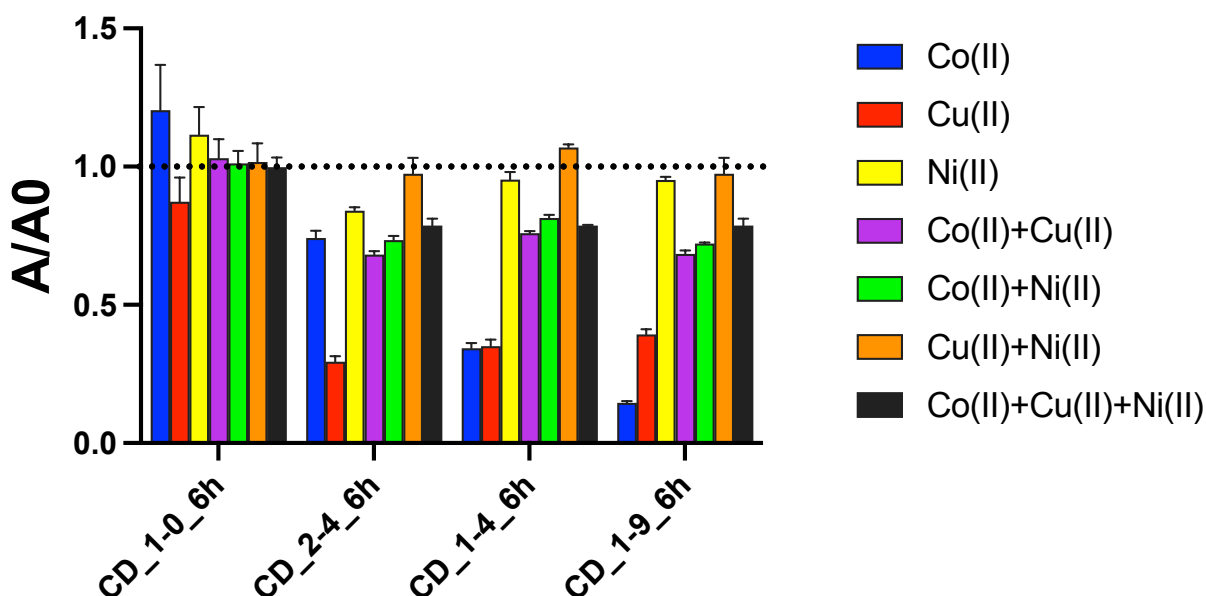


Figure 5.17 The fluorescence variation of each of the CDs by meeting with different the sensing targets, including Co(II), Cu(II), and Ni(II) confidence ellipsoid of binary and ternary mixture

It is important to note that the fluorescence response of the mixture was not a simple superposition of the signals from individual metal ions. For example, on the LDA scatter diagram, the binary mixture of Cu(II) and Ni(II) did not fall between the scatter clusters of single Cu(II) and Ni(II) ions. From Figure 5.18, it was observed that the response of CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h to the binary mixture of Cu(II) and Ni(II) was greater than their response to the individual Cu(II) and Ni(II) ions alone. Additionally, while the fluorescence of CD_1-0_6 h was effective in sensing individual metal ions, it failed to generate a response when exposed to mixtures. These findings suggest that the CDs-based sensing array exhibited complex and selective responses to various metal ion mixtures, surpassing the limitations of traditional fluorescent sensors. The array's ability to discriminate mixtures with high accuracy and capture intricate fluorescence responses highlights its potential for practical applications in analytical chemistry and environmental monitoring.

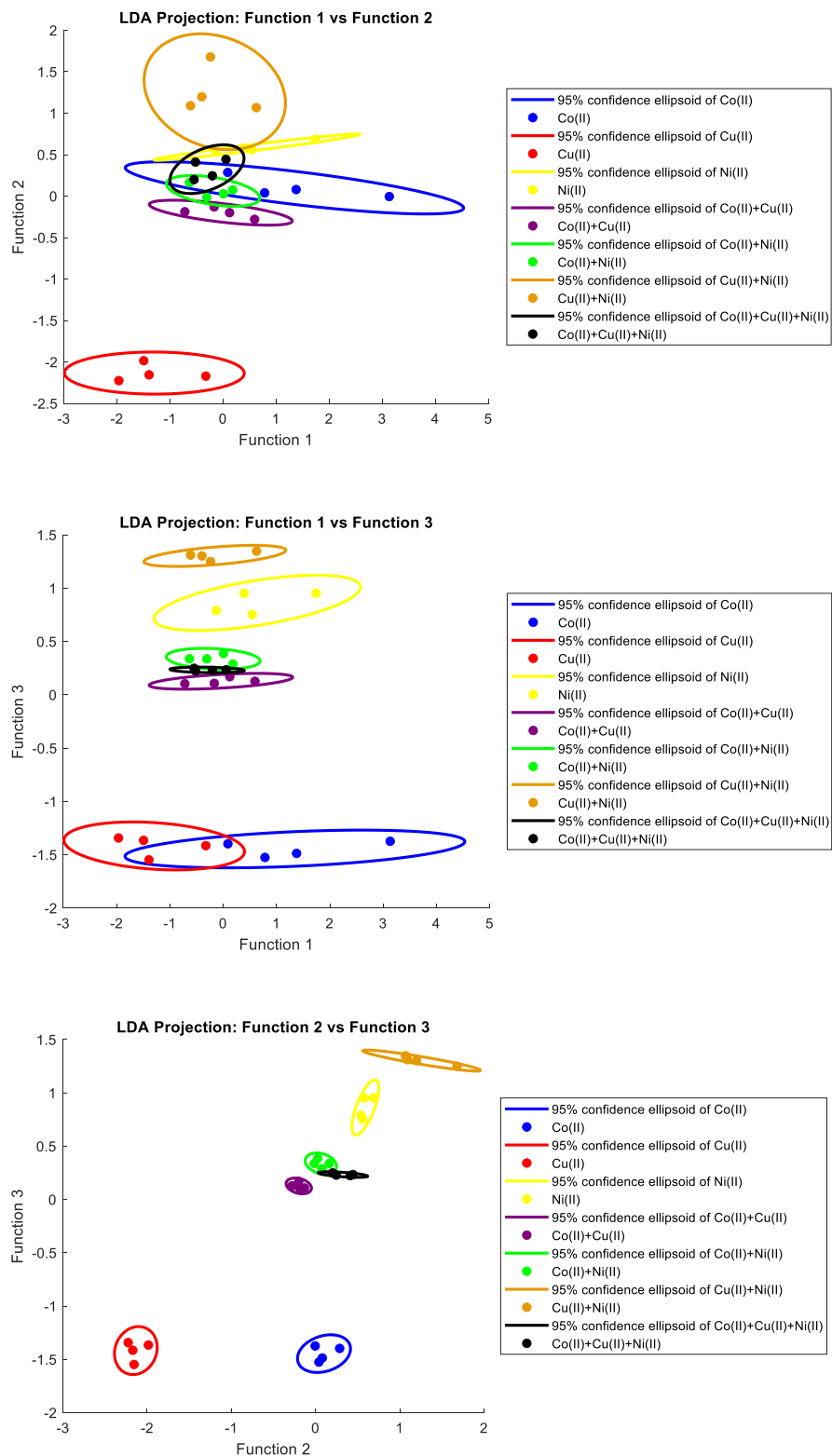


Figure 5.18 Co(II), Cu(II), and Ni(II) confidence ellipsoid of binary and ternary mixture classification based on MATLAB, the 2D LDA plotting between function 1 and 2 (top), function 1 and 3 (middle), function 2 and 3 (bottom), implanted with 95% confidence ellipsoid. (1 mg/mL CDs solutions, with four replicates)

5.2.10.2 Discrimination of Cu(II), Co(II), and Ni(II) with in varying pH environments

The performance of the four CDs-based array in distinguishing Cu(II), Co(II), and Ni(II) was further evaluated. In this study, the four-CDs-based array was tested for its ability to discriminate these three metal ions across three different pH values (3.5, 6.3, and 8.9). The LDA results reveal four canonical discriminant functions. The eigenvalues associated with the first three functions cumulatively explain 99.4% of the total variance. The Wilks' Lambda test was used to assess the statistical significance of the discriminant functions, as shown in

Table 5.13. The test results indicate that all four functions exhibit significant discriminant ability, as evidenced by the extremely low p-values ($p < 0.001$). This suggests that the sensor array's responses provide valuable information for distinguishing between the metal ions. As classification results shown in Figure 5.19, both the original and cross-validated results demonstrate perfect classification, with 100% of the cases being correctly classified. Each metal ion at a specific pH value is assigned to its respective group, indicating accurate classification. These results demonstrate the ability of the sensor array for precise identification and differentiation of the metal ions under investigation.

Table 5.13 Wilks' Lambda test of CDs array-based Cu(II), Co(II), and Ni(II) classification within varying pH environments

Wilks' Lambda				
Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1 through 4	0	385.818	32	0
2 through 4	0	220.507	21	0
3 through 4	0.033	97.28	12	0
4	0.279	36.357	5	0

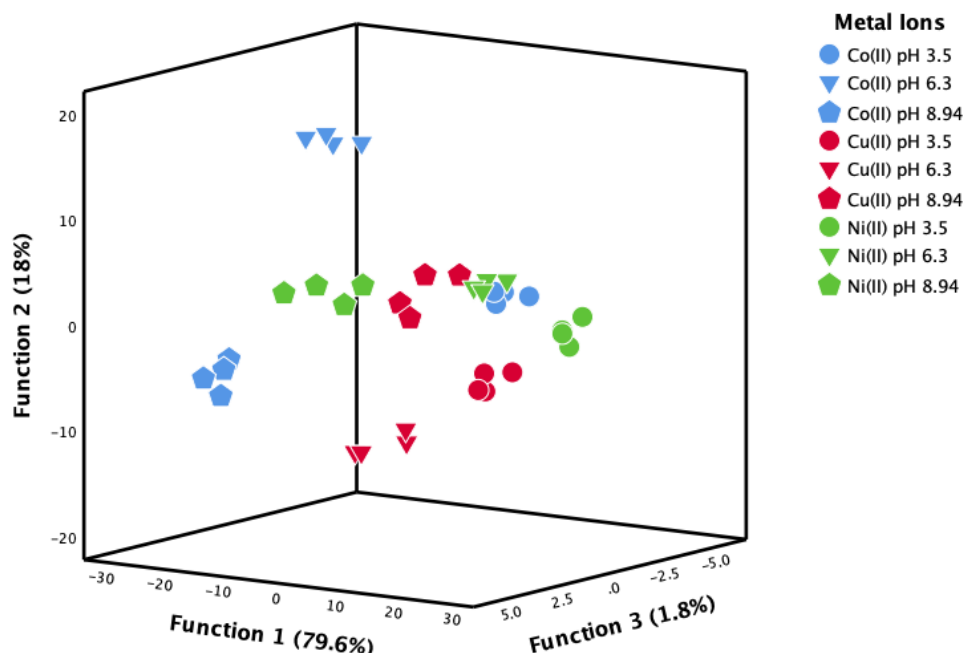


Figure 5.19 LDA score plots for the analysis of Cu(II), Co(II), and Ni(II) (1mg/mL CDs, where $\lambda_{ex} = 370$ nm and $\lambda_{em} = 390-750$ nm) in different buffered Milli-Q water with pH 3.5, 6.3, and 8.9 (80 μ M metal ion, with four replicates).

Lastly, the ability of the four-CDs-based sensing array to classify Cu(II), Co(II), and Ni(II) was carried out to discriminate these metal ions, at three different concentrations, 10, 50, 100 μ M. From the LDA result, as shown in Figure 5.20, both the original and cross-validated results demonstrate a perfect classification, with 100% of the cases being correctly classified. In the LDA for sensing Cu(II), Co(II), and Ni(II) at 5, 10, 50, and 100 μ M, the original classification achieved an overall accuracy of 93.8% correctly classifying most of the cases. The cross-validated results, which assess the performance of the model on unseen data, achieved an overall accuracy of 85.4%. For Cu(II) at 5 μ M concentration, the cross-validated accuracy drops to 50%, which suggests that the classification performance may be slightly reduced at lower concentrations for this particular metal ion.

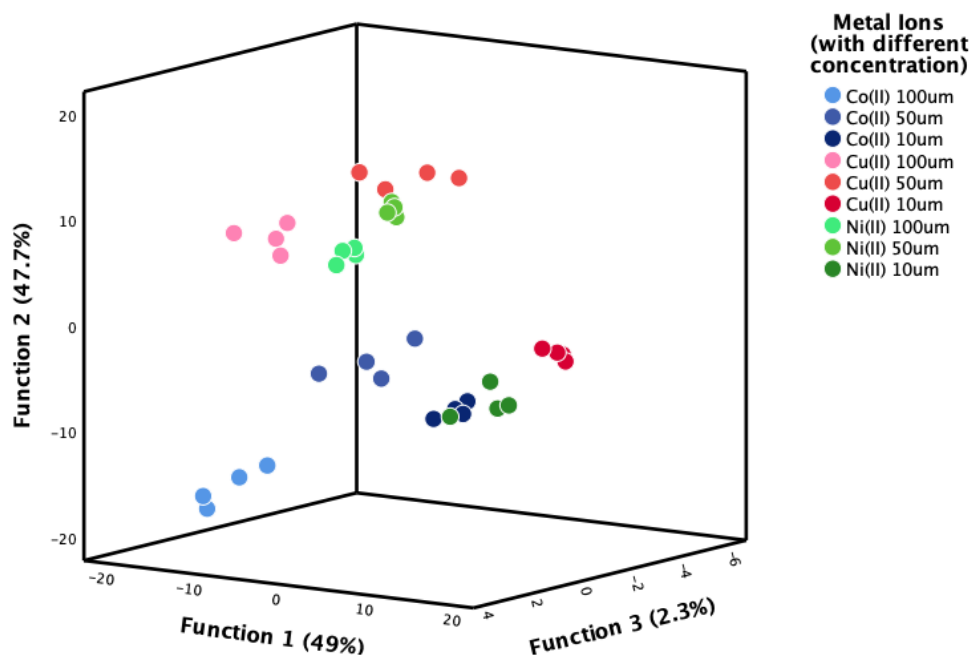


Figure 5.20 LDA score plots for the analysis of Cu(II), Co(II), and Ni(II) (1 mg/mL CDs, where $\lambda_{ex} = 370$ nm and $\lambda_{em} = 390-750$ nm) with different concentration of metal ions (10, 50, 100 μ M). (1 mg/mL CDs solutions, with four replicates)

5.2.10.3 Metal ions discrimination in a park water sample

The practicality of using the four CDs-based sensing array for metal ion identification was assessed using real pond water samples collected from Victoria Park, Sydney, Australia. To mitigate potential effects of environmental water on fluorescence response patterns, the samples were filtered through a 0.22 μ m membrane to remove particulate matter. Fourteen metal ions: Al(III), Ag(I), Ca(II), Co(II), Cu(II), Cd(II), Fe(III), Hg(II), Ni(II), Pb(II), Zn(II), Mg(II), Cr(II), and Mn(II), were added to the environmental water samples. CDs solutions were added to these metal ion-doped samples.

During the plate preparation, the volume percentage of the real sample used was 50% (v/v), and the pH buffer constituted 30% of the solution. The concentration of the metal ions was controlled at 100 μ M, and the CDs concentration was kept consistent at 1 mg/mL.

The LDA analysis of the results achieved an 89.3% classification accuracy, demonstrating the potential application of the four CDs-based array for metal ion detection in real samples, as shown in (Figure 5.21). However, it was observed that Ca(II), Hg(II), Zn(II), and Mg(II) were challenging

to classify accurately in the environmental water samples, likely due to their high concentrations in the lake water sample.

These findings indicate that high concentrations of certain metal ions in environmental water can affect the classification performance of the sensing array. Further optimisation and consideration of factors such as the concentration ranges of the metal ions or specific interferences in the environmental water may be necessary to improve the accuracy of classifying these challenging metal ions.

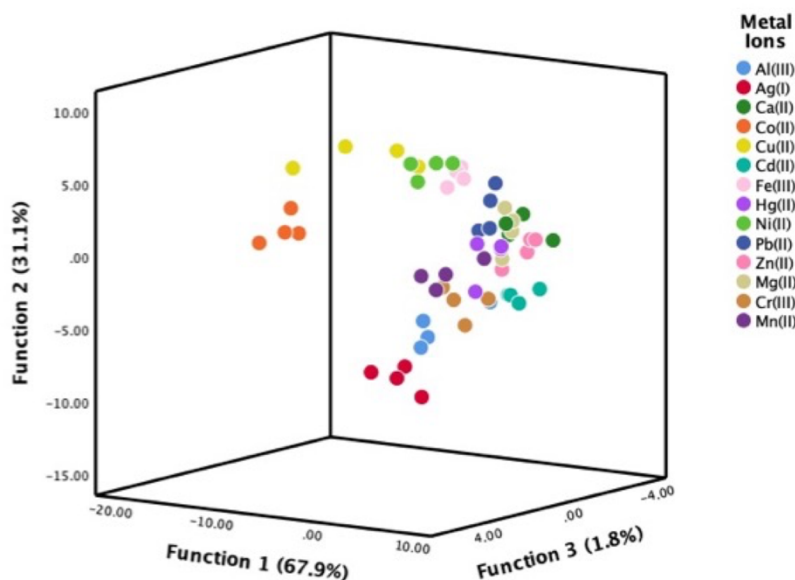


Figure 5.21 LDA score plots for the analysis of metal ions ($80\ \mu\text{M}$ metal ion, 1mg/mL CDs ($\lambda_{\text{ex}} = 370\ \text{nm}$, $\lambda_{\text{em}} = 390\text{--}750\ \text{nm}$)) in buffered Milli-Q water ($20\ \text{mM}$, $\text{pH } 7.0$, $30\%(\text{v/v})$), non-laboratory sample ($50\%(\text{v/v})$) for the array application of environmental sample with Victoria Park lake water (14 metal ions, with four replicates)

5.2.10.4 Metal ion discrimination in a urine sample

The four-CDs-based array was employed to discriminate metal ions in human urine samples. Eight selected metal ions were treated as analytes, they are Al(III) Ag(I) Ca(II) Co(II) Cu(II) Cd(II) Fe(III) Hg(II) Ni(II) and Cr(III) .

The detection of metal ions in urine serves as an indicator of exposure to heavy metals, which pose toxic and genotoxic threats to human health⁶¹. The biological significance and potential toxicity of each selected metal ion underscore the necessity for their assessment. Heavy metal ion exposure is associated with a plethora of health issues including developmental anomalies, cancers,

hypertension, immune deficiencies, and neurological impairments.⁶² Notably, Cu(II) and Fe(III) ions are indispensable for human physiological functions, contributing to enzymatic activities and oxygen transport.⁶³⁻⁶⁵ However, their elevated concentrations can precipitate oxidative stress and subsequent organ damage. The carcinogenic potential of certain metals, such as Cr(III), Cd(II), and Ni(II), is contingent upon their concentration, speciation, and duration of exposure, accentuating the imperative to scrutinise these ions within biological matrices.⁶⁶ Cd(II), acting as a metalloestrogen, plays a role in the aetiology of breast cancer, highlighting the extensive effects of metal ions on human health beyond mere toxicity⁶⁷. The high toxicity of Cd(II) and Hg(II), coupled with their lack of biological utility, precipitates severe health outcomes like renal impairment and neurological dysfunctions upon excessive exposure.^{68, 69} The selection encompasses a wide array of metal ions to evaluate both essential and deleterious elements in human urine, reflecting the diversity of common environmental pollutants.

Regarding environmental applications, for instance, Ni(II), Hg(II), Cr(II) and Cd(II), prevalent in industrial applications, raise concerns regarding human exposure and bioaccumulate through environmental contamination.^{70, 71} Monitoring such ions is vital for acquiring data on environmental exposure and its consequent impact on public health. The selection of metal ions presents varied analytical challenges, including disparities in size, charge, and chemical properties, thereby enabling the comprehensive evaluation and validation of the CDs-based array's adaptability and sensitivity. This variety ensures the applicability of the array across a spectrum of analytical tasks beyond the scope of this study. These ions were chosen based on their commonality and detectability in human urine, a critical excretory pathway for metals, offering a non-invasive approach for the surveillance of metal exposure and bodily accumulation. This selection is aimed at developing a methodology that is both pertinent and feasible for regular health surveillance.

Urine samples were filtered using a 0.22 μm membrane. During plate preparation, the volume percentage of the real sample is 50%(v/v), the pH 7.0 buffer is 30%, with the metal ions' concentration controlled at 100 μM and CDs concentration identical at 1 mg/mL.

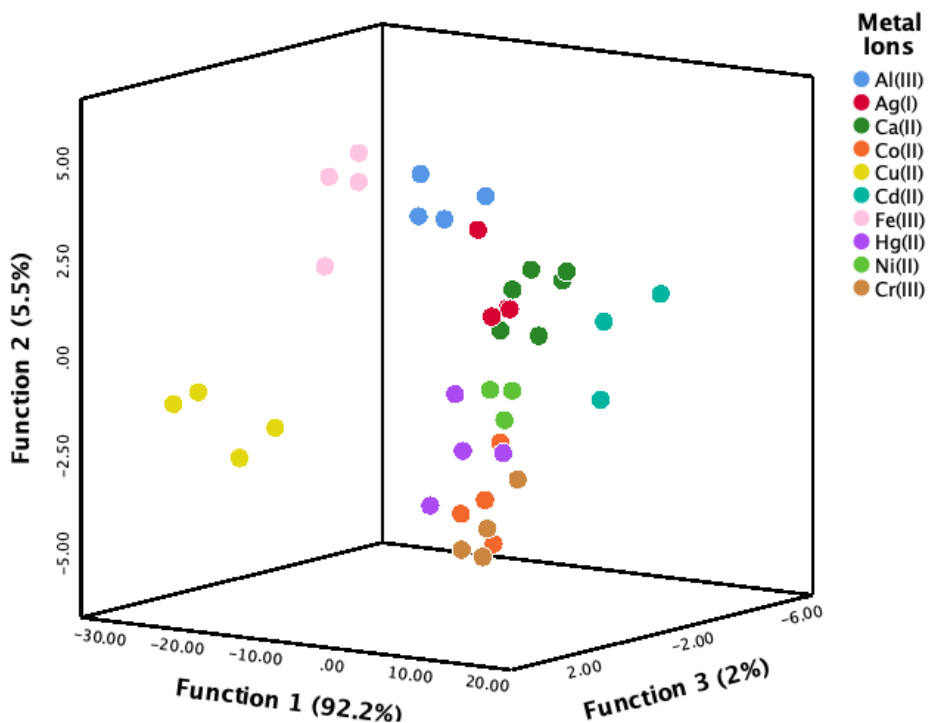


Figure 5.22 LDA score plots for the analysis of metal ions ($80\ \mu\text{M}$ metal ion, 1mg/mL CDs ($\lambda_{\text{ex}} = 370\ \text{nm}$, $\lambda_{\text{em}} = 390\text{--}750\ \text{nm}$)) in buffered Milli-Q water ($20\ \text{mM}$, $\text{pH } 7.0$, $30\%(\text{v/v})$), non-laboratory sample ($50\%(\text{v/v})$), for array application of biological sample with urine sample (10 metal ions, with four replicates)

The LDA classification for our CD-based array's sensing of metal ions in urine samples. From the summary of canonical discriminant functions listed in Table S5.11 in supporting material, the Wilks' Lambda test was utilised to evaluate the equality of group means among different CDs (CD_2-4_6 h, CD_1-0_6 h, CD_1-4_6 h, and CD_1-9_6 h) in terms of their metal ion sensing capabilities. The obtained p-values (Sig.) for all the tests were statistically significant ($p < 0.001$), indicating significant differences between the groups. The pooled within-group matrices displayed covariance and correlation values between the different CDs. The covariance values were close to zero, suggesting low covariance between the CDs. We looked at the eigenvalues to see how much variance each discriminant function explained. The first three canonical discriminant functions accounted for 92.2%, 5.5%, and 2.0% of the total variance, respectively. These three functions collectively explained 99.6% of the total variance.

The standardised canonical discriminant function coefficients were examined to understand the relative importance of the CDs in classifying the metal ions. Higher absolute coefficients suggested stronger contributions to the classification, listed in Table S5.16. Regarding to Table S5.17, The structure matrix revealed correlations between the discriminating variables (CDs) and the

standardised canonical discriminant functions, with variables ordered based on the absolute size of the correlation within each function. Functions at group centroids provided insight into the positions of the metal ions in the discriminant function space, where higher values on a specific function indicated a stronger association with that particular group. Each function is highly correlated with one of the sensors; regarding Function 1 to CD_2-4_6 h (r of 0.679); Function 2 to CD_1-9_6 h (r of 0.900); Function 3 to CD_1-0_6 h (r of 0.974); and Function 4 to CD_1-4_6 h (r of 0.928).

The classification results showed the predicted group membership based on the LDA classification. The original section displayed the count and percentage of correctly classified cases for each metal ion group, while the cross-validated section presented the classification results obtained through cross-validation. The overall classification accuracy was 95.0% for the original grouped cases and 87.5% for the cross-validated cases, indicating a high level of predictive accuracy.

The LDA classification for carbon-dot array sensing of metal ions in urine samples yielded promising results. The analysis revealed significant differences between the CDs in their ability to sense different metal ions. The derived discriminant functions effectively classified the metal ions with high overall accuracy. The coefficients, structure matrix, and group centroids provided valuable insights into the relative importance and relationships between the CDs and the metal ions. These findings suggest that the carbon-dot-based array sensing approach holds potential for reliable and accurate detection and classification of metal ions in urine samples.

5.3 Conclusions

In summary, a robust sensor array based on carbon dots (CDs) has been developed for the identification of essential and toxic metal ions. The CDs were synthesised using fructose and PPD via a hydrothermal reaction under optimised conditions. MATLAB-based SVM and LDA classifiers were created to evaluate and optimise the array performance using parameters including SVM and LDA classification accuracy, as well as ellipsoid volumes and ellipsoid distances of LDA classification. As results the optimised four CDs-based sensing array was determined, and the CDs selection was confirmed by PCA.

These CDs were selected for the sensor array owing to their ability to generate diverse turn-on and turn-off fluorescence emission intensity responses for the 14 metal analytes, based on the functional groups attached to their surface. The initial arrays in Milli-Q water demonstrated 100%

classification accuracy for the discrimination of the 14 metal ions, showing promise for environmental applications and saving time and costs associated with statistical analysis.

Moreover, the array's ability to distinguish Cu(II), Co(II), and Ni(II) under various conditions was evaluated and highlighted. The array successfully classified these three metal ions even in mixture conditions, different pH environments, and various metal concentrations, achieving high classification accuracy. This demonstrates the potential of the four CDs-based array to be used in a wide range of analysis media, including environmental samples and human urine samples. Such applications hold promise for future environmental metal monitoring and potential medical sample testing, providing valuable sensing benefits.

5.4 Materials and methods

5.4.1 Chemicals and characterisation

All chemical reagents and solvents used were laboratory or analytical grade purchased from Sigma-Aldrich, Merck or Combi-blocks. Precursors for carbon dot synthesis were fructose and p-phenylenediamine (PPD). pH 7 buffer (50 mM) was prepared by sodium phosphate dibasic heptahydrate and sodium phosphate monobasic monohydrate. Metal salts used were aluminium(III) nitrate, silver(I) nitrate, calcium(II) chloride, cobalt(II) nitrate hexahydrate, copper(II) nitrate trihydrate, cadmium(II) nitrate tetrahydrate, iron(III) nitrate nonahydrate, mercury(II) nitrate monohydrate, nickel(II) nitrate hexahydrate, lead(II) nitrate, zinc(II) nitrate hexahydrate, magnesium(II) nitrate, chromium(III) chloride hexahydrate, and manganese(II) chloride. All chemicals were used as received without further purification. X-ray photoelectron spectroscopy (XPS) data were obtained using a Kratos Axis Nova spectrometer (Kratos Analytical, UK), equipped with a monochromatised aluminium X-ray source (Al α 1486.6 eV) operating at 10 mA and 15 kV (150 kW). Survey and high-resolution spectra were acquired at detector pass energies of 160 and 20 eV, respectively. The XPS data were analysed with Thermo Advantage processing software (v5. 9902). All photophysical studies were performed on a PerkinElmer EnSpire Multimode Plate Reader in 300 μ L, 96-well polypropylene microplates (Item No.: 655209, Greiner Bio-One).

5.4.2 Preparation of carbon dots

A one-step hydrothermal/solvothermal synthesis route was employed to synthesise carbon dots. Fructose and PPD served as the precursor materials. The synthesis process involved mixing the precursors in 50 mL of Milli-Q water (Millipore, USA), and achieving a homogeneous mixture through ultrasonication. The resulting solution was then transferred into a Teflon-lined stainless-steel autoclave chamber with a 50% filling ratio. The detailed synthetic conditions for each type of CD are provided in Table 5.14.

After synthesis, the CDs series appeared as orange solutions with yellow fluorescence emission. The initial purification step involved passing the solution through a 0.22 μm cellulose acetate Millipore filter to remove any large or agglomerated particles. Subsequently, the sample was concentrated using a rotary evaporator (Heidolph, Germany), and dried using a freeze dryer for 48 h. To prepare 1 mg/mL CD solutions, the dried powder was reconstituted in Milli-Q water.

5.4.3 Array preparation and fluorescence spectroscopy

Arrays for distinguishing various metal ions were prepared by mixing aqueous metal salt stock solutions (80 mM), freshly prepared aqueous carbon dots (CDs) sensor stock solutions (1 mg/mL), and Milli-Q water in 300 μL 96-well polypropylene microplates to achieve the desired concentrations. Each well was adjusted to a final volume of 200 μL using a 20 mM phosphate buffer at pH 7.0. For non-laboratory samples, including Victoria Park lake water and urine samples, the mixtures comprised 80 μM metal ion concentration, 1 mg/mL CDs (with excitation wavelength $\lambda_{\text{ex}} = 370$ nm and emission wavelength $\lambda_{\text{em}} = 390\text{--}750$ nm), in a buffered Milli-Q water solution (20 mM, pH 7.0, 30% v/v), and non-laboratory sample (50% v/v). Wherein, Victoria Park lake water was tested for the presence of 14 different metal ions, and urine samples were analysed for 8 different metal ions. In every sensor replicate, a control well containing no metal ion was included to normalise data for analysis. All array data were collected using a PerkinElmer EnSpire Multimode Plate Reader. The microplate was agitated for 30 seconds prior to measurements. Fluorescence emission spectra for each well were recorded from 390 to 750 nm at 5 nm increments, upon excitation at 370 nm.

Table 5.14 Summary of synthesis condition of fructose and p-phenylenediamine based carbon dots

Carbon dots NO.	Precursors			Synthesis time (h)	Synthesis temperature (°C)	
	Fructose (g)	p-Phenylenediamine (g)	mass ratio of fructose: p-Phenylenediamine			
CD_1-0_6 h	0.5	0	1:00	6	180 °C	
CD_9-1_6 h	0.469	0.031	9:01			
CD_4-1_6 h	0.43	0.07	4:01			
CD_4-2_6 h	0.385	0.115	4:02			
CD_4-3_6 h	0.345	0.155	4:03			
CD_4-4_6 h	0.312	0.188	4:04			
CD_3-4_6 h	0.278	0.222	3:04			
CD_2-4_6 h	0.227	0.273	2:04			
CD_1-4_6 h	0.147	0.353	1:04			
CD_3-17_6 h	0.114	0.386	3:17			
CD_1-9_6 h	0.078	0.422	1:09			
CD_1-0_8 h	0.5	0	1:00	8		
CD_9-1_8 h	0.469	0.031	9:01			
CD_4-1_8 h	0.43	0.07	4:01			
CD_4-2_8 h	0.385	0.115	4:02			
CD_4-3_8 h	0.345	0.155	4:03			
CD_4-4_8 h	0.312	0.188	4:04			
CD_3-4_8 h	0.278	0.222	3:04			
CD_2-4_8 h	0.227	0.273	2:04			
CD_1-4_8 h	0.147	0.353	1:04			
CD_3-17_8 h	0.114	0.386	3:17			
CD_1-9_8 h	0.078	0.422	1:09			

5.4.4 Statistical analysis of fluorescence array data

After the MATLAB program provided the sensor selections, SPSS Statistics version 25 (IBM) was applied for data analysis and verification of the classification accuracy by using linear discriminant analysis (LDA). We subjected the normalised integrated emission spectra to LDA, using the metal analytes as the grouping variable and normalised sensor responses as independent variables. The data were classified as all groups being equal and a combined groups map, territorial maps and classification with leave-one-out cross-validation summary tables were produced for interpretation.

Discriminant scores and predicted membership were also recorded. For classifying unknown samples, data were excluded from the grouping variable, and LDA was employed to predict group membership by assigning discriminant function values for each replicate and assigning them a location on the territorial map. Robustness test was designed based on this operating principle, to verify the stability of the array sensing performance.

5.4.5 Machine learning for array development

The value of the normalised integrated emission spectra used to detect an analyte divided by the normalised integrated emission spectra of the control group is regarded as the sensory response to the analyte. These ratios were summarised and labelled in a table, serving as an input file for our programme, which was implemented using LIBSVM, a library for support vector machines designed by researchers at National Taiwan University ⁴⁹. We developed MATLAB based programme which could systematically processes all feature selections, employing the chosen features for SVM classification. The features in the software of this project correspond to different types of CDs. This program hired support vector machines (SVMs) classification results as a criterion for array feature selection evaluation. And all the array selection with SVM classification accuracy of 100% were listed. Compared to principal component analysis (PCA), commonly used for dimension reduction in reported arrays, our programme provides a more innovative and efficient approach for optimising array feature selection.

Reported common dimension reduction was also performed using principal component analysis, based on SPSS. The normalised sensor responses grouped by metal analyte were entered as variables (no selection variable). The number of principal components extracted was determined through Scree plot interpretation (eigenvalues >1) and the components underwent a varimax rotation. Outputs from PCA such as measures of sampling adequacies (MSAs), communality scores and correlations were used to inform which sensors to exclude from the array upon array simplification. Bases on the PCA guidance the sensor selection, the simplified sensors still required manually test to confirm the classification accuracy. For this array development involved twenty-two sensors can generate more than one million array sensing selections. Manual testing of all possible sensor combinations using PCA analysis is extremely time-consuming. In contrast, our MATLAB-based programme automates sensor selection, requiring no additional operations beyond submitting input data.

5.5 Reference

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6 Conclusion and outlook

In this thesis, novel carbon dot (CD)-based nanoprobe have been prepared and evaluated for their unique fluorescence properties, aiming at effective pH and metal ion sensing applications. The primary objectives of this research were achieved across multiple chapters, each contributing to the advancement of CD-based sensing technologies.

Chapter 3 reports a series of fructose-derived CDs with varying levels of nitrogen doping were synthesised using a one-step hydrothermal method. The impact of surface functional groups on the pH responsiveness of CDs was extensively investigated, revealing how specific groups, such as hydroxyl and amino functionalities, modulate fluorescence behaviour. An optimised pH sensor array was developed using machine learning techniques, including Gaussian process regression (GPR) and linear discriminant analysis (LDA). GPR was utilised to predict pH levels accurately, while LDA enhanced classification accuracy across a wide pH range. These optimised arrays demonstrated high accuracy in differentiating between fresh and expired milk samples based on pH, showcasing the practical potential of CDs in food safety and quality monitoring. This research underscored the utility of integrating nanomaterials with machine learning for advanced sensing applications, offering valuable insights for the future design and optimisation of pH-responsive CDs.

In Chapter 4, fluorescent sensors for Cu(II) and Fe(III) detection were introduced, featuring both biomass-based CDs from Jacaranda leaves and chemically synthesised fructose-based CDs. The study highlighted how nitrogen doping significantly enhanced the CDs' optical properties and broadened their applicability. The N-doped fructose-based CDs exhibited selective quenching for Cu(II) with a detection limit of 36 μM , while the fructose-based CDs effectively detected Fe(III) with a detection limit of 44 μM . Additionally, Jacaranda-derived CDs were found to exhibit dual fluorescence peaks, with Jacaranda_(NaOH)_CDs demonstrating promising full-range pH sensitivity. A ratiometric pH sensor was then developed by combining nitrogen doping with Jacaranda CDs' inherent fluorescence properties, producing a turn-off signal at 430 nm and a turn-on signal at 650 nm, thus establishing a robust methodology for pH sensing across various conditions. This chapter illustrated the versatility of CDs in metal ion sensing and the role of tailored surface functionalisation in enhancing selectivity and sensitivity.

Chapter 5 focused on advancing metal ion detection through the development of a robust CD-based sensor array, capable of identifying 14 different metal ions. Synthesised from fructose and p-phenylenediamine (PPD) under optimised hydrothermal conditions, the selected four-CD array demonstrated excellent classification accuracy through support vector machine (SVM) and LDA analyses. The array effectively distinguished between metal ions, even under complex conditions such as mixed-ion environments and varied pH, with applications tested on environmental and biological samples, including human urine. This array exemplifies a significant improvement over traditional metal ion detection techniques, providing a selective, cost-effective, and reliable approach for environmental and biomedical monitoring.

Overall, this thesis highlights the potential of carbon dots as versatile, high-performance sensing platforms for pH and metal ion detection. The integration of machine learning techniques, such as GPR, LDA, and SVM, enhanced the selectivity and precision of these CD-based sensors, positioning them as valuable tools for real-world applications. Future research should explore further functionalisation strategies and investigate broader applications of CDs in complex environmental and biological matrices, paving the way for CD-based sensors to become integral components in advanced analytical technologies for environmental, industrial, and biomedical settings.

6.1 Outlook

Future research directions for each chapter involve optimising sensing efficacy, expanding biocompatibility and imaging capabilities, and exploring size effects on sensing, all of which aim to develop more robust and versatile sensors for real-world applications.

Chapter 3 explored fructose-based carbon dots with varying levels of nitrogen doping, highlighting their effectiveness in pH sensing across a broad pH range (pH 3 to 10). To expand their utility, future research should investigate the biocompatibility of these CDs through cell viability tests, thereby establishing their safety for biological applications. Additionally, testing these pH-sensitive CDs in cell imaging applications would allow for real-time pH monitoring within cellular environments. Such studies would refine the design of CD-based sensors for biomedical diagnostics and expand their applicability in live-cell imaging and pH-sensitive tracking within biological samples.

In Chapter 4, the development of biomass-derived and fructose-based CDs was demonstrated for selective sensing of Fe(III) and Cu(II) ions, with nitrogen doping shown to enhance

selectivity and fluorescence properties. Future studies could continue to explore additional dopants or functionalisation techniques to expand selectivity for a wider array of metal ions. Computational studies, such as density functional theory (DFT), and advanced spectroscopic analyses could be employed to better understand metal ion interactions at the molecular level. Another promising direction involves conjugating fluorescent probes onto CD scaffolds to improve biocompatibility and multifunctionality. By treating CDs as versatile scaffolds, functional fluorescent probes could be added to enable simultaneous imaging and metal ion sensing, potentially enhancing their utility in cellular and in vivo applications. Testing these functionalised CDs in biological media would validate their dual imaging and sensing potential in biomedical applications.

Chapter 5 advances the field of CD-based array sensing by developing a four-CD sensor array optimised through machine learning for detecting diverse metal ions. Regarding the cross-reactive metal ions sensor array based on CDs, future research should investigate the role of CD size in influencing metal selectivity. Specifically, synthesising CDs under consistent conditions and then separating them by size using dialysis bags could reveal whether size-specific fractions display distinct metal selectivities. Comparative studies of pre- and post-separation CDs would assess how the collective metal selectivity of a mixed-size CD population results from synergy across sizes, potentially providing a new understanding of CD metal sensing behaviour. Enhanced machine learning models such as neural networks could further aid in optimising the array's performance by adapting to more complex sample matrices.

The research presented in this thesis highlights the adaptability and efficacy of carbon dots in chemical sensing, with machine learning playing a crucial role in optimising array performance and enabling practical applications. Future studies should focus on integrating these sensors into automated systems for real-time environmental or diagnostic applications. By expanding training datasets and refining machine learning capabilities, CD-based sensors could ultimately support accessible, accurate, and real-time monitoring in diverse settings. This pathway holds great potential to advance cost-effective, high-performance sensors for environmental safety, biomedical diagnostics, and industrial monitoring.

7 Appendices

7.1 Appendix I: supporting material for chapter two

7.1.1 MATLAB based SVM classification code and protocol

7.1.1.1 Coding

```
% Load training and testing data from excel files
training_data = readtable('training_data.xlsx');
testing_data = readtable('testing_data.xlsx');

% Extract classification labels (first column) and features (other columns)
train_labels = training_data; % Labels are in the first column
train_features = training_data{:,2:end}; % Features are in the subsequent columns

test_labels = testing_data{:,1}; % Testing data labels
test_features = testing_data{:,2:end}; % Testing data features

% Define folder for saving results
results_folder = 'SVM_results';
if ~exist(results_folder, 'dir')
    mkdir(results_folder);
end

% Initialize a variable to store classification results
result_data = {};

% Get the number of features
num_features = size(train_features, 2);

% Ask the user for the minimum and maximum combination sizes
min_combination_size = input('Enter the minimum number of features for combination: ');
max_combination_size = input('Enter the maximum number of features for combination: ');

% Define the list of kernel functions to test
kernels = {'linear', 'rbf', 'polynomial', 'sigmoid'};

% Loop through different sizes of feature combinations
for combination_size = min_combination_size:max_combination_size
    % Get all possible combinations of the specified number of features
    feature_combinations = nchoosek(1:num_features, combination_size);

    for i = 1:size(feature_combinations, 1)
        % Select features based on the current combination
        features_idx = feature_combinations(i,:);
        train_subset = train_features(:, features_idx);
        test_subset = test_features(:, features_idx);
```

```

% Try different kernel functions
for j = 1:length(kernels)
    kernel_function = kernels{j};

    % Start timer for measuring time taken
    tic;

    % Perform parameter optimization using grid search for each kernel function
    svm_template = templateSVM('KernelFunction', kernel_function);
    model = fitsvm(train_subset, train_labels, 'KernelFunction', kernel_function, ...
        'OptimizeHyperparameters', 'auto', ...
        'HyperparameterOptimizationOptions', struct('UseParallel', true));

    % Predict and evaluate on testing set
    predicted_labels = predict(model, test_subset);
    accuracy = sum(strcmp(predicted_labels, test_labels)) / length(test_labels) * 100;

    % End timer and get elapsed time
    time_taken = toc;

    % Store the results
    timestamp = datestr(now, 'yyyy-mm-dd HH:MM:SS'); % Get current date and time
    model_details = model.HyperparameterOptimizationResults; % Get model details

    % Save the relevant information into result_data
    result_data{end+1, 1} = features_idx; % Feature combination
    result_data{end+1, 2} = kernel_function; % Kernel function used
    result_data{end+1, 3} = accuracy; % Classification accuracy
    result_data{end+1, 4} = time_taken; % Time taken for the combination
    result_data{end+1, 5} = timestamp; % Timestamp for the run
    result_data{end+1, 6} = model_details; % Model details from optimization

    % For two-feature combinations, plot the classification result
    if combination_size == 2
        figure;
        gscatter(train_subset(:,1), train_subset(:,2), train_labels);
        hold on;
        sv = model.SupportVectors;
        plot(sv(:,1),sv(:,2),'ko','MarkerSize',10);
        title(['SVM Classification with Features ', num2str(features_idx), ', Kernel: ',
kernel_function]);
        xlabel(['Feature ', num2str(features_idx(1))]);
        ylabel(['Feature ', num2str(features_idx(2))]);

        % Save the plot
        saveas(gcf, fullfile(results_folder, ['SVM_Classification_Features_',
num2str(features_idx), '_Kernel_', kernel_function, '.png']));
        close;
    end
end

```

```

        end
    end
end

% Convert results to a table and save as Excel file
result_table = cell2table(result_data, 'VariableNames', ...
    {'Feature_Combination', 'Kernel_Function', 'Accuracy', 'Time_Taken', 'Timestamp',
    'Model_Details'});
writetable(result_table, fullfile(results_folder, 'SVM_Results_With_Kernels.xlsx'));

disp('SVM classification completed. Results saved in SVM_results folder.');
```

7.1.1.2 Application Protocol

Prepare the Excel Files:

- As before, ensure the training_data.xlsx and testing_data.xlsx are in the same directory as the script.
- The first column should contain the labels, and the remaining columns should contain the features.

Run the Script:

- Input the minimum and maximum combination sizes when prompted. For instance, if you want combinations of 2 to 3 features, enter 2 and 3.

SVM Model Training:

- The code automatically tests different kernel functions for SVM: 'linear', 'rbf', 'polynomial', and 'sigmoid'.
- Each kernel undergoes parameter optimization using MATLAB's fitsvm function with grid search (OptimizeHyperparameters).

Results and Plots:

- The accuracy, kernel type, feature combination, time taken, and model details are stored in the SVM_Results_With_Kernels.xlsx file.
- For two-feature combinations, classification plots are generated and saved as PNG files.

Explore Results:

- Open the results file to view details of each kernel function's performance with different feature combinations.
- Review the saved plots for a visual understanding of how the classification worked for two-feature combinations.

7.1.2 MATLAB based LDA classification code and protocol

7.1.2.1 Coding

```

% Load training and testing data from excel files
training_data = readtable('training_data.xlsx');
testing_data = readtable('testing_data.xlsx');

% Extract classification labels (first column) and features (other columns)
train_labels = training_data{:,1}; % Labels are in the first column
```

```

train_features = training_data{:,2:end}; % Features are in the subsequent columns

test_labels = testing_data{:,1}; % Testing data labels
test_features = testing_data{:,2:end}; % Testing data features

% Define folder for saving results
results_folder = 'LDA_results';
if ~exist(results_folder, 'dir')
    mkdir(results_folder);
end

% Initialize a variable to store classification results
result_data = {};

% Get the number of features
num_features = size(train_features, 2);

% Ask the user for the minimum and maximum combination sizes
min_combination_size = input('Enter the minimum number of features for combination: ');
max_combination_size = input('Enter the maximum number of features for combination: ');

% Loop through different sizes of feature combinations
for combination_size = min_combination_size:max_combination_size
    % Get all possible combinations of the specified number of features
    feature_combinations = nchoosek(1:num_features, combination_size);

    for i = 1:size(feature_combinations, 1)
        % Select features based on the current combination
        features_idx = feature_combinations(i,:);
        train_subset = train_features(:, features_idx);
        test_subset = test_features(:, features_idx);

        % Start timer for measuring time taken
        tic;

        % Train the LDA model
        model = fitcdiscr(train_subset, train_labels, 'DiscrimType', 'linear');

        % Predict and evaluate on testing set
        predicted_labels = predict(model, test_subset);
        accuracy = sum(strcmp(predicted_labels, test_labels)) / length(test_labels) * 100;

        % End timer and get elapsed time
        time_taken = toc;

        % Store the results
        timestamp = datestr(now, 'yyyy-mm-dd HH:MM:SS'); % Get current date and time
        model_details = model; % Store the model itself for reference

        % Save the relevant information into result_data

```

```

result_data{end+1, 1} = features_idx; % Feature combination
result_data{end+1, 2} = accuracy; % Classification accuracy
result_data{end+1, 3} = time_taken; % Time taken for the combination
result_data{end+1, 4} = timestamp; % Timestamp for the run
result_data{end+1, 5} = model_details; % Model details (LDA model)

% For two-feature combinations, plot the classification result
if combination_size == 2
    figure;
    gscatter(train_subset(:,1), train_subset(:,2), train_labels);
    hold on;

    % Plot decision boundary
    K = model.Coeffs(1,2).Const;
    L = model.Coeffs(1,2).Linear;
    f = @(x1, x2) K + L(1)*x1 + L(2)*x2;
    h2 = fimplicit(f, [min(train_subset(:,1)) max(train_subset(:,1)) min(train_subset(:,2))
max(train_subset(:,2))]);
    h2.Color = 'k';
    h2.LineWidth = 2;

    title(['LDA Classification with Features ', num2str(features_idx)]);
    xlabel(['Feature ', num2str(features_idx(1))]);
    ylabel(['Feature ', num2str(features_idx(2))]);

    % Save the plot
    saveas(gcf, fullfile(results_folder, ['LDA_Classification_Features_',
num2str(features_idx), '.png']));
    close;
end
end
end

```

```

% Convert results to a table and save as Excel file
result_table = cell2table(result_data, 'VariableNames', ...
{'Feature_Combination', 'Accuracy', 'Time_Taken', 'Timestamp', 'Model_Details'});
writetable(result_table, fullfile(results_folder, 'LDA_Results.xlsx'));

```

```

disp('LDA classification completed. Results saved in LDA_results folder.');
```

7.1.2.2 Application Protocol

Prepare the Excel Files:

- As in the previous versions, ensure the training_data.xlsx and testing_data.xlsx files are in the same directory as the script.
- The first column of each file should contain the classification labels (target labels), and the remaining columns should contain numerical features for classification.

Run the Script:

- When prompted, input the minimum and maximum combination sizes for the feature combinations. For example, if you want to explore combinations of 2 to 3 features, enter 2 and 3.

LDA Model Training:

- The script will perform LDA-based classification on the training data for each combination of features, using `fitcdiscr` with the `DiscrimType` set to 'linear'.
- It will then evaluate the model on the testing data and compute classification accuracy.

Results and Plots:

- The classification results (accuracy, time taken, timestamp, and model details) are saved in the `LDA_Results.xlsx` file within the `LDA_results` folder.
- For two-feature combinations, a classification plot with the decision boundary is generated and saved as a PNG file in the same folder.

Explore Results:

- You can review the saved results by opening the `LDA_Results.xlsx` file.
- Examine the plots for a visual understanding of how well the LDA model separates the classes using two features.

7.2 Appendix II: supporting material for chapter three

7.2.1 Characterisation of carbon dots

7.2.1.1 Absorption

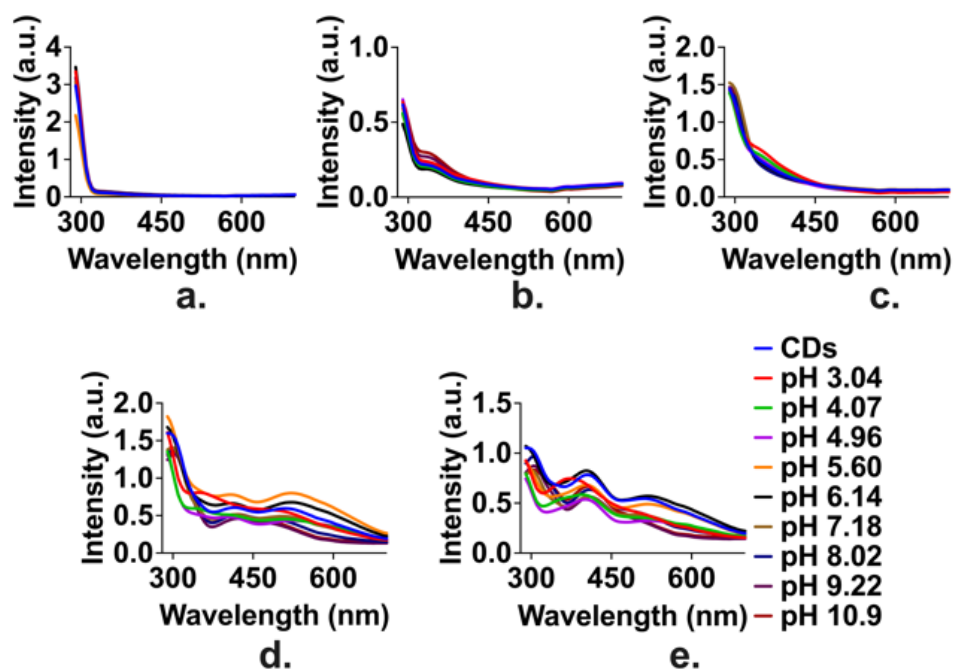


Figure S3. 1: UV-vis absorption of five CDs under different pH buffers (a. CD100, b. CD92, c. CD69, d. CD43, and e. CD25, f. summary of CDs, at 0.25 mg/mL respectively.)

7.2.1.2 Naked eye observation

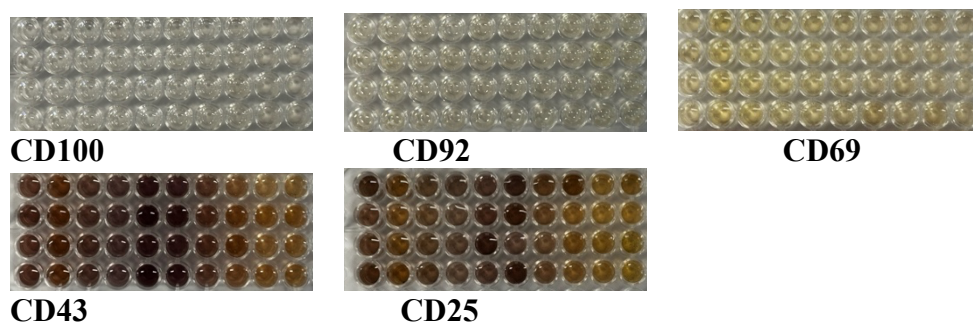


Figure S3. 2 Naked eye visual observation of CDs pH probes under varying pH conditions (Each photo contains four replicates, with each row representing one replicate. From left to right, the columns represent CDs in MilliQ water, followed by CDs exposed to pH levels 3.04, 4.07, 4.96, 5.60, 6.14, 7.18, 8.02, 9.22, and 10.89).

7.2.2 Nine CDs based array overview

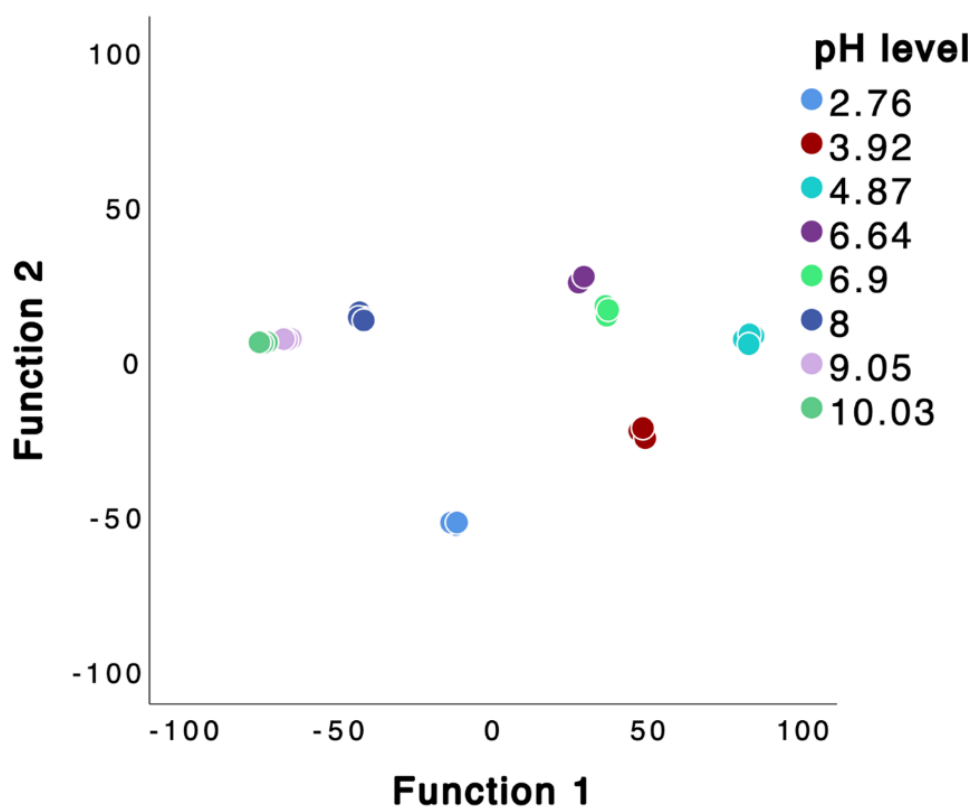


Figure S3. 3 The LDA classification results by using nine CDs based array as pH sensor array to discriminate 8 different pH environments.

Table S43. 10 LDA classification results by using nine CDs as pH sensor array

Classification Results ^{a,c}											
Items		pH	Predicted Group Membership							10	Total
			2.8	3.9	4.9	6.6	6.9	8	9.1		
Original	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
a. 100.0% of original grouped cases correctly classified.											
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.											
c. 100.0% of cross-validated grouped cases correctly classified.											

7.2.3 MATLAB based LDA and SVM

7.2.3.1 MATLAB code development

Here we introduce the development of MATLAB based array classification and optimisation program. This MATLAB code was designed to develop and evaluate both support vector machine (SVM) and linear discriminant analysis (LDA) models for classifying pH levels based on feature data.

Data loading and preprocessing: The code began by clearing the workspace and setting up directories and files for storing results. It then loaded the dataset from an Excel file and displayed a message indicating the successful data load:

```
clear;
%% Load data
files = strcat('Data.xlsx');
mkdir result
fileID = fopen('result/SVM_LDA_result.txt','w');
[data,txt,row] = xlsread(files); % load data
disp('Data loaded successfully');
disp(['Size of data: ', num2str(size(data))]);

% Extract pH levels and features
pH_levels = txt(2:end, 1); % Assuming the first row is the header
features = data(:, 1:end); % Features are from the first column to the last column
```

The feature data were then normalised, and the normalised data were saved to a file:

```
% Normalize the feature data
normalized_features = normalize(features);

% Save the normalized data to a file
normalized_data_filename = 'result/normalized_data.xlsx';
normalized_data = [pH_levels, num2cell(normalized_features)];
writecell([txt(1, :); normalized_data], normalized_data_filename);
disp(['Normalized data saved to ', normalized_data_filename]);
```

Data conversion and cross-validation setup: The pH levels were converted to categorical labels, and a 4-fold cross-validation setup was prepared:

```
% Convert pH levels to categorical labels
[pH_levels, ~, labels] = unique(pH_levels);

% Determine the number of unique pH levels
numClasses = length(unique(labels));

indices = crossvalind('Kfold', labels, 4); % Develop 4-fold cross-validation
```

Model training and performance evaluation: For each combination of features, the script trained SVM and LDA models and evaluated their performance using cross-validation. The performance metrics were stored in a table and written to a result file:

```

% Initialize a table to store performance metrics
columnNames = {'FeatureSet', 'SVM_Accuracy', 'SVM_Precision', 'SVM_Recall', 'SVM_F1', 'LDA_Accuracy',
'LDA_Precision', 'LDA_Recall', 'LDA_F1'};
perfMetrics = table('Size', [0, 9], 'VariableTypes', {'string', 'double', 'double', 'double', 'double', 'double', 'double',
'double', 'double'}, 'VariableNames', columnNames);

for feature_num = 1:9 % Assuming 9 features to represent the CDs
    disp(['Processing feature number: ', num2str(feature_num)]);
    C = nchoosek(1:9, feature_num); % Combinations of features
    for i = 1:size(C, 1)
        disp(['Processing combination: ', num2str(i), ' of feature_num: ', num2str(feature_num)]);
        feature_select = C(i,:);
        feature_data = normalized_features(:, feature_select); % Select features

        % Initialize variables to accumulate metrics
        metrics_svm = zeros(4, 4); % 4 metrics (accuracy, precision, recall, f1) for each fold
        metrics_lda = zeros(4, 4);
        for k = 1:4 % 4-fold cross-validation
            disp(['Cross-validation fold: ', num2str(k)]);
            test = (indices == k);
            train = ~test;
            train_data = feature_data(train, :);
            train_label = labels(train);
            test_data = feature_data(test, :);
            test_label = labels(test);

            % Train SVM model using fitcecoc for multi-class classification
            svm_model = fitcecoc(train_data, train_label, 'Learners', templateSVM('KernelFunction', 'linear',
'BoxConstraint', 1));
            predicted_label_svm = predict(svm_model, test_data);
            [acc_svm, prec_svm, rec_svm, f1_svm] = calculateMetrics(predicted_label_svm, test_label,
numClasses);
            metrics_svm(k, :) = [acc_svm, prec_svm, rec_svm, f1_svm];
            disp(['SVM accuracy on fold ', num2str(k), ': ', num2str(acc_svm)]);

            % Train LDA model with 'pseudoLinear' type
            lda_model = fitcdiscr(train_data, train_label, 'DiscrimType', 'pseudoLinear');
            predicted_label_lda = predict(lda_model, test_data);
            [acc_lda, prec_lda, rec_lda, f1_lda] = calculateMetrics(predicted_label_lda, test_label, numClasses);
            metrics_lda(k, :) = [acc_lda, prec_lda, rec_lda, f1_lda];
            disp(['LDA accuracy on fold ', num2str(k), ': ', num2str(acc_lda)]);
        end
        disp(['Completed processing feature set: ', num2str(feature_select)]);
        % Calculate average metrics over folds
        avgMetrics_svm = mean(metrics_svm, 1);
        avgMetrics_lda = mean(metrics_lda, 1);
        % Store in the performance metrics table
        newRow = {mat2str(feature_select), avgMetrics_svm(1), avgMetrics_svm(2), avgMetrics_svm(3),
avgMetrics_svm(4), avgMetrics_lda(1), avgMetrics_lda(2), avgMetrics_lda(3), avgMetrics_lda(4)};
        perfMetrics = [perfMetrics; newRow];

        % Write performance metrics to file after each feature set processing
        fprintf(fileID, 'Feature set: %s, SVM: Acc: %f, Prec: %f, Rec: %f, F1: %f, LDA: Acc: %f, Prec: %f,
Rec: %f, F1: %f\n', ...
            perfMetrics.FeatureSet{end}, perfMetrics.SVM_Accuracy(end), perfMetrics.SVM_Precision(end),
perfMetrics.SVM_Recall(end), perfMetrics.SVM_F1(end), ...
            perfMetrics.LDA_Accuracy(end), perfMetrics.LDA_Precision(end), perfMetrics.LDA_Recall(end),
perfMetrics.LDA_F1(end));
    end
end

```

```
fclose(fileID);
disp('Program completed.');
```

Best performing feature set identification: The script identified the best performing feature set for both SVM and LDA based on the accuracy metric:

```
% Identify the best performing combination for SVM
[~, best_svm_idx] = max(perfMetrics.SVM_Accuracy);
best_svm_features = perfMetrics.FeatureSet{best_svm_idx};
best_svm_accuracy = perfMetrics.SVM_Accuracy(best_svm_idx);
best_svm_precision = perfMetrics.SVM_Precision(best_svm_idx);
best_svm_recall = perfMetrics.SVM_Recall(best_svm_idx);
best_svm_f1 = perfMetrics.SVM_F1(best_svm_idx);
```

```
disp('Best SVM Feature Set:');
disp(['Feature Set: ', best_svm_features]);
disp(['Accuracy: ', num2str(best_svm_accuracy)]);
disp(['Precision: ', num2str(best_svm_precision)]);
disp(['Recall: ', num2str(best_svm_recall)]);
disp(['F1 Score: ', num2str(best_svm_f1)]);
```

```
% Identify the best performing combination for LDA
[~, best_lda_idx] = max(perfMetrics.LDA_Accuracy);
best_lda_features = perfMetrics.FeatureSet{best_lda_idx};
best_lda_accuracy = perfMetrics.LDA_Accuracy(best_lda_idx);
best_lda_precision = perfMetrics.LDA_Precision(best_lda_idx);
best_lda_recall = perfMetrics.LDA_Recall(best_lda_idx);
best_lda_f1 = perfMetrics.LDA_F1(best_lda_idx);
```

```
disp('Best LDA Feature Set:');
disp(['Feature Set: ', best_lda_features]);
disp(['Accuracy: ', num2str(best_lda_accuracy)]);
disp(['Precision: ', num2str(best_lda_precision)]);
disp(['Recall: ', num2str(best_lda_recall)]);
disp(['F1 Score: ', num2str(best_lda_f1)]);
```

Utility function for metric calculation: A utility function was included to calculate the accuracy, precision, recall, and F1 score for the predicted labels:

```
function [accuracy, precision, recall, f1score] = calculateMetrics(predicted, actual, numClasses)
% Calculate confusion matrix
cm = confusionmat(actual, predicted);

% Initialize metrics
classPrecision = zeros(numClasses, 1);
classRecall = zeros(numClasses, 1);

% Calculate precision and recall for each class
for i = 1:numClasses
    truePositives = cm(i, i);
    falsePositives = sum(cm(:, i)) - truePositives;
    falseNegatives = sum(cm(i, :)) - truePositives;

    classPrecision(i) = truePositives / (truePositives + falsePositives);
    classRecall(i) = truePositives / (truePositives + falseNegatives);
end
```

```

% Handle NaN cases (if any division by zero occurs)
classPrecision(isnan(classPrecision)) = 0;
classRecall(isnan(classRecall)) = 0;

% Calculate macro-averaged precision, recall, and F1 score
precision = mean(classPrecision);
recall = mean(classRecall);
f1score = 2 * (precision * recall) / (precision + recall);

% Calculate overall accuracy
accuracy = sum(diag(cm)) / sum(cm(:));
end

```

This structured approach ensured a thorough evaluation of different feature combinations, providing insights into the optimal configuration for accurate pH level classification using SVM and LDA models.

7.2.3.2 MATLAB based array optimisation results

Table S3. 11 The features numbers matched for carbon dots

Feature number	Carbon dot
1	CD100
2	CD100 (-OH protected)
3	CD69
4	CD69 (-OH protected)
5	CD69 (-NH ₂ protected)
6	CD25
7	CD25 (-NH ₂ protected)
8	CD92
9	CD43

Table S3. 12 Summary of the optimisation of SVM and LDA using all combinations of features shown in Table S6. Acc = accuracy; Prec = precision; Rec = recall; F1 = F1 score (F-measure).

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1	0.750000	0.672619	0.750000	0.708981	0.785714	0.732143	0.785714	0.757475
2	0.250000	0.098214	0.250000	0.139751	0.214286	0.113095	0.214286	0.144558
3	0.785714	0.714286	0.785714	0.747239	0.750000	0.696429	0.750000	0.720842
4	0.428571	0.297619	0.428571	0.346918	0.500000	0.386905	0.500000	0.434917
5	0.571429	0.470238	0.571429	0.515699	0.714286	0.595238	0.714286	0.649093
6	0.571429	0.488095	0.571429	0.526084	0.642857	0.535714	0.642857	0.580525
7	0.464286	0.375000	0.464286	0.412245	0.571429	0.470238	0.571429	0.515306
8	0.714286	0.642857	0.714286	0.675604	0.750000	0.732143	0.750000	0.740683
9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2	0.678571	0.559524	0.678571	0.612186	0.785714	0.726190	0.785714	0.754012
1 3	0.857143	0.803571	0.857143	0.829111	0.857143	0.821429	0.857143	0.838509
1 4	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 5	0.892857	0.851190	0.892857	0.870785	0.892857	0.851190	0.892857	0.870785
1 6	0.928571	0.892857	0.928571	0.909938	0.928571	0.892857	0.928571	0.909938
1 7	0.750000	0.696429	0.750000	0.721679	0.750000	0.678571	0.750000	0.712444
1 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3	0.571429	0.410714	0.571429	0.472666	0.714286	0.678571	0.714286	0.695445
2 4	0.607143	0.455357	0.607143	0.518857	0.535714	0.458333	0.535714	0.492515
2 5	0.535714	0.389881	0.535714	0.451220	0.678571	0.565476	0.678571	0.615058
2 6	0.607143	0.508929	0.607143	0.553476	0.607143	0.547619	0.607143	0.573436
2 7	0.500000	0.392857	0.500000	0.438424	0.500000	0.434524	0.500000	0.464054
2 8	0.642857	0.511905	0.642857	0.569444	0.892857	0.839286	0.892857	0.863699
2 9	0.535714	0.410714	0.535714	0.463946	1.000000	1.000000	1.000000	1.000000
3 4	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 5	0.928571	0.892857	0.928571	0.909938	0.928571	0.892857	0.928571	0.909938
3 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 7	0.785714	0.750000	0.785714	0.767081	0.750000	0.732143	0.750000	0.740683
3 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
4 5	0.642857	0.523810	0.642857	0.577201	0.750000	0.660714	0.750000	0.702045
4 6	0.642857	0.494048	0.642857	0.557221	0.642857	0.494048	0.642857	0.557221
4 7	0.714286	0.625000	0.714286	0.666205	0.785714	0.678571	0.785714	0.727398
4 8	0.892857	0.857143	0.892857	0.874142	0.928571	0.892857	0.928571	0.909938
4 9	0.821429	0.732143	0.821429	0.773637	1.000000	1.000000	1.000000	1.000000
5 6	0.678571	0.580357	0.678571	0.625111	0.642857	0.541667	0.642857	0.586789
5 7	0.678571	0.583333	0.678571	0.626946	0.821429	0.750000	0.821429	0.784080
5 8	0.857143	0.797619	0.857143	0.825754	0.892857	0.851190	0.892857	0.870785
5 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
6 7	0.714286	0.625000	0.714286	0.666205	0.714286	0.642857	0.714286	0.675306

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
6 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
6 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
7 8	0.821429	0.732143	0.821429	0.768620	0.964286	0.946429	0.964286	0.954969
7 9	0.892857	0.857143	0.892857	0.874142	1.000000	1.000000	1.000000	1.000000
8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3	0.714286	0.625000	0.714286	0.666622	0.857143	0.821429	0.857143	0.838509
1 2 4	0.892857	0.875000	0.892857	0.883540	0.928571	0.892857	0.928571	0.909938
1 2 5	0.714286	0.595238	0.714286	0.649093	0.821429	0.750000	0.821429	0.784080
1 2 6	0.857143	0.821429	0.857143	0.838509	0.892857	0.839286	0.892857	0.864907
1 2 7	0.678571	0.613095	0.678571	0.643663	0.714286	0.636905	0.714286	0.673141
1 2 8	0.821429	0.750000	0.821429	0.784080	0.964286	0.946429	0.964286	0.954969
1 2 9	0.821429	0.803571	0.821429	0.812112	1.000000	1.000000	1.000000	1.000000
1 3 4	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5	0.857143	0.797619	0.857143	0.825754	0.928571	0.892857	0.928571	0.909938
1 3 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 7	0.821429	0.750000	0.821429	0.784080	0.821429	0.767857	0.821429	0.793478
1 3 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 5	0.964286	0.946429	0.964286	0.954969	0.928571	0.910714	0.928571	0.919173
1 4 6	0.964286	0.946429	0.964286	0.954969	0.928571	0.892857	0.928571	0.909938
1 4 7	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 4 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 4 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 5 6	0.928571	0.892857	0.928571	0.909938	0.928571	0.892857	0.928571	0.909938
1 5 7	0.892857	0.851190	0.892857	0.870785	0.892857	0.851190	0.892857	0.870785
1 5 8	0.892857	0.839286	0.892857	0.864907	0.964286	0.946429	0.964286	0.954969
1 5 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 6 7	0.928571	0.892857	0.928571	0.909938	0.928571	0.892857	0.928571	0.909938
1 6 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4	0.714286	0.607143	0.714286	0.655763	1.000000	1.000000	1.000000	1.000000
2 3 5	0.642857	0.517857	0.642857	0.572549	0.857143	0.821429	0.857143	0.838509
2 3 6	0.785714	0.714286	0.785714	0.747032	1.000000	1.000000	1.000000	1.000000
2 3 7	0.714286	0.625000	0.714286	0.666622	0.714286	0.714286	0.714286	0.714286
2 3 8	0.750000	0.642857	0.750000	0.691117	1.000000	1.000000	1.000000	1.000000
2 3 9	0.821429	0.767857	0.821429	0.792063	1.000000	1.000000	1.000000	1.000000
2 4 5	0.607143	0.508929	0.607143	0.553476	0.642857	0.529762	0.642857	0.580664
2 4 6	0.607143	0.502976	0.607143	0.550012	0.607143	0.511905	0.607143	0.552022
2 4 7	0.750000	0.714286	0.750000	0.731159	0.714286	0.625000	0.714286	0.666205
2 4 8	0.750000	0.666667	0.750000	0.704417	0.928571	0.892857	0.928571	0.909938
2 4 9	0.714286	0.625000	0.714286	0.664512	1.000000	1.000000	1.000000	1.000000
2 5 6	0.607143	0.508929	0.607143	0.553476	0.571429	0.467262	0.571429	0.513212
2 5 7	0.535714	0.446429	0.535714	0.487013	0.714286	0.660714	0.714286	0.685128
2 5 8	0.607143	0.476190	0.607143	0.533498	0.857143	0.797619	0.857143	0.824546
2 5 9	0.714286	0.607143	0.714286	0.655763	1.000000	1.000000	1.000000	1.000000
2 6 7	0.607143	0.547619	0.607143	0.575331	0.607143	0.547619	0.607143	0.574546
2 6 8	0.714286	0.619048	0.714286	0.662849	0.892857	0.851190	0.892857	0.870785
2 6 9	0.750000	0.660714	0.750000	0.702001	1.000000	1.000000	1.000000	1.000000
2 7 8	0.642857	0.553571	0.642857	0.593055	0.928571	0.904762	0.928571	0.915816
2 7 9	0.821429	0.785714	0.821429	0.802713	1.000000	1.000000	1.000000	1.000000
2 8 9	0.892857	0.839286	0.892857	0.863699	1.000000	1.000000	1.000000	1.000000
3 4 5	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 5 6	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 7	0.892857	0.839286	0.892857	0.864907	0.892857	0.875000	0.892857	0.883540
3 5 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
3 5 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 6 7	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 6 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 7 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 7 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
4 5 6	0.678571	0.583333	0.678571	0.626903	0.750000	0.654762	0.750000	0.698688
4 5 7	0.750000	0.696429	0.750000	0.721843	0.821429	0.732143	0.821429	0.773637
4 5 8	0.892857	0.857143	0.892857	0.874142	0.892857	0.839286	0.892857	0.864907
4 5 9	0.785714	0.714286	0.785714	0.748240	1.000000	1.000000	1.000000	1.000000
4 6 7	0.821429	0.732143	0.821429	0.773637	0.750000	0.660714	0.750000	0.701001
4 6 8	0.892857	0.857143	0.892857	0.874142	0.964286	0.946429	0.964286	0.954969
4 6 9	0.821429	0.732143	0.821429	0.773637	1.000000	1.000000	1.000000	1.000000
4 7 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
4 7 9	0.785714	0.696429	0.785714	0.737841	1.000000	1.000000	1.000000	1.000000
4 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
5 6 7	0.785714	0.714286	0.785714	0.747239	0.750000	0.672619	0.750000	0.708087
5 6 8	0.892857	0.839286	0.892857	0.864907	0.928571	0.892857	0.928571	0.909938
5 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
5 7 8	0.857143	0.785714	0.857143	0.818668	0.964286	0.946429	0.964286	0.954969
5 7 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
6 7 8	0.857143	0.785714	0.857143	0.817460	0.964286	0.946429	0.964286	0.954969
6 7 9	0.928571	0.892857	0.928571	0.908730	1.000000	1.000000	1.000000	1.000000
6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 5	0.714286	0.625000	0.714286	0.666622	0.857143	0.785714	0.857143	0.819876
1 2 3 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 7	0.714286	0.625000	0.714286	0.666622	0.821429	0.767857	0.821429	0.793478

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 3 8	0.857143	0.785714	0.857143	0.819876	1.000000	1.000000	1.000000	1.000000
1 2 3 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 4 5	0.857143	0.833333	0.857143	0.844388	0.928571	0.892857	0.928571	0.909938
1 2 4 6	0.892857	0.875000	0.892857	0.883540	0.892857	0.839286	0.892857	0.864907
1 2 4 7	0.857143	0.821429	0.857143	0.838509	0.928571	0.892857	0.928571	0.909938
1 2 4 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 2 4 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 5 6	0.857143	0.833333	0.857143	0.844388	0.892857	0.839286	0.892857	0.864907
1 2 5 7	0.714286	0.625000	0.714286	0.666622	0.857143	0.803571	0.857143	0.829111
1 2 5 8	0.821429	0.750000	0.821429	0.784080	0.928571	0.892857	0.928571	0.909938
1 2 5 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 6 7	0.857143	0.821429	0.857143	0.838509	0.892857	0.839286	0.892857	0.864907
1 2 6 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 2 6 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 2 7 8	0.785714	0.714286	0.785714	0.748284	0.964286	0.946429	0.964286	0.954969
1 2 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 7	0.857143	0.797619	0.857143	0.825754	0.928571	0.892857	0.928571	0.909938
1 3 5 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 3 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 4 5 6	0.964286	0.946429	0.964286	0.954969	0.892857	0.857143	0.892857	0.874142
1 4 5 7	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 4 5 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 4 5 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 6 7	0.928571	0.892857	0.928571	0.909938	0.928571	0.892857	0.928571	0.909938
1 4 6 8	1.000000	1.000000	1.000000	1.000000	0.964286	0.946429	0.964286	0.954969
1 4 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 4 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 5 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 5 6 8	0.964286	0.946429	0.964286	0.954969	0.928571	0.892857	0.928571	0.909938
1 5 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 5 7 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 5 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 6 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 5	0.785714	0.714286	0.785714	0.747032	1.000000	1.000000	1.000000	1.000000
2 3 4 6	0.785714	0.714286	0.785714	0.747032	1.000000	1.000000	1.000000	1.000000
2 3 4 7	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 3 4 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 4 9	0.821429	0.767857	0.821429	0.793271	1.000000	1.000000	1.000000	1.000000
2 3 5 6	0.785714	0.714286	0.785714	0.747032	1.000000	1.000000	1.000000	1.000000
2 3 5 7	0.714286	0.625000	0.714286	0.666622	0.892857	0.875000	0.892857	0.883540
2 3 5 8	0.750000	0.642857	0.750000	0.691117	1.000000	1.000000	1.000000	1.000000
2 3 5 9	0.785714	0.714286	0.785714	0.747032	1.000000	1.000000	1.000000	1.000000
2 3 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
2 3 6 8	0.892857	0.839286	0.892857	0.863699	1.000000	1.000000	1.000000	1.000000
2 3 6 9	0.857143	0.785714	0.857143	0.817460	1.000000	1.000000	1.000000	1.000000
2 3 7 8	0.785714	0.714286	0.785714	0.748284	1.000000	1.000000	1.000000	1.000000
2 3 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 4 5 6	0.642857	0.556548	0.642857	0.596251	0.678571	0.583333	0.678571	0.626946
2 4 5 7	0.750000	0.714286	0.750000	0.731203	0.714286	0.642857	0.714286	0.675729
2 4 5 8	0.750000	0.666667	0.750000	0.704417	0.821429	0.744048	0.821429	0.780723
2 4 5 9	0.714286	0.642857	0.714286	0.675397	1.000000	1.000000	1.000000	1.000000
2 4 6 7	0.785714	0.750000	0.785714	0.766999	0.678571	0.571429	0.678571	0.619967
2 4 6 8	0.821429	0.761905	0.821429	0.789808	0.892857	0.839286	0.892857	0.864907
2 4 6 9	0.750000	0.660714	0.750000	0.700794	1.000000	1.000000	1.000000	1.000000
2 4 7 8	0.857143	0.797619	0.857143	0.824546	0.964286	0.946429	0.964286	0.954969
2 4 7 9	0.821429	0.785714	0.821429	0.802713	1.000000	1.000000	1.000000	1.000000
2 4 8 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
2 5 6 7	0.642857	0.601190	0.642857	0.620785	0.678571	0.619048	0.678571	0.645975
2 5 6 8	0.750000	0.654762	0.750000	0.698688	0.857143	0.797619	0.857143	0.825754
2 5 6 9	0.714286	0.607143	0.714286	0.655763	1.000000	1.000000	1.000000	1.000000
2 5 7 8	0.678571	0.607143	0.678571	0.640674	0.892857	0.857143	0.892857	0.874142
2 5 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 5 8 9	0.892857	0.839286	0.892857	0.863699	1.000000	1.000000	1.000000	1.000000
2 6 7 8	0.785714	0.761905	0.785714	0.772959	0.964286	0.946429	0.964286	0.954969
2 6 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 6 8 9	0.892857	0.839286	0.892857	0.863699	1.000000	1.000000	1.000000	1.000000
2 7 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 5 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 5 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
3 4 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 6 7	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 5 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 6 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 7 8	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
3 5 7 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 5 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 6 7 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
4 5 6 7	0.785714	0.714286	0.785714	0.747239	0.821429	0.732143	0.821429	0.773637
4 5 6 8	0.892857	0.857143	0.892857	0.874142	0.892857	0.839286	0.892857	0.864907
4 5 6 9	0.785714	0.714286	0.785714	0.748240	1.000000	1.000000	1.000000	1.000000
4 5 7 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
4 5 7 9	0.821429	0.767857	0.821429	0.793478	1.000000	1.000000	1.000000	1.000000
4 5 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
4 6 7 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
4 6 7 9	0.857143	0.785714	0.857143	0.818668	1.000000	1.000000	1.000000	1.000000
4 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
4 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
5 6 7 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
5 6 7 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
5 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 3 4 7	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 3 4 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6	0.928571	0.928571	0.928571	0.928571	1.000000	1.000000	1.000000	1.000000
1 2 3 5 7	0.714286	0.625000	0.714286	0.666622	0.892857	0.839286	0.892857	0.864907
1 2 3 5 8	0.857143	0.785714	0.857143	0.819876	1.000000	1.000000	1.000000	1.000000
1 2 3 5 9	0.928571	0.928571	0.928571	0.928571	1.000000	1.000000	1.000000	1.000000
1 2 3 6 7	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
1 2 3 6 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 6 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 7 8	0.785714	0.714286	0.785714	0.748284	1.000000	1.000000	1.000000	1.000000
1 2 3 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 5 6	0.892857	0.875000	0.892857	0.883540	0.892857	0.839286	0.892857	0.864907
1 2 4 5 7	0.857143	0.821429	0.857143	0.838509	0.928571	0.892857	0.928571	0.909938
1 2 4 5 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 2 4 5 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 4 6 7	0.857143	0.821429	0.857143	0.838509	0.892857	0.839286	0.892857	0.864907
1 2 4 6 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 2 4 6 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 4 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 7 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
1 2 4 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 5 6 7	0.857143	0.821429	0.857143	0.838509	0.928571	0.892857	0.928571	0.909938
1 2 5 6 8	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 2 5 6 9	0.928571	0.928571	0.928571	0.928571	1.000000	1.000000	1.000000	1.000000
1 2 5 7 8	0.785714	0.714286	0.785714	0.748284	0.964286	0.946429	0.964286	0.954969
1 2 5 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 6 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 2 6 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 5 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 5 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 5 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 5 6 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 6 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 5 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 7 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 5 6 7	0.928571	0.892857	0.928571	0.909938	0.964286	0.946429	0.964286	0.954969
1 4 5 6 8	1.000000	1.000000	1.000000	1.000000	0.928571	0.892857	0.928571	0.909938
1 4 5 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 5 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 5 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 4 5 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 4 6 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 4 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 4 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 4 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 5 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 5 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6	0.785714	0.714286	0.785714	0.747032	1.000000	1.000000	1.000000	1.000000
2 3 4 5 7	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 4 5 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 4 5 9	0.821429	0.767857	0.821429	0.793271	1.000000	1.000000	1.000000	1.000000
2 3 4 6 7	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
2 3 4 6 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 4 6 9	0.857143	0.821429	0.857143	0.838302	1.000000	1.000000	1.000000	1.000000
2 3 4 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 3 4 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 5 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 5 6 8	0.892857	0.839286	0.892857	0.863699	1.000000	1.000000	1.000000	1.000000
2 3 5 6 9	0.821429	0.767857	0.821429	0.792271	1.000000	1.000000	1.000000	1.000000
2 3 5 7 8	0.785714	0.714286	0.785714	0.748284	1.000000	1.000000	1.000000	1.000000
2 3 5 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 3 5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 6 7 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 6 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 3 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 4 5 6 7	0.750000	0.732143	0.750000	0.740602	0.714286	0.642857	0.714286	0.675729
2 4 5 6 8	0.750000	0.666667	0.750000	0.704417	0.821429	0.744048	0.821429	0.780723
2 4 5 6 9	0.750000	0.660714	0.750000	0.700794	1.000000	1.000000	1.000000	1.000000
2 4 5 7 8	0.857143	0.797619	0.857143	0.824546	0.892857	0.839286	0.892857	0.863699
2 4 5 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
2 4 5 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 4 6 7 8	0.892857	0.851190	0.892857	0.870785	0.964286	0.946429	0.964286	0.954969
2 4 6 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 4 6 8 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
2 4 7 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 5 6 7 8	0.750000	0.708333	0.750000	0.727928	0.928571	0.892857	0.928571	0.909938
2 5 6 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 5 6 8 9	0.892857	0.839286	0.892857	0.863699	1.000000	1.000000	1.000000	1.000000
2 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 5 6 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 5 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 5 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 6 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 5 6 7 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
4 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
4 5 6 7 9	0.821429	0.767857	0.821429	0.793478	1.000000	1.000000	1.000000	1.000000
4 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
4 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
4 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 3 4 5 6	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 7	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6 7	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 2 3 4 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 3 4 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 7	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 9	0.928571	0.928571	0.928571	0.928571	1.000000	1.000000	1.000000	1.000000
1 2 3 5 7 8	0.785714	0.714286	0.785714	0.748284	1.000000	1.000000	1.000000	1.000000
1 2 3 5 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 3 5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 2 3 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 5 6 7	0.857143	0.821429	0.857143	0.838509	0.928571	0.892857	0.928571	0.909938
1 2 4 5 6 8	0.928571	0.892857	0.928571	0.909938	0.928571	0.892857	0.928571	0.909938
1 2 4 5 6 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 4 5 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 2 4 5 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 4 5 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 6 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 2 4 6 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 4 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 4 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 5 6 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 5 6 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
1 2 5 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 7	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 5 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 5 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 6 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 6 7 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 5 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 4 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 5 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 4 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 4 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 4 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 7	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 9	0.821429	0.767857	0.821429	0.793271	1.000000	1.000000	1.000000	1.000000
2 3 4 5 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 5 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 4 5 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
2 3 4 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 6 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 4 6 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 4 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 5 6 7 8	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 5 6 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 3 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 3 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 4 5 6 7 8	0.892857	0.851190	0.892857	0.870785	0.964286	0.946429	0.964286	0.954969
2 4 5 6 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
2 4 5 6 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 4 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 4 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 6 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 5 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
3 4 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 5 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 4 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
3 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
4 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 7	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 7 9	0.857143	0.821429	0.857143	0.838509	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6 7 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
1 2 3 4 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000

Feature set	SVM				LDA			
	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 3 4 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 5 6 7 8	0.964286	0.946429	0.964286	0.954969	0.964286	0.946429	0.964286	0.954969
1 2 4 5 6 7 9	0.857143	0.821429	0.857143	0.838509	0.964286	0.946429	0.964286	0.954969
1 2 4 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 4 5 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 7 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 5 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 4 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 3 5 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 4 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 7 8	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 7 9	0.892857	0.875000	0.892857	0.883540	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 8 9	0.928571	0.892857	0.928571	0.909938	1.000000	1.000000	1.000000	1.000000
2 3 4 5 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 3 4 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 3 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 4 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
3 4 5 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 7 8	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 7 9	0.892857	0.839286	0.892857	0.864907	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
1 2 3 4 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 3 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 2 4 5 6 7 8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
1 3 4 5 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
2 3 4 5 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 2 3 4 5 6 7 8 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000

Table S3. 13 SVM and LDA data for 1- and 2-feature combinations including CD43 (feature 9). Combinations showing 100% accuracy are highlighted.

	SVM				LDA			
Feature set	Acc.	Prec.	Rec.	F1	Acc.	Prec.	Rec.	F1
9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
1 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
2 9	0.535714	0.410714	0.535714	0.463946	1.000000	1.000000	1.000000	1.000000
3 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
4 9	0.821429	0.732143	0.821429	0.773637	1.000000	1.000000	1.000000	1.000000
5 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000
6 9	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000	1.000000
7 9	0.892857	0.857143	0.892857	0.874142	1.000000	1.000000	1.000000	1.000000
8 9	0.964286	0.946429	0.964286	0.954969	1.000000	1.000000	1.000000	1.000000

7.2.3.3 SPSS-based LDA

Table S3. 14 LDA classification results using CD43 only

Classification Results ^{a,c}											
Items		pH	Predicted Group Membership								Total
			2.8	3.9	4.9	6.6	6.9	8	9.1	10	
Original	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
a. 100.0% of original grouped cases correctly classified.											
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.											
c. 100.0% of cross-validated grouped cases correctly classified.											

Table S3. 15 Summary of canonical discriminant function by using CD43 only

Eigenvalues				
Function	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation
1	2866.052	100	100	1
Wilks' Lambda				
Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1	0	210.968	7	<.001

Table S3. 16 Functions at group centroids using CD43 only

pH level	Function
2.76	-2.288
3.92	44.289
4.87	69.607
6.64	21.624
6.9	28.512
8	-38.346
9.05	-58.543
10.03	-64.855

Table S3. 17 LDA classification of array built by CD43 and CD69.

Classification Results ^{a,c}											
Items		pH	Predicted Group Membership							10	Total
			2.8	3.9	4.9	6.6	6.9	8	9.1		
Original	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
a. 100.0% of original grouped cases correctly classified.											
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.											
c. 100.0% of cross-validated grouped cases correctly classified.											

Table S3. 18 Summary of canonical discriminant function using array built by CD43 and CD69.

Eigenvalues				
Function	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation
1	2867.290 ^a	84.3	84.3	1
2	534.396 ^a	15.7	100	0.999
a. First 2 canonical discriminant functions were used in the analysis.				
Wilks' Lambda				
Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1 through 2	0	370.356	14	<.001
2	0.002	163.358	6	<.001

Table S3. 19 Functions at group centroids using array built by CD43 and CD69.

pH level	Function	
	1	2
2.76	-3.252	-41.79
3.92	43.839	-20.034
4.87	69.764	6.006
6.64	22.226	25.888
6.9	28.856	14.576
8	-38.091	11.478
9.05	-58.501	2.504
10.03	-64.841	1.372

Table S3. 20 LDA classification results by using array built by CD43 and CD25.

Classification Results ^{a,c}											
Items		pH	Predicted Group Membership								Total
			2.8	3.9	4.9	6.6	6.9	8	9.1	10	
Original	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	2.76	4	0	0	0	0	0	0	0	4
		3.92	0	4	0	0	0	0	0	0	4
		4.87	0	0	4	0	0	0	0	0	4
		6.64	0	0	0	4	0	0	0	0	4
		6.9	0	0	0	0	4	0	0	0	4
		8	0	0	0	0	0	4	0	0	4
		9.05	0	0	0	0	0	0	4	0	4
		10	0	0	0	0	0	0	0	4	4
	%	2.76	100	0	0	0	0	0	0	0	100
		3.92	0	100	0	0	0	0	0	0	100
		4.87	0	0	100	0	0	0	0	0	100
		6.64	0	0	0	100	0	0	0	0	100
		6.9	0	0	0	0	100	0	0	0	100
		8	0	0	0	0	0	100	0	0	100
		9.05	0	0	0	0	0	0	100	0	100
		10	0	0	0	0	0	0	0	100	100
a. 100.0% of original grouped cases correctly classified.											
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.											
c. 100.0% of cross-validated grouped cases correctly classified.											

Table S3. 21 LDA classification results by using array built by CD43 and CD25.

Eigenvalues				
Function	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation
1	3357.485 ^a	98.2	98.2	1
2	61.584 ^a	1.8	100	0.992
a. First 2 canonical discriminant functions were used in the analysis.				
Wilks' Lambda				
Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1 through 2	0	318.65	14	<.001
2	0.016	107.549	6	<.001

Table S3. 22 Functions at group centroids by using array built by CD43 and CD25.

pH level	Function	
	1	2
2.76	-8.219	13.707
3.92	45.033	7.119
4.87	75.528	-0.162
6.64	26.668	-7.706
6.9	34.036	-7.47
8	-40.047	-3.638
9.05	-62.812	-1.562
10.03	-70.187	-0.288

7.2.4 MATLAB based GPR

7.2.4.1 MATLAB GPR code development

For developing a pH sensing array for GPR prediction application, we developed the GPR based MATLAB. GPR is a non-parametric, Bayesian machine learning method that can be used for regression problems. It is based on the Gaussian Process, a collection of random variables where any finite subset of these variables follows a multivariate Gaussian distribution. GPR is particularly useful in cases where the relationship between the input features and the target variable is complex and non-linear. In GPR, the objective is to learn a function that best maps the input features to the target variable. This function is modelled as a Gaussian Process with a mean function and a covariance function (also called the kernel function). The kernel function measures the similarity between the input data points and encodes the correlation between the target values. The choice of the kernel function can have a significant impact on the performance of the model. Here introduces the MATLAB code development based on the code flow. A step-by-step explanation is provided in supporting material. The script performed data preprocessing, model training, prediction, and performance evaluation using various feature combinations.

Data Loading and Preprocessing: The code began by clearing the workspace, closing all figures, and turning off warnings. It then loaded the training and test datasets from Excel files:

```
clc
clear all
close all
warning off

%% Load data
train_data = xlsread('DataHaobo.xlsx');
test_data_full = xlsread('Test.xlsx');
```

The features ('X_train' and 'X_test') and target variables ('y_train' and 'y_test_actual') were extracted from the datasets. The target variable for training was also converted to a logarithmic scale to improve the performance of the GPR model:

```
% Features and target variables
X_train = train_data(:, 2:end); % Features for training
y_train = train_data(:, 1); % Target for training

X_test = test_data_full(:, 2:end); % Features for testing
y_test_actual = test_data_full(:, 1); % Actual pH for testing

% Convert target to logarithmic scale for GPR
y_train_log = log(y_train);
```

Feature Combinations and Directory Setup: The script generated all possible combinations of features to be used for model training and initialised directories for storing the results:

```
% Initialize combinations and results storage
num_features = size(X_train, 2);
combinations = cell(num_features, 1);

    combinations{k} = nchoosek(1:num_features, k);
end

% Create directories for results if they don't exist
if ~exist('GPR_result', 'dir')
    mkdir('GPR_result')
end
if ~exist('GPR_result/Figures', 'dir')
    mkdir('GPR_result/Figures')
end
if ~exist('GPR_result/EditableFigures', 'dir')
    mkdir('GPR_result/EditableFigures')
end
```

Model Training and Prediction: For each combination of features, the script trained a GPR model and made predictions on the test data. The Kernel Function with SquaredExponential was applied during the GPR trainer modelling. There were 4 test set for each of the array selection. It then computed various evaluation metrics and saved the results:

```
% Loop through all feature combinations
for k = 1:num_features
    for i = 1:size(combinations{k}, 1)
        feature_combination = combinations{k}(i, :);
        X_train_comb = X_train(:, feature_combination);
        X_test_comb = X_test(:, feature_combination);

        % Train the GPR model
        gprMdl = fitrgp(X_train_comb, y_train_log);

        % Predict target variable for test data
        y_pred_log = predict(gprMdl, X_test_comb);
        y_pred = exp(y_pred_log);

        % Compute predictions for all training data points
        [ypred_log, ~, yci_log] = predict(gprMdl, X_train_comb);
        ypred = exp(ypred_log);
        yci = exp(yci_log);

        % Calculate average predicted pH value and standard deviation for n=4
        mean_y_pred = mean(y_pred);
        std_y_pred = std(y_pred);
        prediction_range = [mean_y_pred - std_y_pred, mean_y_pred + std_y_pred];

        % Convert kernel information to string
```

```

kernelInfoStr = struct2str(gprMdl.KernelInformation);

% Save the results in a text file
result_filename = ['GPR_result/FeatureCombination_', mat2str(feature_combination), '.txt'];
fileID = fopen(result_filename, 'w');

fprintf(fileID, 'Feature Combination: %s\n', mat2str(feature_combination));
fprintf(fileID, 'Kernel Information: %s\n', kernelInfoStr);
fprintf(fileID, 'Kernel Function: %s\n', gprMdl.KernelFunction);
fprintf(fileID, 'Standard deviation of predicted values (n=4): %.10f\n', std_y_pred);
fprintf(fileID, 'Test pH of %.2f gave an average prediction of %.10f ± %.10f (standard deviation, n=%d)\n',
y_test_actual(1), mean_y_pred, std_y_pred, size(X_test_comb, 1));
fprintf(fileID, 'Prediction range: %.10f to %.10f\n', prediction_range(1), prediction_range(2));

% Calculate and display the coefficient of determination (R2), MSE, and MAE
R2 = 1 - sum((y_train - ypred).^2) / sum((y_train - mean(y_train)).^2);
MSE = mean((y_train - ypred).^2);
MAE = mean(abs(y_train - ypred));

fprintf(fileID, 'Coefficient of Determination (R2): %.10f\n', R2);
fprintf(fileID, 'Mean Squared Error (MSE): %.10f\n', MSE);
fprintf(fileID, 'Mean Absolute Error (MAE): %.10f\n', MAE);

fclose(fileID);

% Append results to the result table
result_table = [result_table; {mat2str(feature_combination), kernelInfoStr, gprMdl.KernelFunction,
std_y_pred, mean_y_pred, std_y_pred, size(X_test_comb, 1), prediction_range(1), prediction_range(2), R2,
MSE, MAE}];

```

Visualisation and Results Saving: The code visualised the predicted versus actual pH values for each feature combination and saved the figures in both TIFF and MATLAB editable formats. It also saved the results to an Excel file for further analysis:

```

% Plot predicted vs actual values
figure;
hold on;
% Add error bars to the training data
error_bar = (yci(:, 2) - yci(:, 1)) / 2;
errorbar(y_train, ypred, error_bar, 'bo');
plot(y_train, y_train, 'k-');
xlabel('Actual pH');
ylabel('Predicted pH');
title(['GPR - Feature Combination: ', mat2str(feature_combination)]);
legend('Predicted vs actual', 'Ideal line', 'Location', 'Northwest');
% Plot predicted test points
for j = 1:size(X_test_comb, 1)
    plot(y_test_actual(j), y_pred(j), 'r*', 'MarkerSize', 10);
end

% Save figure in TIFF format

```

```

    print('-dtiff', '-r1200', ['GPR_result/Figures/GPR_FeatureCombination_', mat2str(feature_combination),
    '.tiff']);

    % Save figure in MATLAB editable format
    saveas(gcf, ['GPR_result/EditableFigures/GPR_FeatureCombination_', mat2str(feature_combination),
    '.fig']);

    % Close the plot window
    close(gcf);
end
end

% Save results to Excel file
result_headers = {'Feature Combination', 'Kernel Information', 'Kernel Function', 'Std Deviation', 'Mean
Prediction', 'Std Dev', 'Num Test Points', 'Prediction Range Min', 'Prediction Range Max', 'R^2', 'MSE', 'MAE'};
result_table = cell2table(result_table, 'VariableNames', result_headers);
writetable(result_table, 'GPR_result/GPR_results.xlsx');

```

Utility Functions: The script included utility functions for converting structures and arrays to strings, which were used in the process of saving the results:

```

% Function to convert structure to string
function str = struct2str(structVar)
    str = "";
    fields = fieldnames(structVar);
    for i = 1:numel(fields)
        fieldName = fields{i};
        fieldValue = structVar.(fieldName);
        if isstruct(fieldValue)
            fieldValueStr = struct2str(fieldValue);
        else
            fieldValueStr = array2str(fieldValue);
        end
        str = [str, fieldName, ': ', fieldValueStr, ' '];
    end
end

```

```

% Function to convert array to string
function str = array2str(array)
    if isnumeric(array) || islogical(array)
        str = mat2str(array);
    elseif ischar(array)
        str = array;
    elseif isstring(array)
        str = char(array);
    else
        str = 'Unsupported data type';
    end
end

```

```

% Function to extract and display kernel parameters names and values
function getKernelParameters(gprMdl)

```

```

kernelInfo = gprMdl.KernelInformation;
paramName = gprMdl.KernelInformation.KernelParameterNames;
paramValues = kernelInfo.KernelParameters;

disp('Kernel parameter names:');
disp(paramName);

disp('Kernel parameter values:');
for i = 1:numel(paramValues)
    fprintf('%0.10f\n', paramValues(i));
end
end

```

In summary, this paper presents a well-structured, modular, and efficient MATLAB code implementation of Gaussian Process Regression (GPR) for predicting pH values. The code adheres to design principles, ensuring an organized and systematic approach to addressing the regression problem. This comprehensive approach ensured robust evaluation of different feature combinations, providing insights into the optimal configuration for accurate pH prediction using GPR. Modularity is a key feature of the code, with distinct sections dedicated to data loading, pre-processing, model training, prediction, statistical analysis, and visualization. This separation not only improves readability and maintainability but also makes it more user-friendly. The code's flexibility allows for easy adaptation to other datasets or regression problems, requiring only minor adjustments to the input data file and feature/target variable assignments. One of the code's main strengths is its user-friendliness, enabling users with limited coding and MATLAB experience to quickly adapt it for specific regression problems with minimal modifications. Overall, the MATLAB code described in this paper offers a practical, organized, and user-friendly solution for implementing GPR, making it a valuable tool for a wide range of applications and users.

7.2.4.2 GPR results

Table S3. 23 The most promising feature combination is highlighted associate with their GPR performance

Feature Combination	Test pH	Kernel Parameters	Std Deviation	Mean Prediction	Num Test Points	Prediction Range Min	Prediction Range Max	R ²	MSE	MAE
9	6.9	84.75922314882;0.362353037454844	0.120	7.259	4	7.139	7.379	1.000	0.000	0.009
3 9	6.9	427.6691970988;0.316333780807546	0.110	6.795	4	6.685	6.904	1.000	0.000	0.009
6 9	6.9	13068.578066183;0.346680829348127	0.069	7.154	4	7.085	7.223	1.000	0.002	0.024
3 9	4.07	86829.8011803673;0.468063851297936	0.106	4.039	4	3.934	4.145	0.987	0.055	0.165
3 9	9.05	75299.2182486293;0.437275578262553	0.035	8.970	4	8.934	9.006	0.996	0.019	0.102

7.2.4.3 GPR based pH sensing application with milk

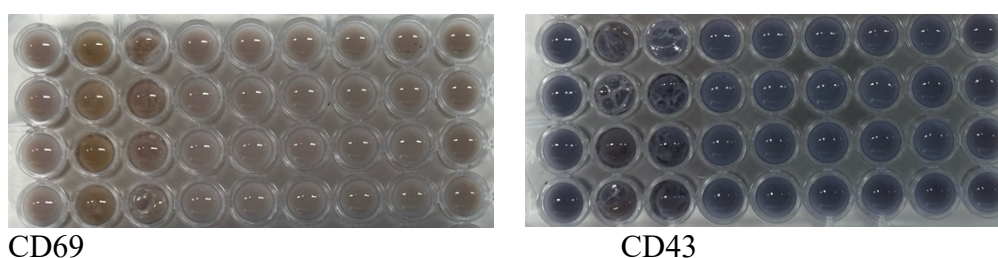


Figure S3. 4 Naked eye visual observation of CDs sensor array applied with milk under varying pH conditions (Each photo contains four replicates, with each row representing one replicate. From left to right, the columns represent CDs in MilliQ water and spoiled milk, followed by CDs with spoiled milk exposed to pH levels 3.06, 4.04, 5.55, 6.15, 7.2, 8.05, 9.25, and 10.09).

7.2.5 Carbon dots stability

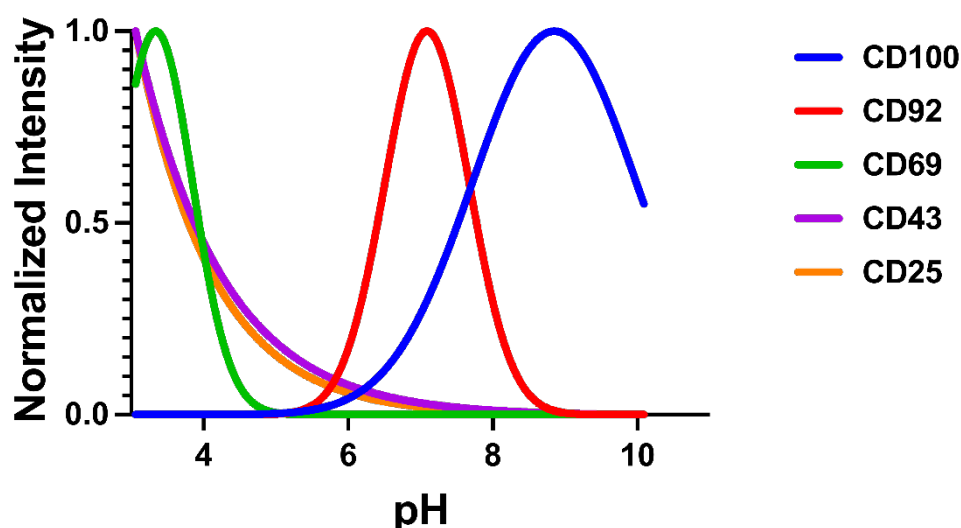


Figure S3. 5 Overview of CDs (three months after solution prepared) with variable pH response patterns (1 mg/mL CDs; $\lambda_{ex} = 370$ nm, λ_{em} integrated across 390–750 nm) in buffered Milli-Q water (25 mM buffers, pH 3–10): pH responses of individual CDs: CD100 (blue), CD92 (red), CD69 (green), CD43 (purple), and CD25 (orange), respectively.

7.3 Appendix III supporting material for chapter five

7.3.1 Carbon dots characterisation

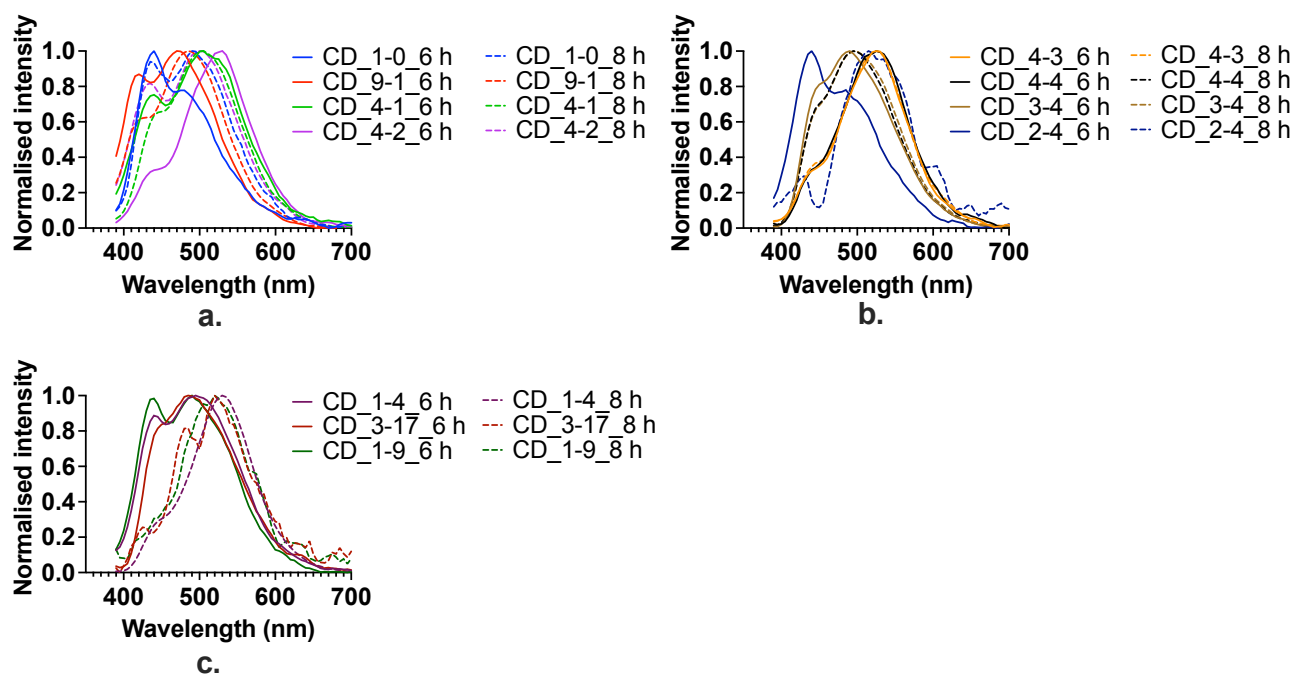
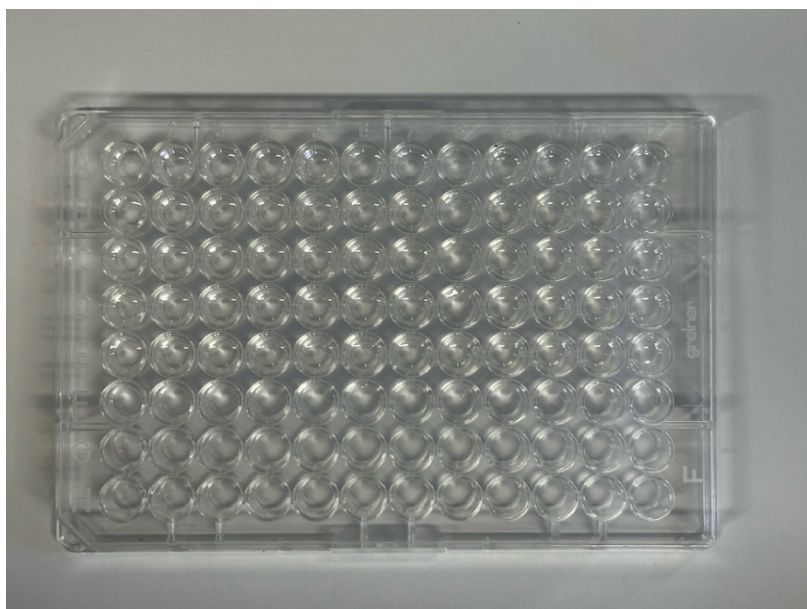


Figure S5. 1 The normalised fluorescence emission spectra of all the 22 CDs (1mg/mL CDs ($\lambda_{ex} = 370 \text{ nm}$, $\lambda_{em} = 390\text{--}700 \text{ nm}$))

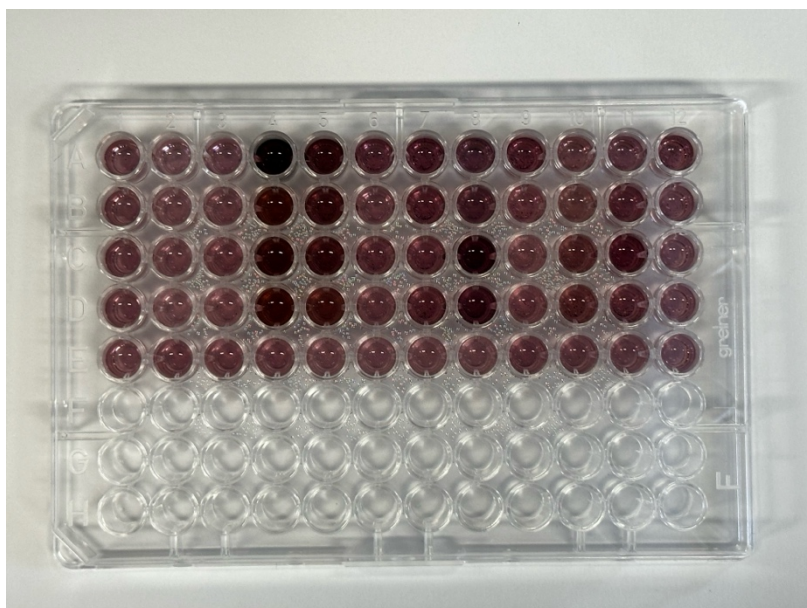
The appearance of the CDs meeting with different metal ions were listed in the following group of photos (Figure S5.2), with 0.25 mg/mL CDs and 20 μM metal ions in a total volume of 200 μL .

	1	2	3	4	5	6	7	8	9	10	11	12
A	CD	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)
B	CD	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)
C	CD	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)
D	CD	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)
E	Mg(II)	Mg(II)	Mg(II)	Mg(II)	Cr(III)	Cr(III)	Cr(III)	Cr(III)	Mn(II)	Mn(II)	Mn(II)	Mn(II)
F												
G												
H												

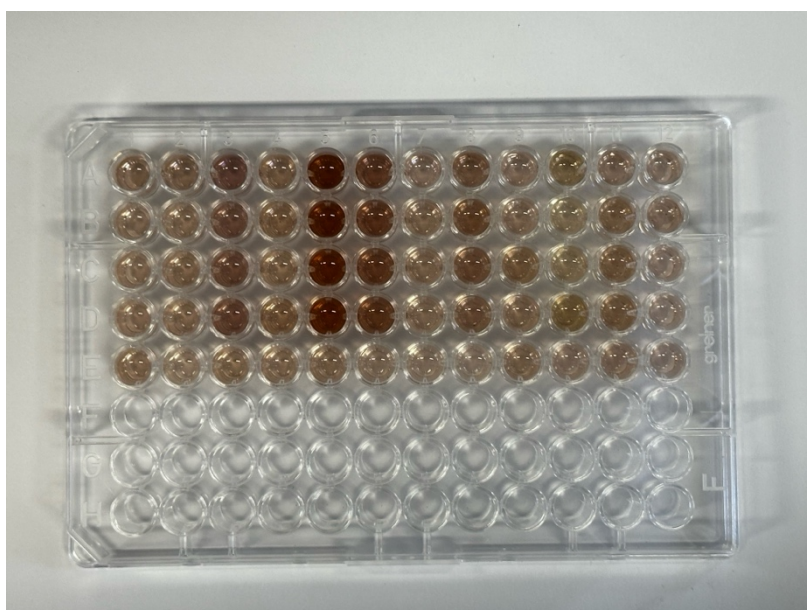
a.



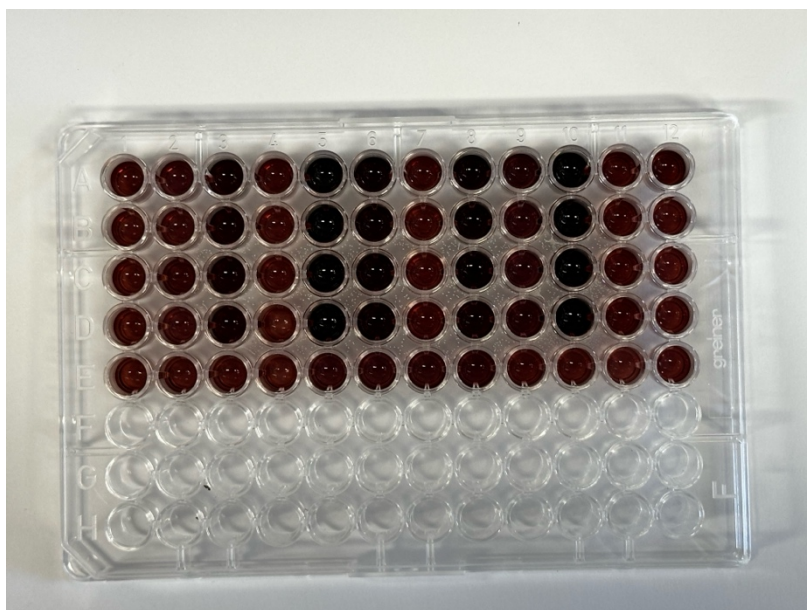
b.



c.



d.



e.

Figure S5.2 The appearances variation of Carbon dots by meeting with different metal ions (a. lay out demonstration b. CD_1-0_6 h, c. CD_2-4_6 h, d. CD_1-4_6 h, e. CD_1-9_6 h at 0.25 mg/mL, with 20 μ M metal ion).

7.3.2 SPSS based LDA classification

7.3.2.1 SPSS based LDA analysis for 22-CDs-based array

Table S5.1 LDA classification results using 22-CDs-based array

Classification Results ^{a,c}																	
		Predicted Group Membership															
		Metal ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4

			Zn(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	4
			Mg(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	4
			Mn(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	4
			Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	4	4
		%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100
			Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100
			Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	100
			Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	100
			Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	100
			Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	100
			Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	100
			Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	100
			Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	100
			Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	100
			Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	100
			Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	100
			Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	100	100
			Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	100	100
		Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	4
			Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	4
			Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	4
			Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	4
			Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	4
			Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	4
			Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	4
			Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	4
			Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	4
			Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	4
			Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	4
			Mg(II)	0	0	1	0	0	0	0	0	0	0	0	3	0	4
			Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	4	4
			Cr(III)	0	0	0	0	0	0	0	0	0	0	0	1	3	4
		%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100
			Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100
			Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	100
			Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	100
			Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	100
			Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	100
			Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	100
			Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	100
			Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	100

		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	25	0	0	0	0	0	0	0	0	75	0	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	100	0	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	25	75	100
a. 100.0% of original grouped cases correctly classified.																	
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																	
c. 96.4% of cross-validated grouped cases correctly classified.																	

7.3.2.2 SPSS based LDA analysis for three-CDs-based array selections listed by MATLAB
based SVM classifier

Table S5. 1 LDA classification results using three-CDs-based array (CD_4-0_6 h, CD_2-4_6 h, and CD_1-4_6 h)

Classification Results ^{a,c}																
		Predicted Group Membership														
		Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	1	2	1	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	1	3	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	1	2	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	25	50	25	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	25	75	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	25	50	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	3	0	0	0	0	0	0	0	0	1	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	3	1	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	3	1	0	0	4

		Zn(II)	0	0	0	0	0	0	0	0	0	1	2	1	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	1	3	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	2	1	1	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	1	3	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	75	0	0	0	0	0	0	0	0	25	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	75	25	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	75	25	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	25	50	25	0	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	25	75	0	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	50	25	25	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	25	75	100
	a. 91.1% of original grouped cases correctly classified.																
	b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																
	c. 82.1% of cross-validated grouped cases correctly classified.																

Table S5.2 LDA classification results using three-CDs-based array (CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h)

Classification Results ^{a,c}																	
		Predicted Group Membership															
		Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	3	1	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	75	25	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	3	0	1	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	1	2	1	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	4	4

	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100	
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100	
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	100	
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	100	
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	100	
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	100	
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	100	
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	100	
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	100	
		Pb(II)	0	0	0	0	0	0	0	0	0	75	0	25	0	100	
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	100	
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	100	
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	25	50	25	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	100	100
a. 98.2% of original grouped cases correctly classified.																	
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																	
c. 94.6% of cross-validated grouped cases correctly classified.																	

Table S5.3 LDA classification results using three-CDs-based array (CD_2-4_6 h, CD_1-9_6 h, and CD_9-1_8 h)

Classification Results ^{a,c}																	
		Predicted Group Membership															
		Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	3	1	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	1	3	0	4
		Cr(III)	0	0	1	0	0	0	0	0	0	0	0	0	0	3	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	75	25	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	25	75	0	100
		Cr(III)	0	0	25	0	0	0	0	0	0	0	0	0	0	75	100
Cross-validated ^b	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	2	0	0	0	0	0	0	0	0	0	0	2	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	1	0	0	0	0	0	0	0	0	2	1	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	1	3	0	4
		Cr(III)	0	0	1	0	0	0	0	0	0	0	0	0	0	3	4

	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100	
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100	
		Ca(II)	0	0	50	0	0	0	0	0	0	0	0	0	0	50	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	25	0	0	0	0	0	0	0	0	50	25	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	25	75	0	100
		Cr(III)	0	0	25	0	0	0	0	0	0	0	0	0	0	75	100
a. 94.6% of original grouped cases correctly classified.																	
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																	
c. 89.3% of cross-validated grouped cases correctly classified.																	

Table S5.4 LDA classification results using three-CDs-based array (CD_2-4_6 h, CD_1-9_6 h, and CD_1-4_6 h)

Classification Results ^{a,c}																	
		Predicted Group Membership															
		Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	4	0	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	100	0	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	Al(III)	3	0	0	0	0	0	0	0	0	0	0	0	0	1	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	3	1	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	4	4

%	Al(III)	75	0	0	0	0	0	0	0	0	0	0	0	0	25	100
	Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
	Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	0	100
	Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
	Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	0	100
	Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
	Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
	Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	0	100
	Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
	Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
	Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
	Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	0	100
	Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	75	25	100
	Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	100	100
a. 100.0% of original grouped cases correctly classified.																
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																
c. 96.4% of cross-validated grouped cases correctly classified.																

7.3.3 MATLAB based classification

7.3.3.1 Background

For the MATLAB projects developed, the focus was on utilising carbon dots-based sensors for metal ion detection. The dataset incorporated consists of 22 distinct carbon dots sensors, enumerated in an accompanying **Table S5.5**. These sensors were deployed to distinguish between 14 metal ions, each tested across four replicates. In the outputs of our MATLAB projects, for enhanced clarity and simplification, the carbon dots are denoted by their corresponding numbers as outlined in the **Table S5.5**. These MATLAB projects have good versatility and applicability, enabling the projects to be seamlessly adapted for a variety of array sensing scenarios.

Table S5.5 Carbon dots numbering

No.	Carbon dot probe	No.	Carbon dot probe
1	CD_1-0_6h	12	CD_1-0_8h
2	CD_9-1_6h	13	CD_9-1_8h
3	CD_4-1_6h	14	CD_4-1_8h
4	CD_4-2_6h	15	CD_4-2_8h
5	CD_4-3_6h	16	CD_4-3_8h
6	CD_4-4_6h	17	CD_4-4_8h
7	CD_3-4_6h	18	CD_3-4_8h
8	CD_2_4_6h	19	CD_2_4_8h
9	CD_1-4_6h	20	CD_1-4_8h
10	CD_3-17_6h	21	CD_3-17_8h
11	CD_1-9_6h	22	CD_1-9_8h

7.3.3.2 MATLAB based SVM classifier

Aims and working mechanism

The MATLAB code is meticulously crafted to conduct an exhaustive analysis and classification employing Support Vector Machines (SVM) on data sourced from an Excel file

titled SVMdata.xlsx. The code's objective is multifaceted, encompassing data pre-processing, feature selection, model training with hyperparameter optimisation, cross-validation, and the visualisation of results. Below is a detailed overview of the code's primary objectives and functionalities:

Data Loading and Preprocessing: Initiating with a clean workspace, the script prepares the environment and imports data from SVMdata.xlsx. It further establishes a directory named 'result' for storing output files and a text file 'result.txt' to log the findings.

Labeling and Cross-validation Setup: The script assigns labels to the dataset, presuming that each of the 14 categories comprises 4 samples, and orchestrates a 4-fold cross-validation to assess the model's efficacy.

Feature Selection and SVM Parameter Optimisation: The script employs a bespoke function, SVMcgForClass, to determine the optimal parameters (C and gamma) for the SVM classifier across each possible combination of features (up to 22 features). This step is vital for improving the model's precision through the adjustment of the cost parameter (C) and the gamma parameter for the radial basis function (RBF) kernel.

Model Training and Evaluation: Utilising the optimised parameters, the script trains an SVM model with the training data and evaluates its performance on the testing data within the cross-validation framework. This is facilitated by the libsvmtrain and libsvmpredict functions from the LIBSVM library, renowned for its SVM implementations.

Visualization: Specifically for a scenario utilising two features, the script generates a 2D plot to illustrate the SVM model's decision boundaries. By creating a grid of points via meshgrid, predicting labels for these points to delineate the decision surface, and superimposing the actual

data points, this visualisation aids in comprehending the SVM model's ability to differentiate between various classes in a bidimensional feature space.

Logging Results: The script meticulously records the accuracy of diverse feature combinations and the overarching accuracy per feature count in the 'result.txt' file, highlighting instances where a feature combination achieves 100% accuracy.

Custom SVM Parameter Search Function (SVMcgForClass): This bespoke function is integral to the script, designed to navigate through a spectrum of C and gamma values to pinpoint the optimum parameters for the SVM classifier. It seeks to maximise cross-validation accuracy while ensuring computational efficiency by eliminating parameter combinations that yield low accuracy.

Script and functions

```
clear;
%%
files = strcat('SVMdata.xlsx');
mkdir result
fileID = fopen('result.txt','w');

[data,txt,raw] = xlsread(files); % Loading data
label = repelem(1:14,4); % Label data from 1-14
indices = crossvalind('Kfold', label, 4); % Develop 4-fold cross validation

clr = hsv(30); % Define colours
legends1 = strsplit(num2str(1:14));
legends2 = strsplit(num2str(1:14));
for i = 1:14
    legends1{i} = txt{i*4,2} + " region";
    legends2{i} = "observed " + txt{i*4,2};
end
%%
for feature_num = 1:22 % The number of CDs involved in array development, chosen from 1 to 22.
    C = nchoosek(1:22, feature_num);
    cp = classperf(label);
    for i = 1:size(C, 1) % Iterate through every feature combination
        feature_select = C(i,:);
        feature_data = data(:, feature_select); % Current feature selections, feature numbers are matching with
        % CDs according to the CDs numbering table.
        [bestacc, bestc, bestg] = SVMcgForClass(label, feature_data, -2,4,-4,4,4,0.5,0.5,0.9); % find the best
        % parameters, c and g
        cp_feature = classperf(label);
        for k = 1:4 % Perform 4-fold cross validation
```

```

test = (indices == k); % Testing set
train = ~test; % Training set
train_data = feature_data(train, :); % Train the training set
train_label = label(train); % training label
test_data = feature_data(test, :); % test features
test_label = label(test); % testing label
model = libsvmtrain(train_label, train_data, ['-s 0 -t 2 -c ', num2str(bestc), '-g ', num2str(bestg), '-q']); % Develop SVM classifier with tunable parameters % model = libsvmtrain(train_label, train_data, '-s 0 -t 0 -q');
[predicted_label, ~, ~] = libsvmpredict(test_label, test_data, model, '-q'); % Test the model on the
test set
classperf(cp, predicted_label, test);
classperf(cp_feature, predicted_label, test);
if feature_num == 2 && k == 1 % Plot the 2D figure
[X1, X2] = meshgrid(min(feature_data(:,1)):1e-3:max(feature_data(:,1)), ...
    min(feature_data(:,2)):1e-3:max(feature_data(:,2))); % Generate grid points (the area
of the sensing targets)
X_grid = [X1(:), X2(:)];
grid_label = ones(size(X_grid, 1), 1);

[pre_label, ~, ~] = libsvmpredict(grid_label, X_grid, model, '-q'); % Make predictions on grid
points to draw classification surfaces
h = figure(1);
h.WindowState = 'maximized';
gscatter(X_grid(:,1), X_grid(:,2), pre_label, clr(1:14,:)); % Plotting grid points
hold on
axis tight
gscatter(feature_data(:,1), feature_data(:,2), label+14, clr(15:28,:)); % Plotting sample points
xlabel(sprintf('feature %s', txt{2, feature_select(1)+2})) % The first feature (CD sensor) is the X-
axis
ylabel(sprintf('feature %s', txt{2, feature_select(2)+2})) % The second feature (CD sensor) is the
Y-axis
legend([legends1, legends2]);
hold off
saveas(gcf, sprintf('./result/%d-%d.png', feature_select))
end
end
if cp_feature.CorrectRate == 1
fprintf(fileID, 'accuracy of features %s is 100.00%%\r\n', num2str(feature_select)); % 100%
predicted feature selection documented
end
end
fprintf(fileID, 'accuracy of %d features is %f\r\n', feature_num, cp.CorrectRate); % The results of each
feature quantity are recorded into a file
end

fclose(fileID);

function [bestacc,bestc,bestg] =
SVMcgForClass(train_label,train,cmin,cmax,gmin,gmax,v,cstep,gstep,accstep)
%%% about the parameters of SVMcg
if nargin < 10
accstep = 4.5;
end
if nargin < 8
cstep = 0.8;
gstep = 0.8;
end
if nargin < 7
v = 5;

```

```

end
if nargin < 5
    gmax = 8;
    gmin = -8;
end
if nargin < 3
    cmax = 8;
    cmin = -8;
end
%% X:c Y:g cg:CVaccuracy
[X,Y] = meshgrid(cmin:cstep:cmax,gmin:gstep:gmax); % Generate grid points of c and g according to the
given parameters
[m,n] = size(X);
cg = zeros(m,n);
eps = 1e-1;
%% record acc with different c & g,and find the bestacc with the smallest c
bestc = 1;
bestg = 0.1;
bestacc = 0;
basenum = 2;
for i = 1:m
    for j = 1:n
        cmd = ['-v ',num2str(v),' -c ',num2str( basenum^X(i,j) ),' -g ',num2str( basenum^Y(i,j) ) ' -q'];
        cg(i,j) = libsvmtrain(train_label, train, cmd); % Train svm according to the given parameters

        if cg(i,j) <= 55 % Those with an accuracy rate lower than 55% will be discarded.
            continue;
        end
        if cg(i,j) > bestacc % If the effect is better than the current one, record the current c and g parameters.
            bestacc = cg(i,j);
            bestc = basenum^X(i,j);
            bestg = basenum^Y(i,j);
        end
        if abs( cg(i,j)-bestacc ) <= eps && bestc > basenum^X(i,j) % If the effect is very close to the best effect
and the current c is smaller, record the current c
            bestacc = cg(i,j);
            bestc = basenum^X(i,j);
            bestg = basenum^Y(i,j);
        end
    end
end
end
end

```

Results

MATLAB based SVM classification and array optimization results

Listed here are CD selections optimized for a list of CDs based metal sensing array via the SVM classification MATLAB program. The results used 1 to 22 to represent 22 CDs. For the carbon dots number reference, checking Table. And accuracy of N features represents the average SVM classification accuracy by using N-CDs based array, where N represent the number of CDs involved in the array. For example, as the first line of the results said,

accuracy of features 1	8	11	16	20	is 100.00%	accuracy of features 2	9	10	13	15	is 100.00%	accuracy of features 1	2	4	8	9	21	is 100.00%
accuracy of features 1	8	11	17	20	is 100.00%	accuracy of features 2	9	11	13	15	is 100.00%	accuracy of features 1	2	4	8	9	22	is 100.00%
accuracy of features 1	8	11	17	21	is 100.00%	accuracy of features 3	8	9	11	21	is 100.00%	accuracy of features 1	2	4	8	11	15	is 100.00%
accuracy of features 1	8	11	17	22	is 100.00%	accuracy of features 3	8	10	20	21	is 100.00%	accuracy of features 1	2	4	8	11	21	is 100.00%
accuracy of features 1	8	11	18	22	is 100.00%	accuracy of features 3	8	18	20	21	is 100.00%	accuracy of features 1	2	4	8	11	22	is 100.00%
accuracy of features 1	8	11	19	20	is 100.00%	accuracy of features 3	9	18	20	21	is 100.00%	accuracy of features 1	2	4	8	18	21	is 100.00%
accuracy of features 1	8	11	19	21	is 100.00%	accuracy of features 3	10	20	21	22	is 100.00%	accuracy of features 1	2	4	9	10	11	is 100.00%
accuracy of features 1	8	11	19	22	is 100.00%	accuracy of features 3	11	18	20	21	is 100.00%	accuracy of features 1	2	4	9	10	22	is 100.00%
accuracy of features 1	8	11	20	21	is 100.00%	accuracy of features 4	5	11	13	20	is 100.00%	accuracy of features 1	2	4	9	11	21	is 100.00%
accuracy of features 1	8	11	20	22	is 100.00%	accuracy of features 4	8	9	11	15	is 100.00%	accuracy of features 1	2	4	9	11	22	is 100.00%
accuracy of features 1	8	11	21	22	is 100.00%	accuracy of features 4	9	10	11	21	is 100.00%	accuracy of features 1	2	4	9	13	20	is 100.00%
accuracy of features 1	8	17	18	21	is 100.00%	accuracy of features 4	9	11	13	20	is 100.00%	accuracy of features 1	2	4	9	13	21	is 100.00%
accuracy of features 1	9	10	11	20	is 100.00%	accuracy of features 4	9	11	14	20	is 100.00%	accuracy of features 1	2	4	9	13	22	is 100.00%
accuracy of features 1	9	10	11	22	is 100.00%	accuracy of features 5	8	9	10	21	is 100.00%	accuracy of features 1	2	4	9	14	21	is 100.00%
accuracy of features 1	9	10	13	22	is 100.00%	accuracy of features 5	8	10	11	21	is 100.00%	accuracy of features 1	2	4	9	15	21	is 100.00%
accuracy of features 1	9	10	20	22	is 100.00%	accuracy of features 5	9	10	11	21	is 100.00%	accuracy of features 1	2	4	9	15	22	is 100.00%
accuracy of features 1	9	10	21	22	is 100.00%	accuracy of features 6	9	11	13	20	is 100.00%	accuracy of features 1	2	4	9	16	21	is 100.00%
accuracy of features 1	9	11	12	13	is 100.00%	accuracy of features 7	8	9	10	21	is 100.00%	accuracy of features 1	2	4	9	16	22	is 100.00%
accuracy of features 1	9	11	13	14	is 100.00%	accuracy of features 7	8	10	11	21	is 100.00%	accuracy of features 1	2	4	9	17	21	is 100.00%
accuracy of features 1	9	11	13	15	is 100.00%	accuracy of features 7	9	10	11	21	is 100.00%	accuracy of features 1	2	4	9	17	22	is 100.00%
accuracy of features 1	9	11	13	21	is 100.00%	accuracy of features 7	9	10	13	21	is 100.00%	accuracy of features 1	2	4	9	18	21	is 100.00%
accuracy of features 1	9	11	13	22	is 100.00%	accuracy of features 7	9	10	14	21	is 100.00%	accuracy of features 1	2	4	9	18	22	is 100.00%
accuracy of features 1	9	11	14	16	is 100.00%	accuracy of features 7	9	10	15	21	is 100.00%	accuracy of features 1	2	4	9	19	21	is 100.00%
accuracy of features 1	9	11	14	20	is 100.00%	accuracy of features 7	9	10	17	21	is 100.00%	accuracy of features 1	2	4	9	19	22	is 100.00%
accuracy of features 1	9	11	15	22	is 100.00%	accuracy of features 7	9	10	20	21	is 100.00%	accuracy of features 1	2	4	9	20	21	is 100.00%
accuracy of features 1	9	11	16	20	is 100.00%	accuracy of features 7	9	10	21	22	is 100.00%	accuracy of features 1	2	4				

[illegible]

accuracy of features	1	3	4	8	9	21	is	100.00%
accuracy of features	1	3	4	8	9	22	is	100.00%
accuracy of features	1	3	4	9	11	21	is	100.00%
accuracy of features	1	3	4	9	11	22	is	100.00%
accuracy of features	1	3	4	9	13	22	is	100.00%
accuracy of features	1	3	4	9	14	22	is	100.00%
accuracy of features	1	3	4	9	16	22	is	100.00%
accuracy of features	1	3	4	9	17	22	is	100.00%
accuracy of features	1	3	4	9	18	22	is	100.00%
accuracy of features	1	3	4	9	19	21	is	100.00%
accuracy of features	1	3	4	9	20	21	is	100.00%
accuracy of features	1	3	4	9	20	22	is	100.00%
accuracy of features	1	3	4	11	13	20	is	100.00%
accuracy of features	1	3	4	11	14	20	is	100.00%
accuracy of features	1	3	4	11	16	20	is	100.00%
accuracy of features	1	3	4	11	17	21	is	100.00%
accuracy of features	1	3	4	18	19	21	is	100.00%
accuracy of features	1	3	4	18	20	21	is	100.00%
accuracy of features	1	3	5	6	11	20	is	100.00%
accuracy of features	1	3	5	8	9	21	is	100.00%
accuracy of features	1	3	5	8	11	22	is	100.00%
accuracy of features	1	3	5	8	18	21	is	100.00%
accuracy of features	1	3	5	8	20	22	is	100.00%
accuracy of features	1	3	5	9	11	21	is	100.00%
accuracy of features	1	3	5	9	20	22	is	100.00%
accuracy of features	1	3	5	11	13	20	is	100.00%
accuracy of features	1	3	5	11	16	20	is	100.00%
accuracy of features	1	3	5	11	20	22	is	100.00%
accuracy of features	1	3	6	7	11	20	is	100.00%
accuracy of features	1	3	6	8	9	22	is	100.00%
accuracy of features	1	3	6	8	11	20	is	100.00%
accuracy of features	1	3	6	8	11	22	is	100.00%
accuracy of features	1	3	6	9	20	22	is	100.00%
accuracy of features	1	3	6	11	13	20	is	100.00%
accuracy of features	1	3	6	11	14	20	is	100.00%
accuracy of features	1	3	6	11	15	20	is	100.00%
accuracy of features	1	3	6	11	16	20	is	100.00%
accuracy of features	1	3	6	11	17	20	is	100.00%
accuracy of features	1	3	6	11	20	22	is	100.00%
accuracy of features	1	3	6	18	20	22	is	100.00%
accuracy of features	1	3	7	8	9	21	is	100.00%
accuracy of features	1	3	7	8	9	22	is	100.00%
accuracy of features	1	3	7	8	11	20	is	100.00%
accuracy of features	1	3	7	8	11	22	is	100.00%
accuracy of features	1	3	7	8	18	21	is	100.00%
accuracy of features	1	3	7	9	11	21	is	100.00%
accuracy of features	1	3	7	9	13	22	is	100.00%
accuracy of features	1	3	7	9	17	22	is	100.00%
accuracy of features	1	3	7	9	20	22	is	100.00%
accuracy of features	1	3	7	9	21	22	is	100.00%
accuracy of features	1	3	7	11	13	20	is	100.00%
accuracy of features	1	3	7	11	14	20	is	100.00%
accuracy of features	1	3	7	11	15	20	is	100.00%
accuracy of features	1	3	7	11	16	20	is	100.00%
accuracy of features	1	3	7	11	20	22	is	100.00%
accuracy of features	1	3	7	18	20	22	is	100.00%
accuracy of features	1	3	8	9	11	22	is	100.00%
accuracy of features	1	3	8	9	13	21	is	100.00%
accuracy of features	1	3	8	9	14	21	is	100.00%
accuracy of features	1	3	8	9	14	22	is	100.00%
accuracy of features	1	3	8	9	15	20	is	10

accuracy of features	1	4	9	10	20	22	is	100.00%	accuracy of features	1	5	7	9	11	22	is	100.00%	accuracy of features	1	6	7	11	15	20	is	100.00%
accuracy of features	1	4	9	11	13	14	is	100.00%	accuracy of features	1	5	7	11	13	20	is	100.00%	accuracy of features	1	6	7	11	16	20	is	100.00%
accuracy of features	1	4	9	11	13	15	is	100.00%	accuracy of features	1	5	7	11	13	22	is	100.00%	accuracy of features	1	6	7	11	17	20	is	100.00%
accuracy of features	1	4	9	11	13	16	is	100.00%	accuracy of features	1	5	7	11	14	20	is	100.00%	accuracy of features	1	6	7	11	17	22	is	100.00%
accuracy of features	1	4	9	11	13	17	is	100.00%	accuracy of features	1	5	7	11	14	22	is	100.00%	accuracy of features	1	6	7	11	20	22	is	100.00%
accuracy of features	1	4	9	11	13	20	is	100.00%	accuracy of features	1	5	7	11	16	20	is	100.00%	accuracy of features	1	6	7	18	20	22	is	100.00%
accuracy of features	1	4	9	11	13	21	is	100.00%	accuracy of features	1	5	7	11	17	22	is	100.00%	accuracy of features	1	6	8	9	10	22	is	100.00%
accuracy of features	1	4	9	11	13	22	is	100.00%	accuracy of features	1	5	7	10	22	is	100.00%	accuracy of features	1	6	8	9	11	15	is	100.00%	
accuracy of features	1	4	9	11	14	20	is	100.00%	accuracy of features	1	5	8	9	11	12	is	100.00%	accuracy of features	1	6	8	9	11	19	is	100.00%
accuracy of features	1	4	9	11	14	21	is	100.00%	accuracy of features	1	5	8	9	11	13	is	100.00%	accuracy of features	1	6	8	9	11	20	is	100.00%
accuracy of features	1	4	9	11	14	22	is	100.00%	accuracy of features	1	5	8	9	11	15	is	100.00%	accuracy of features	1	6	8	9	11	22	is	100.00%
accuracy of features	1	4	9	11	15	16	is	100.00%	accuracy of features	1	5	8	9	11	16	is	100.00%	accuracy of features	1	6	8	9	18	22	is	100.00%
accuracy of features	1	4	9	11	15	20	is	100.00%	accuracy of features	1	5	8	9	11	19	is	100.00%	accuracy of features	1	6	8	10	11	22	is	100.00%
accuracy of features	1	4	9	11	15	21	is	100.00%	accuracy of features	1	5	8	9	11	20	is	100.00%	accuracy of features	1	6	8	11	13	15	is	100.00%
accuracy of features	1	4	9	11	15	22	is	100.00%	accuracy of features	1	5	8	9	11	21	is	100.00%	accuracy of features	1	6	8	11	13	19	is	100.00%
accuracy of features	1	4	9	11	16	20	is	100.00%	accuracy of features	1	5	8	9	11	22	is	100.00%	accuracy of features	1	6	8	11	13	20	is	100.00%
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accuracy of features	1	4	9	11	17	21	is	100.00%	accuracy of features	1	5	8	11	13	16	is	100.00%	accuracy of features	1	6	8	11	14	19	is	100.00%
accuracy of features	1	4	9	11	19	21	is	100.00%	accuracy of features	1	5	8	11	13	17	is	100.00%	accuracy of features	1	6	8	11	14	20	is	100.00%
accuracy of features	1	4	9	11	19	22	is	100.00%	accuracy of features	1	5	8	11	13	19	is	100.00%	accuracy of features	1	6	8	11	14	22	is	100.00%

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accuracy of features	1	8	13	18	19	21		is 100.00%	accuracy of features	1	9	11	15	19	22		is 100.00%	accuracy of features	1	11	13	16	20	22		is 100.00%
accuracy of features	1	8	13	18	20	21		is 100.00%	accuracy of features	1	9	11	15	20	22		is 100.00%	accuracy of features	1	11	13	17	18	20		is 100.00%
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accuracy of features	1	8	14	18	21	22		is 100.00%	accuracy of features	1	9	11	16	17	21		is 100.00%	accuracy of features	1	11	13	17	20	21		is 100.00%
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accuracy of features 1	2	5	7	9	10	11	is	100.00%
accuracy of features 1	2	5	7	9	10	22	is	100.00%
accuracy of features 1	2	5	7	9	11	21	is	100.00%
accuracy of features 1	2	5	7	9	19	22	is	100.00%
accuracy of features 1	2	5	7	9	20	22	is	100.00%
accuracy of features 1	2	5	7	10	11	20	is	100.00%
accuracy of features 1	2	5	7	10	11	22	is	100.00%
accuracy of features 1	2	5	7	11	13	20	is	100.00%
accuracy of features 1	2	5	7	11	14	20	is	100.00%
accuracy of features 1	2	5	7	11	16	20	is	100.00%
accuracy of features 1	2	5	7	11	20	22	is	100.00%
accuracy of features 1	2	5	8	9	10	11	is	100.00%
accuracy of features 1	2	5	8	9	10	19	is	100.00%
accuracy of features 1	2	5	8	9	10	20	is	100.00%
accuracy of features 1	2	5	8	9	10	22	is	100.00%
accuracy of features 1	2	5	8	9	11	21	is	100.00%
accuracy of features 1	2	5	8	9	11	22	is	100.00%
accuracy of features 1	2	5	8	9	13	21	is	100.00%
accuracy of features 1	2	5	8	9	13	22	is	100.00%
accuracy of features 1	2	5	8	9	14	22	is	100.00%
accuracy of features 1	2	5	8	9	15	21	is	100.00%
accuracy of features 1	2	5	8	9	15	22	is	100.00%
accuracy of features 1	2	5	8	9	16	22	is	100.00%
accuracy of features 1	2	5	8	9	17	21	is	100.00%
accuracy of features 1	2	5	8	9	17	22	is	100.00%
accuracy of features 1	2	5	8	9	18	21	is	100.00%
accuracy of features 1	2	5	8	9	19	21	is	100.00%
accuracy of features 1	2	5	8	9	19	22	is	100.00%
accuracy of features 1	2	5	8	9	20	22	is	100.00%
accuracy of features 1	2	5	8	9	21	22	is	100.00%
accuracy of features 1	2	5	8	10	11	21	is	100.00%
accuracy of features 1	2	5	8	10	11	22	is	100.00%
accuracy of features 1	2	5	8	10	20	22	is	100.00%
accuracy of features 1	2	5	8	10	21	22	is	100.00%
accuracy of features 1	2	5	8	11	13	15	is	100.00%
accuracy of features 1	2	5	8	11	13	19	is	100.00%
accuracy of features 1	2	5	8	11	13	20	is	100.00%
accuracy of features 1	2	5	8	11	13	21	is	100.00%
accuracy of features 1	2	5	8	11	13	22	is	100.00%
accuracy of features 1	2	5	8	11	14	20	is	100.00%
accuracy of features 1	2	5	8	11	14	22	is	100.00%
accuracy of features 1	2	5	8	11	15	22	is	100.00%
accuracy of features 1	2	5	8	11	16	20	is	100.00%
accuracy of features 1	2	5	8	11	16	22	is	100.00%

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accuracy of features 1	3	5	6	11	17	20	is	100.00%
accuracy of features 1	3	5	6	11	20	22	is	100.00%
accuracy of features 1	3	5	7	8	9	22	is	100.00%
accuracy of features 1	3	5	7	8	11	20	is	100.00%
accuracy of features 1	3	5	7	8	11	22	is	100.00%
accuracy of features 1	3	5	7	9	10	11	is	100.00%
accuracy of features 1	3	5	7	9	11	21	is	100.00%
accuracy of features 1	3	5	7	9	11	22	is	100.00%
accuracy of features 1	3	5	7	9	20	22	is	100.00%
accuracy of features 1	3	5	7	11	13	20	is	100.00%
accuracy of features 1	3	5	7	11	14	20	is	100.00%
accuracy of features 1	3	5	7	11	16	20	is	100.00%
accuracy of features 1	3	5	7	11	20	22	is	100.00%
accuracy of features 1	3	5	7	18	20	22	is	100.00%
accuracy of features 1	3	5	8	9	11	21	is	100.00%
accuracy of features 1	3	5	8	9	11	22	is	100.00%
accuracy of features 1	3	5	8	9	13	21	is	100.00%
accuracy of features 1	3	5	8	9	14	22	is	100.00%
accuracy of features 1	3	5	8	9	15	22	is	100.00%
accuracy of features 1	3	5	8	9	16	22	is	100.00%
accuracy of features 1	3	5	8	9	17	21	is	100.00%
accuracy of features 1	3	5	8	9	17	22	is	100.00%
accuracy of features 1	3	5	8	9	18	21	is	100.00%
accuracy of features 1	3	5	8	9	20	22	is	100.00%
accuracy of features 1	3	5	8	11	13	20	is	100.00%
accuracy of features 1	3	5	8	11	13	22	is	100.00%
accuracy of features 1	3	5	8	11	14	20	is	100.00%
accuracy of features 1	3	5	8	11	14	22	is	100.00%
accuracy of features 1	3	5	8	11	15	22	is	100.00%
accuracy of features 1	3	5	8	11	16	20	is	100.00%
accuracy of features 1	3	5	8	11	16	22	is	100.00%
accuracy of features 1	3	5	8	11	17	22	is	100.00%
accuracy of features 1	3	5	8	11	18	21	is	100.00%
accuracy of features 1	3	5	8	11	18	22	is	100.00%
accuracy of features 1	3	5	8	11	20	22	is	100.00%
accuracy of features 1	3	5	8	14	18	22	is	100.00%
accuracy of features 1	3	5	8	17	18	21	is	100.00%
accuracy of features 1	3	5	8	17	20	22	is	100.00%
accuracy of features 1	3	5	9	10	11	14	is	100.00%
accuracy of features 1	3	5	9	10	11	20	is	100.00%
accuracy of features 1	3	5	9	10	11	21	is	100.00%
accuracy of features 1	3	5	9	10	11	22	is	100.00%
accuracy of features 1	3	5	9	10	20	22	is	100.00%
accuracy of features 1	3	5	9	10	21	22	is	100.00%
accuracy of features 1	3	5	9	11	13	21	is	100.00%
accuracy of features 1	3	5	9	11	15	21	is	100.00%
accuracy of features 1	3	5	9	11	16	20	is	100.00%
accuracy of features 1	3	5	9	11	17	21	is	100.00%
accuracy of features 1	3	5	9	11	18	21	is	100.00%
accuracy of features 1	3	5	9	11	19	21	is	100.00%
accuracy of features 1	3	5	9	11	20	21	is	100.00%
accuracy of features 1	3	5	9	11	20	22	is	100.00%
accuracy of features 1	3	5	9	11	21	22	is	100.00%
accuracy of features 1	3	5	9	13	20	22	is	100.00%
accuracy of features 1	3	5	9	14	20	22	is	100.00%
accuracy of features 1	3	5	9	14	21	22	is	100.00%
accuracy of features 1	3	5	9	15	16	22	is	100.00%
accuracy of features 1	3	5	9	15	17	22	is	100.00%
accuracy of features 1	3	5	9	15	20	22		

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accuracy of features 1	100.00%	1	5	8	13	15	18	22	is	100.00%	1	5	9	10	18	20	22	is	100.00%	1	5	9	13	16	20	22	is
accuracy of features 1	100.00%	1	5	8	13	17	18	21	is	100.00%	1	5	9	11	12	13	15	is	100.00%	1	5	9	13	17	20	22	is
accuracy of features 1	100.00%	1	5	8	13	17	18	22	is	100.00%	1	5	9	11	12	13	20	is	100.00%	1	5	9	13	18	20	22	is
accuracy of features 1	100.00%	1	5	8	13	17	20	22	is	100.00%	1	5	9	11	12	13	22	is	100.00%	1	5	9	15	19	20	22	is
accuracy of features 1	100.00%	1	5	8	13	18	20	22	is	100.00%	1	5	9	11	12	14	20	is	100.00%	1	5	9	16	18	19	22	is
accuracy of features 1	100.00%	1	5	8	14	15	18	22	is	100.00%	1	5	9	11	12	17	22	is	100.00%	1	5	10	11	13	18	20	is
accuracy of features 1	100.00%	1	5	8	14	18	19	22	is	100.00%	1	5	9	11	13	14	15	is	100.00%	1	5	10	11	13	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	12	14	is	100.00%	1	5	9	11	13	14	16	is	100.00%	1	5	10	11	13	21	22	is
accuracy of features 1	100.00%	1	5	9	10	11	12	19	is	100.00%	1	5	9	11	13	14	17	is	100.00%	1	5	10	11	14	15	22	is
accuracy of features 1	100.00%	1	5	9	10	11	12	20	is	100.00%	1	5	9	11	13	14	19	is	100.00%	1	5	10	11	14	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	13	19	is	100.00%	1	5	9	11	13	14	20	is	100.00%	1	5	10	11	15	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	13	20	is	100.00%	1	5	9	11	13	14	21	is	100.00%	1	5	10	11	15	21	22	is
accuracy of features 1	100.00%	1	5	9	10	11	13	22	is	100.00%	1	5	9	11	13	15	17	is	100.00%	1	5	10	11	16	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	14	15	is	100.00%	1	5	9	11	13	15	20	is	100.00%	1	5	10	11	17	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	14	19	is	100.00%	1	5	9	11	13	15	21	is	100.00%	1	5	10	11	17	21	22	is
accuracy of features 1	100.00%	1	5	9	10	11	14	20	is	100.00%	1	5	9	11	13	15	22	is	100.00%	1	5	10	11	18	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	14	22	is	100.00%	1	5	9	11	13	16	17	is	100.00%	1	5	10	14	18	20	22	is
accuracy of features 1	100.00%	1	5	9	10	11	15	19	is	100.00%	1	5	9	11	13	17	20	is	100.00%	1	5	11	12	13	18	20	is
accuracy of features 1	100.00%	1	5	9	10	11	15	20	is	100.00%	1	5	9	11	13	17	21	is	100.00%	1	5	11	13	14	15	20	is
accuracy of features 1	100.00%	1	5	9	10	11	15	22	is	100.00%	1	5	9	11	13	18	21	is	100.00%	1	5	11	13	14	15	22	is
accuracy of features 1	100.0																										

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accuracy of features 1 100.00%	8	9	15	16	19	21	is	accuracy of features 1 100.00%	8	10	11	18	21	22	is	accuracy of features 1 100.00%	8	11	13	15	18	20	is
accuracy of features 1 100.00%	8	9	15	17	18	22	is	accuracy of features 1 100.00%	8	10	11	19	20	21	is	accuracy of features 1 100.00%	8	11	13	15	18	22	is
accuracy of features 1 100.00%	8	9	15	18	19	22	is	accuracy of features 1 100.00%	8	10	11	19	20	22	is	accuracy of features 1 100.00%	8	11	13	15	19	20	is
accuracy of features 1 100.00%	8	9	15	19	21	22	is	accuracy of features 1 100.00%	8	10	11	19	21	22	is	accuracy of features 1 100.00%	8	11	13	15	19	21	is
accuracy of features 1 100.00%	8	9	16	17	18	22	is	accuracy of features 1 100.00%	8	10	11	20	21	22	is	accuracy of features 1 100.00%	8	11	13	15	19	22	is
accuracy of features 1 100.00%	8	9	16	18	19	22	is	accuracy of features 1 100.00%	8	10	13	14	21	22	is	accuracy of features 1 100.00%	8	11	13	15	20	21	is
accuracy of features 1 100.00%	8	9	16	18	20	22	is	accuracy of features 1 100.00%	8	10	13	20	21	22	is	accuracy of features 1 100.00%	8	11	13	15	20	22	is
accuracy of features 1 100.00%	8	9	16	18	21	22	is	accuracy of features 1 100.00%	8	10	14	18	19	21	is	accuracy of features 1 100.00%	8	11	13	16	17	19	is
accuracy of features 1 100.00%	8	9	16	19	21	22	is	accuracy of features 1 100.00%	8	10	14	18	19	22	is	accuracy of features 1 100.00%	8	11	13	16	17	20	is
accuracy of features 1 100.00%	8	9	17	18	20	22	is	accuracy of features 1 100.00%	8	10	14	18	20	22	is	accuracy of features 1 100.00%	8	11	13	16	17	22	is
accuracy of features 1 100.00%	8	9	18	19	21	22	is	accuracy of features 1 100.00%	8	10	14	18	21	22	is	accuracy of features 1 100.00%	8	11	13	16	18	20	is
accuracy of features 1 100.00%	8	10	11	12	14	18	is	accuracy of features 1 100.00%	8	10	14	19	20	22	is	accuracy of features 1 100.00%	8	11	13	16	18	21	is
accuracy of features 1 100.00%	8	10	11	12	18	20	is	accuracy of features 1 100.00%	8	10	18	19	20	21	is	accuracy of features 1 100.00%	8	11	13	16	18	22	is
accuracy of features 1 100.00%	8	10	11	13	14	21	is	accuracy of features 1 100.00%	8	10	18	19	20	22	is	accuracy of features 1 100.00%	8	11	13	16	19	20	is
accuracy of features 1 100.00%	8	10	11	13	20	21	is	accuracy of features 1 100.00%	8	10	18	19	21	22	is	accuracy of features 1 100.00%	8	11	13	16	19	21	is
accuracy of features 1 100.00%	8	10	11	13	20	22	is	accuracy of features 1 100.00%	8	10	18	20	21	22	is	accuracy of features 1 100.00%	8	11	13	16	19	22	is
accuracy of features 1 100.00%	8	10	11	13	21	22	is	accuracy of features 1 100.00%	8	11	13	14	15	17	is	accuracy of features 1 100.00%	8	11	13	16	20	21	is
accuracy of features 1 100.00%	8	10	11	14	15	21	is	accuracy of features 1 100.00%	8	11	13	14	15	19	is	accuracy of features 1 100.00%	8	11	13	17	18	20	is
accuracy of features 1 100.00%	8	10	11	14	15	22	is	accuracy of features 1 100.00%	8	11	13	14	15	20	is	accuracy of features 1 100.00%	8	11	13	17	18	22	is
accuracy of features 1 100.00%	8	10	11	14	17	21	is	accuracy of features 1 100.00%	8	11	13	14	15	21	is	accuracy of features 1 100.00%	8	11	13	17	19	20	is
accuracy of features 1 100.00%	8	10	11	14	17	22	is	accuracy of features 1 100.00%	8	11	13	14	15	22									

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accuracy of features 2	9	10	11	17	20	21	is	100.00%	accuracy of features 2	10	11	13	15	20	21	is	100.00%	accuracy of features 3	5	10	11	15	21	22	is	100.00%
accuracy of features 2	9	10	11	19	20	21	is	100.00%	accuracy of features 2	10	11	13	17	20	21	is	100.00%	accuracy of features 3	5	10	11	18	20	21	is	100.00%
accuracy of features 2	9	10	11	20	21	22	is	100.00%	accuracy of features 2	10	11	13	20	21	22	is	100.00%	accuracy of features 3	6	9	11	13	15	20	is	100.00%
accuracy of features 2	9	10	13	14	15	21	is	100.00%	accuracy of features 2	10	11	14	20	21	22	is	100.00%	accuracy of features 3	7	8	9	10	15	21	is	100.00%
accuracy of features 2	9	10	13	14	17	21	is	100.00%	accuracy of features 2	10	11	15	19	20	21	is	100.00%	accuracy of features 3	7	8	9	10	20	21	is	100.00%
accuracy of features 2	9	10	13	14	20	21	is	100.00%	accuracy of features 2	10	11	15	20	21	22	is	100.00%	accuracy of features 3	7	8	9	15	21	22	is	100.00%
accuracy of features 2	9	10	13	15	19	21	is	100.00%	accuracy of features 2	10	11	17	20	21	22	is	100.00%	accuracy of features 3	7	8	10	11	15	21	is	100.00%
accuracy of features 2	9	10	13	15	20	21	is	100.00%	accuracy of features 2	11	13	15	18	20	21	is	100.00%	accuracy of features 3	7	8	10	15	21	22	is	100.00%
accuracy of features 2	9	10	13	15	21	22	is	100.00%	accuracy of features 2	11	13	17	20	21	22	is	100.00%	accuracy of features 3	7	8	10	18	20	21	is	100.00%
accuracy of features 2	9	10	13	17	20	21	is	100.00%	accuracy of features 2	11	14	17	20	21	22	is	100.00%	accuracy of features 3	7	9	10	11	15	21	is	100.00%
accuracy of features 2	9	10	13	20	21	22	is	100.00%	accuracy of features 2	11	15	17	20	21	22	is	100.00%	accuracy of features 3	7	9	10	11	20	21	is	100.00%
accuracy of features 2	9	10	14	15	19	21	is	100.00%	accuracy of features 3	4	6	9	11	15	20	is	100.00%	accuracy of features 3	7	9	10	13	15	21	is	100.00%
accuracy of features 2	9	10	14	15	20	21	is	100.00%	accuracy of features 3	4	8	9	10	15	21	is	100.00%	accuracy of features 3	7	9	10	15	17	21	is	100.00%
accuracy of features 2	9	10	14	15	21	22	is	100.00%	accuracy of features 3	4	8	10	13	20	21	is	100.00%	accuracy of features 3	7	9	10	18	20	21	is	100.00%
accuracy of features 2	9	10	14	17	20	21	is	100.00%	accuracy of features 3	4	8	10	15	20	21	is	100.00%	accuracy of features 3	7	9	10	20	21	22	is	100.00%
accuracy of features 2	9	10	14	19	20	21	is	100.00%	accuracy of features 3	4	8	10	17	20	21	is	100.00%	accuracy of features 3	7	10	11	13	15	21	is	100.00%
accuracy of features 2	9	10	14	20	21	22	is	100.00%	accuracy of features 3	4	8	10	19	20	21	is	100.00%	accuracy of features 3	7	10	11	15	17	21	is	100.00%
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accuracy of features 2	9	10	15	17	20	21	is	100.00%	accuracy of features 3	4	9	10	11	15	21	is	100.00%	accuracy of features 3	7	10	11	20	21	22	is	100.00%
accuracy of features 2	9	10	15	17	21	22	is	100.00%	accuracy of features 3	4	9	10	13	15	21	is	100.00%	accuracy of features 3	7	10						

accuracy of features 3	8	9	15	17	19	21	is	100.00%	accuracy of features 3	9	10	12	13	20	21	is	100.00%	accuracy of features 3	11	18	19	20	21	22	is	100.00%	
accuracy of features 3	8	9	15	17	21	22	is	100.00%	accuracy of features 3	9	10	13	14	20	21	is	100.00%	accuracy of features 4	5	6	9	11	13	20	is	100.00%	
accuracy of features 3	8	9	15	18	19	21	is	100.00%	accuracy of features 3	9	10	13	15	21	22	is	100.00%	accuracy of features 4	5	7	9	10	11	15	is	100.00%	
accuracy of features 3	8	9	15	18	21	22	is	100.00%	accuracy of features 3	9	10	14	15	21	22	is	100.00%	accuracy of features 4	5	8	9	10	11	13	is	100.00%	
accuracy of features 3	8	9	15	20	21	22	is	100.00%	accuracy of features 3	9	10	15	17	21	22	is	100.00%	accuracy of features 4	5	8	9	10	11	17	is	100.00%	
accuracy of features 3	8	9	18	19	20	21	is	100.00%	accuracy of features 3	9	10	15	18	20	21	is	100.00%	accuracy of features 4	5	8	9	10	11	20	is	100.00%	
accuracy of features 3	8	9	18	20	21	22	is	100.00%	accuracy of features 3	9	10	15	19	20	21	is	100.00%	accuracy of features 4	5	8	9	10	13	20	is	100.00%	
accuracy of features 3	8	9	19	20	21	22	is	100.00%	accuracy of features 3	9	10	15	19	21	22	is	100.00%	accuracy of features 4	5	8	9	10	13	21	is	100.00%	
accuracy of features 3	8	10	11	13	20	21	is	100.00%	accuracy of features 3	9	10	15	20	21	22	is	100.00%	accuracy of features 4	5	8	9	11	17	20	is	100.00%	
accuracy of features 3	8	10	11	14	15	21	is	100.00%	accuracy of features 3	9	10	18	20	21	22	is	100.00%	accuracy of features 4	5	8	10	11	14	15	is	100.00%	
accuracy of features 3	8	10	11	14	20	21	is	100.00%	accuracy of features 3	9	10	19	20	21	22	is	100.00%	accuracy of features 4	5	8	10	11	19	20	is	100.00%	
accuracy of features 3	8	10	11	15	19	21	is	100.00%	accuracy of features 3	9	11	13	18	20	21	is	100.00%	accuracy of features 4	5	8	11	13	17	20	is	100.00%	
accuracy of features 3	8	10	11	15	20	21	is	100.00%	accuracy of features 3	9	11	13	20	21	22	is	100.00%	accuracy of features 4	5	8	11	13	17	20	is	100.00%	
accuracy of features 3	8	10	11	15	21	22	is	100.00%	accuracy of features 3	9	11	14	18	20	21	is	100.00%	accuracy of features 4	6	8	9	10	11	15	is	100.00%	
accuracy of features 3	8	10	11	17	20	21	is	100.00%	accuracy of features 3	9	11	17	18	20	21	is	100.00%	accuracy of features 4	6	8	9	11	18	20	is	100.00%	
accuracy of features 3	8	10	13	14	20	21	is	100.00%	accuracy of features 3	9	11	18	20	21	22	is	100.00%	accuracy of features 4	6	8	10	11	19	20	is	100.00%	
accuracy of features 3	8	10	13	15	20	21	is	100.00%	accuracy of features 3	9	13	18	20	21	22	is	100.00%	accuracy of features 4	6	8	10	11	19	20	is	100.00%	
accuracy of features 3	8	10	13	17	20	21	is	100.00%	accuracy of features 3	9	14	15	20	21	22	is	100.00%	accuracy of features 4	6	8	9	10	13	15	20	is	100.00%
accuracy of features 3	8	10	13	19	20	21	is	100.00%	accuracy of features 3	9	15	17	18	20	21	is	100.00%	accuracy of features 4	6	8	9	10	13	17	20	is	100.00%
accuracy of features 3	8	10	13	20	21	22	is	100.00%	accuracy of features 3	9	15	19	20	21	22	is	100.00%	accuracy of features 4	6	8	9						

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Aims and working mechanism

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- **Setup and Data Loading:** The process initiates by configuring the environment, establishing a results directory (should it not already exist), and opening a file for logging outcomes. It proceeds to load the dataset from 'data.xlsx', under the assumption that it comprises samples from 14 categories.
- **Labeling and Cross-validation Preparation:** The dataset is systematically labelled, followed by the establishment of a 4-fold cross-validation framework to enable reliable assessment of the model's efficacy.
- **Feature Selection and LDA Classification:** The code explores the impact of different feature combinations on classification accuracy by selecting subsets of features (either 1 or 4 features from a pool of 22). For each subset, it conducts a 4-fold cross-validation, wherein an LDA model is trained on the training set and subsequently used to predict the labels of the testing set.
- **Performance Evaluation:** It calculates and logs the mean accuracy for each feature set, proceeding to compute and log detailed performance metrics such as precision, recall, and F1 score for feature sets where the mean accuracy surpasses 50%.
- **Special Handling for High-Performing Feature Sets:** For those sets of 4 features that achieve a mean accuracy of 100%, additional steps are taken:
 - **Dimensionality Reduction and Visualization:** Implements PCA to reduce dimensionality to 2D and visualizes the classification outcome via a scatter plot, facilitating a visual appraisal of the LDA model's class-separation capability in a reduced-dimensional space.
 - **Ellipse Property Calculation:** Computes and records the properties of ellipses (encompassing area, centroid, and angle) that could delineate the distribution of each class within the feature space, presumably for subsequent data distribution and classification boundary analysis.

- **Utility Functions:** Incorporated within the code are functions designed to depict the 2D LDA classification outcomes, compute ellipse properties for optimally performing feature sets, and evaluate classification results through performance metrics (accuracy, precision, recall, F1 score).

Script and functions

```

clear;
files = 'data.xlsx'; % Assuming the data file is named 'data.xlsx'
resultsDir = 'Testing_LDA_Array_results';
if ~exist(resultsDir, 'dir')
    mkdir(resultsDir);
end
fileID = fopen(fullfile(resultsDir, 'lda_WITHOUTN_array_result.txt'), 'w');

% Load data
[data, txt, ~] = xlsread(files);
label = repelem(1:14, 4); % Assuming each metal ion has the same number of samples
indices = crossvalind('Kfold', label, 4);

for feature_num = [1, 4]
    C = nchoosek(1:22, feature_num); % Generate combinations of features
    for i = 1:size(C, 1)
        feature_select = C(i, :);
        feature_data = data(:, feature_select);
        accuracy = zeros(4, 1); % To store accuracy for each fold
        for k = 1:4 % 4-fold cross-validation
            test = (indices == k);
            train = ~test;
            lda = fitcdiscr(feature_data(train,:), label(train));
            predictions = predict(lda, feature_data(test,:));
            accuracy(k) = mean(predictions == label(test));
        end
        mean_accuracy = mean(accuracy);
        fprintf(fileID, 'Feature Set: %s, Mean Accuracy: %f\n', mat2str(feature_select), mean_accuracy);

        if mean_accuracy > 0.5
            metrics = CalculatePerformanceMetrics(label(test), predictions);
            fprintf(fileID, 'Feature Set: %s, Accuracy: %f, Precision: %f, Recall: %f, F1 Score: %f\n', ...
                mat2str(feature_select), metrics(1), metrics(2), metrics(3), metrics(4));
        end

        if feature_num == 4 && mean_accuracy == 1.0
            % Plotting and calculation for 4 features with 100% accuracy
            plotLDA2D(lda, feature_data, label, txt, resultsDir, feature_select);
            calculateEllipseProperties(feature_data, label, resultsDir, feature_select);
        end
    end
end
fclose(fileID);

```

```

function plotLDA2D(lda, data, label, txt, resultsDir, feature_select)
    % Perform PCA for dimensionality reduction to 2D
    [coeff, score, ~, ~, explained] = pca(data);
    reducedData = data * coeff(:,1:2); % Projecting to 2D
    figure;
    gscatter(reducedData(:,1), reducedData(:,2), label, hsv(14));
    xlabel('First Principal Component');
    ylabel('Second Principal Component');
    title(sprintf('LDA Classification with Features %s', mat2str(feature_select)));
    legend(txt(4:4:end,2), 'Location', 'bestoutside');
    saveas(gcf, fullfile(resultsDir, sprintf('LDA_2DPlot_Features_%s.png', mat2str(feature_select))));
    close(gcf); % Close the figure window after saving the plot

end

function calculateEllipseProperties(data, label, resultsDir, feature_select)
    numTargets = max(label);
    properties = struct();
    for i = 1:numTargets
        idx = find(label == i);
        selectedData = data(idx, :);
        [eigenvec, eigenval] = eig(cov(selectedData));
        angle = atan2(eigenvec(2,1), eigenvec(1,1)) * (180/pi);
        properties(i).Area = pi * sqrt(eigenval(1,1) * eigenval(2,2));
        properties(i).Centroid = mean(selectedData, 1);
        properties(i).Angle = angle;
    end
    save(fullfile(resultsDir, sprintf('EllipseProperties_Features_%s.mat', mat2str(feature_select))),
    'properties');
end

function metrics = CalculatePerformanceMetrics(true_labels, predicted_labels)
    % Calculate confusion matrix
    [C,~] = confusionmat(true_labels, predicted_labels);
    TP = diag(C);
    FP = sum(C, 1) - TP;
    FN = sum(C, 2) - TP;
    TN = sum(C(:)) - TP - FP - FN;

    Accuracy = sum(TP + TN) / sum(TP + FP + FN + TN);
    Precision = TP ./ (TP + FP);
    Recall = TP ./ (TP + FN);
    F1 = 2 .* (Precision .* Recall) ./ (Precision + Recall);

    % Return the mean of the metrics across all classes
    metrics = [mean(Accuracy), mean(nanmean(Precision)), mean(nanmean(Recall)), mean(nanmean(F1))];
end

```

Result of the one feature and four features-based CDs array studies

Feature Set: 1, Mean Accuracy: 0.446429
 Feature Set: 2, Mean Accuracy: 0.214286
 Feature Set: 3, Mean Accuracy: 0.142857
 Feature Set: 4, Mean Accuracy: 0.446429
 Feature Set: 5, Mean Accuracy: 0.339286
 Feature Set: 6, Mean Accuracy: 0.321429
 Feature Set: 7, Mean Accuracy: 0.339286
 Feature Set: 8, Mean Accuracy: 0.678571
 Feature Set: 8, Accuracy: 0.928571, Precision: 0.538462, Recall: 0.500000, F1 Score: 1.000000
 Feature Set: 9, Mean Accuracy: 0.464286

Feature Set: 10, Mean Accuracy: 0.571429
 Feature Set: 10, Accuracy: 0.928571, Precision: 0.500000, Recall: 0.500000, F1 Score: 0.809524
 Feature Set: 11, Mean Accuracy: 0.696429
 Feature Set: 11, Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: 12, Mean Accuracy: 0.285714
 Feature Set: 13, Mean Accuracy: 0.250000
 Feature Set: 14, Mean Accuracy: 0.196429
 Feature Set: 15, Mean Accuracy: 0.250000
 Feature Set: 16, Mean Accuracy: 0.321429

Feature Set: [1 2 6 17], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
Feature Set: [1 2 6 18], Mean Accuracy: 0.678571
Feature Set: [1 2 6 18], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [1 2 6 19], Mean Accuracy: 0.696429
Feature Set: [1 2 6 19], Accuracy: 0.928571, Precision: 0.541667, Recall: 0.500000, F1 Score: 0.952381
Feature Set: [1 2 6 20], Mean Accuracy: 0.803571
Feature Set: [1 2 6 20], Accuracy: 0.938776, Precision: 0.611111, Recall: 0.571429, F1 Score: 0.937500
Feature Set: [1 2 6 21], Mean Accuracy: 0.839286
Feature Set: [1 2 6 21], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [1 2 6 22], Mean Accuracy: 0.839286
Feature Set: [1 2 6 22], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
Feature Set: [1 2 7 8], Mean Accuracy: 0.892857
Feature Set: [1 2 7 8], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 2 7 9], Mean Accuracy: 0.910714
Feature Set: [1 2 7 9], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 2 7 10], Mean Accuracy: 0.910714
Feature Set: [1 2 7 10], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 2 7 11], Mean Accuracy: 0.964286
Feature Set: [1 2 7 11], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 2 7 12], Mean Accuracy: 0.750000
Feature Set: [1 2 7 12], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 2 7 13], Mean Accuracy: 0.714286
Feature Set: [1 2 7 13], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
Feature Set: [1 2 7 14], Mean Accuracy: 0.767857
Feature Set: [1 2 7 14], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [1 2 7 15], Mean Accuracy: 0.785714
Feature Set: [1 2 7 15], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
Feature Set: [1 2 7 16], Mean Accuracy: 0.732143
Feature Set: [1 2 7 16], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 2 7 17], Mean Accuracy: 0.750000
Feature Set: [1 2 7 17], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
Feature Set: [1 2 7 18], Mean Accuracy: 0.732143
Feature Set: [1 2 7 18], Accuracy: 0.969388, Precision: 0.931818, Recall: 0.785714, F1 Score: 0.945455
Feature Set: [1 2 7 19], Mean Accuracy: 0.785714
Feature Set: [1 2 7 19], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 2 7 20], Mean Accuracy: 0.857143
Feature Set: [1 2 7 20], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 2 7 21], Mean Accuracy: 0.875000
Feature Set: [1 2 7 21], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 2 7 22], Mean Accuracy: 0.857143
Feature Set: [1 2 7 22], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 2 8 9], Mean Accuracy: 0.910714
Feature Set: [1 2 8 9], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 2 8 10], Mean Accuracy: 0.964286
Feature Set: [1 2 8 10], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 2 8 11], Mean Accuracy: 0.982143
Feature Set: [1 2 8 11], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 2 8 12], Mean Accuracy: 0.875000
Feature Set: [1 2 8 12], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 2 8 13], Mean Accuracy: 0.892857
Feature Set: [1 2 8 13], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 2 8 14], Mean Accuracy: 0.875000
Feature Set: [1 2 8 14], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 2 8 15], Mean Accuracy: 0.928571
Feature Set: [1 2 8 15], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 2 8 16], Mean Accuracy: 0.839286
Feature Set: [1 2 8 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 2 8 17], Mean Accuracy: 0.892857
Feature Set: [1 2 8 17], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 2 8 18], Mean Accuracy: 0.857143
Feature Set: [1 2 8 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 2 8 19], Mean Accuracy: 0.857143
Feature Set: [1 2 8 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 2 8 20], Mean Accuracy: 0.910714
Feature Set: [1 2 8 20], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [1 2 8 21], Mean Accuracy: 0.964286
Feature Set: [1 2 8 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

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Feature Set: [1 3 11 16], Mean Accuracy: 0.964286
Feature Set: [1 3 11 16], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 3 11 17], Mean Accuracy: 0.946429
Feature Set: [1 3 11 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 3 11 18], Mean Accuracy: 0.928571
Feature Set: [1 3 11 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 3 11 19], Mean Accuracy: 0.946429
Feature Set: [1 3 11 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 3 11 20], Mean Accuracy: 0.946429
Feature Set: [1 3 11 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 3 11 21], Mean Accuracy: 0.982143
Feature Set: [1 3 11 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 3 11 22], Mean Accuracy: 0.946429
Feature Set: [1 3 11 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 3 12 13], Mean Accuracy: 0.696429
Feature Set: [1 3 12 13], Accuracy: 0.959184, Precision: 0.866667, Recall: 0.714286, F1 Score: 0.900000
Feature Set: [1 3 12 14], Mean Accuracy: 0.625000
Feature Set: [1 3 12 14], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 3 12 15], Mean Accuracy: 0.732143
Feature Set: [1 3 12 15], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
Feature Set: [1 3 12 16], Mean Accuracy: 0.660714
Feature Set: [1 3 12 16], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 3 12 17], Mean Accuracy: 0.625000
Feature Set: [1 3 12 17], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 3 12 18], Mean Accuracy: 0.696429
Feature Set: [1 3 12 18], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 3 12 19], Mean Accuracy: 0.696429
Feature Set: [1 3 12 19], Accuracy: 0.928571, Precision: 0.590909, Recall: 0.500000, F1 Score: 0.952381
Feature Set: [1 3 12 20], Mean Accuracy: 0.785714
Feature Set: [1 3 12 20], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 3 12 21], Mean Accuracy: 0.839286
Feature Set: [1 3 12 21], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [1 3 12 22], Mean Accuracy: 0.839286
Feature Set: [1 3 12 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 3 13 14], Mean Accuracy: 0.660714
Feature Set: [1 3 13 14], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 3 13 15], Mean Accuracy: 0.678571
Feature Set: [1 3 13 15], Accuracy: 0.948980, Precision: 0.653846, Recall: 0.642857, F1 Score: 0.962963
Feature Set: [1 3 13 16], Mean Accuracy: 0.625000
Feature Set: [1 3 13 16], Accuracy: 0.959184, Precision: 0.727272, Recall: 0.714286, F1 Score: 0.900000
Feature Set: [1 3 13 17], Mean Accuracy: 0.607143
Feature Set: [1 3 13 17], Accuracy: 0.948980, Precision: 0.653846, Recall: 0.642857, F1 Score: 0.962963
Feature Set: [1 3 13 18], Mean Accuracy: 0.696429
Feature Set: [1 3 13 18], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [1 3 13 19], Mean Accuracy: 0.660714
Feature Set: [1 3 13 19], Accuracy: 0.928571, Precision: 0.600000, Recall: 0.500000, F1 Score: 0.904762
Feature Set: [1 3 13 20], Mean Accuracy: 0.785714
Feature Set: [1 3 13 20], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
Feature Set: [1 3 13 21], Mean Accuracy: 0.785714
Feature Set: [1 3 13 21], Accuracy: 0.959184, Precision: 0.714286, Recall: 0.714286, F1 Score: 1.000000
Feature Set: [1 3 13 22], Mean Accuracy: 0.785714
Feature Set: [1 3 13 22], Accuracy: 0.948980, Precision: 0.692308, Recall: 0.642857, F1 Score: 1.000000
Feature Set: [1 3 14 15], Mean Accuracy: 0.732143
Feature Set: [1 3 14 15], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 3 14 16], Mean Accuracy: 0.642857
Feature Set: [1 3 14 16], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 3 14 17], Mean Accuracy: 0.678571
Feature Set: [1 3 14 17], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 3 14 18], Mean Accuracy: 0.714286
Feature Set: [1 3 14 18], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [1 3 14 19], Mean Accuracy: 0.732143
Feature Set: [1 3 14 19], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [1 3 14 20], Mean Accuracy: 0.821429
Feature Set: [1 3 14 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 3 14 21], Mean Accuracy: 0.821429
Feature Set: [1 3 14 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 3 14 22], Mean Accuracy: 0.839286

Feature Set: [1 4 7 13], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
Feature Set: [1 4 7 14], Mean Accuracy: 0.732143
Feature Set: [1 4 7 14], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 4 7 15], Mean Accuracy: 0.785714
Feature Set: [1 4 7 15], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 4 7 16], Mean Accuracy: 0.732143
Feature Set: [1 4 7 16], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 4 7 17], Mean Accuracy: 0.714286
Feature Set: [1 4 7 17], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 4 7 18], Mean Accuracy: 0.785714
Feature Set: [1 4 7 18], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 4 7 19], Mean Accuracy: 0.750000
Feature Set: [1 4 7 19], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
Feature Set: [1 4 7 20], Mean Accuracy: 0.839286
Feature Set: [1 4 7 20], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [1 4 7 21], Mean Accuracy: 0.875000
Feature Set: [1 4 7 21], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 4 7 22], Mean Accuracy: 0.910714
Feature Set: [1 4 7 22], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 4 8 9], Mean Accuracy: 0.910714
Feature Set: [1 4 8 9], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 10], Mean Accuracy: 0.964286
Feature Set: [1 4 8 10], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 11], Mean Accuracy: 1.000000
Feature Set: [1 4 8 11], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 4 8 12], Mean Accuracy: 0.857143
Feature Set: [1 4 8 12], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 4 8 13], Mean Accuracy: 0.839286
Feature Set: [1 4 8 13], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 4 8 14], Mean Accuracy: 0.839286
Feature Set: [1 4 8 14], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 15], Mean Accuracy: 0.892857
Feature Set: [1 4 8 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 16], Mean Accuracy: 0.857143
Feature Set: [1 4 8 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 17], Mean Accuracy: 0.821429
Feature Set: [1 4 8 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 18], Mean Accuracy: 0.875000
Feature Set: [1 4 8 18], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 19], Mean Accuracy: 0.875000
Feature Set: [1 4 8 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 20], Mean Accuracy: 0.857143
Feature Set: [1 4 8 20], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [1 4 8 21], Mean Accuracy: 0.910714
Feature Set: [1 4 8 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 8 22], Mean Accuracy: 0.910714
Feature Set: [1 4 8 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 9 10], Mean Accuracy: 0.982143
Feature Set: [1 4 9 10], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 4 9 11], Mean Accuracy: 0.964286
Feature Set: [1 4 9 11], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 9 12], Mean Accuracy: 0.892857
Feature Set: [1 4 9 12], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 4 9 13], Mean Accuracy: 0.910714
Feature Set: [1 4 9 13], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 9 14], Mean Accuracy: 0.910714
Feature Set: [1 4 9 14], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 9 15], Mean Accuracy: 0.928571
Feature Set: [1 4 9 15], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [1 4 9 16], Mean Accuracy: 0.892857
Feature Set: [1 4 9 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 4 9 17], Mean Accuracy: 0.875000
Feature Set: [1 4 9 17], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 4 9 18], Mean Accuracy: 0.910714
Feature Set: [1 4 9 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 4 9 19], Mean Accuracy: 0.928571
Feature Set: [1 4 9 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

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Feature Set: [1 7 18 2], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 7 19 20], Mean Accuracy: 0.785714
Feature Set: [1 7 19 20], Accuracy: 0.948980, Precision: 0.694444, Recall: 0.642857, F1 Score: 0.944444
Feature Set: [1 7 19 21], Mean Accuracy: 0.857143
Feature Set: [1 7 19 21], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
Feature Set: [1 7 19 22], Mean Accuracy: 0.857143
Feature Set: [1 7 19 22], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
Feature Set: [1 7 20 21], Mean Accuracy: 0.928571
Feature Set: [1 7 20 21], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 7 20 22], Mean Accuracy: 0.928571
Feature Set: [1 7 20 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 7 21 22], Mean Accuracy: 0.875000
Feature Set: [1 7 21 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 8 9 10], Mean Accuracy: 0.964286
Feature Set: [1 8 9 10], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 9 11], Mean Accuracy: 1.000000
Feature Set: [1 8 9 11], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 9 12], Mean Accuracy: 0.910714
Feature Set: [1 8 9 12], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 9 13], Mean Accuracy: 0.892857
Feature Set: [1 8 9 13], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
Feature Set: [1 8 9 14], Mean Accuracy: 0.910714
Feature Set: [1 8 9 14], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 9 15], Mean Accuracy: 0.946429
Feature Set: [1 8 9 15], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 9 16], Mean Accuracy: 0.892857
Feature Set: [1 8 9 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 9 17], Mean Accuracy: 0.910714
Feature Set: [1 8 9 17], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [1 8 9 18], Mean Accuracy: 0.928571
Feature Set: [1 8 9 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 9 19], Mean Accuracy: 0.910714
Feature Set: [1 8 9 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 9 20], Mean Accuracy: 0.964286
Feature Set: [1 8 9 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 9 21], Mean Accuracy: 0.964286
Feature Set: [1 8 9 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 9 22], Mean Accuracy: 0.982143
Feature Set: [1 8 9 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 10 11], Mean Accuracy: 1.000000
Feature Set: [1 8 10 11], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 10 12], Mean Accuracy: 0.982143
Feature Set: [1 8 10 12], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 10 13], Mean Accuracy: 0.964286
Feature Set: [1 8 10 13], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 10 14], Mean Accuracy: 0.964286
Feature Set: [1 8 10 14], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 10 15], Mean Accuracy: 0.946429
Feature Set: [1 8 10 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 10 16], Mean Accuracy: 0.928571
Feature Set: [1 8 10 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 10 17], Mean Accuracy: 0.928571
Feature Set: [1 8 10 17], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [1 8 10 18], Mean Accuracy: 0.946429
Feature Set: [1 8 10 18], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 10 19], Mean Accuracy: 0.946429
Feature Set: [1 8 10 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 10 20], Mean Accuracy: 0.964286
Feature Set: [1 8 10 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 10 21], Mean Accuracy: 0.964286
Feature Set: [1 8 10 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 10 22], Mean Accuracy: 0.946429
Feature Set: [1 8 10 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 8 11 12], Mean Accuracy: 1.000000
Feature Set: [1 8 11 12], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 8 11 13], Mean Accuracy: 1.000000
Feature Set: [1 8 11 13], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

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Feature Set: [1 9 18 21], Mean Accuracy: 0.928571
Feature Set: [1 9 18 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 9 18 22], Mean Accuracy: 0.910714
Feature Set: [1 9 18 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 9 19 20], Mean Accuracy: 0.946429
Feature Set: [1 9 19 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 9 19 21], Mean Accuracy: 0.964286
Feature Set: [1 9 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 9 19 22], Mean Accuracy: 0.857143
Feature Set: [1 9 19 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 9 20 21], Mean Accuracy: 0.982143
Feature Set: [1 9 20 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 9 20 22], Mean Accuracy: 0.946429
Feature Set: [1 9 20 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 9 21 22], Mean Accuracy: 0.946429
Feature Set: [1 9 21 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 11 12], Mean Accuracy: 0.982143
Feature Set: [1 10 11 12], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 11 13], Mean Accuracy: 1.000000
Feature Set: [1 10 11 13], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 11 14], Mean Accuracy: 1.000000
Feature Set: [1 10 11 14], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 11 15], Mean Accuracy: 0.982143
Feature Set: [1 10 11 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 11 16], Mean Accuracy: 1.000000
Feature Set: [1 10 11 16], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 11 17], Mean Accuracy: 0.982143
Feature Set: [1 10 11 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 11 18], Mean Accuracy: 0.964286
Feature Set: [1 10 11 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 11 19], Mean Accuracy: 0.982143
Feature Set: [1 10 11 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 11 20], Mean Accuracy: 1.000000
Feature Set: [1 10 11 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 11 21], Mean Accuracy: 1.000000
Feature Set: [1 10 11 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 11 22], Mean Accuracy: 1.000000
Feature Set: [1 10 11 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [1 10 12 13], Mean Accuracy: 0.910714
Feature Set: [1 10 12 13], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [1 10 12 14], Mean Accuracy: 0.928571
Feature Set: [1 10 12 14], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 12 15], Mean Accuracy: 0.875000
Feature Set: [1 10 12 15], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
Feature Set: [1 10 12 16], Mean Accuracy: 0.910714
Feature Set: [1 10 12 16], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [1 10 12 17], Mean Accuracy: 0.875000
Feature Set: [1 10 12 17], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [1 10 12 18], Mean Accuracy: 0.910714
Feature Set: [1 10 12 18], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 12 19], Mean Accuracy: 0.839286
Feature Set: [1 10 12 19], Accuracy: 0.948980, Precision: 0.692308, Recall: 0.642857, F1 Score: 1.000000
Feature Set: [1 10 12 20], Mean Accuracy: 0.946429
Feature Set: [1 10 12 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 12 21], Mean Accuracy: 0.928571
Feature Set: [1 10 12 21], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
Feature Set: [1 10 12 22], Mean Accuracy: 0.910714
Feature Set: [1 10 12 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [1 10 13 14], Mean Accuracy: 0.910714
Feature Set: [1 10 13 14], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [1 10 13 15], Mean Accuracy: 0.910714
Feature Set: [1 10 13 15], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [1 10 13 16], Mean Accuracy: 0.964286
Feature Set: [1 10 13 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [1 10 13 17], Mean Accuracy: 0.875000
Feature Set: [1 10 13 17], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [1 10 13 18], Mean Accuracy: 0.875000

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Feature Set: [2 3 4 12], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [2 3 4 13], Mean Accuracy: 0.589286
 Feature Set: [2 3 4 13], Accuracy: 0.938776, Precision: 0.681818, Recall: 0.571429, F1 Score: 0.958333
 Feature Set: [2 3 4 14], Mean Accuracy: 0.535714
 Feature Set: [2 3 4 14], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [2 3 4 15], Mean Accuracy: 0.678571
 Feature Set: [2 3 4 15], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [2 3 4 16], Mean Accuracy: 0.571429
 Feature Set: [2 3 4 16], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
 Feature Set: [2 3 4 17], Mean Accuracy: 0.607143
 Feature Set: [2 3 4 17], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [2 3 4 18], Mean Accuracy: 0.625000
 Feature Set: [2 3 4 18], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [2 3 4 19], Mean Accuracy: 0.678571
 Feature Set: [2 3 4 19], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [2 3 4 20], Mean Accuracy: 0.696429
 Feature Set: [2 3 4 20], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [2 3 4 21], Mean Accuracy: 0.785714
 Feature Set: [2 3 4 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 4 22], Mean Accuracy: 0.767857
 Feature Set: [2 3 4 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 5 6], Mean Accuracy: 0.482143
 Feature Set: [2 3 5 7], Mean Accuracy: 0.553571
 Feature Set: [2 3 5 7], Accuracy: 0.918367, Precision: 0.611111, Recall: 0.428571, F1 Score: 0.944444
 Feature Set: [2 3 5 8], Mean Accuracy: 0.785714
 Feature Set: [2 3 5 8], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [2 3 5 9], Mean Accuracy: 0.839286
 Feature Set: [2 3 5 9], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 5 10], Mean Accuracy: 0.696429
 Feature Set: [2 3 5 10], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [2 3 5 11], Mean Accuracy: 0.892857
 Feature Set: [2 3 5 11], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 5 12], Mean Accuracy: 0.535714
 Feature Set: [2 3 5 12], Accuracy: 0.938776, Precision: 0.683333, Recall: 0.571429, F1 Score: 0.895833
 Feature Set: [2 3 5 13], Mean Accuracy: 0.428571
 Feature Set: [2 3 5 14], Mean Accuracy: 0.375000
 Feature Set: [2 3 5 15], Mean Accuracy: 0.589286
 Feature Set: [2 3 5 15], Accuracy: 0.948980, Precision: 0.681818, Recall: 0.642857, F1 Score: 0.888889
 Feature Set: [2 3 5 16], Mean Accuracy: 0.482143
 Feature Set: [2 3 5 17], Mean Accuracy: 0.357143
 Feature Set: [2 3 5 18], Mean Accuracy: 0.500000
 Feature Set: [2 3 5 19], Mean Accuracy: 0.553571
 Feature Set: [2 3 5 19], Accuracy: 0.928571, Precision: 0.590909, Recall: 0.500000, F1 Score: 0.952381
 Feature Set: [2 3 5 20], Mean Accuracy: 0.642857
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 Feature Set: [2 3 5 21], Mean Accuracy: 0.696429
 Feature Set: [2 3 5 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [2 3 5 22], Mean Accuracy: 0.642857
 Feature Set: [2 3 5 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [2 3 6 7], Mean Accuracy: 0.607143
 Feature Set: [2 3 6 7], Accuracy: 0.938776, Precision: 0.590909, Recall: 0.571429, F1 Score: 0.875000
 Feature Set: [2 3 6 8], Mean Accuracy: 0.785714
 Feature Set: [2 3 6 8], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [2 3 6 9], Mean Accuracy: 0.821429
 Feature Set: [2 3 6 9], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [2 3 6 10], Mean Accuracy: 0.678571
 Feature Set: [2 3 6 10], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [2 3 6 11], Mean Accuracy: 0.857143
 Feature Set: [2 3 6 11], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [2 3 6 12], Mean Accuracy: 0.571429
 Feature Set: [2 3 6 12], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 0.880952
 Feature Set: [2 3 6 13], Mean Accuracy: 0.410714
 Feature Set: [2 3 6 14], Mean Accuracy: 0.392857
 Feature Set: [2 3 6 15], Mean Accuracy: 0.535714
 Feature Set: [2 3 6 15], Accuracy: 0.928571, Precision: 0.566667, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [2 3 6 16], Mean Accuracy: 0.482143
 Feature Set: [2 3 6 17], Mean Accuracy: 0.446429
 Feature Set: [2 3 6 18], Mean Accuracy: 0.482143
 Feature Set: [2 3 6 19], Mean Accuracy: 0.553571
 Feature Set: [2 3 6 19], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
 Feature Set: [2 3 6 20], Mean Accuracy: 0.625000

Feature Set: [2 3 6 20], Accuracy: 0.918367, Precision: 0.444444, Recall: 0.428571, F1 Score: 0.916667
 Feature Set: [2 3 6 21], Mean Accuracy: 0.678571
 Feature Set: [2 3 6 21], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [2 3 6 22], Mean Accuracy: 0.607143
 Feature Set: [2 3 6 22], Accuracy: 0.948980, Precision: 0.783333, Recall: 0.642857, F1 Score: 0.907407
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 Feature Set: [2 3 7 8], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [2 3 7 9], Mean Accuracy: 0.839286
 Feature Set: [2 3 7 9], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [2 3 7 10], Mean Accuracy: 0.732143
 Feature Set: [2 3 7 10], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [2 3 7 11], Mean Accuracy: 0.875000
 Feature Set: [2 3 7 11], Accuracy: 0.969388, Precision: 0.875000, Recall: 0.785714, F1 Score: 0.969697
 Feature Set: [2 3 7 12], Mean Accuracy: 0.589286
 Feature Set: [2 3 7 12], Accuracy: 0.938776, Precision: 0.636364, Recall: 0.571429, F1 Score: 0.916667
 Feature Set: [2 3 7 13], Mean Accuracy: 0.500000
 Feature Set: [2 3 7 14], Mean Accuracy: 0.500000
 Feature Set: [2 3 7 15], Mean Accuracy: 0.607143
 Feature Set: [2 3 7 15], Accuracy: 0.938776, Precision: 0.621212, Recall: 0.571429, F1 Score: 0.895833
 Feature Set: [2 3 7 16], Mean Accuracy: 0.482143
 Feature Set: [2 3 7 17], Mean Accuracy: 0.517857
 Feature Set: [2 3 7 17], Accuracy: 0.918367, Precision: 0.533333, Recall: 0.428571, F1 Score: 0.916667
 Feature Set: [2 3 7 18], Mean Accuracy: 0.553571
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 Feature Set: [2 3 7 19], Mean Accuracy: 0.642857
 Feature Set: [2 3 7 19], Accuracy: 0.938776, Precision: 0.648148, Recall: 0.571429, F1 Score: 0.812500
 Feature Set: [2 3 7 20], Mean Accuracy: 0.714286
 Feature Set: [2 3 7 20], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [2 3 7 21], Mean Accuracy: 0.714286
 Feature Set: [2 3 7 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [2 3 7 22], Mean Accuracy: 0.696429
 Feature Set: [2 3 7 22], Accuracy: 0.959184, Precision: 0.848485, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [2 3 8 9], Mean Accuracy: 0.892857
 Feature Set: [2 3 8 9], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 8 10], Mean Accuracy: 0.875000
 Feature Set: [2 3 8 10], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [2 3 8 11], Mean Accuracy: 0.946429
 Feature Set: [2 3 8 11], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
 Feature Set: [2 3 8 12], Mean Accuracy: 0.750000
 Feature Set: [2 3 8 12], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [2 3 8 13], Mean Accuracy: 0.750000
 Feature Set: [2 3 8 13], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [2 3 8 14], Mean Accuracy: 0.785714
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 Feature Set: [2 3 8 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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 Feature Set: [2 3 8 18], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 8 19], Mean Accuracy: 0.696429
 Feature Set: [2 3 8 19], Accuracy: 0.959184, Precision: 0.800000, Recall: 0.714286, F1 Score: 0.866667
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 Feature Set: [2 3 8 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [2 3 8 21], Mean Accuracy: 0.785714
 Feature Set: [2 3 8 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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 Feature Set: [2 3 8 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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 Feature Set: [2 3 9 10], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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 Feature Set: [2 3 9 13], Mean Accuracy: 0.750000
 Feature Set: [2 3 9 13], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091

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 Feature Set: [2 6 10 19], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [2 6 10 20], Mean Accuracy: 0.892857
 Feature Set: [2 6 10 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [2 6 10 21], Mean Accuracy: 0.750000
 Feature Set: [2 6 10 21], Accuracy: 0.948980, Precision: 0.708333, Recall: 0.642857, F1 Score: 0.962963
 Feature Set: [2 6 10 22], Mean Accuracy: 0.839286
 Feature Set: [2 6 10 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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 Feature Set: [2 6 11 13], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
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 Feature Set: [2 6 11 14], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
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 Feature Set: [2 6 11 15], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [2 6 11 16], Mean Accuracy: 0.892857
 Feature Set: [2 6 11 16], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [2 6 11 17], Mean Accuracy: 0.875000
 Feature Set: [2 6 11 17], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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 Feature Set: [2 6 11 18], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
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 Feature Set: [2 6 11 20], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
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 Feature Set: [2 6 11 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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 Feature Set: [2 6 12 18], Mean Accuracy: 0.535714
 Feature Set: [2 6 12 18], Accuracy: 0.938776, Precision: 0.600000, Recall: 0.571429, F1 Score: 0.833333
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 Feature Set: [2 6 12 20], Accuracy: 0.928571, Precision: 0.530303, Recall: 0.500000, F1 Score: 0.880952
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 Feature Set: [2 6 13 15], Mean Accuracy: 0.625000
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 Feature Set: [2 6 13 19], Accuracy: 0.928571, Precision: 0.530303, Recall: 0.500000, F1 Score: 0.880952
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 Feature Set: [2 6 13 22], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
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 Feature Set: [2 6 14 17], Mean Accuracy: 0.428571

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 Feature Set: [2 6 14 21], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
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 Feature Set: [2 6 14 22], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
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 Feature Set: [2 6 15 20], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
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 Feature Set: [2 6 15 21], Accuracy: 0.938776, Precision: 0.600000, Recall: 0.571429, F1 Score: 0.833333
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 Feature Set: [2 6 16 19], Mean Accuracy: 0.607143
 Feature Set: [2 6 16 19], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
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 Feature Set: [2 6 16 22], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
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 Feature Set: [2 6 17 18], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
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 Feature Set: [2 6 18 22], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
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 Feature Set: [2 6 21 22], Accuracy: 0.938776, Precision: 0.633333, Recall: 0.571429, F1 Score: 0.854167
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Feature Set: [2 7 15 17], Accuracy: 0.938776, Precision: 0.621212, Recall: 0.571429, F1 Score: 0.895833
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Feature Set: [2 7 15 19], Accuracy: 0.938776, Precision: 0.633333, Recall: 0.571429, F1 Score: 0.854167
Feature Set: [2 7 15 20], Mean Accuracy: 0.714286
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Feature Set: [2 7 17 20], Accuracy: 0.928571, Precision: 0.604167, Recall: 0.500000, F1 Score: 0.785714
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Feature Set: [2 8 9 11], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
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Feature Set: [2 8 9 13], Accuracy: 0.969388, Precision: 0.846154, Recall: 0.785714, F1 Score: 1.000000

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F1 Score: 0.974359
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F1 Score: 0.974359
Feature Set: [2 8 9 18], Mean Accuracy: 0.857143
F1 Score: 0.974359
Feature Set: [2 8 9 19], Mean Accuracy: 0.857143
F1 Score: 0.958333
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F1 Score: 1.000000
Feature Set: [2 8 9 21], Mean Accuracy: 0.892857
F1 Score: 1.000000
Feature Set: [2 8 9 22], Mean Accuracy: 0.946429
F1 Score: 1.000000
Feature Set: [2 8 10 11], Mean Accuracy: 0.964286
F1 Score: 0.974359
Feature Set: [2 8 10 12], Mean Accuracy: 0.857143
F1 Score: 0.969697
Feature Set: [2 8 10 13], Mean Accuracy: 0.839286
F1 Score: 0.933333
Feature Set: [2 8 10 14], Mean Accuracy: 0.857143
F1 Score: 0.969697
Feature Set: [2 8 10 15], Mean Accuracy: 0.875000
F1 Score: 0.944444
Feature Set: [2 8 10 16], Mean Accuracy: 0.892857
F1 Score: 0.974359
Feature Set: [2 8 10 17], Mean Accuracy: 0.839286
F1 Score: 0.944444
Feature Set: [2 8 10 18], Mean Accuracy: 0.839286
F1 Score: 0.944444
Feature Set: [2 8 10 19], Mean Accuracy: 0.803571
F1 Score: 0.939394
Feature Set: [2 8 10 20], Mean Accuracy: 0.946429
F1 Score: 1.000000
Feature Set: [2 8 10 21], Mean Accuracy: 0.928571
F1 Score: 1.000000
Feature Set: [2 8 10 22], Mean Accuracy: 0.821429
F1 Score: 0.969697
Feature Set: [2 8 11 12], Mean Accuracy: 0.928571
F1 Score: 0.974359
Feature Set: [2 8 11 13], Mean Accuracy: 0.982143
F1 Score: 1.000000
Feature Set: [2 8 11 14], Mean Accuracy: 0.946429
F1 Score: 1.000000
Feature Set: [2 8 11 15], Mean Accuracy: 0.946429
F1 Score: 0.974359
Feature Set: [2 8 11 16], Mean Accuracy: 0.982143
F1 Score: 1.000000
Feature Set: [2 8 11 17], Mean Accuracy: 0.928571
F1 Score: 1.000000
Feature Set: [2 8 11 18], Mean Accuracy: 0.946429
F1 Score: 1.000000
Feature Set: [2 8 11 19], Mean Accuracy: 0.910714
F1 Score: 0.944444
Feature Set: [2 8 11 20], Mean Accuracy: 0.982143
F1 Score: 1.000000
Feature Set: [2 8 11 21], Mean Accuracy: 0.964286
F1 Score: 1.000000
Feature Set: [2 8 11 22], Mean Accuracy: 0.946429
F1 Score: 1.000000
Feature Set: [2 8 12 13], Mean Accuracy: 0.714286
F1 Score: 0.925926
Feature Set: [2 8 12 14], Mean Accuracy: 0.767857

Feature Set: [2 9 11 13], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 9 11 14], Mean Accuracy: 0.946429
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Feature Set: [2 9 11 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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Feature Set: [2 9 11 16], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 9 11 17], Mean Accuracy: 0.928571
Feature Set: [2 9 11 17], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 9 11 18], Mean Accuracy: 0.892857
Feature Set: [2 9 11 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 9 11 19], Mean Accuracy: 0.892857
Feature Set: [2 9 11 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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Feature Set: [2 9 11 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 9 11 22], Mean Accuracy: 0.910714
Feature Set: [2 9 11 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
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Feature Set: [2 9 12 14], Mean Accuracy: 0.803571
Feature Set: [2 9 12 14], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [2 9 12 15], Mean Accuracy: 0.857143
Feature Set: [2 9 12 15], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
Feature Set: [2 9 12 16], Mean Accuracy: 0.803571
Feature Set: [2 9 12 16], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [2 9 12 17], Mean Accuracy: 0.839286
Feature Set: [2 9 12 17], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [2 9 12 18], Mean Accuracy: 0.839286
Feature Set: [2 9 12 18], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [2 9 12 19], Mean Accuracy: 0.767857
Feature Set: [2 9 12 19], Accuracy: 0.959184, Precision: 0.875000, Recall: 0.714286, F1 Score: 0.906667
Feature Set: [2 9 12 20], Mean Accuracy: 0.928571
Feature Set: [2 9 12 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 9 12 21], Mean Accuracy: 0.821429
Feature Set: [2 9 12 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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Feature Set: [2 9 12 22], Accuracy: 0.959184, Precision: 0.800000, Recall: 0.714286, F1 Score: 0.866667
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Feature Set: [2 9 13 15], Mean Accuracy: 0.875000
Feature Set: [2 9 13 15], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [2 9 13 16], Mean Accuracy: 0.785714
Feature Set: [2 9 13 16], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [2 9 13 17], Mean Accuracy: 0.839286
Feature Set: [2 9 13 17], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [2 9 13 18], Mean Accuracy: 0.821429
Feature Set: [2 9 13 18], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [2 9 13 19], Mean Accuracy: 0.732143
Feature Set: [2 9 13 19], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
Feature Set: [2 9 13 20], Mean Accuracy: 0.910714
Feature Set: [2 9 13 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 9 13 21], Mean Accuracy: 0.892857
Feature Set: [2 9 13 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 9 13 22], Mean Accuracy: 0.803571
Feature Set: [2 9 13 22], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [2 9 14 15], Mean Accuracy: 0.892857
Feature Set: [2 9 14 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 9 14 16], Mean Accuracy: 0.839286
Feature Set: [2 9 14 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 9 14 17], Mean Accuracy: 0.857143
Feature Set: [2 9 14 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 9 14 18], Mean Accuracy: 0.857143
Feature Set: [2 9 14 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 9 14 19], Mean Accuracy: 0.821429
Feature Set: [2 9 14 19], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333

[illegible]

[illegible]

Feature Set: [2 11 16 20], Mean Accuracy: 0.910714
Feature Set: [2 11 16 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 11 16 21], Mean Accuracy: 0.982143
Feature Set: [2 11 16 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 11 16 22], Mean Accuracy: 0.875000
Feature Set: [2 11 16 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 11 17 18], Mean Accuracy: 0.910714
Feature Set: [2 11 17 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 11 17 19], Mean Accuracy: 0.875000
Feature Set: [2 11 17 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 11 17 20], Mean Accuracy: 0.928571
Feature Set: [2 11 17 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 11 17 21], Mean Accuracy: 0.946429
Feature Set: [2 11 17 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 11 17 22], Mean Accuracy: 0.910714
Feature Set: [2 11 17 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
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Feature Set: [2 11 18 19], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [2 11 18 20], Mean Accuracy: 0.892857
Feature Set: [2 11 18 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 11 18 21], Mean Accuracy: 0.964286
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Feature Set: [2 11 18 22], Mean Accuracy: 0.910714
Feature Set: [2 11 18 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
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Feature Set: [2 11 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
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Feature Set: [2 11 19 22], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
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Feature Set: [2 11 20 22], Mean Accuracy: 0.892857
Feature Set: [2 11 20 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [2 11 21 22], Mean Accuracy: 0.946429
Feature Set: [2 11 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [2 12 13 14], Mean Accuracy: 0.553571
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Feature Set: [2 12 13 18], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 0.819048
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Feature Set: [2 12 13 20], Mean Accuracy: 0.607143
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Feature Set: [2 12 13 22], Mean Accuracy: 0.642857
Feature Set: [2 12 13 22], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
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Feature Set: [2 12 14 18], Mean Accuracy: 0.535714
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Feature Set: [2 12 14 19], Mean Accuracy: 0.625000
Feature Set: [2 12 14 19], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 0.819048
Feature Set: [2 12 14 20], Mean Accuracy: 0.714286
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Feature Set: [2 12 14 21], Mean Accuracy: 0.732143
Feature Set: [2 12 14 21], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [2 12 14 22], Mean Accuracy: 0.696429

Feature Set: [2 12 14 22], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [2 12 15 16], Mean Accuracy: 0.553571
 Feature Set: [2 12 15 16], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 0.819048
 Feature Set: [2 12 15 17], Mean Accuracy: 0.607143
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 Feature Set: [2 12 15 18], Mean Accuracy: 0.517857
 Feature Set: [2 12 15 18], Accuracy: 0.938776, Precision: 0.675000, Recall: 0.571429, F1 Score: 0.883333
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 Feature Set: [2 12 15 19], Accuracy: 0.928571, Precision: 0.712500, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [2 12 15 20], Mean Accuracy: 0.714286
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 Feature Set: [2 12 16 18], Accuracy: 0.948980, Precision: 0.775000, Recall: 0.642857, F1 Score: 0.896296
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 Feature Set: [2 12 16 22], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
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 Feature Set: [2 12 17 18], Accuracy: 0.938776, Precision: 0.675000, Recall: 0.571429, F1 Score: 0.883333
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 Feature Set: [2 13 14 22], Accuracy: 0.928571, Precision: 0.555556, Recall: 0.500000, F1 Score: 0.809524
 Feature Set: [2 13 15 16], Mean Accuracy: 0.589286
 Feature Set: [2 13 15 16], Accuracy: 0.938776, Precision: 0.625000, Recall: 0.571429, F1 Score: 0.958333
 Feature Set: [2 13 15 17], Mean Accuracy: 0.589286
 Feature Set: [2 13 15 17], Accuracy: 0.928571, Precision: 0.550000, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [2 13 15 18], Mean Accuracy: 0.571429
 Feature Set: [2 13 15 18], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [2 13 15 19], Mean Accuracy: 0.714286
 Feature Set: [2 13 15 19], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
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 Feature Set: [2 13 17 18], Accuracy: 0.918367, Precision: 0.500000, Recall: 0.428571, F1 Score: 0.833333
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 Feature Set: [2 13 17 20], Mean Accuracy: 0.642857
 Feature Set: [2 13 17 20], Accuracy: 0.897959, Precision: 0.287037, Recall: 0.285714, F1 Score: 0.725000
 Feature Set: [2 13 17 21], Mean Accuracy: 0.714286
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 Feature Set: [2 13 17 22], Mean Accuracy: 0.571429
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 Feature Set: [2 13 18 20], Accuracy: 0.928571, Precision: 0.574074, Recall: 0.500000, F1 Score: 0.809524
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 Feature Set: [2 13 18 22], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
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 Feature Set: [2 13 19 22], Accuracy: 0.938776, Precision: 0.683333, Recall: 0.571429, F1 Score: 0.895833
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 Feature Set: [2 13 20 22], Mean Accuracy: 0.767857
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 Feature Set: [3 4 16 19], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
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 Feature Set: [3 4 16 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [3 4 17 18], Mean Accuracy: 0.589286
 Feature Set: [3 4 17 18], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [3 4 17 19], Mean Accuracy: 0.696429
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 Feature Set: [3 4 18 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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 Feature Set: [3 4 19 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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 Feature Set: [3 4 19 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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 Feature Set: [3 5 6 15], Mean Accuracy: 0.553571
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Feature Set: [3 5 6 21], Accuracy: 0.948980, Precision: 0.796296, Recall: 0.642857, F1 Score: 0.851852
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 Feature Set: [3 5 8 16], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
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 Feature Set: [3 5 8 18], Mean Accuracy: 0.714286
 Feature Set: [3 5 8 18], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
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 Feature Set: [3 5 9 12], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
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 Feature Set: [3 5 9 15], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [3 5 9 16], Mean Accuracy: 0.803571

Feature Set: [3 5 9 16], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
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 Feature Set: [3 5 11 15], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
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 Feature Set: [3 5 14 19], Mean Accuracy: 0.553571
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 Feature Set: [3 7 13 16], Mean Accuracy: 0.357143
 Feature Set: [3 7 13 17], Mean Accuracy: 0.357143
 Feature Set: [3 7 13 18], Mean Accuracy: 0.410714
 Feature Set: [3 7 13 19], Mean Accuracy: 0.607143
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 Feature Set: [3 7 13 20], Accuracy: 0.928571, Precision: 0.537037, Recall: 0.500000, F1 Score: 0.785714
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 Feature Set: [3 7 14 17], Mean Accuracy: 0.446429
 Feature Set: [3 7 14 18], Mean Accuracy: 0.428571
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 Feature Set: [3 7 14 21], Mean Accuracy: 0.678571
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 Feature Set: [3 7 14 22], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
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Feature Set: [3 8 15 17], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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Feature Set: [3 8 16 18], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
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Feature Set: [3 8 16 19], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [3 8 16 20], Mean Accuracy: 0.767857
Feature Set: [3 8 16 20], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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Feature Set: [3 8 17 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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Feature Set: [3 8 17 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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Feature Set: [3 8 21 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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[illegible]

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Feature Set: [3 12 14 18], Mean Accuracy: 0.464286
Feature Set: [3 12 14 19], Mean Accuracy: 0.571429
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Feature Set: [4 8 15 16], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [4 8 15 17], Mean Accuracy: 0.750000
Feature Set: [4 8 15 17], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [4 8 15 18], Mean Accuracy: 0.714286
Feature Set: [4 8 15 18], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [4 8 15 19], Mean Accuracy: 0.839286
Feature Set: [4 8 15 19], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [4 8 15 20], Mean Accuracy: 0.785714
Feature Set: [4 8 15 20], Accuracy: 0.959184, Precision: 0.818182, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [4 8 15 21], Mean Accuracy: 0.875000
Feature Set: [4 8 15 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 8 15 22], Mean Accuracy: 0.875000
Feature Set: [4 8 15 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 8 16 17], Mean Accuracy: 0.714286
Feature Set: [4 8 16 17], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [4 8 16 18], Mean Accuracy: 0.678571
Feature Set: [4 8 16 18], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [4 8 16 19], Mean Accuracy: 0.785714

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Feature Set: [4 9 11 15], Mean Accuracy: 0.928571
Feature Set: [4 9 11 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 11 16], Mean Accuracy: 0.964286
Feature Set: [4 9 11 16], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 9 11 17], Mean Accuracy: 0.964286
Feature Set: [4 9 11 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 11 18], Mean Accuracy: 0.928571
Feature Set: [4 9 11 18], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 11 19], Mean Accuracy: 0.928571
Feature Set: [4 9 11 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 11 20], Mean Accuracy: 0.964286
Feature Set: [4 9 11 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 11 21], Mean Accuracy: 0.982143
Feature Set: [4 9 11 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 11 22], Mean Accuracy: 0.946429
Feature Set: [4 9 11 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 9 12 13], Mean Accuracy: 0.803571
Feature Set: [4 9 12 13], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
Feature Set: [4 9 12 14], Mean Accuracy: 0.892857
Feature Set: [4 9 12 14], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 9 12 15], Mean Accuracy: 0.839286
Feature Set: [4 9 12 15], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [4 9 12 16], Mean Accuracy: 0.857143
Feature Set: [4 9 12 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 12 17], Mean Accuracy: 0.839286
Feature Set: [4 9 12 17], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [4 9 12 18], Mean Accuracy: 0.857143
Feature Set: [4 9 12 18], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [4 9 12 19], Mean Accuracy: 0.875000
Feature Set: [4 9 12 19], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 9 12 20], Mean Accuracy: 0.928571
Feature Set: [4 9 12 20], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [4 9 12 21], Mean Accuracy: 0.839286
Feature Set: [4 9 12 21], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [4 9 12 22], Mean Accuracy: 0.857143
Feature Set: [4 9 12 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [4 9 13 14], Mean Accuracy: 0.857143
Feature Set: [4 9 13 14], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
Feature Set: [4 9 13 15], Mean Accuracy: 0.892857
Feature Set: [4 9 13 15], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.912222
Feature Set: [4 9 13 16], Mean Accuracy: 0.839286
Feature Set: [4 9 13 16], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 13 17], Mean Accuracy: 0.875000
Feature Set: [4 9 13 17], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [4 9 13 18], Mean Accuracy: 0.857143
Feature Set: [4 9 13 18], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [4 9 13 19], Mean Accuracy: 0.875000
Feature Set: [4 9 13 19], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.912222
Feature Set: [4 9 13 20], Mean Accuracy: 0.928571
Feature Set: [4 9 13 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [4 9 13 21], Mean Accuracy: 0.928571
Feature Set: [4 9 13 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 13 22], Mean Accuracy: 0.928571
Feature Set: [4 9 13 22], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [4 9 14 15], Mean Accuracy: 0.910714
Feature Set: [4 9 14 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 14 16], Mean Accuracy: 0.839286
Feature Set: [4 9 14 16], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [4 9 14 17], Mean Accuracy: 0.857143
Feature Set: [4 9 14 17], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 14 18], Mean Accuracy: 0.857143
Feature Set: [4 9 14 18], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 9 14 19], Mean Accuracy: 0.892857
Feature Set: [4 9 14 19], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [4 9 14 20], Mean Accuracy: 0.964286
Feature Set: [4 9 14 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 9 14 21], Mean Accuracy: 0.910714

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Feature Set: [4 10 11 16], Mean Accuracy: 0.946429
Feature Set: [4 10 11 16], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 10 11 17], Mean Accuracy: 0.946429
Feature Set: [4 10 11 17], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 10 11 18], Mean Accuracy: 0.946429
Feature Set: [4 10 11 18], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 10 11 19], Mean Accuracy: 0.928571
Feature Set: [4 10 11 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 10 11 20], Mean Accuracy: 0.982143
Feature Set: [4 10 11 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 10 11 21], Mean Accuracy: 1.000000
Feature Set: [4 10 11 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 10 11 22], Mean Accuracy: 0.964286
Feature Set: [4 10 11 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [4 10 12 13], Mean Accuracy: 0.803571
Feature Set: [4 10 12 13], Accuracy: 0.948980, Precision: 0.681818, Recall: 0.642857, F1 Score: 0.888889
Feature Set: [4 10 12 14], Mean Accuracy: 0.839286
Feature Set: [4 10 12 14], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 12 15], Mean Accuracy: 0.875000
Feature Set: [4 10 12 15], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
Feature Set: [4 10 12 16], Mean Accuracy: 0.821429
Feature Set: [4 10 12 16], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
Feature Set: [4 10 12 17], Mean Accuracy: 0.839286
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Feature Set: [4 10 12 18], Mean Accuracy: 0.857143
Feature Set: [4 10 12 18], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [4 10 12 19], Mean Accuracy: 0.857143
Feature Set: [4 10 12 19], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [4 10 12 20], Mean Accuracy: 0.928571
Feature Set: [4 10 12 20], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 12 21], Mean Accuracy: 0.803571
Feature Set: [4 10 12 21], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [4 10 12 22], Mean Accuracy: 0.839286
Feature Set: [4 10 12 22], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [4 10 13 14], Mean Accuracy: 0.803571
Feature Set: [4 10 13 14], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [4 10 13 15], Mean Accuracy: 0.857143
Feature Set: [4 10 13 15], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 13 16], Mean Accuracy: 0.750000
Feature Set: [4 10 13 16], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
Feature Set: [4 10 13 17], Mean Accuracy: 0.803571
Feature Set: [4 10 13 17], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [4 10 13 18], Mean Accuracy: 0.803571
Feature Set: [4 10 13 18], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
Feature Set: [4 10 13 19], Mean Accuracy: 0.910714
Feature Set: [4 10 13 19], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 13 20], Mean Accuracy: 0.910714
Feature Set: [4 10 13 20], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 13 21], Mean Accuracy: 0.875000
Feature Set: [4 10 13 21], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [4 10 13 22], Mean Accuracy: 0.839286
Feature Set: [4 10 13 22], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [4 10 14 15], Mean Accuracy: 0.875000
Feature Set: [4 10 14 15], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 10 14 16], Mean Accuracy: 0.839286
Feature Set: [4 10 14 16], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 14 17], Mean Accuracy: 0.821429
Feature Set: [4 10 14 17], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 14 18], Mean Accuracy: 0.839286
Feature Set: [4 10 14 18], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 14 19], Mean Accuracy: 0.875000
Feature Set: [4 10 14 19], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 14 20], Mean Accuracy: 0.928571
Feature Set: [4 10 14 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [4 10 14 21], Mean Accuracy: 0.857143
Feature Set: [4 10 14 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [4 10 14 22], Mean Accuracy: 0.875000

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Feature Set: [5 6 8 20], Mean Accuracy: 0.732143
 Feature Set: [5 6 8 20], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [5 6 8 21], Mean Accuracy: 0.821429
 Feature Set: [5 6 8 21], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [5 6 8 22], Mean Accuracy: 0.785714
 Feature Set: [5 6 8 22], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: [5 6 9 10], Mean Accuracy: 0.910714
 Feature Set: [5 6 9 10], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [5 6 9 11], Mean Accuracy: 0.964286
 Feature Set: [5 6 9 11], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [5 6 9 12], Mean Accuracy: 0.714286
 Feature Set: [5 6 9 12], Accuracy: 0.948980, Precision: 0.783333, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [5 6 9 13], Mean Accuracy: 0.750000
 Feature Set: [5 6 9 13], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [5 6 9 14], Mean Accuracy: 0.714286
 Feature Set: [5 6 9 14], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [5 6 9 15], Mean Accuracy: 0.732143
 Feature Set: [5 6 9 15], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [5 6 9 16], Mean Accuracy: 0.714286
 Feature Set: [5 6 9 16], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [5 6 9 17], Mean Accuracy: 0.732143
 Feature Set: [5 6 9 17], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [5 6 9 18], Mean Accuracy: 0.803571
 Feature Set: [5 6 9 18], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [5 6 9 19], Mean Accuracy: 0.732143
 Feature Set: [5 6 9 19], Accuracy: 0.959184, Precision: 0.848485, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [5 6 9 20], Mean Accuracy: 0.803571
 Feature Set: [5 6 9 20], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: [5 6 9 21], Mean Accuracy: 0.821429
 Feature Set: [5 6 9 21], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
 Feature Set: [5 6 9 22], Mean Accuracy: 0.767857
 Feature Set: [5 6 9 22], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
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 Feature Set: [5 6 10 12], Accuracy: 0.928571, Precision: 0.575758, Recall: 0.500000, F1 Score: 0.928571
 Feature Set: [5 6 10 13], Mean Accuracy: 0.696429
 Feature Set: [5 6 10 13], Accuracy: 0.928571, Precision: 0.575758, Recall: 0.500000, F1 Score: 0.928571
 Feature Set: [5 6 10 14], Mean Accuracy: 0.660714
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 Feature Set: [5 6 10 16], Mean Accuracy: 0.642857
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 Feature Set: [5 6 10 17], Mean Accuracy: 0.642857
 Feature Set: [5 6 10 17], Accuracy: 0.938776, Precision: 0.636364, Recall: 0.571429, F1 Score: 0.916667
 Feature Set: [5 6 10 18], Mean Accuracy: 0.660714
 Feature Set: [5 6 10 18], Accuracy: 0.938776, Precision: 0.625000, Recall: 0.571429, F1 Score: 0.958333
 Feature Set: [5 6 10 19], Mean Accuracy: 0.750000
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 Feature Set: [5 6 10 20], Mean Accuracy: 0.821429
 Feature Set: [5 6 10 20], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [5 6 10 21], Mean Accuracy: 0.750000
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 Feature Set: [5 6 10 22], Mean Accuracy: 0.767857
 Feature Set: [5 6 10 22], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [5 6 11 12], Mean Accuracy: 0.821429
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 Feature Set: [5 6 11 13], Mean Accuracy: 0.839286
 Feature Set: [5 6 11 13], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [5 6 11 14], Mean Accuracy: 0.803571
 Feature Set: [5 6 11 14], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [5 6 11 15], Mean Accuracy: 0.785714
 Feature Set: [5 6 11 15], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [5 6 11 16], Mean Accuracy: 0.803571
 Feature Set: [5 6 11 16], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [5 6 11 17], Mean Accuracy: 0.821429

Feature Set: [5 6 11 17], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: [5 6 11 18], Mean Accuracy: 0.732143
 Feature Set: [5 6 11 18], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [5 6 11 19], Mean Accuracy: 0.839286
 Feature Set: [5 6 11 19], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [5 6 11 20], Mean Accuracy: 0.803571
 Feature Set: [5 6 11 20], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [5 6 11 21], Mean Accuracy: 0.910714
 Feature Set: [5 6 11 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [5 6 11 22], Mean Accuracy: 0.857143
 Feature Set: [5 6 11 22], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [5 6 12 13], Mean Accuracy: 0.482143
 Feature Set: [5 6 12 14], Mean Accuracy: 0.303571
 Feature Set: [5 6 12 15], Mean Accuracy: 0.482143
 Feature Set: [5 6 12 16], Mean Accuracy: 0.339286
 Feature Set: [5 6 12 17], Mean Accuracy: 0.464286
 Feature Set: [5 6 12 18], Mean Accuracy: 0.392857
 Feature Set: [5 6 12 19], Mean Accuracy: 0.482143
 Feature Set: [5 6 12 20], Mean Accuracy: 0.607143
 Feature Set: [5 6 12 20], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
 Feature Set: [5 6 12 21], Mean Accuracy: 0.625000
 Feature Set: [5 6 12 21], Accuracy: 0.938776, Precision: 0.685185, Recall: 0.571429, F1 Score: 0.833333
 Feature Set: [5 6 12 22], Mean Accuracy: 0.589286
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 Feature Set: [5 6 13 16], Mean Accuracy: 0.339286
 Feature Set: [5 6 13 17], Mean Accuracy: 0.428571
 Feature Set: [5 6 13 18], Mean Accuracy: 0.410714
 Feature Set: [5 6 13 19], Mean Accuracy: 0.464286
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 Feature Set: [5 6 14 16], Mean Accuracy: 0.375000
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 Feature Set: [5 6 14 18], Mean Accuracy: 0.321429
 Feature Set: [5 6 14 19], Mean Accuracy: 0.392857
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 Feature Set: [6 12 18 22], Mean Accuracy: 0.660714
 Feature Set: [6 12 18 22], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [6 12 19 20], Mean Accuracy: 0.642857
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 Feature Set: [6 12 21 22], Accuracy: 0.938776, Precision: 0.685185, Recall: 0.571429, F1 Score: 0.833333
 Feature Set: [6 13 14 15], Mean Accuracy: 0.553571
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 Feature Set: [6 13 14 17], Mean Accuracy: 0.392857
 Feature Set: [6 13 14 18], Mean Accuracy: 0.428571
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 Feature Set: [6 13 14 19], Accuracy: 0.908163, Precision: 0.500000, Recall: 0.357143, F1 Score: 0.933333
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 Feature Set: [6 13 18 22], Accuracy: 0.938776, Precision: 0.621212, Recall: 0.571429, F1 Score: 0.895833
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 Feature Set: [6 14 15 19], Accuracy: 0.918367, Precision: 0.550000, Recall: 0.428571, F1 Score: 0.944444
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 Feature Set: [6 19 21 22], Accuracy: 0.938776, Precision: 0.683333, Recall: 0.571429, F1 Score: 0.895833
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 Feature Set: [7 10 15 20], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
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 Feature Set: [7 10 15 21], Accuracy: 0.938776, Precision: 0.675000, Recall: 0.571429, F1 Score: 0.883333
 Feature Set: [7 10 15 22], Mean Accuracy: 0.839286
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Feature Set: [7 16 18 22], Accuracy: 0.938776, Precision: 0.648148, Recall: 0.571429, F1 Score: 0.812500
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Feature Set: [7 16 20 21], Accuracy: 0.959184, Precision: 0.875000, Recall: 0.714286, F1 Score: 0.906667
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Feature Set: [8 9 10 12], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [8 9 10 13], Mean Accuracy: 1.000000
Feature Set: [8 9 10 13], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [8 9 10 14], Mean Accuracy: 0.982143
Feature Set: [8 9 10 14], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [8 9 10 15], Mean Accuracy: 0.982143

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Feature Set: [8 12 13 20], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [8 12 13 21], Mean Accuracy: 0.839286
Feature Set: [8 12 13 21], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [8 12 13 22], Mean Accuracy: 0.839286
Feature Set: [8 12 13 22], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [8 12 14 15], Mean Accuracy: 0.732143
Feature Set: [8 12 14 15], Accuracy: 0.938776, Precision: 0.625000, Recall: 0.571429, F1 Score: 0.958333
Feature Set: [8 12 14 16], Mean Accuracy: 0.696429
Feature Set: [8 12 14 16], Accuracy: 0.938776, Precision: 0.703704, Recall: 0.571429, F1 Score: 0.854167
Feature Set: [8 12 14 17], Mean Accuracy: 0.696429
Feature Set: [8 12 14 17], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
Feature Set: [8 12 14 18], Mean Accuracy: 0.607143
Feature Set: [8 12 14 18], Accuracy: 0.948980, Precision: 0.653846, Recall: 0.642857, F1 Score: 0.962963
Feature Set: [8 12 14 19], Mean Accuracy: 0.696429
Feature Set: [8 12 14 19], Accuracy: 0.938776, Precision: 0.750000, Recall: 0.571429, F1 Score: 0.883333
Feature Set: [8 12 14 20], Mean Accuracy: 0.732143
Feature Set: [8 12 14 20], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
Feature Set: [8 12 14 21], Mean Accuracy: 0.857143
Feature Set: [8 12 14 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [8 12 14 22], Mean Accuracy: 0.803571
Feature Set: [8 12 14 22], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [8 12 15 16], Mean Accuracy: 0.714286
Feature Set: [8 12 15 16], Accuracy: 0.938776, Precision: 0.611111, Recall: 0.571429, F1 Score: 0.937500
Feature Set: [8 12 15 17], Mean Accuracy: 0.714286
Feature Set: [8 12 15 17], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
Feature Set: [8 12 15 18], Mean Accuracy: 0.607143
Feature Set: [8 12 15 18], Accuracy: 0.938776, Precision: 0.611111, Recall: 0.571429, F1 Score: 0.937500
Feature Set: [8 12 15 19], Mean Accuracy: 0.714286
Feature Set: [8 12 15 19], Accuracy: 0.928571, Precision: 0.600000, Recall: 0.500000, F1 Score: 0.904762
Feature Set: [8 12 15 20], Mean Accuracy: 0.767857
Feature Set: [8 12 15 20], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
Feature Set: [8 12 15 21], Mean Accuracy: 0.821429
Feature Set: [8 12 15 21], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [8 12 15 22], Mean Accuracy: 0.839286
Feature Set: [8 12 15 22], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [8 12 16 17], Mean Accuracy: 0.678571
Feature Set: [8 12 16 17], Accuracy: 0.938776, Precision: 0.666667, Recall: 0.571429, F1 Score: 0.937500
Feature Set: [8 12 16 18], Mean Accuracy: 0.660714
Feature Set: [8 12 16 18], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
Feature Set: [8 12 16 19], Mean Accuracy: 0.732143
Feature Set: [8 12 16 19], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 0.761905
Feature Set: [8 12 16 20], Mean Accuracy: 0.750000
Feature Set: [8 12 16 20], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [8 12 16 21], Mean Accuracy: 0.857143
Feature Set: [8 12 16 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [8 12 16 22], Mean Accuracy: 0.803571
Feature Set: [8 12 16 22], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [8 12 17 18], Mean Accuracy: 0.696429
Feature Set: [8 12 17 18], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
Feature Set: [8 12 17 19], Mean Accuracy: 0.625000
Feature Set: [8 12 17 19], Accuracy: 0.918367, Precision: 0.583333, Recall: 0.428571, F1 Score: 0.900000
Feature Set: [8 12 17 20], Mean Accuracy: 0.732143
Feature Set: [8 12 17 20], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
Feature Set: [8 12 17 21], Mean Accuracy: 0.767857
Feature Set: [8 12 17 21], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
Feature Set: [8 12 17 22], Mean Accuracy: 0.767857
Feature Set: [8 12 17 22], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
Feature Set: [8 12 18 19], Mean Accuracy: 0.696429
Feature Set: [8 12 18 19], Accuracy: 0.938776, Precision: 0.685185, Recall: 0.571429, F1 Score: 0.833333
Feature Set: [8 12 18 20], Mean Accuracy: 0.750000
Feature Set: [8 12 18 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [8 12 18 21], Mean Accuracy: 0.821429
Feature Set: [8 12 18 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [8 12 18 22], Mean Accuracy: 0.785714
Feature Set: [8 12 18 22], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
Feature Set: [8 12 19 20], Mean Accuracy: 0.785714
Feature Set: [8 12 19 20], Accuracy: 0.938776, Precision: 0.744444, Recall: 0.571429, F1 Score: 0.875000

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Feature Set: [9 11 17 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 11 17 22], Mean Accuracy: 0.946429
Feature Set: [9 11 17 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [9 11 18 19], Mean Accuracy: 0.857143
Feature Set: [9 11 18 19], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [9 11 18 20], Mean Accuracy: 0.910714
Feature Set: [9 11 18 20], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 11 18 21], Mean Accuracy: 0.928571
Feature Set: [9 11 18 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 11 18 22], Mean Accuracy: 0.928571
Feature Set: [9 11 18 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 11 19 20], Mean Accuracy: 0.892857
Feature Set: [9 11 19 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [9 11 19 21], Mean Accuracy: 0.982143
Feature Set: [9 11 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 11 19 22], Mean Accuracy: 0.910714
Feature Set: [9 11 19 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [9 11 20 21], Mean Accuracy: 0.982143
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Feature Set: [9 11 20 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 11 21 22], Mean Accuracy: 0.964286
Feature Set: [9 11 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [9 12 13 14], Mean Accuracy: 0.767857
Feature Set: [9 12 13 14], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [9 12 13 15], Mean Accuracy: 0.875000
Feature Set: [9 12 13 15], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [9 12 13 16], Mean Accuracy: 0.785714
Feature Set: [9 12 13 16], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [9 12 13 17], Mean Accuracy: 0.767857
Feature Set: [9 12 13 17], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [9 12 13 18], Mean Accuracy: 0.767857
Feature Set: [9 12 13 18], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [9 12 13 19], Mean Accuracy: 0.732143
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Feature Set: [9 12 13 20], Mean Accuracy: 0.928571
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Feature Set: [9 12 13 22], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [9 12 14 15], Mean Accuracy: 0.785714
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Feature Set: [9 12 14 18], Mean Accuracy: 0.821429
Feature Set: [9 12 14 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [9 12 14 19], Mean Accuracy: 0.732143
Feature Set: [9 12 14 19], Accuracy: 0.959184, Precision: 0.920000, Recall: 0.714286, F1 Score: 0.933333
Feature Set: [9 12 14 20], Mean Accuracy: 0.875000
Feature Set: [9 12 14 20], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
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Feature Set: [9 12 14 22], Mean Accuracy: 0.767857
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Feature Set: [9 12 15 17], Mean Accuracy: 0.767857
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Feature Set: [9 12 15 18], Mean Accuracy: 0.750000
Feature Set: [9 12 15 18], Accuracy: 0.959184, Precision: 0.791667, Recall: 0.714286, F1 Score: 0.966667
Feature Set: [9 12 15 19], Mean Accuracy: 0.767857
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Feature Set: [9 14 16 18], Mean Accuracy: 0.767857
 Feature Set: [9 14 16 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [9 14 16 19], Mean Accuracy: 0.732143
 Feature Set: [9 14 16 19], Accuracy: 0.959184, Precision: 0.875000, Recall: 0.714286, F1 Score: 0.906667
 Feature Set: [9 14 16 20], Mean Accuracy: 0.857143
 Feature Set: [9 14 16 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 14 16 21], Mean Accuracy: 0.839286
 Feature Set: [9 14 16 21], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [9 14 16 22], Mean Accuracy: 0.785714
 Feature Set: [9 14 16 22], Accuracy: 0.969388, Precision: 0.863636, Recall: 0.785714, F1 Score: 0.909091
 Feature Set: [9 14 17 18], Mean Accuracy: 0.767857
 Feature Set: [9 14 17 18], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [9 14 17 19], Mean Accuracy: 0.750000
 Feature Set: [9 14 17 19], Accuracy: 0.969388, Precision: 0.931818, Recall: 0.785714, F1 Score: 0.945455
 Feature Set: [9 14 17 20], Mean Accuracy: 0.910714
 Feature Set: [9 14 17 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 14 17 21], Mean Accuracy: 0.839286
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 Feature Set: [9 14 17 22], Mean Accuracy: 0.857143
 Feature Set: [9 14 17 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 14 18 19], Mean Accuracy: 0.767857
 Feature Set: [9 14 18 19], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [9 14 18 20], Mean Accuracy: 0.892857
 Feature Set: [9 14 18 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 14 18 21], Mean Accuracy: 0.839286
 Feature Set: [9 14 18 21], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [9 14 18 22], Mean Accuracy: 0.875000
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 Feature Set: [9 14 19 21], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [9 14 19 22], Mean Accuracy: 0.821429
 Feature Set: [9 14 19 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [9 14 20 21], Mean Accuracy: 0.910714
 Feature Set: [9 14 20 21], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [9 14 20 22], Mean Accuracy: 0.910714
 Feature Set: [9 14 20 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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 Feature Set: [9 14 21 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [9 15 16 17], Mean Accuracy: 0.785714
 Feature Set: [9 15 16 17], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [9 15 16 18], Mean Accuracy: 0.821429
 Feature Set: [9 15 16 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [9 15 16 19], Mean Accuracy: 0.803571
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 Feature Set: [9 15 16 21], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
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 Feature Set: [9 15 17 18], Mean Accuracy: 0.767857
 Feature Set: [9 15 17 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [9 15 17 19], Mean Accuracy: 0.785714
 Feature Set: [9 15 17 19], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: [9 15 17 20], Mean Accuracy: 0.857143
 Feature Set: [9 15 17 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
 Feature Set: [9 15 17 21], Mean Accuracy: 0.875000
 Feature Set: [9 15 17 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 15 17 22], Mean Accuracy: 0.910714
 Feature Set: [9 15 17 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 15 18 19], Mean Accuracy: 0.803571
 Feature Set: [9 15 18 19], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: [9 15 18 20], Mean Accuracy: 0.857143
 Feature Set: [9 15 18 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [9 15 18 21], Mean Accuracy: 0.839286

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Feature Set: [11 13 18 20], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
Feature Set: [11 13 18 21], Mean Accuracy: 0.946429
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Feature Set: [11 13 18 22], Mean Accuracy: 0.892857
Feature Set: [11 13 18 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [11 13 19 20], Mean Accuracy: 0.839286
Feature Set: [11 13 19 20], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
Feature Set: [11 13 19 21], Mean Accuracy: 0.946429
Feature Set: [11 13 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [11 13 19 22], Mean Accuracy: 0.857143
Feature Set: [11 13 19 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
Feature Set: [11 13 20 21], Mean Accuracy: 0.982143
Feature Set: [11 13 20 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [11 13 20 22], Mean Accuracy: 0.875000
Feature Set: [11 13 20 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [11 13 21 22], Mean Accuracy: 0.964286
Feature Set: [11 13 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [11 14 15 16], Mean Accuracy: 0.857143
Feature Set: [11 14 15 16], Accuracy: 0.969388, Precision: 0.807692, Recall: 0.785714, F1 Score: 0.969697
Feature Set: [11 14 15 17], Mean Accuracy: 0.875000
Feature Set: [11 14 15 17], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [11 14 15 18], Mean Accuracy: 0.857143
Feature Set: [11 14 15 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 15 19], Mean Accuracy: 0.857143
Feature Set: [11 14 15 19], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
Feature Set: [11 14 15 20], Mean Accuracy: 0.875000
Feature Set: [11 14 15 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 15 21], Mean Accuracy: 0.946429
Feature Set: [11 14 15 21], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 15 22], Mean Accuracy: 0.910714
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Feature Set: [11 14 16 17], Mean Accuracy: 0.803571
Feature Set: [11 14 16 17], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
Feature Set: [11 14 16 18], Mean Accuracy: 0.857143
Feature Set: [11 14 16 18], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 16 19], Mean Accuracy: 0.892857
Feature Set: [11 14 16 19], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
Feature Set: [11 14 16 20], Mean Accuracy: 0.875000
Feature Set: [11 14 16 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
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Feature Set: [11 14 16 22], Mean Accuracy: 0.928571
Feature Set: [11 14 16 22], Accuracy: 0.979592, Precision: 0.857143, Recall: 0.857143, F1 Score: 1.000000
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Feature Set: [11 14 17 18], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [11 14 17 19], Mean Accuracy: 0.839286
Feature Set: [11 14 17 19], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
Feature Set: [11 14 17 20], Mean Accuracy: 0.892857
Feature Set: [11 14 17 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
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Feature Set: [11 14 17 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [11 14 17 22], Mean Accuracy: 0.892857
Feature Set: [11 14 17 22], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
Feature Set: [11 14 18 19], Mean Accuracy: 0.839286
Feature Set: [11 14 18 19], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 18 20], Mean Accuracy: 0.839286
Feature Set: [11 14 18 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 18 21], Mean Accuracy: 0.964286
Feature Set: [11 14 18 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [11 14 18 22], Mean Accuracy: 0.892857
Feature Set: [11 14 18 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
Feature Set: [11 14 19 20], Mean Accuracy: 0.875000
Feature Set: [11 14 19 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222
Feature Set: [11 14 19 21], Mean Accuracy: 0.982143
Feature Set: [11 14 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000
Feature Set: [11 14 19 22], Mean Accuracy: 0.928571
Feature Set: [11 14 19 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

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Feature Set: [11 16 19 20], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 16 19 21], Mean Accuracy: 0.964286

Feature Set: [11 16 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 16 19 22], Mean Accuracy: 0.910714

Feature Set: [11 16 19 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444

Feature Set: [11 16 20 21], Mean Accuracy: 0.982143

Feature Set: [11 16 20 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 16 20 22], Mean Accuracy: 0.910714

Feature Set: [11 16 20 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 16 21 22], Mean Accuracy: 0.964286

Feature Set: [11 16 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 17 18 19], Mean Accuracy: 0.821429

Feature Set: [11 17 18 19], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444

Feature Set: [11 17 18 20], Mean Accuracy: 0.839286

Feature Set: [11 17 18 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222

Feature Set: [11 17 18 21], Mean Accuracy: 0.892857

Feature Set: [11 17 18 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 17 18 22], Mean Accuracy: 0.875000

Feature Set: [11 17 18 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 17 19 20], Mean Accuracy: 0.821429

Feature Set: [11 17 19 20], Accuracy: 0.959184, Precision: 0.818182, Recall: 0.714286, F1 Score: 0.933333

Feature Set: [11 17 19 21], Mean Accuracy: 0.964286

Feature Set: [11 17 19 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 17 19 22], Mean Accuracy: 0.892857

Feature Set: [11 17 19 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444

Feature Set: [11 17 20 21], Mean Accuracy: 0.928571

Feature Set: [11 17 20 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 17 20 22], Mean Accuracy: 0.910714

Feature Set: [11 17 20 22], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222

Feature Set: [11 17 21 22], Mean Accuracy: 0.946429

Feature Set: [11 17 21 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 18 19 20], Mean Accuracy: 0.839286

Feature Set: [11 18 19 20], Accuracy: 0.979592, Precision: 0.884615, Recall: 0.857143, F1 Score: 0.972222

Feature Set: [11 18 19 21], Mean Accuracy: 0.946429

Feature Set: [11 18 19 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 18 19 22], Mean Accuracy: 0.892857

Feature Set: [11 18 19 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 18 20 21], Mean Accuracy: 0.928571

Feature Set: [11 18 20 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 18 20 22], Mean Accuracy: 0.892857

Feature Set: [11 18 20 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359

Feature Set: [11 18 21 22], Mean Accuracy: 0.928571

Feature Set: [11 18 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 19 20 21], Mean Accuracy: 0.964286

Feature Set: [11 19 20 21], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 19 20 22], Mean Accuracy: 0.875000

Feature Set: [11 19 20 22], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394

Feature Set: [11 19 21 22], Mean Accuracy: 0.964286

Feature Set: [11 19 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [11 20 21 22], Mean Accuracy: 0.946429

Feature Set: [11 20 21 22], Accuracy: 1.000000, Precision: 1.000000, Recall: 1.000000, F1 Score: 1.000000

Feature Set: [12 13 14 15], Mean Accuracy: 0.517857

Feature Set: [12 13 14 15], Accuracy: 0.918367, Precision: 0.500000, Recall: 0.428571, F1 Score: 0.833333

Feature Set: [12 13 14 16], Mean Accuracy: 0.500000

Feature Set: [12 13 14 17], Mean Accuracy: 0.464286

Feature Set: [12 13 14 18], Mean Accuracy: 0.535714

Feature Set: [12 13 14 18], Accuracy: 0.938776, Precision: 0.683333, Recall: 0.571429, F1 Score: 0.895833

Feature Set: [12 13 14 19], Mean Accuracy: 0.589286

Feature Set: [12 13 14 19], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333

Feature Set: [12 13 14 20], Mean Accuracy: 0.660714

Feature Set: [12 13 14 20], Accuracy: 0.938776, Precision: 0.648148, Recall: 0.571429, F1 Score: 0.812500

Feature Set: [12 13 14 21], Mean Accuracy: 0.642857

Feature Set: [12 13 14 21], Accuracy: 0.938776, Precision: 0.648148, Recall: 0.571429, F1 Score: 0.812500

Feature Set: [12 13 14 22], Mean Accuracy: 0.732143

Feature Set: [12 13 14 22], Accuracy: 0.948980, Precision: 0.796296, Recall: 0.642857, F1 Score: 0.851852

Feature Set: [12 13 15 16], Mean Accuracy: 0.535714

Feature Set: [12 13 15 16], Accuracy: 0.928571, Precision: 0.648148, Recall: 0.500000, F1 Score: 0.880952

Feature Set: [12 13 15 17], Mean Accuracy: 0.482143

Feature Set: [12 16 17 21], Mean Accuracy: 0.642857
 Feature Set: [12 16 17 21], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [12 16 17 22], Mean Accuracy: 0.642857
 Feature Set: [12 16 17 22], Accuracy: 0.948980, Precision: 0.783333, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [12 16 18 19], Mean Accuracy: 0.571429
 Feature Set: [12 16 18 19], Accuracy: 0.928571, Precision: 0.656250, Recall: 0.500000, F1 Score: 0.819048
 Feature Set: [12 16 18 20], Mean Accuracy: 0.571429
 Feature Set: [12 16 18 20], Accuracy: 0.928571, Precision: 0.590909, Recall: 0.500000, F1 Score: 0.952381
 Feature Set: [12 16 18 21], Mean Accuracy: 0.660714
 Feature Set: [12 16 18 21], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
 Feature Set: [12 16 18 22], Mean Accuracy: 0.660714
 Feature Set: [12 16 18 22], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
 Feature Set: [12 16 19 20], Mean Accuracy: 0.625000
 Feature Set: [12 16 19 20], Accuracy: 0.918367, Precision: 0.587500, Recall: 0.428571, F1 Score: 0.833333
 Feature Set: [12 16 19 21], Mean Accuracy: 0.678571
 Feature Set: [12 16 19 21], Accuracy: 0.918367, Precision: 0.509259, Recall: 0.428571, F1 Score: 0.816667
 Feature Set: [12 16 19 22], Mean Accuracy: 0.625000
 Feature Set: [12 16 19 22], Accuracy: 0.928571, Precision: 0.625000, Recall: 0.500000, F1 Score: 0.785714
 Feature Set: [12 16 20 21], Mean Accuracy: 0.839286
 Feature Set: [12 16 20 21], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [12 16 20 22], Mean Accuracy: 0.732143
 Feature Set: [12 16 20 22], Accuracy: 0.969388, Precision: 0.861111, Recall: 0.785714, F1 Score: 0.954545
 Feature Set: [12 16 21 22], Mean Accuracy: 0.714286
 Feature Set: [12 16 21 22], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
 Feature Set: [12 17 18 19], Mean Accuracy: 0.500000
 Feature Set: [12 17 18 20], Mean Accuracy: 0.571429
 Feature Set: [12 17 18 20], Accuracy: 0.918367, Precision: 0.483333, Recall: 0.428571, F1 Score: 0.861111
 Feature Set: [12 17 18 21], Mean Accuracy: 0.625000
 Feature Set: [12 17 18 21], Accuracy: 0.928571, Precision: 0.592593, Recall: 0.500000, F1 Score: 0.833333
 Feature Set: [12 17 18 22], Mean Accuracy: 0.607143
 Feature Set: [12 17 18 22], Accuracy: 0.938776, Precision: 0.633333, Recall: 0.571429, F1 Score: 0.854167
 Feature Set: [12 17 19 20], Mean Accuracy: 0.625000
 Feature Set: [12 17 19 20], Accuracy: 0.918367, Precision: 0.545455, Recall: 0.428571, F1 Score: 1.000000
 Feature Set: [12 17 19 21], Mean Accuracy: 0.571429
 Feature Set: [12 17 19 21], Accuracy: 0.918367, Precision: 0.509259, Recall: 0.428571, F1 Score: 0.816667
 Feature Set: [12 17 19 22], Mean Accuracy: 0.607143
 Feature Set: [12 17 19 22], Accuracy: 0.908163, Precision: 0.385417, Recall: 0.357143, F1 Score: 0.713333
 Feature Set: [12 17 20 21], Mean Accuracy: 0.767857
 Feature Set: [12 17 20 21], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [12 17 20 22], Mean Accuracy: 0.750000
 Feature Set: [12 17 20 22], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [12 17 21 22], Mean Accuracy: 0.696429
 Feature Set: [12 17 21 22], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [12 18 19 20], Mean Accuracy: 0.625000
 Feature Set: [12 18 19 20], Accuracy: 0.908163, Precision: 0.450000, Recall: 0.357143, F1 Score: 0.933333
 Feature Set: [12 18 19 21], Mean Accuracy: 0.714286
 Feature Set: [12 18 19 21], Accuracy: 0.938776, Precision: 0.675000, Recall: 0.571429, F1 Score: 0.883333
 Feature Set: [12 18 19 22], Mean Accuracy: 0.660714
 Feature Set: [12 18 19 22], Accuracy: 0.928571, Precision: 0.687500, Recall: 0.500000, F1 Score: 0.828571
 Feature Set: [12 18 20 21], Mean Accuracy: 0.875000
 Feature Set: [12 18 20 21], Accuracy: 0.979592, Precision: 0.944444, Recall: 0.857143, F1 Score: 0.958333
 Feature Set: [12 18 20 22], Mean Accuracy: 0.839286
 Feature Set: [12 18 20 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [12 18 21 22], Mean Accuracy: 0.678571
 Feature Set: [12 18 21 22], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [12 19 20 21], Mean Accuracy: 0.714286
 Feature Set: [12 19 20 21], Accuracy: 0.928571, Precision: 0.568182, Recall: 0.500000, F1 Score: 0.914286
 Feature Set: [12 19 20 22], Mean Accuracy: 0.732143
 Feature Set: [12 19 20 22], Accuracy: 0.938776, Precision: 0.731481, Recall: 0.571429, F1 Score: 0.862500
 Feature Set: [12 19 21 22], Mean Accuracy: 0.678571
 Feature Set: [12 19 21 22], Accuracy: 0.928571, Precision: 0.566667, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [12 20 21 22], Mean Accuracy: 0.767857
 Feature Set: [12 20 21 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [13 14 15 16], Mean Accuracy: 0.589286
 Feature Set: [13 14 15 16], Accuracy: 0.938776, Precision: 0.590909, Recall: 0.571429, F1 Score: 0.875000
 Feature Set: [13 14 15 17], Mean Accuracy: 0.464286
 Feature Set: [13 14 15 18], Mean Accuracy: 0.535714
 Feature Set: [13 14 15 18], Accuracy: 0.928571, Precision: 0.600000, Recall: 0.500000, F1 Score: 0.904762

Feature Set: [13 14 15 19], Mean Accuracy: 0.571429
 Feature Set: [13 14 15 19], Accuracy: 0.908163, Precision: 0.363636, Recall: 0.357143, F1 Score: 0.866667
 Feature Set: [13 14 15 20], Mean Accuracy: 0.696429
 Feature Set: [13 14 15 20], Accuracy: 0.948980, Precision: 0.681818, Recall: 0.642857, F1 Score: 0.888889
 Feature Set: [13 14 15 21], Mean Accuracy: 0.732143
 Feature Set: [13 14 15 21], Accuracy: 0.948980, Precision: 0.694444, Recall: 0.642857, F1 Score: 0.944444
 Feature Set: [13 14 15 22], Mean Accuracy: 0.732143
 Feature Set: [13 14 15 22], Accuracy: 0.959184, Precision: 0.818182, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [13 14 16 17], Mean Accuracy: 0.375000
 Feature Set: [13 14 16 18], Mean Accuracy: 0.446429
 Feature Set: [13 14 16 19], Mean Accuracy: 0.607143
 Feature Set: [13 14 16 19], Accuracy: 0.918367, Precision: 0.483333, Recall: 0.428571, F1 Score: 0.861111
 Feature Set: [13 14 16 20], Mean Accuracy: 0.660714
 Feature Set: [13 14 16 20], Accuracy: 0.928571, Precision: 0.500000, Recall: 0.500000, F1 Score: 0.761905
 Feature Set: [13 14 16 21], Mean Accuracy: 0.642857
 Feature Set: [13 14 16 21], Accuracy: 0.928571, Precision: 0.638889, Recall: 0.500000, F1 Score: 0.866667
 Feature Set: [13 14 16 22], Mean Accuracy: 0.589286
 Feature Set: [13 14 16 22], Accuracy: 0.928571, Precision: 0.611111, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [13 14 17 18], Mean Accuracy: 0.482143
 Feature Set: [13 14 17 19], Mean Accuracy: 0.517857
 Feature Set: [13 14 17 19], Accuracy: 0.908163, Precision: 0.361111, Recall: 0.357143, F1 Score: 0.900000
 Feature Set: [13 14 17 20], Mean Accuracy: 0.589286
 Feature Set: [13 14 17 20], Accuracy: 0.928571, Precision: 0.537037, Recall: 0.500000, F1 Score: 0.785714
 Feature Set: [13 14 17 21], Mean Accuracy: 0.571429
 Feature Set: [13 14 17 21], Accuracy: 0.928571, Precision: 0.592593, Recall: 0.500000, F1 Score: 0.833333
 Feature Set: [13 14 17 22], Mean Accuracy: 0.607143
 Feature Set: [13 14 17 22], Accuracy: 0.918367, Precision: 0.454545, Recall: 0.428571, F1 Score: 0.888889
 Feature Set: [13 14 18 19], Mean Accuracy: 0.607143
 Feature Set: [13 14 18 19], Accuracy: 0.928571, Precision: 0.604167, Recall: 0.500000, F1 Score: 0.785714
 Feature Set: [13 14 18 20], Mean Accuracy: 0.625000
 Feature Set: [13 14 18 20], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [13 14 18 21], Mean Accuracy: 0.535714
 Feature Set: [13 14 18 21], Accuracy: 0.928571, Precision: 0.611111, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [13 14 18 22], Mean Accuracy: 0.642857
 Feature Set: [13 14 18 22], Accuracy: 0.938776, Precision: 0.700000, Recall: 0.571429, F1 Score: 0.916667
 Feature Set: [13 14 19 20], Mean Accuracy: 0.696429
 Feature Set: [13 14 19 20], Accuracy: 0.928571, Precision: 0.590909, Recall: 0.500000, F1 Score: 0.952381
 Feature Set: [13 14 19 21], Mean Accuracy: 0.696429
 Feature Set: [13 14 19 21], Accuracy: 0.938776, Precision: 0.666667, Recall: 0.571429, F1 Score: 0.875000
 Feature Set: [13 14 19 22], Mean Accuracy: 0.696429
 Feature Set: [13 14 19 22], Accuracy: 0.938776, Precision: 0.703704, Recall: 0.571429, F1 Score: 0.854167
 Feature Set: [13 14 20 21], Mean Accuracy: 0.839286
 Feature Set: [13 14 20 21], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
 Feature Set: [13 14 20 22], Mean Accuracy: 0.785714
 Feature Set: [13 14 20 22], Accuracy: 0.959184, Precision: 0.803030, Recall: 0.714286, F1 Score: 0.916667
 Feature Set: [13 14 21 22], Mean Accuracy: 0.660714
 Feature Set: [13 14 21 22], Accuracy: 0.938776, Precision: 0.648148, Recall: 0.571429, F1 Score: 0.812500
 Feature Set: [13 15 16 17], Mean Accuracy: 0.410714
 Feature Set: [13 15 16 18], Mean Accuracy: 0.535714
 Feature Set: [13 15 16 18], Accuracy: 0.948980, Precision: 0.750000, Recall: 0.642857, F1 Score: 0.888889
 Feature Set: [13 15 16 19], Mean Accuracy: 0.625000
 Feature Set: [13 15 16 19], Accuracy: 0.918367, Precision: 0.454545, Recall: 0.428571, F1 Score: 0.888889
 Feature Set: [13 15 16 20], Mean Accuracy: 0.589286
 Feature Set: [13 15 16 20], Accuracy: 0.938776, Precision: 0.633333, Recall: 0.571429, F1 Score: 0.854167
 Feature Set: [13 15 16 21], Mean Accuracy: 0.750000
 Feature Set: [13 15 16 21], Accuracy: 0.948980, Precision: 0.653846, Recall: 0.642857, F1 Score: 0.962963
 Feature Set: [13 15 16 22], Mean Accuracy: 0.696429
 Feature Set: [13 15 16 22], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [13 15 17 18], Mean Accuracy: 0.464286
 Feature Set: [13 15 17 19], Mean Accuracy: 0.517857
 Feature Set: [13 15 17 19], Accuracy: 0.908163, Precision: 0.393939, Recall: 0.357143, F1 Score: 0.900000
 Feature Set: [13 15 17 20], Mean Accuracy: 0.589286
 Feature Set: [13 15 17 20], Accuracy: 0.908163, Precision: 0.416667, Recall: 0.357143, F1 Score: 0.766667
 Feature Set: [13 15 17 21], Mean Accuracy: 0.660714
 Feature Set: [13 15 17 21], Accuracy: 0.928571, Precision: 0.545455, Recall: 0.500000, F1 Score: 0.904762
 Feature Set: [13 15 17 22], Mean Accuracy: 0.607143
 Feature Set: [13 15 17 22], Accuracy: 0.928571, Precision: 0.530303, Recall: 0.500000, F1 Score: 0.880952
 Feature Set: [13 15 18 19], Mean Accuracy: 0.607143
 Feature Set: [13 15 18 19], Accuracy: 0.928571, Precision: 0.500000, Recall: 0.500000, F1 Score: 0.809524

Feature Set: [13 15 18 20], Mean Accuracy: 0.607143
 Feature Set: [13 15 18 20], Accuracy: 0.948980, Precision: 0.733333, Recall: 0.642857, F1 Score: 0.870370
 Feature Set: [13 15 18 21], Mean Accuracy: 0.767857
 Feature Set: [13 15 18 21], Accuracy: 0.938776, Precision: 0.625000, Recall: 0.571429, F1 Score: 0.958333
 Feature Set: [13 15 18 22], Mean Accuracy: 0.678571
 Feature Set: [13 15 18 22], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [13 15 19 20], Mean Accuracy: 0.625000
 Feature Set: [13 15 19 20], Accuracy: 0.928571, Precision: 0.650000, Recall: 0.500000, F1 Score: 0.952381
 Feature Set: [13 15 19 21], Mean Accuracy: 0.750000
 Feature Set: [13 15 19 21], Accuracy: 0.948980, Precision: 0.681818, Recall: 0.642857, F1 Score: 0.888889
 Feature Set: [13 15 19 22], Mean Accuracy: 0.607143
 Feature Set: [13 15 19 22], Accuracy: 0.918367, Precision: 0.518519, Recall: 0.428571, F1 Score: 0.833333
 Feature Set: [13 15 20 21], Mean Accuracy: 0.839286
 Feature Set: [13 15 20 21], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [13 15 20 22], Mean Accuracy: 0.803571
 Feature Set: [13 15 20 22], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [13 15 21 22], Mean Accuracy: 0.767857
 Feature Set: [13 15 21 22], Accuracy: 0.948980, Precision: 0.653846, Recall: 0.642857, F1 Score: 0.962963
 Feature Set: [13 16 17 18], Mean Accuracy: 0.375000
 Feature Set: [13 16 17 19], Mean Accuracy: 0.428571
 Feature Set: [13 16 17 20], Mean Accuracy: 0.535714
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 Feature Set: [13 16 17 21], Accuracy: 0.918367, Precision: 0.533333, Recall: 0.428571, F1 Score: 0.916667
 Feature Set: [13 16 17 22], Mean Accuracy: 0.553571
 Feature Set: [13 16 17 22], Accuracy: 0.928571, Precision: 0.500000, Recall: 0.500000, F1 Score: 0.904762
 Feature Set: [13 16 18 19], Mean Accuracy: 0.607143
 Feature Set: [13 16 18 19], Accuracy: 0.918367, Precision: 0.483333, Recall: 0.428571, F1 Score: 0.861111
 Feature Set: [13 16 18 20], Mean Accuracy: 0.571429
 Feature Set: [13 16 18 20], Accuracy: 0.938776, Precision: 0.685185, Recall: 0.571429, F1 Score: 0.833333
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 Feature Set: [13 16 19 20], Mean Accuracy: 0.660714
 Feature Set: [13 16 19 20], Accuracy: 0.938776, Precision: 0.611111, Recall: 0.571429, F1 Score: 0.937500
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 Feature Set: [13 16 19 22], Mean Accuracy: 0.607143
 Feature Set: [13 16 19 22], Accuracy: 0.928571, Precision: 0.527778, Recall: 0.500000, F1 Score: 0.928571
 Feature Set: [13 16 20 21], Mean Accuracy: 0.892857
 Feature Set: [13 16 20 21], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [13 16 20 22], Mean Accuracy: 0.767857
 Feature Set: [13 16 20 22], Accuracy: 0.959184, Precision: 0.772727, Recall: 0.714286, F1 Score: 0.900000
 Feature Set: [13 16 21 22], Mean Accuracy: 0.625000
 Feature Set: [13 16 21 22], Accuracy: 0.938776, Precision: 0.636364, Recall: 0.571429, F1 Score: 0.916667
 Feature Set: [13 17 18 19], Mean Accuracy: 0.571429
 Feature Set: [13 17 18 19], Accuracy: 0.908163, Precision: 0.411111, Recall: 0.357143, F1 Score: 0.800000
 Feature Set: [13 17 18 20], Mean Accuracy: 0.625000
 Feature Set: [13 17 18 20], Accuracy: 0.938776, Precision: 0.760417, Recall: 0.571429, F1 Score: 0.820833
 Feature Set: [13 17 18 21], Mean Accuracy: 0.642857
 Feature Set: [13 17 18 21], Accuracy: 0.938776, Precision: 0.666667, Recall: 0.571429, F1 Score: 0.875000
 Feature Set: [13 17 18 22], Mean Accuracy: 0.553571
 Feature Set: [13 17 18 22], Accuracy: 0.918367, Precision: 0.454545, Recall: 0.428571, F1 Score: 0.888889
 Feature Set: [13 17 19 20], Mean Accuracy: 0.607143
 Feature Set: [13 17 19 20], Accuracy: 0.918367, Precision: 0.509259, Recall: 0.428571, F1 Score: 0.816667
 Feature Set: [13 17 19 21], Mean Accuracy: 0.678571
 Feature Set: [13 17 19 21], Accuracy: 0.928571, Precision: 0.620370, Recall: 0.500000, F1 Score: 0.842857
 Feature Set: [13 17 19 22], Mean Accuracy: 0.625000
 Feature Set: [13 17 19 22], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
 Feature Set: [13 17 20 21], Mean Accuracy: 0.767857
 Feature Set: [13 17 20 21], Accuracy: 0.938776, Precision: 0.611111, Recall: 0.571429, F1 Score: 0.937500
 Feature Set: [13 17 20 22], Mean Accuracy: 0.732143
 Feature Set: [13 17 20 22], Accuracy: 0.928571, Precision: 0.541667, Recall: 0.500000, F1 Score: 0.952381
 Feature Set: [13 17 21 22], Mean Accuracy: 0.607143
 Feature Set: [13 17 21 22], Accuracy: 0.918367, Precision: 0.484848, Recall: 0.428571, F1 Score: 0.916667
 Feature Set: [13 18 19 20], Mean Accuracy: 0.696429
 Feature Set: [13 18 19 20], Accuracy: 0.948980, Precision: 0.694444, Recall: 0.642857, F1 Score: 0.944444

Feature Set: [13 18 19 21], Mean Accuracy: 0.750000
 Feature Set: [13 18 19 21], Accuracy: 0.928571, Precision: 0.620370, Recall: 0.500000, F1 Score: 0.842857
 Feature Set: [13 18 19 22], Mean Accuracy: 0.732143
 Feature Set: [13 18 19 22], Accuracy: 0.938776, Precision: 0.675000, Recall: 0.571429, F1 Score: 0.883333
 Feature Set: [13 18 20 21], Mean Accuracy: 0.857143
 Feature Set: [13 18 20 21], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [13 18 20 22], Mean Accuracy: 0.803571
 Feature Set: [13 18 20 22], Accuracy: 0.969388, Precision: 0.893939, Recall: 0.785714, F1 Score: 0.924242
 Feature Set: [13 18 21 22], Mean Accuracy: 0.714286
 Feature Set: [13 18 21 22], Accuracy: 0.948980, Precision: 0.759259, Recall: 0.642857, F1 Score: 0.833333
 Feature Set: [13 19 20 21], Mean Accuracy: 0.839286
 Feature Set: [13 19 20 21], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [13 19 20 22], Mean Accuracy: 0.785714
 Feature Set: [13 19 20 22], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [13 19 21 22], Mean Accuracy: 0.714286
 Feature Set: [13 19 21 22], Accuracy: 0.938776, Precision: 0.675000, Recall: 0.571429, F1 Score: 0.883333
 Feature Set: [13 20 21 22], Mean Accuracy: 0.839286
 Feature Set: [13 20 21 22], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 0.883333
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 Feature Set: [14 15 16 18], Mean Accuracy: 0.517857
 Feature Set: [14 15 16 18], Accuracy: 0.928571, Precision: 0.545455, Recall: 0.500000, F1 Score: 0.904762
 Feature Set: [14 15 16 19], Mean Accuracy: 0.607143
 Feature Set: [14 15 16 19], Accuracy: 0.918367, Precision: 0.454545, Recall: 0.428571, F1 Score: 0.888889
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 Feature Set: [14 15 16 21], Mean Accuracy: 0.678571
 Feature Set: [14 15 16 21], Accuracy: 0.948980, Precision: 0.775000, Recall: 0.642857, F1 Score: 0.896296
 Feature Set: [14 15 16 22], Mean Accuracy: 0.660714
 Feature Set: [14 15 16 22], Accuracy: 0.948980, Precision: 0.708333, Recall: 0.642857, F1 Score: 0.962963
 Feature Set: [14 15 17 18], Mean Accuracy: 0.517857
 Feature Set: [14 15 17 18], Accuracy: 0.918367, Precision: 0.500000, Recall: 0.428571, F1 Score: 0.888889
 Feature Set: [14 15 17 19], Mean Accuracy: 0.607143
 Feature Set: [14 15 17 19], Accuracy: 0.908163, Precision: 0.350000, Recall: 0.357143, F1 Score: 0.800000
 Feature Set: [14 15 17 20], Mean Accuracy: 0.642857
 Feature Set: [14 15 17 20], Accuracy: 0.928571, Precision: 0.533333, Recall: 0.500000, F1 Score: 0.833333
 Feature Set: [14 15 17 21], Mean Accuracy: 0.660714
 Feature Set: [14 15 17 21], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
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 Feature Set: [14 15 17 22], Accuracy: 0.948980, Precision: 0.681818, Recall: 0.642857, F1 Score: 0.888889
 Feature Set: [14 15 18 19], Mean Accuracy: 0.571429
 Feature Set: [14 15 18 19], Accuracy: 0.928571, Precision: 0.545455, Recall: 0.500000, F1 Score: 0.904762
 Feature Set: [14 15 18 20], Mean Accuracy: 0.535714
 Feature Set: [14 15 18 20], Accuracy: 0.938776, Precision: 0.590909, Recall: 0.571429, F1 Score: 0.875000
 Feature Set: [14 15 18 21], Mean Accuracy: 0.696429
 Feature Set: [14 15 18 21], Accuracy: 0.948980, Precision: 0.783333, Recall: 0.642857, F1 Score: 0.907407
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 Feature Set: [14 15 18 22], Accuracy: 0.948980, Precision: 0.727273, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [14 15 19 20], Mean Accuracy: 0.660714
 Feature Set: [14 15 19 20], Accuracy: 0.918367, Precision: 0.423077, Recall: 0.428571, F1 Score: 0.944444
 Feature Set: [14 15 19 21], Mean Accuracy: 0.660714
 Feature Set: [14 15 19 21], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 1.000000
 Feature Set: [14 15 19 22], Mean Accuracy: 0.696429
 Feature Set: [14 15 19 22], Accuracy: 0.938776, Precision: 0.694444, Recall: 0.571429, F1 Score: 0.841667
 Feature Set: [14 15 20 21], Mean Accuracy: 0.785714
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 Feature Set: [14 15 20 22], Mean Accuracy: 0.750000
 Feature Set: [14 15 20 22], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [14 15 21 22], Mean Accuracy: 0.714286
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 Feature Set: [14 16 17 20], Mean Accuracy: 0.500000
 Feature Set: [14 16 17 21], Mean Accuracy: 0.517857
 Feature Set: [14 16 17 21], Accuracy: 0.918367, Precision: 0.527778, Recall: 0.428571, F1 Score: 0.844444
 Feature Set: [14 16 17 22], Mean Accuracy: 0.553571
 Feature Set: [14 16 17 22], Accuracy: 0.928571, Precision: 0.600000, Recall: 0.500000, F1 Score: 0.904762
 Feature Set: [14 16 18 19], Mean Accuracy: 0.535714
 Feature Set: [14 16 18 19], Accuracy: 0.908163, Precision: 0.361111, Recall: 0.357143, F1 Score: 0.746667
 Feature Set: [14 16 18 20], Mean Accuracy: 0.589286

Feature Set: [16 17 19 21], Accuracy: 0.928571, Precision: 0.566667, Recall: 0.500000, F1 Score: 0.857143
 Feature Set: [16 17 19 22], Mean Accuracy: 0.625000
 Feature Set: [16 17 19 22], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [16 17 20 21], Mean Accuracy: 0.767857
 Feature Set: [16 17 20 21], Accuracy: 0.959184, Precision: 0.833333, Recall: 0.714286, F1 Score: 1.000000
 Feature Set: [16 17 20 22], Mean Accuracy: 0.750000
 Feature Set: [16 17 20 22], Accuracy: 0.969388, Precision: 0.833333, Recall: 0.785714, F1 Score: 0.939394
 Feature Set: [16 17 21 22], Mean Accuracy: 0.660714
 Feature Set: [16 17 21 22], Accuracy: 0.938776, Precision: 0.600000, Recall: 0.571429, F1 Score: 0.833333
 Feature Set: [16 18 19 20], Mean Accuracy: 0.660714
 Feature Set: [16 18 19 20], Accuracy: 0.928571, Precision: 0.583333, Recall: 0.500000, F1 Score: 0.880952
 Feature Set: [16 18 19 21], Mean Accuracy: 0.696429
 Feature Set: [16 18 19 21], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [16 18 19 22], Mean Accuracy: 0.678571
 Feature Set: [16 18 19 22], Accuracy: 0.948980, Precision: 0.712121, Recall: 0.642857, F1 Score: 0.907407
 Feature Set: [16 18 20 21], Mean Accuracy: 0.857143
 Feature Set: [16 18 20 21], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [16 18 20 22], Mean Accuracy: 0.803571
 Feature Set: [16 18 20 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [16 18 21 22], Mean Accuracy: 0.714286
 Feature Set: [16 18 21 22], Accuracy: 0.979592, Precision: 0.916667, Recall: 0.857143, F1 Score: 0.944444
 Feature Set: [16 19 20 21], Mean Accuracy: 0.767857
 Feature Set: [16 19 20 21], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [16 19 20 22], Mean Accuracy: 0.696429
 Feature Set: [16 19 20 22], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [16 19 21 22], Mean Accuracy: 0.660714
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 Feature Set: [16 20 21 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [17 18 19 20], Mean Accuracy: 0.678571
 Feature Set: [17 18 19 20], Accuracy: 0.928571, Precision: 0.575000, Recall: 0.500000, F1 Score: 0.866667
 Feature Set: [17 18 19 21], Mean Accuracy: 0.660714
 Feature Set: [17 18 19 21], Accuracy: 0.918367, Precision: 0.509259, Recall: 0.428571, F1 Score: 0.816667
 Feature Set: [17 18 19 22], Mean Accuracy: 0.589286
 Feature Set: [17 18 19 22], Accuracy: 0.918367, Precision: 0.566667, Recall: 0.428571, F1 Score: 0.805556
 Feature Set: [17 18 20 21], Mean Accuracy: 0.821429
 Feature Set: [17 18 20 21], Accuracy: 0.959184, Precision: 0.777778, Recall: 0.714286, F1 Score: 0.950000
 Feature Set: [17 18 20 22], Mean Accuracy: 0.750000
 Feature Set: [17 18 20 22], Accuracy: 0.948980, Precision: 0.666667, Recall: 0.642857, F1 Score: 0.925926
 Feature Set: [17 18 21 22], Mean Accuracy: 0.660714
 Feature Set: [17 18 21 22], Accuracy: 0.918367, Precision: 0.527778, Recall: 0.428571, F1 Score: 0.844444
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 Feature Set: [17 19 21 22], Accuracy: 0.928571, Precision: 0.620370, Recall: 0.500000, F1 Score: 0.842857
 Feature Set: [17 20 21 22], Mean Accuracy: 0.839286
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 Feature Set: [18 19 20 21], Mean Accuracy: 0.857143
 Feature Set: [18 19 20 21], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333
 Feature Set: [18 19 20 22], Mean Accuracy: 0.785714
 Feature Set: [18 19 20 22], Accuracy: 0.948980, Precision: 0.694444, Recall: 0.642857, F1 Score: 0.944444
 Feature Set: [18 19 21 22], Mean Accuracy: 0.660714
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 Feature Set: [18 20 21 22], Mean Accuracy: 0.875000
 Feature Set: [18 20 21 22], Accuracy: 0.989796, Precision: 0.961538, Recall: 0.928571, F1 Score: 0.974359
 Feature Set: [19 20 21 22], Mean Accuracy: 0.839286
 Feature Set: [19 20 21 22], Accuracy: 0.959184, Precision: 0.750000, Recall: 0.714286, F1 Score: 0.933333

7.3.3.4 MATLAB based LDA classifier (B) for one feature-based CDs array accuracy study and plotting

Aims and working mechanism

The MATLAB LDA classifier (B) was engineered to assess the efficacy of Linear Discriminant Analysis (LDA) in classifying a dataset that is believed to encompass measurements pertinent to 14 distinct metal ions, with each ion represented by 4 samples. Notably, the dataset is sourced from a file named 'CDdata.xlsx', despite an apparent discrepancy indicated by the variable 'files', which suggests the filename 'data.xlsx'. The script is primarily aimed at executing LDA classification for each feature individually, illustrating the one-dimensional (1D) distributions of the feature values through plotting, and quantifying performance metrics for each feature within a 4-fold cross-validation framework. The script's functionalities are elaborated below:

- **Environment Setup and Data Loading:** Initiates by configuring the environment, establishing a directory for results (if non-existent), and opening a file to log the LDA classification outcomes. Subsequently, it loads the dataset from the designated Excel file.
- **Data Preparation:** Constructs a label vector for the 14 metal ions, predicated on the assumption that each ion is denoted by 4 samples. A 4-fold cross-validation index is devised to enable the evaluation of the classifier's performance across varied data subsets.
- **Visualization Preparation:** An array of colours is determined to visually segregate the 14 metal ions in the ensuing plots.
- **Classification and Evaluation:** Pertaining to each of the 22 features within the dataset, the script undertakes the ensuing procedures:
 - Segregates the dataset into training and testing sets in alignment with the cross-validation indices.

- Trains an LDA model leveraging the training data for the focal feature.
 - Forecasts the metal ion labels for the testing set.
 - Fabricates a 1D plot for the current feature, exhibiting the distribution of feature values for each metal ion, with a distinct colour allocated to each ion.
 - Computes performance metrics (Accuracy, Precision, Recall, F1 Score) for the predictions and archives these metrics in the results file.
- **Performance Metrics Calculation:** Incorporates a bespoke function, 'CalculatePerformanceMetrics', designed to derive the confusion matrix from the actual and predicted labels, thereafter, calculating and returning the mean values of accuracy, precision, recall, and F1 score.
- **Output:** For each feature, the script engenders a 1D plot, encapsulated as a PNG file, elucidating the distribution of feature values across the various metal ions. Moreover, it chronicles the performance metrics of each feature's classification efficacy in a text file, thereby offering insights into the proficiency of each feature in differentiating the metal ions.

Script and functions

```
clear;
files = 'data.xlsx'; % Assuming the input file is named 'CDdata.xlsx'
resultsDir = 'WithoutN_OneD_LDA_results';
if ~exist(resultsDir, 'dir')
    mkdir(resultsDir); % Create results directory if it does not exist
end
fileID = fopen(fullfile(resultsDir, 'lda_result_CD.txt'), 'w');
[data, txt, raw] = xlsread(files);

% Assuming the label vector for 14 metal ions, each with 4 replicates
label = repelem(1:14, 4);
indices = crossvalind('Kfold', label, 4); % 4-fold cross-validation

% Define colors for each metal ion
colors = [
    1, 0, 0; % Red
    0, 1, 0; % Green
```

```

0, 0, 1; % Blue
1, 1, 0; % Yellow
1, 0, 1; % Magenta
0, 1, 1; % Cyan
0.5, 0.5, 0.5; % Gray
0.9, 0.6, 0; % Orange
0.5, 0, 0.5; % Purple
0, 0.5, 0.5; % Teal
0.4, 0.4, 0.2; % Olive
0.6, 0.2, 0.2; % Brown
0, 0.5, 0; % Dark Green
0.5, 0, 0; % Maroon
]; % Generates 14 distinct colors
%%
% Sensing targets (metal ions)
sensingTargets = {'Al(III)', 'Ag(I)', 'Ca(II)', 'Co(II)', 'Cu(II)', 'Cd(II)', 'Fe(III)', 'Hg(II)', 'Ni(II)', 'Pb(II)', 'Zn(II)',
'Mg(II)', 'Mn(II)', 'Cr(III)'};

% Perform LDA classification using one feature at a time
for feature_num = 1:22 % For each of the 22 features
    feature_select = feature_num;
    feature_data = data(:, feature_select);

    for k = 1:4 % For each fold
        test = (indices == k);
        train = ~test;
        train_data = feature_data(train);
        train_label = label(train);
        test_data = feature_data(test);
        test_label = label(test);

        lda = fitcdiscr(train_data, train_label);
        predicted_label = predict(lda, test_data);

        % Plotting 1D data
        figure;
        gscatter(feature_data, zeros(size(feature_data, 1), 1), label, colors);
        xlabel(sprintf('Feature %d', feature_num));
        ylabel('');
        title(sprintf('1D Plot for Feature %d', feature_num));
        legend(sensingTargets, 'Location', 'bestoutside');
        saveas(gcf, fullfile(resultsDir, sprintf('1DPlot_Feature_%d.png', feature_num)));

        % Calculate and report performance metrics
        metrics = CalculatePerformanceMetrics(test_label, predicted_label);
        fprintf(fileID, 'Feature %d - Accuracy: %f, Precision: %f, Recall: %f, F1 Score: %f\r\n', feature_num,
metrics(1), metrics(2), metrics(3), metrics(4));
    end
end
fclose(fileID);

function metrics = CalculatePerformanceMetrics(true_labels, predicted_labels)
% This function calculates the performance metrics
[C,~] = confusionmat(true_labels, predicted_labels);
TP = diag(C);
FP = sum(C, 1) - TP;
FN = sum(C, 2) - TP;
TN = sum(C(:)) - TP - FP - FN;

```

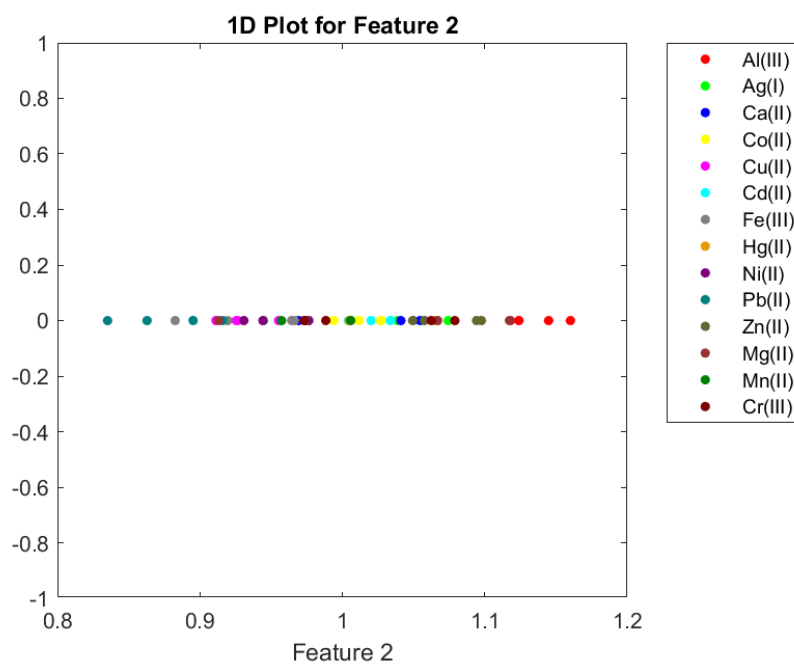
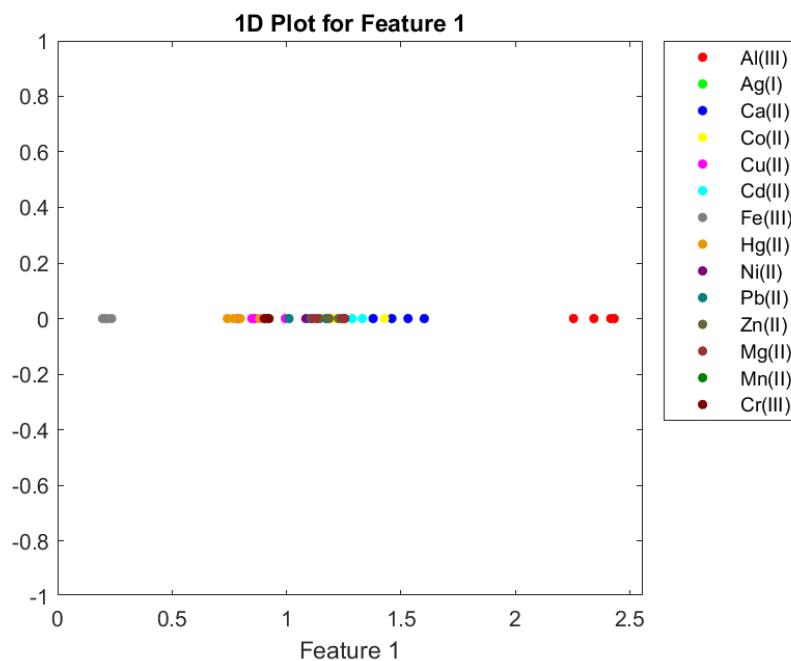


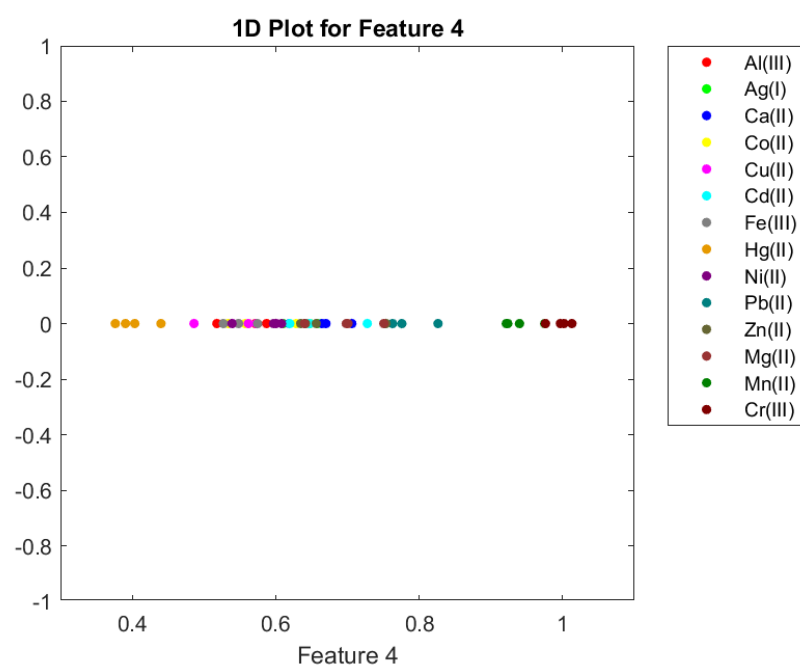
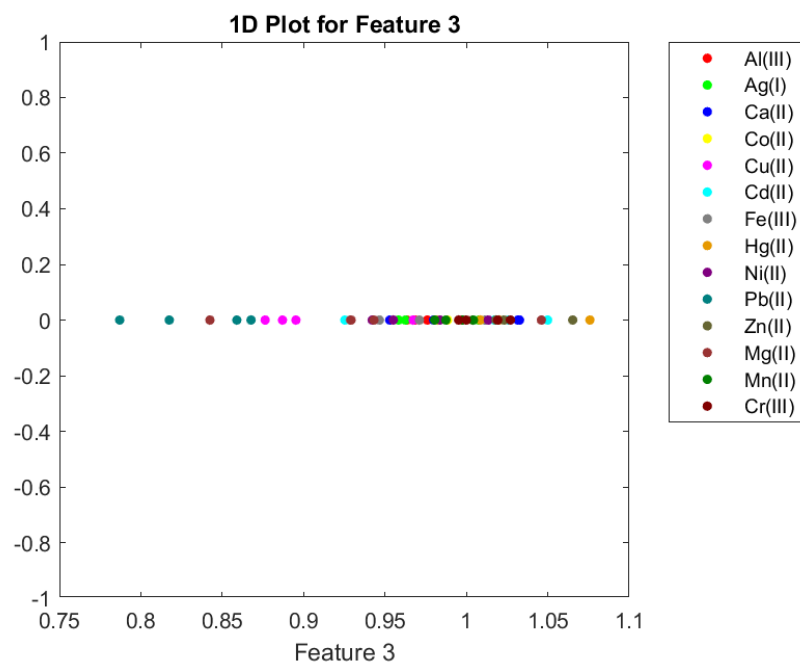
```

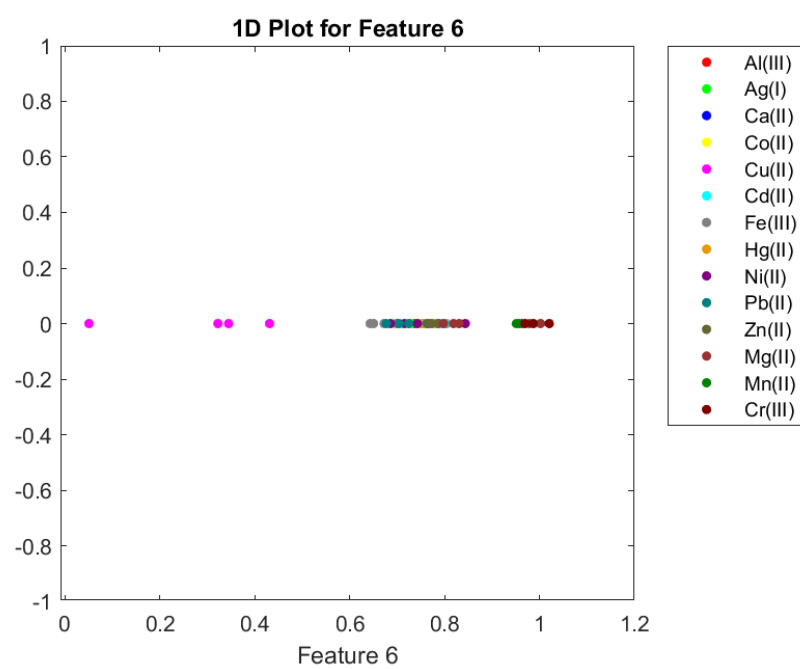
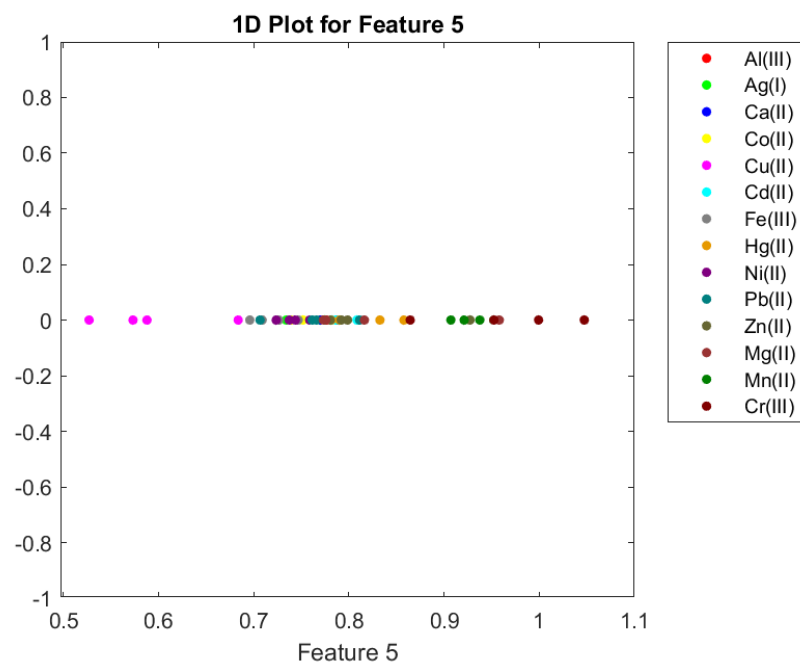
Accuracy = (TP + TN) ./ (TP + FP + FN + TN);
Precision = TP ./ (TP + FP);
Recall = TP ./ (TP + FN);
F1 = 2 .* (Precision .* Recall) ./ (Precision + Recall);
metrics = [mean(Accuracy), mean(Precision), mean(Recall), mean(F1)];
end

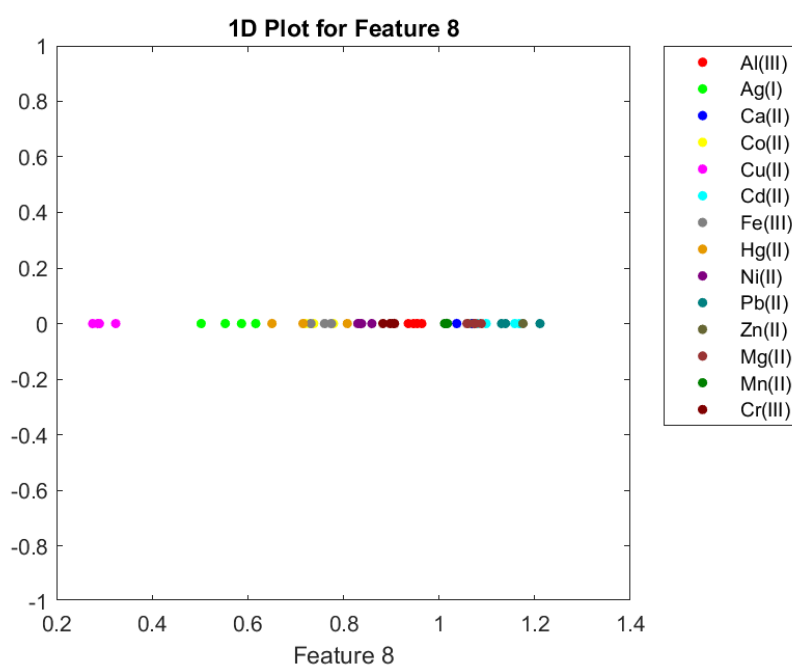
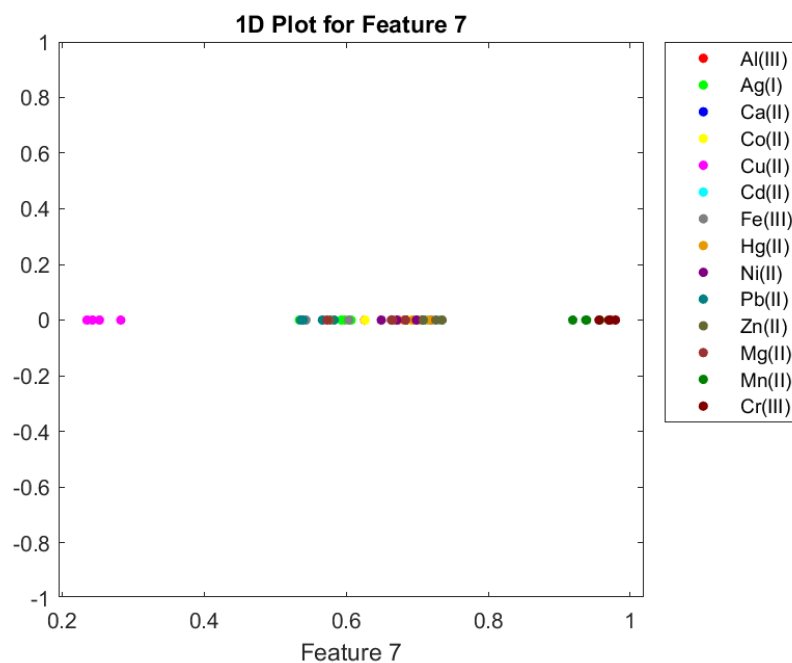
```

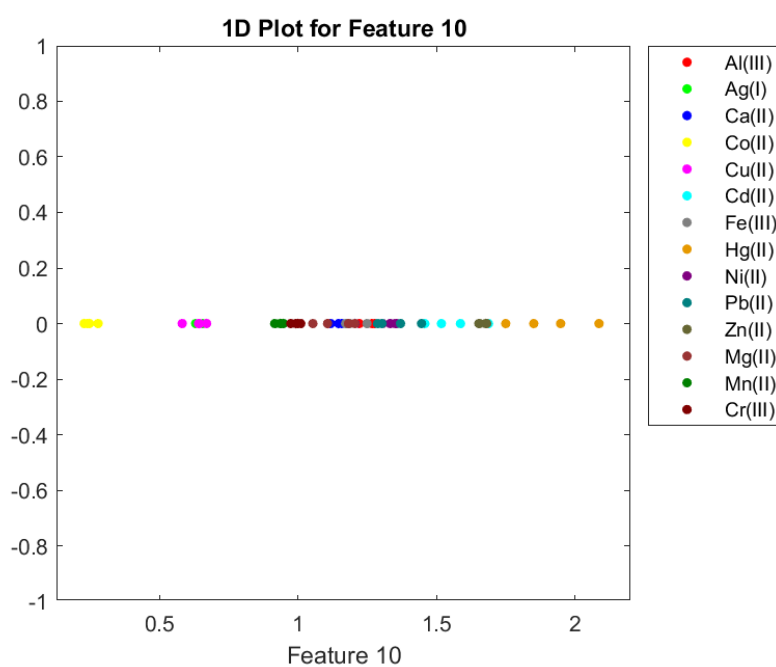
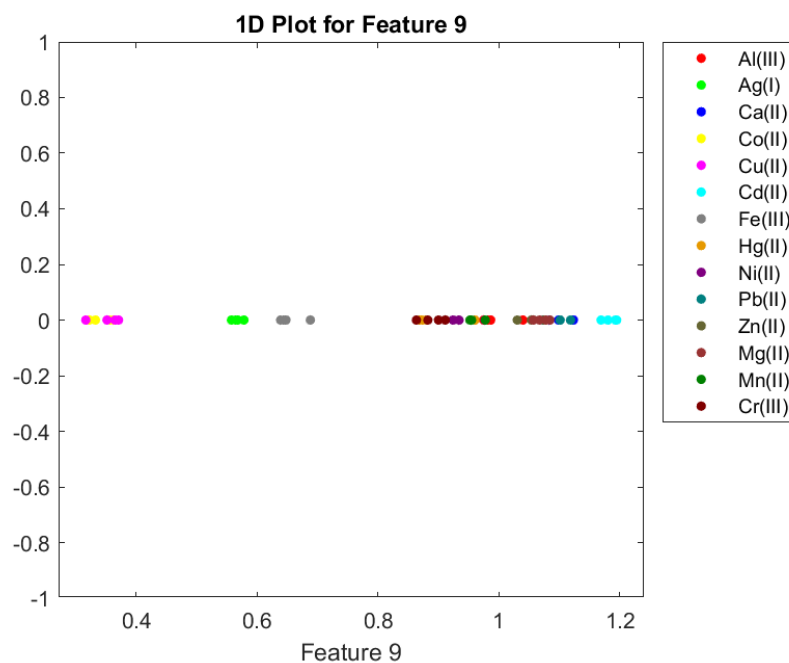
Results of one CD based LDA classification

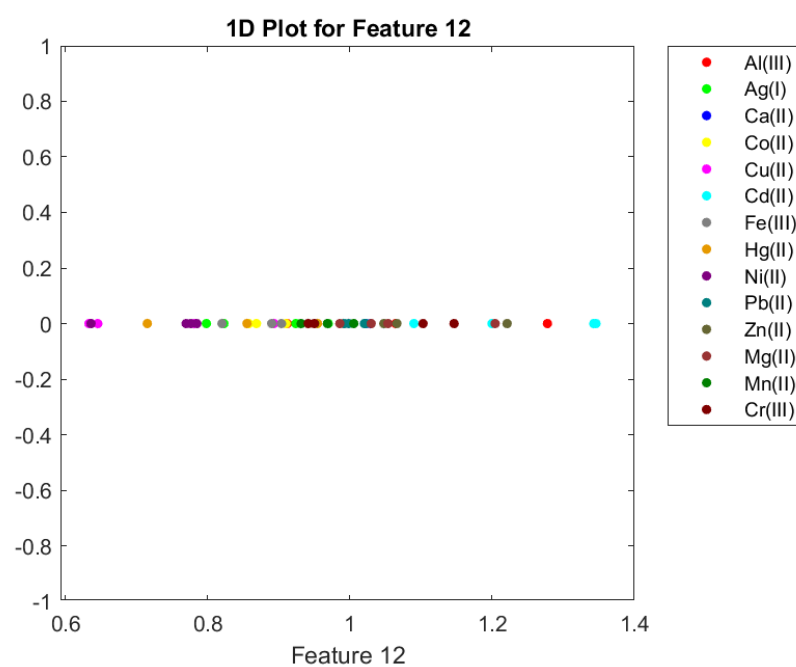
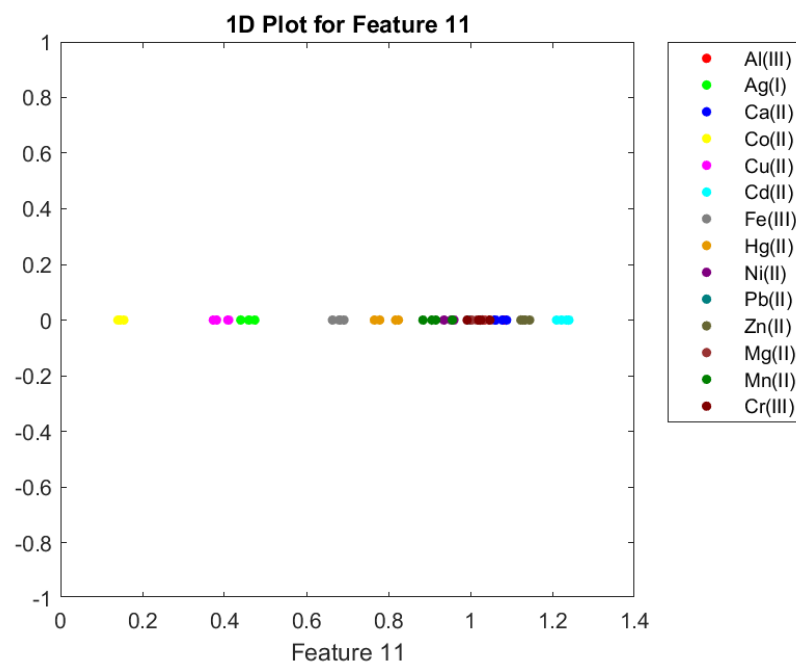


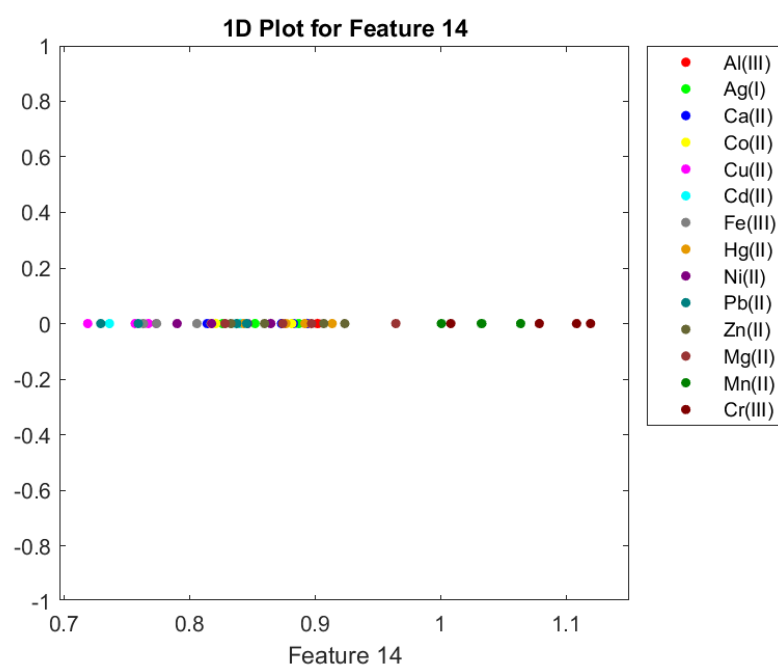
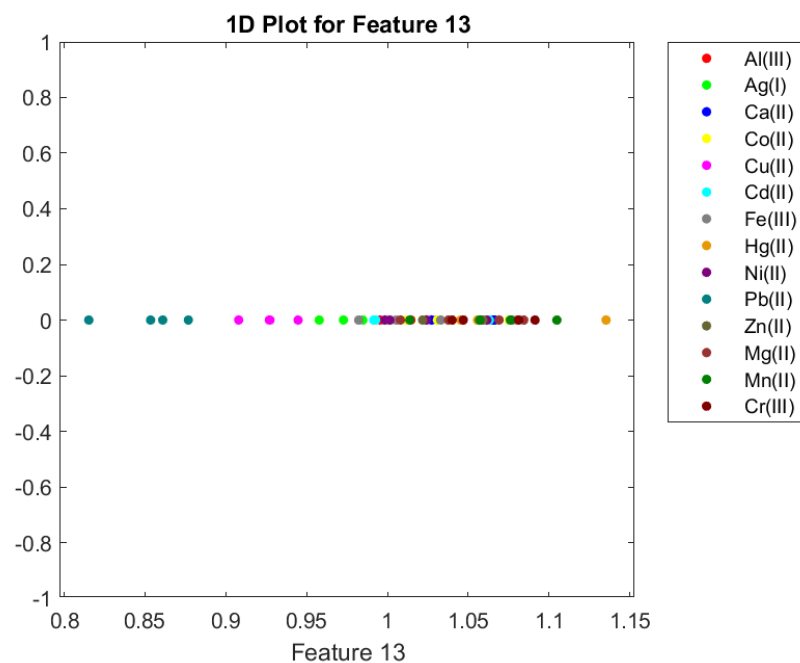


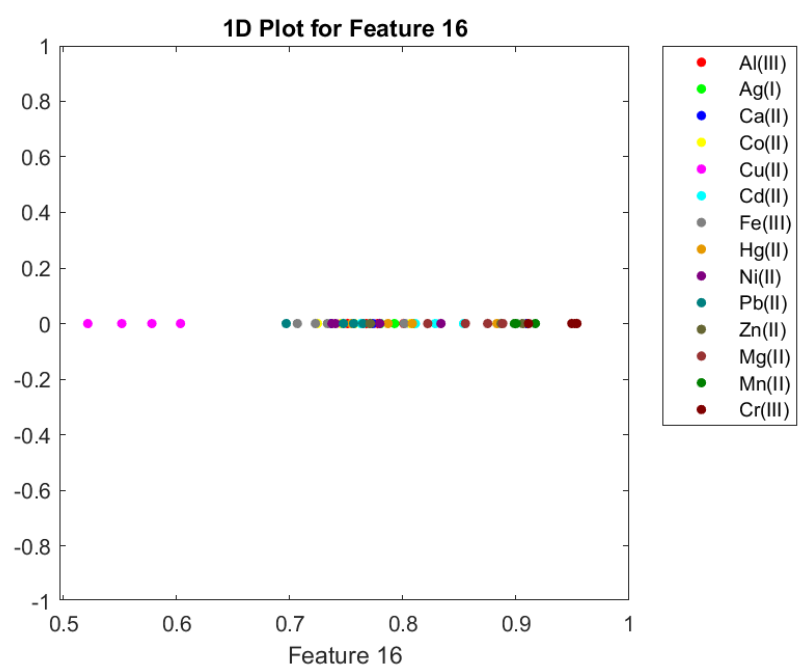
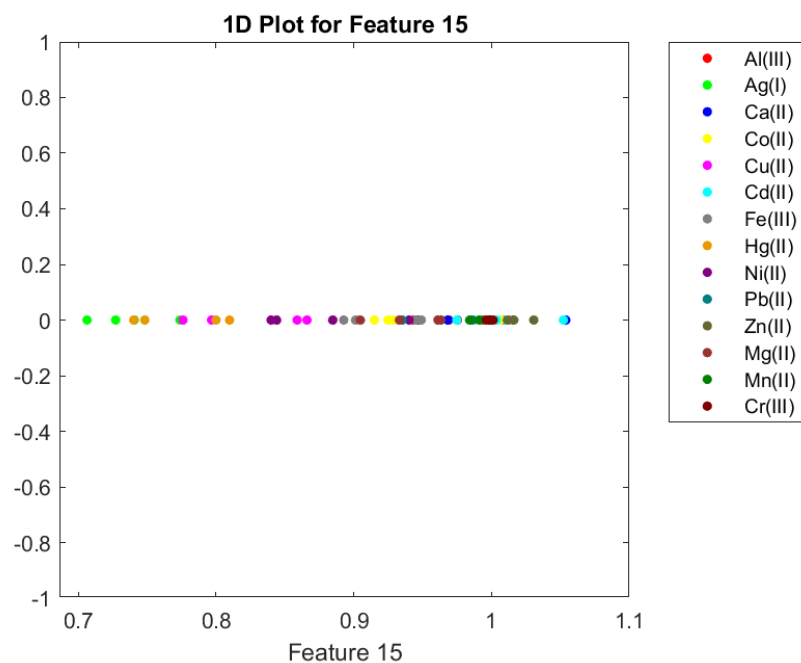


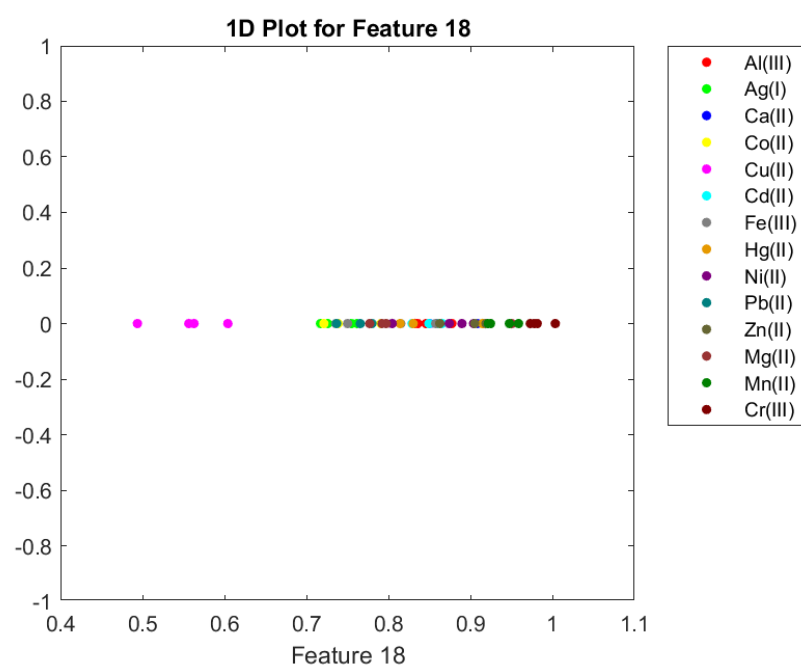
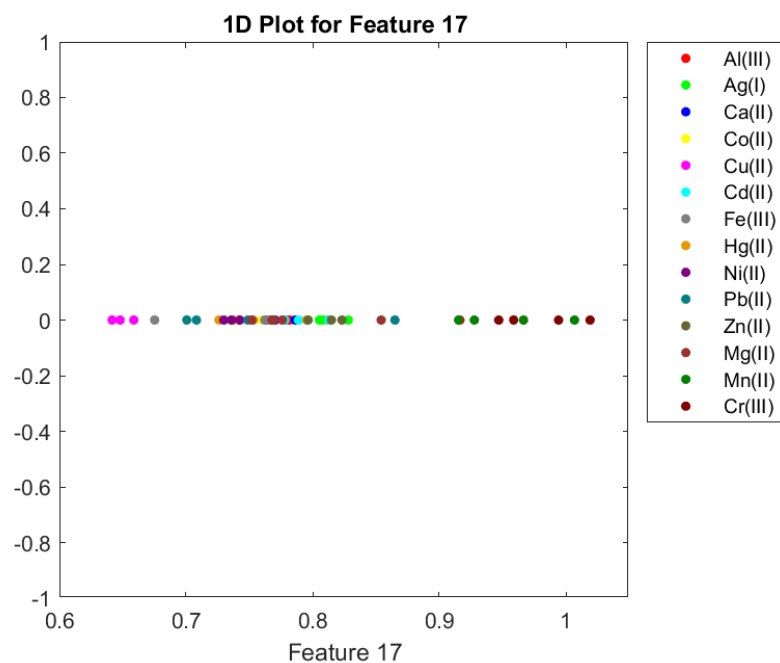


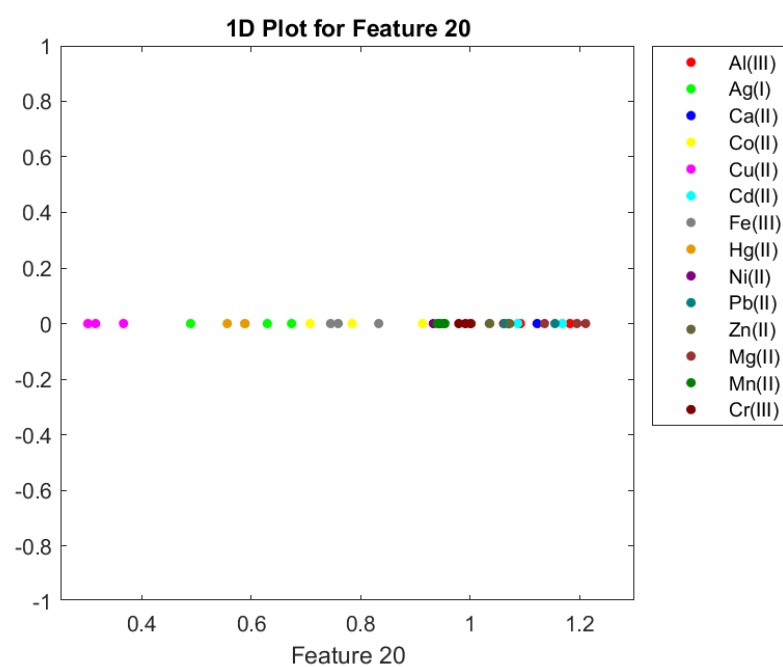
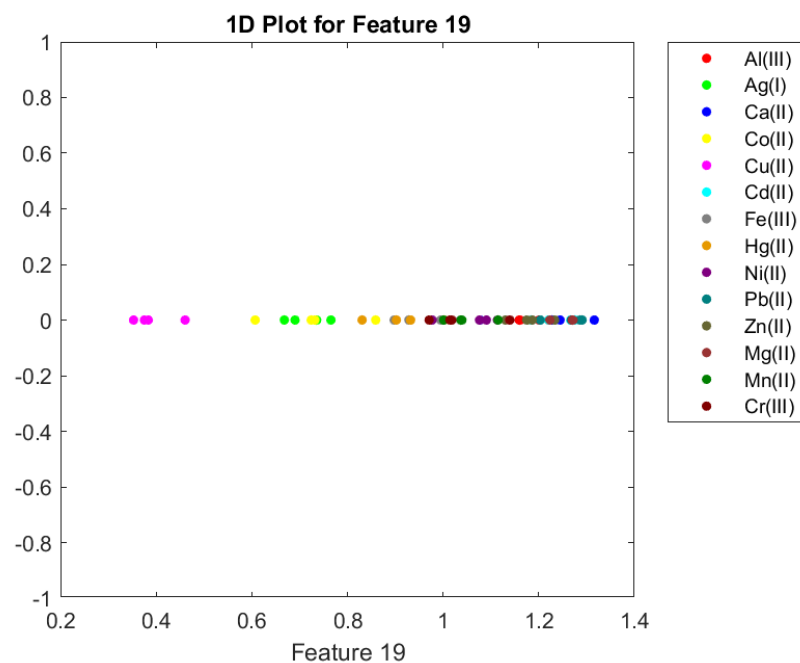












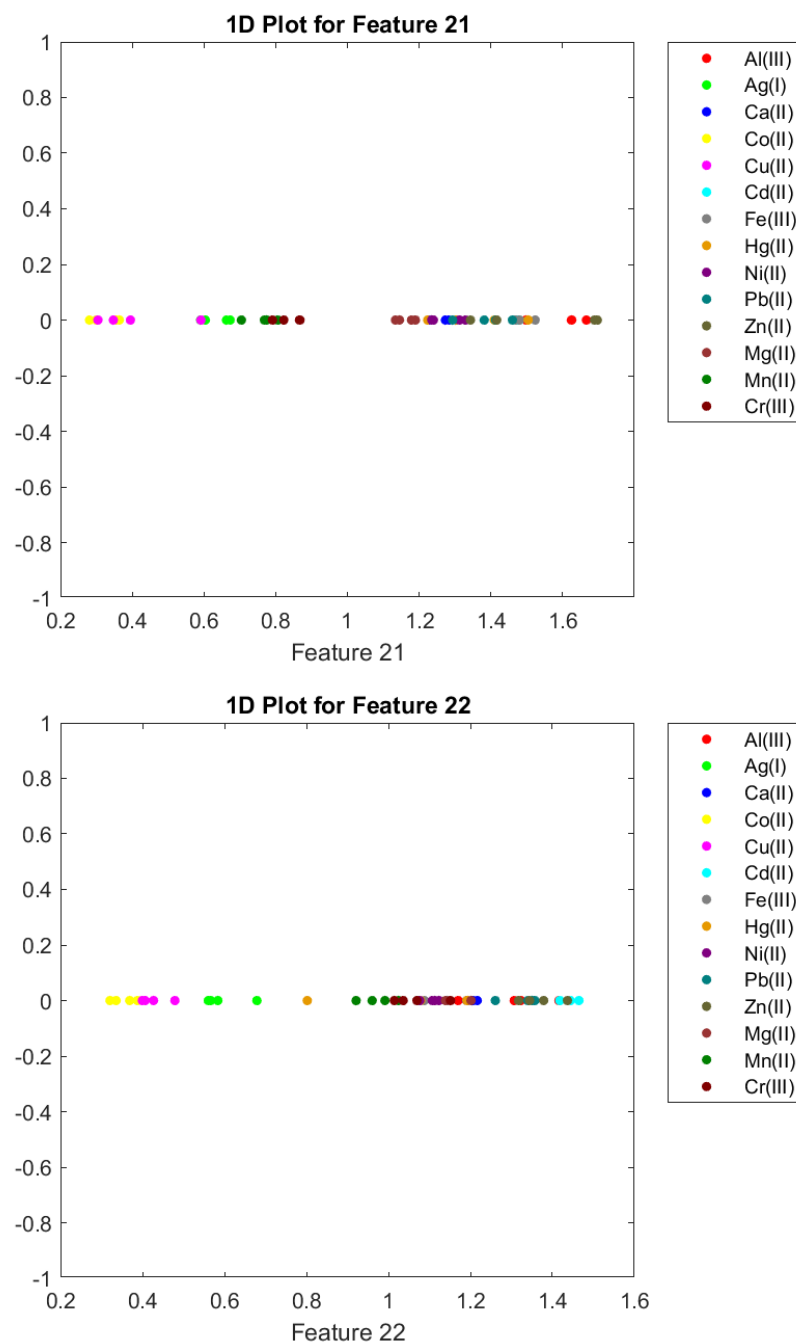


Figure S5. 3 Plots of one CD based LDA classification

7.3.3.5 *MATLAB based LDA classifier (C) for four features-based CDs array plotting*

Aims and working mechanism

MATLAB LDA classifier (C) is devised to execute Linear Discriminant Analysis (LDA) on a dataset comprising features for 14 distinct metal ions, each represented by four replicates. The overarching goal is to scrutinize the dataset's features in quartets to pinpoint combinations that yield impeccable classification (100% accuracy) of the metal ions. An elaborate elucidation of the script's functionalities is provided below:

- **Environment Setup and Data Preparation:** Commencing operations, the script generates a directory for archiving results, assuming its nonexistence. Following this, it loads the dataset from 'data.xlsx', anticipated to encompass measurements pertinent to metal ions. Despite discussions around normalizing the feature data, the actual line of code for normalization remains commented out.
- **Colour and Label Preparation:** A colour scheme is established to visually differentiate the 14 metal ions in subsequent plots, and meaningful labels are attributed to each metal ion category to facilitate the interpretation of outcomes.
- **Feature Combination Selection:** Leveraging the 'nchoosek' function, the script concocts all conceivable combinations of four features out of the 22 available. It meticulously iterates over these combinations to administer LDA.
- **LDA and Performance Evaluation:** For every combination of features, it undertakes the following:
 - Executes LDA classification on the collated dataset.
 - Utilizes the trained LDA model to prognosticate the class labels.

- Assesses the classification performance employing a tailored ‘CalculatePerformanceMetrics’ function, which tabulates accuracy, precision, recall, and F1 score.
- Visualization for Perfect Classifications: Upon a feature combination attaining 100% accuracy, the script visualizes the classification results by illustrating 2D projections of the data for each feature pair within the combination. It employs another bespoke function, ‘plot2DProjectionsAndEllipses’, for rendering these plots and for circumscribing confidence ellipses around the data points of each metal ion, thereby accentuating the class distinctions.
- 2D Projection and Confidence Ellipse Drawing: The ‘plot2DProjectionsAndEllipses’ function orchestrates scatter plots for 2D projections of the feature combinations and delineates confidence ellipses around the clusters of points corresponding to each metal ion. These confidence ellipses furnish a visual testament to the data dispersion and classification confidence for each ion.
- Utility Functions: Included in the script are two ancillary utility functions:
 - ‘CalculatePerformanceMetrics’ ascertains the confusion matrix and extrapolates performance metrics therefrom.
 - ‘drawConfidenceEllipse’ computes and illustrates confidence ellipses for the data points of each metal ion in the 2D projection plots, offering insights into data distribution and inter-class overlaps.

Script and functions

```
clear;
files = 'data.xlsx';
resultsDir = ['Newn_FourD_LDA_results'];
if ~exist(resultsDir, 'dir')
    mkdir(resultsDir); % Create results directory if it does not exist
end
```

```

[data, txt, raw] = xlsread(files);
label = repelem(1:14,4); % 14 metal ions, each with 4 replicates
%data(:, 1:22) = normalize(data(:, 1:22)); % Normalize the feature data

colors = [
    1, 0, 0; % Red
    0, 1, 0; % Green
    0, 0, 1; % Blue
    1, 1, 0; % Yellow
    1, 0, 1; % Magenta
    0, 1, 1; % Cyan
    0.5, 0.5, 0.5; % Gray
    0.9, 0.6, 0; % Orange
    0.5, 0, 0.5; % Purple
    0, 0.5, 0.5; % Teal
    0.4, 0.4, 0.2; % Olive
    0.6, 0.2, 0.2; % Brown
    0, 0.5, 0; % Dark Green
    0.5, 0, 0; % Maroon
]; % MATLAB's 'lines' colormap for distinct colors
sensingTargets = {'Al(III)', 'Ag(I)', 'Ca(II)', 'Co(II)', 'Cu(II)', 'Cd(II)', 'Fe(III)', 'Hg(II)', 'Ni(II)', 'Pb(II)', 'Zn(II)',
'Mg(II)', 'Mn(II)', 'Cr(III)'};

% Select 4 features for LDA classification
C = nchoosek(1:22, 4); % All combinations of 4 features out of 22

for i = 1:size(C, 1)
    feature_select = C(i, :);
    feature_data = data(:, feature_select);

    % Perform LDA and check for 100% accuracy
    lda = fitcdiscr(feature_data, label);
    predicted_label = predict(lda, feature_data);
    metrics = CalculatePerformanceMetrics(label, predicted_label);

    if metrics(1) == 1.0 % If accuracy is 100%
        % Plot 2D projections, draw ellipses, and calculate properties
        plot2DProjectionsAndEllipses(lda, feature_data, label, sensingTargets, colors, feature_select, resultsDir);
    end
end

function metrics = CalculatePerformanceMetrics(true_labels, predicted_labels)
    C = confusionmat(true_labels, predicted_labels);
    TP = diag(C);
    FP = sum(C, 1)' - TP;
    FN = sum(C, 2) - TP;
    TN = sum(C(:)) - TP - FP - FN;
    Accuracy = sum(TP) / sum(C(:));
    Precision = mean(TP ./ (TP + FP));
    Recall = mean(TP ./ (TP + FN));
    F1 = 2 * (Precision * Recall) / (Precision + Recall);
    metrics = [Accuracy, Precision, Recall, F1];
end

function plot2DProjectionsAndEllipses(lda, feature_data, label, sensingTargets, colors, feature_select,
resultsDir)
    combinations = [1 2; 2 3; 1 3];

```

```

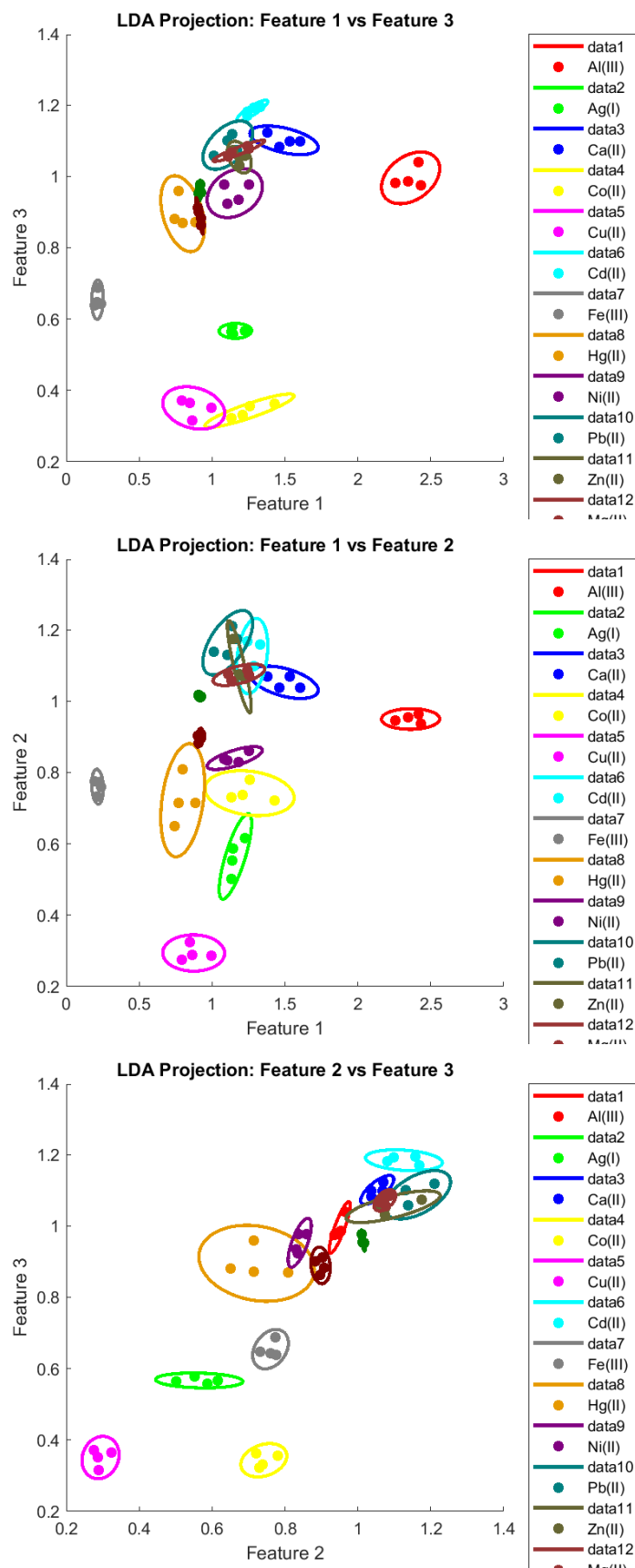
for combo = combinations'
    hFig = figure;
    hold on;
    for i = 1:length(sensingTargets)
        idx = label == i;
        [area, center] = drawConfidenceEllipse(feature_data(idx, combo), colors(i,:));
        scatter(feature_data(idx, combo(1)), feature_data(idx, combo(2)), 36, 'filled', 'MarkerEdgeColor',
colors(i,:), 'MarkerFaceColor', colors(i,:), 'DisplayName', sensingTargets{i});
    end
    xlabel(sprintf('Feature %d', combo(1)));
    ylabel(sprintf('Feature %d', combo(2)));
    title(sprintf('LDA Projection: Feature %d vs Feature %d', combo(1), combo(2)));
    legend('Location', 'eastoutside', 'Orientation', 'vertical');
    hold off;
    saveas(hFig, fullfile(resultsDir, sprintf('LDA_Projection_Features_%s_Features_%d_vs_%d.png',
mat2str(feature_select), combo(1), combo(2))));
    close(hFig); % Close the figure window after saving the plot
end
end

function [area, center] = drawConfidenceEllipse(data, color)
    meanData = mean(data, 1);
    [eigenvec, eigenval] = eig(cov(data));
    angle = atan2(eigenvec(2,1), eigenvec(1,1));
    chiSquareVal = 5.991;
    theta = linspace(0, 2*pi, 100);
    radiusX = sqrt(chiSquareVal*eigenval(1,1));
    radiusY = sqrt(chiSquareVal*eigenval(2,2));
    ellipseX = radiusX * cos(theta);
    ellipseY = radiusY * sin(theta);
    rotationMatrix = [cos(angle) -sin(angle); sin(angle) cos(angle)];
    ellipse = rotationMatrix * [ellipseX; ellipseY];
    plot(ellipse(1,:) + meanData(1), ellipse(2,:) + meanData(2), 'Color', color, 'LineWidth', 2);
    area = pi * radiusX * radiusY;
    center = meanData;
end

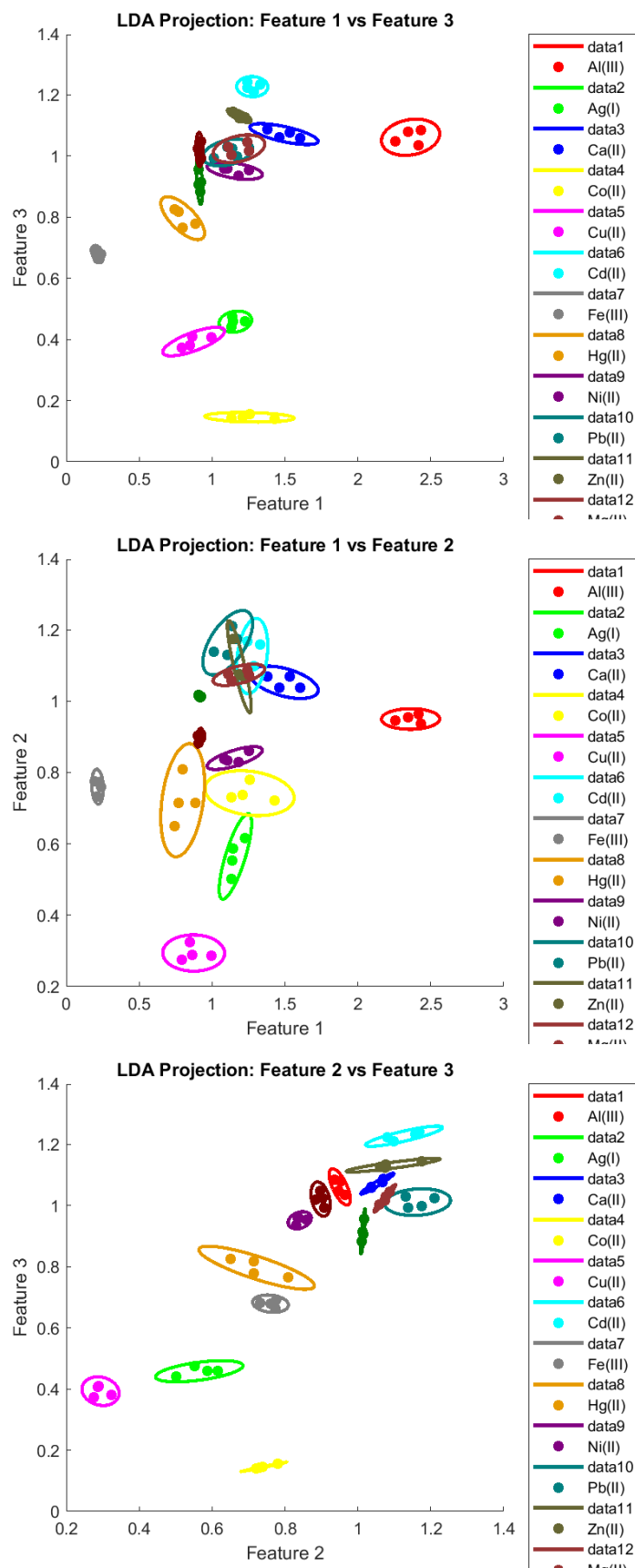
```

Results of 2D plots

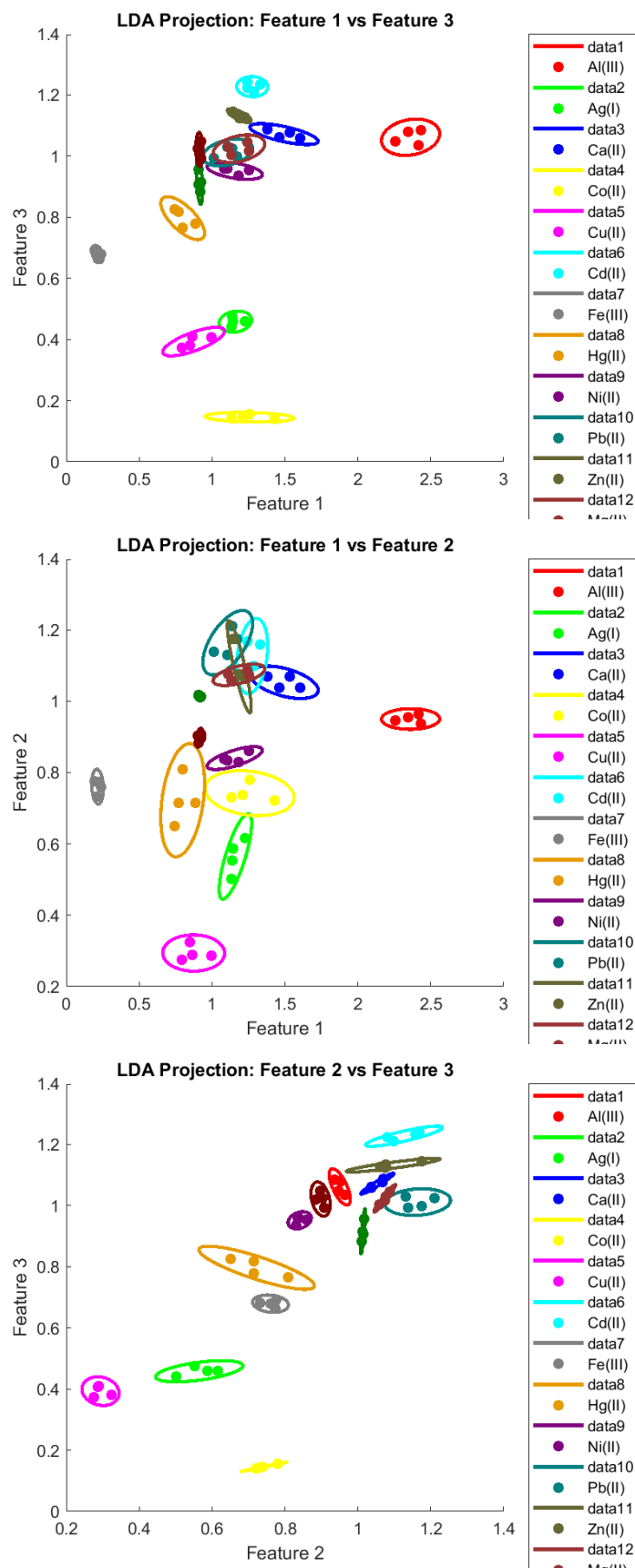
Feature Set: [1 8 9 11]



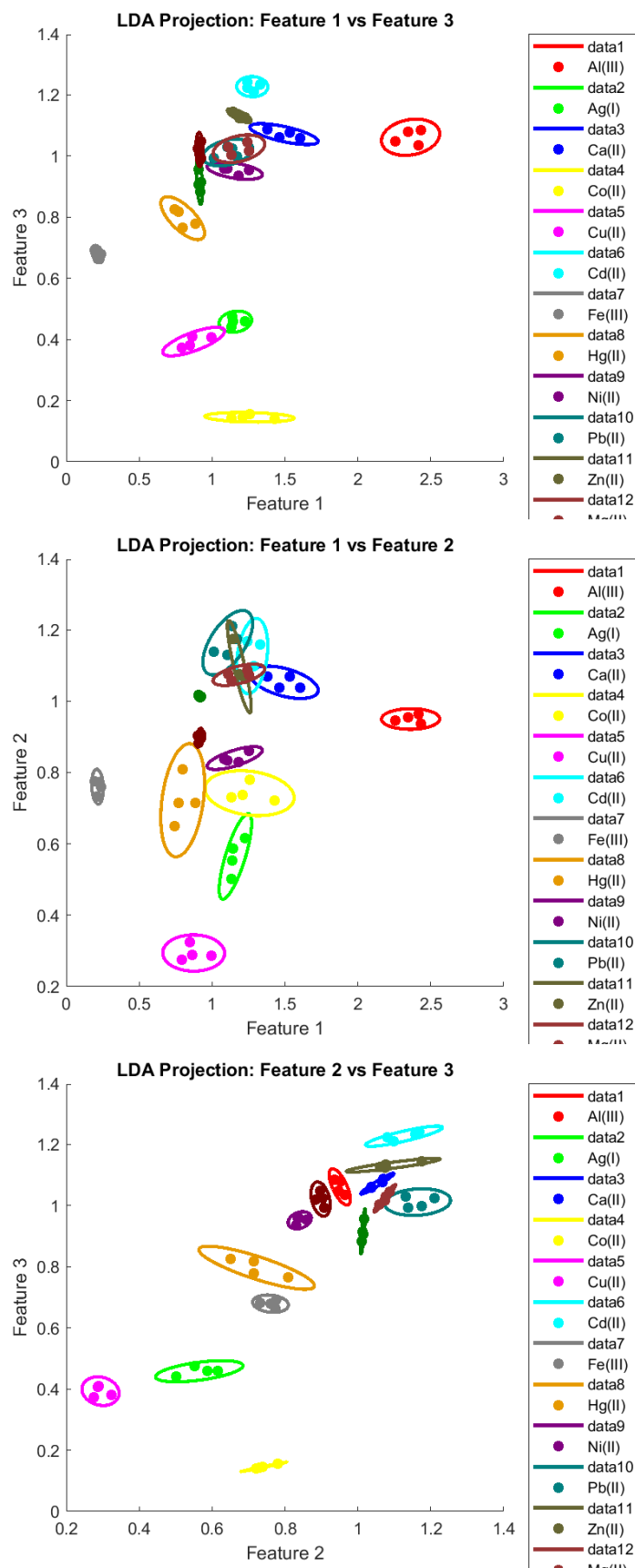
Feature Set: [1 8 11 13]

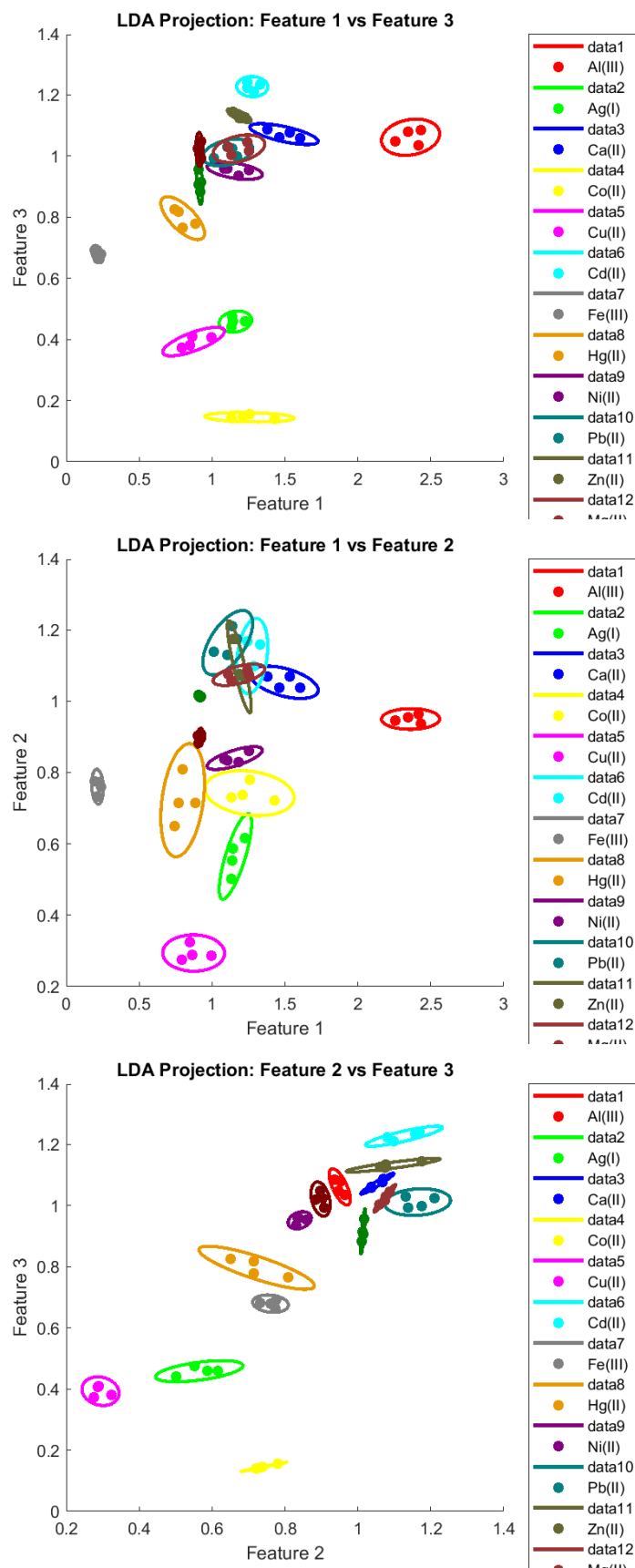


Feature Set: [1 8 11 15]

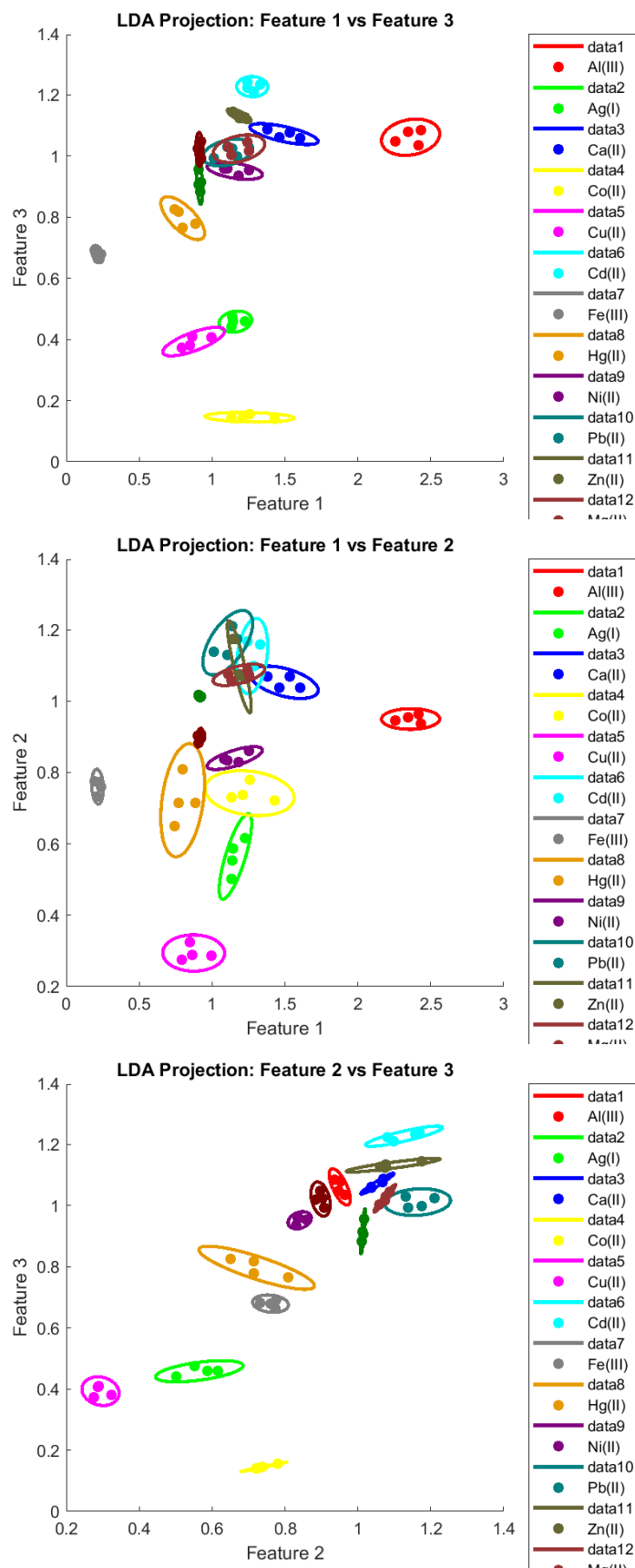


Feature Set: [1 8 11 19]

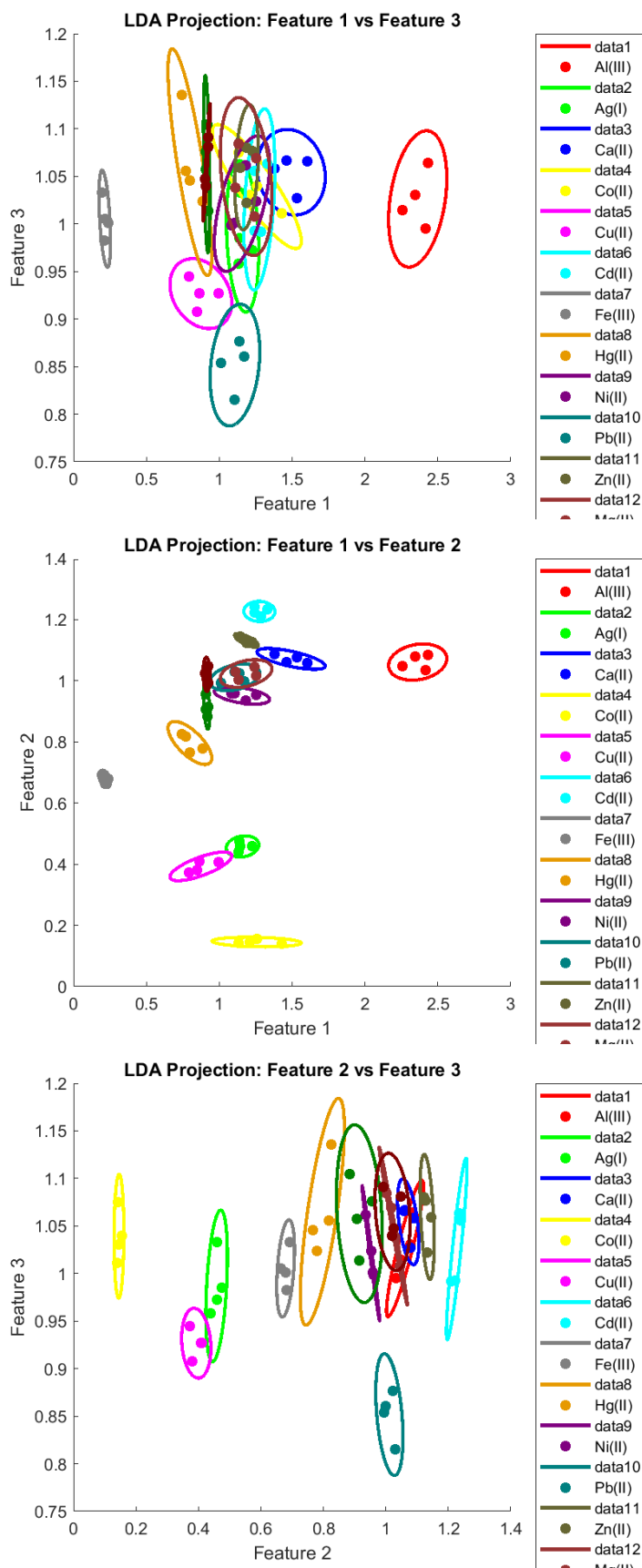




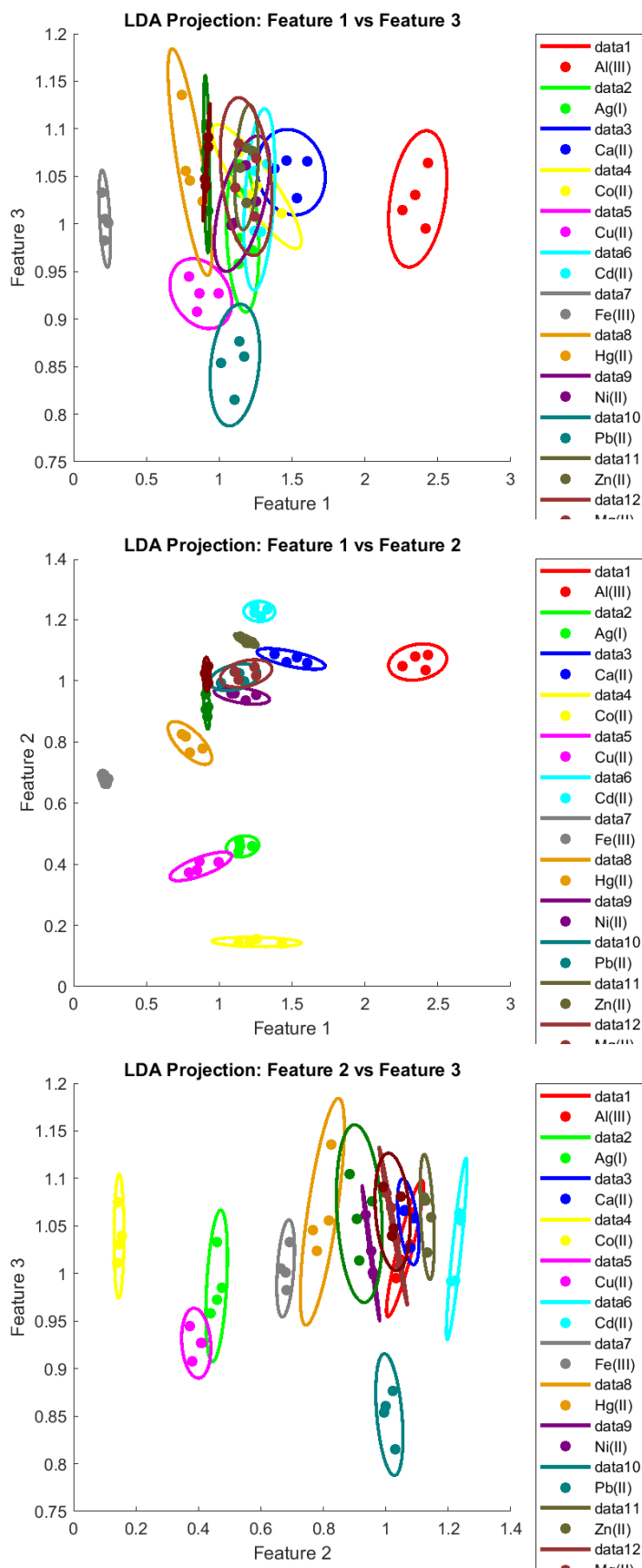
Feature Set: [1 8 11 22]

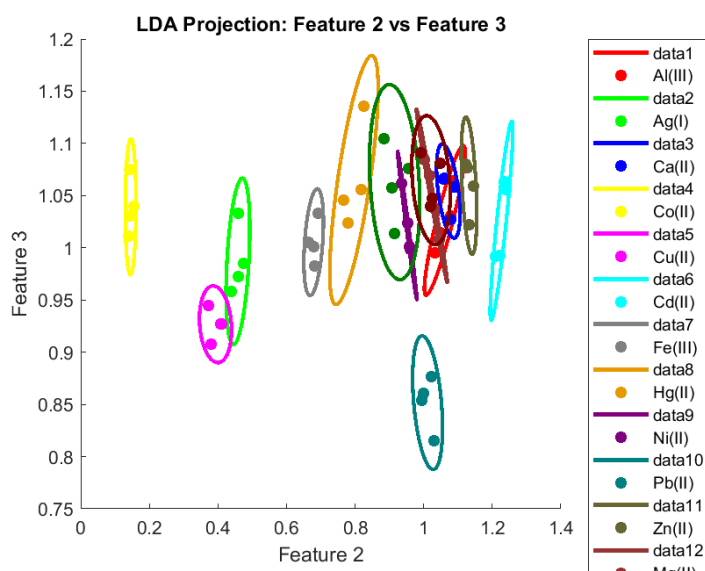
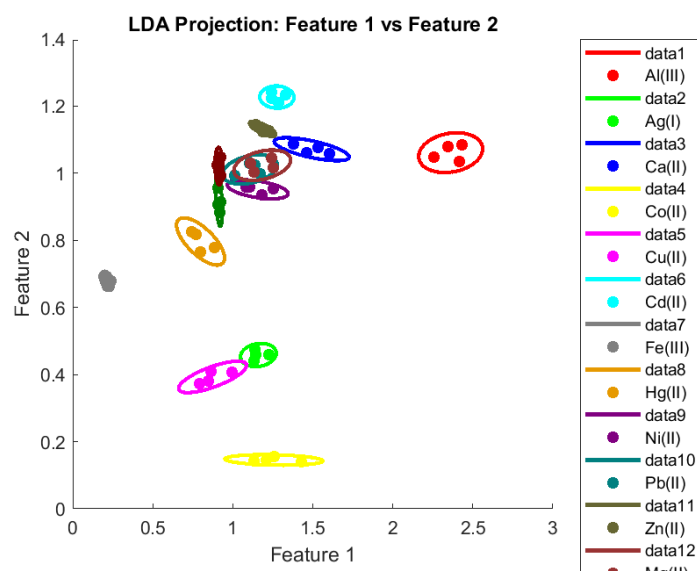
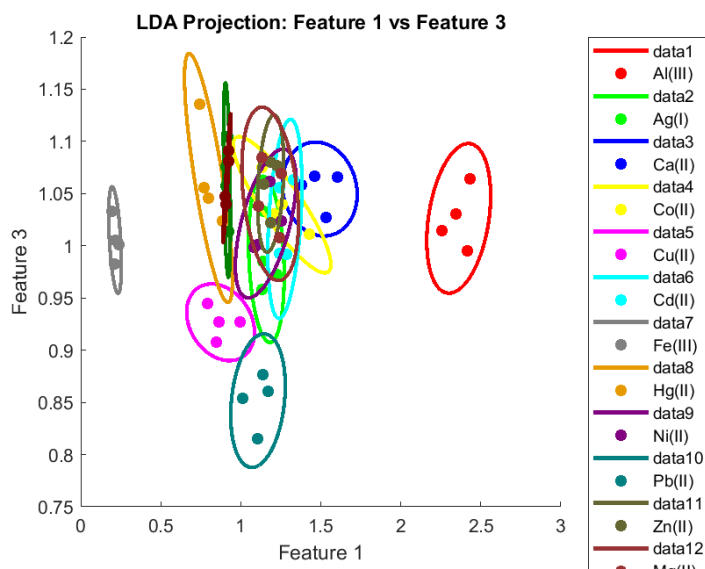


Feature Set: [1 11 13 14]

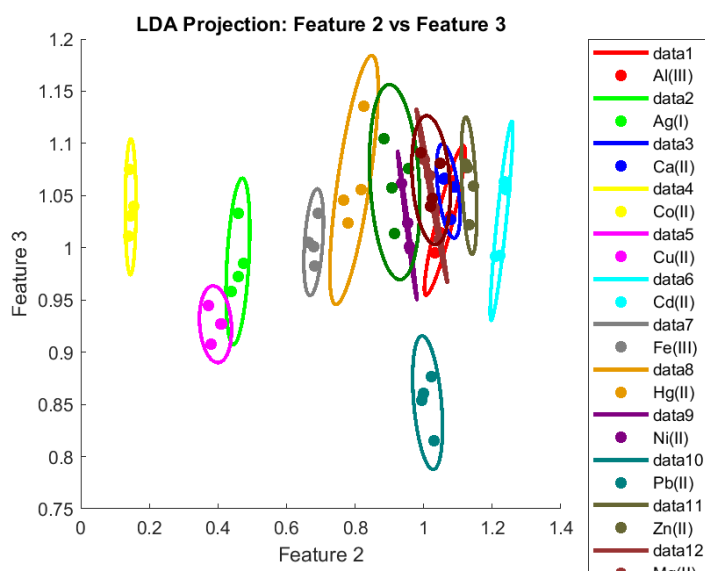
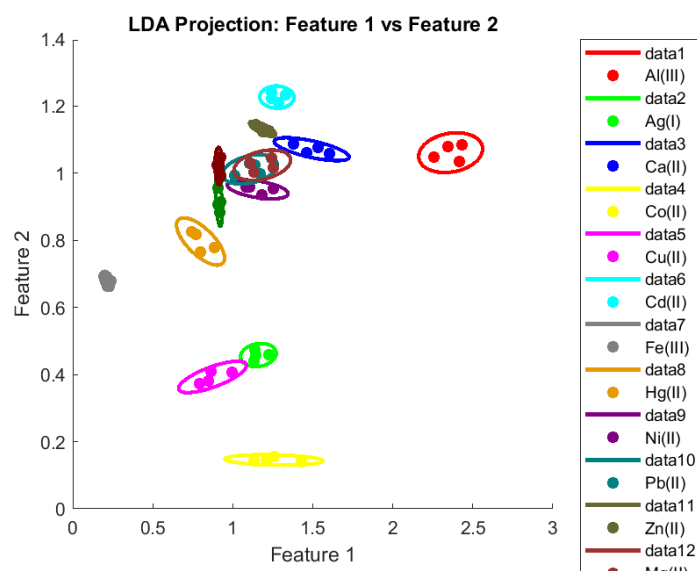
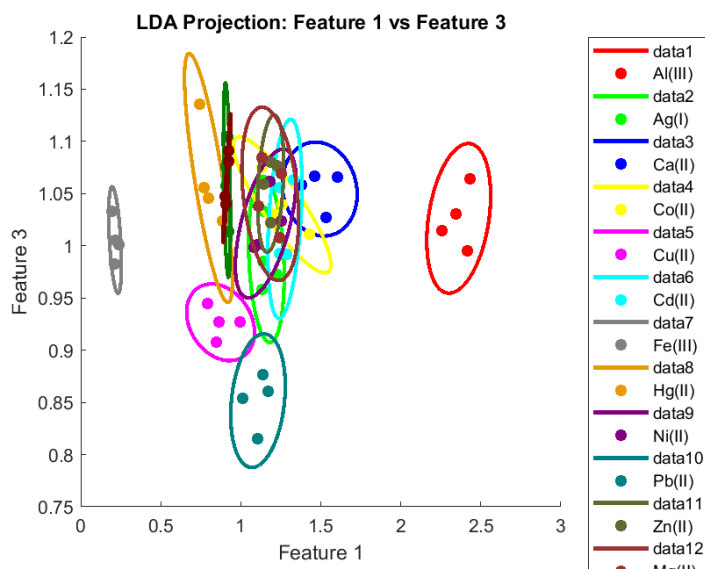


Feature Set: [1 11 13 17]

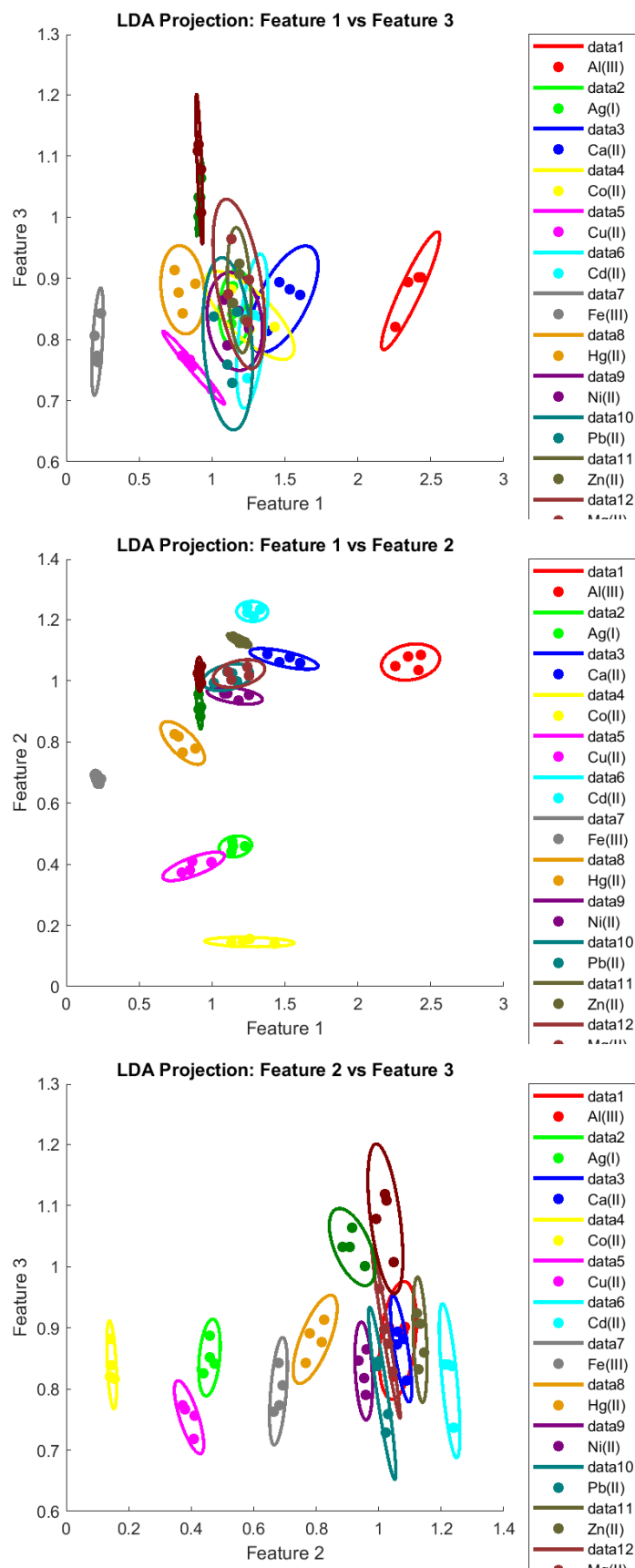




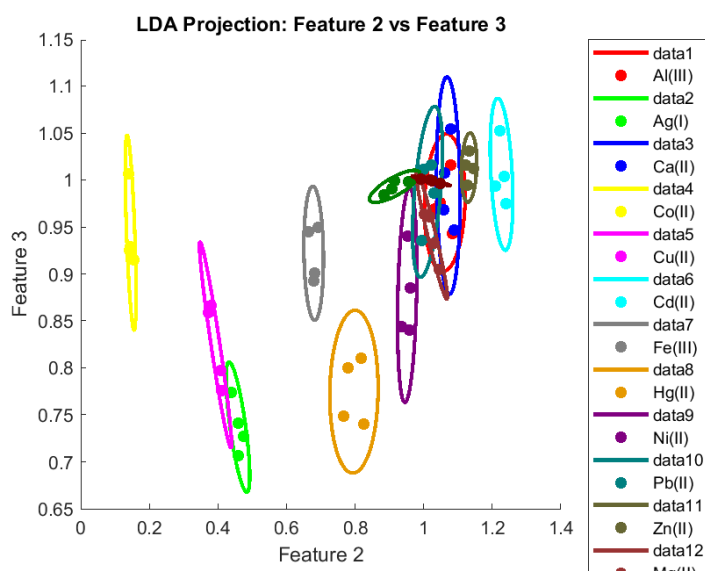
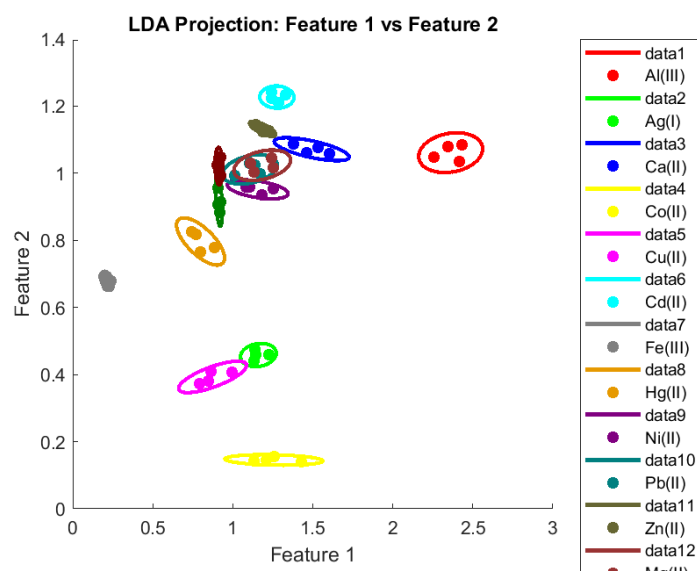
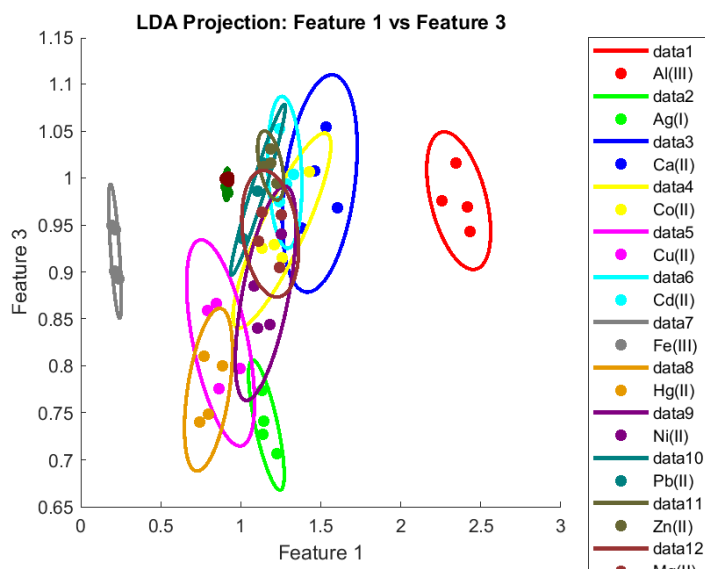
Feature Set: [1 11 13 21]



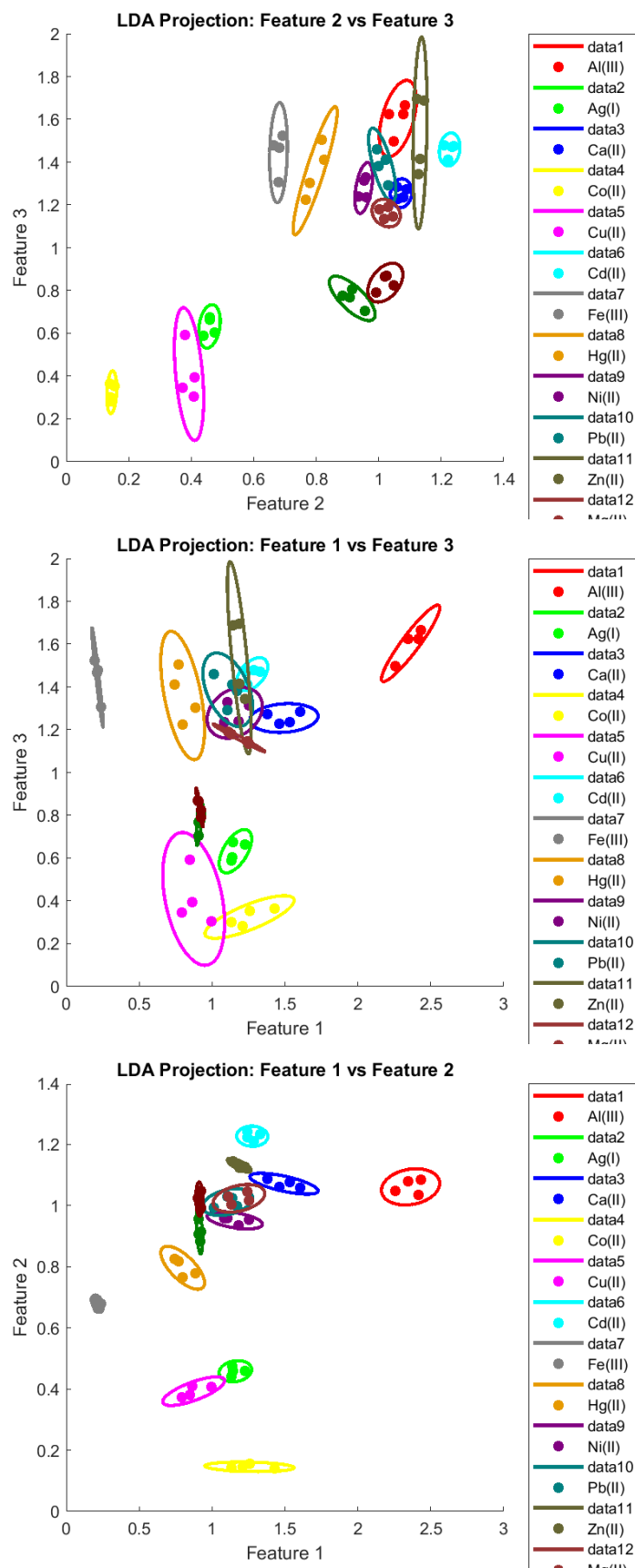
Feature Set: [1 11 14 20]

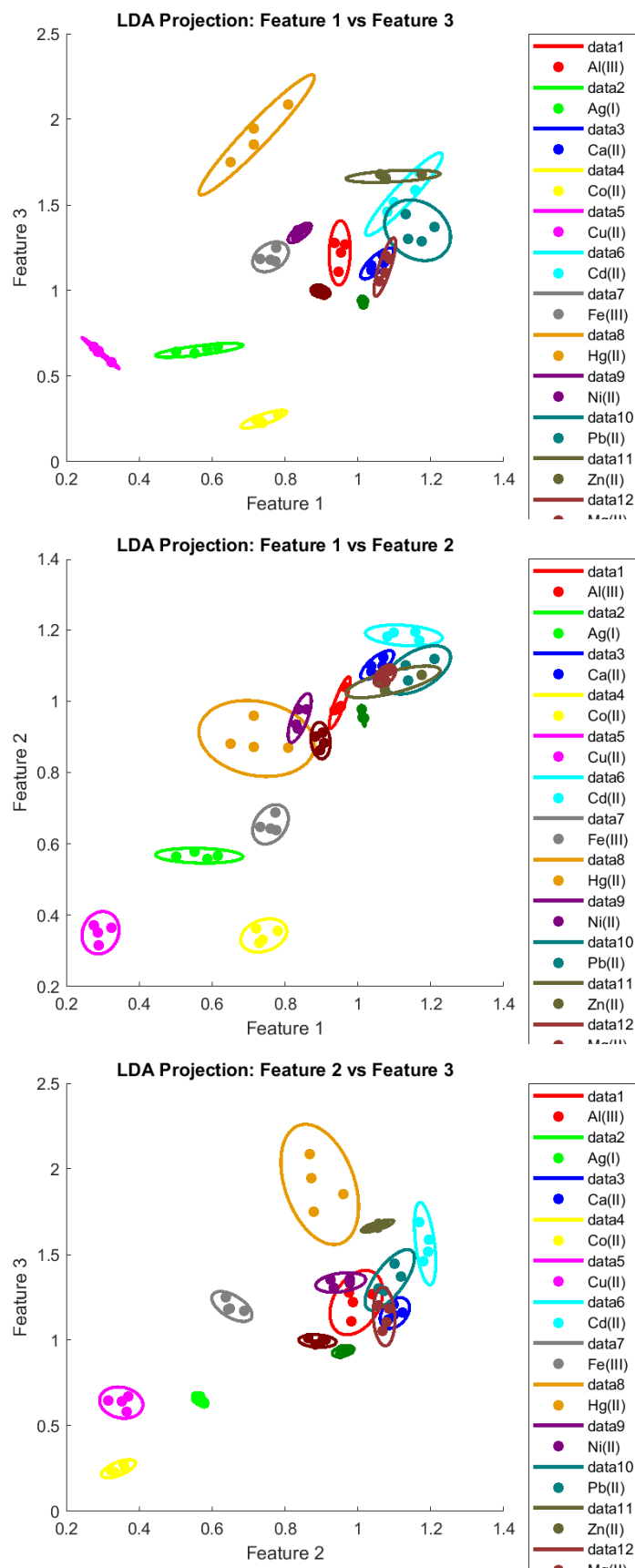


Feature Set: [1 11 15 22]

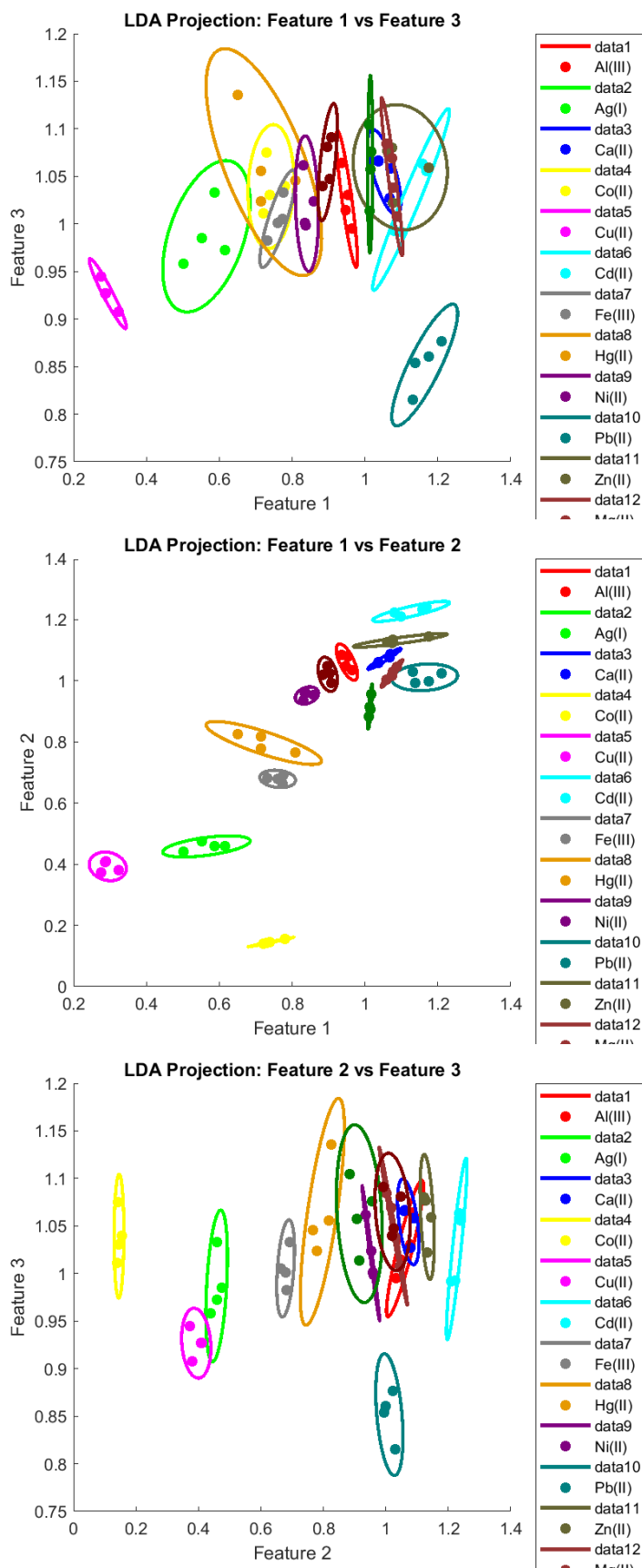


Feature Set: [1 11 21 22]

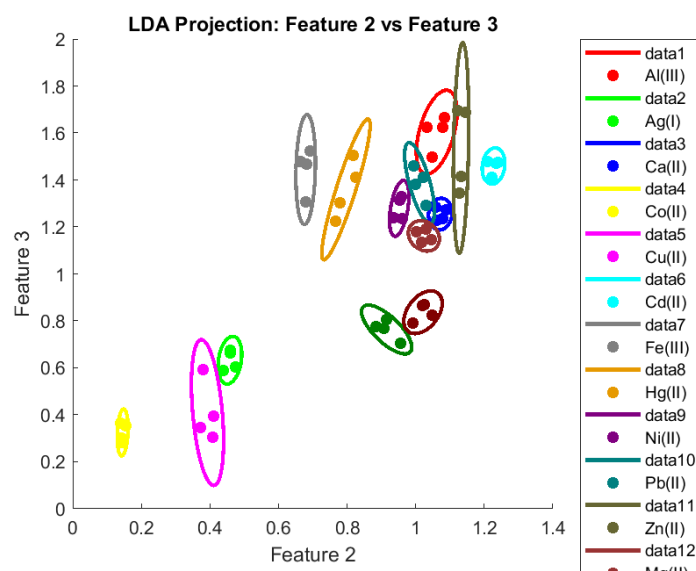
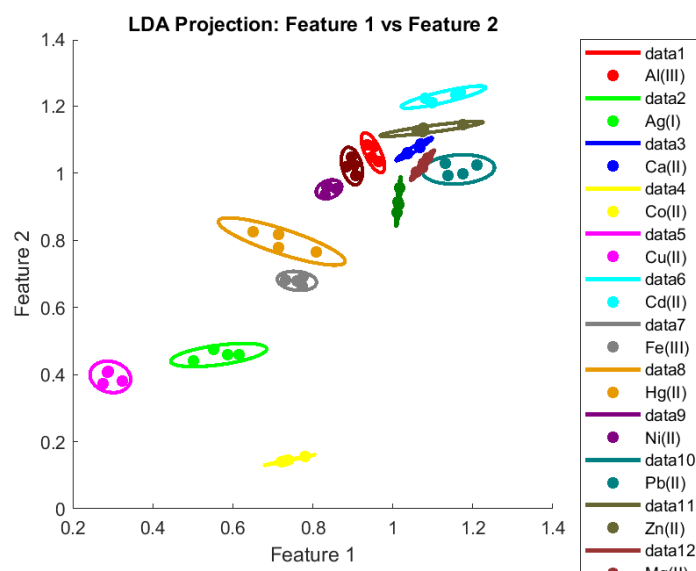
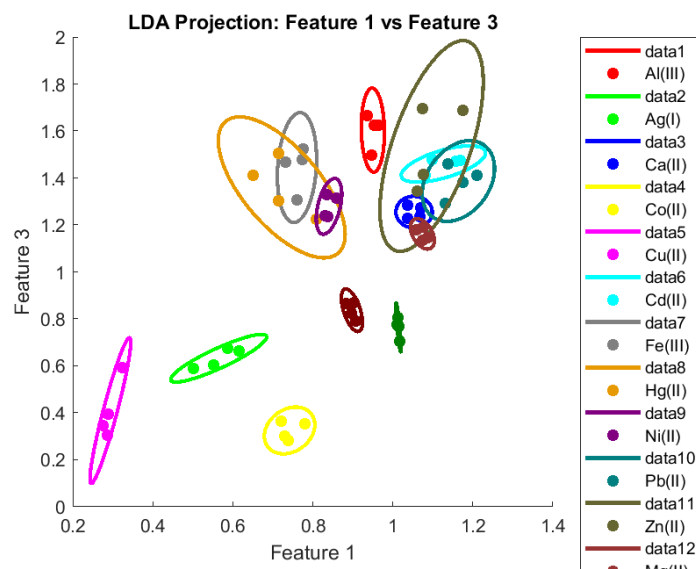




Feature Set: [8 11 13 15]



Feature Set: [8 11 21 22]



7.3.3.6 *MATLAB based LDA classifier (D) for four features-based CDs array classification evaluation by the distances between analyse groups and size of analyse groups*

Aims and working mechanism

MATLAB LDA (D) is designed to leverage Linear Discriminant Analysis (LDA) for a detailed examination of a dataset that includes 22 features representing 14 metal ions, each ion depicted by four replicates. The primary objective is to discern the combinations of four features that enable a flawless classification (100% accuracy) of these metal ions. Upon identifying such combinations, the script undertakes an in-depth analysis and visualization of the data in a reduced-dimensional space, with a particular focus on the properties and distances of ellipsoids that encapsulate the data points for each class within a three-dimensional projection space. The script's architecture and its core functionalities are elucidated as follows:

- Data Preparation and LDA
 - Initiates by loading the dataset from 'data.xlsx' and setting up labels for the 14 metal ions.
 - It meticulously cycles through all possible combinations of four features out of the twenty-two, applying LDA to each set to pinpoint those achieving 100% classification accuracy.
- Feature Combination Directory Creation
 - For each combination that attains perfect accuracy, the script autonomously generates a unique directory within a predetermined results directory, thereby systematizing the storage of results pertinent to each successful feature set.
- Ellipsoid Calculation and Visualization

- A function matrix derived from the LDA model projects the data into a space delineated by the linear discriminants, with the script focusing on the first three principal functions for visualization.
 - It calculates ellipsoids in this tri-dimensional space that enshroud the data points of each class, computing their centres and volumes. These ellipsoids illuminate the distribution and relative positioning of the classes within the reduced space, shedding light on the separation efficacy of the LDA.
- Saving Results
- The properties of these ellipsoids, including centres and volumes, are documented into Excel files within the directory allocated for each successful feature combination. The script generates two pivotal files:
 - ‘EllipsoidVolumes.xlsx’: Chronicles the volumes of the ellipsoids corresponding to each metal ion, providing a quantitative insight into the dispersion of each class in the reduced-dimensional space.
 - ‘EllipsoidCenterDistances.xlsx’: Catalogs the pairwise distances between the ellipsoid centers, elucidating the class separation within the reduced-dimensional space.
- Utility Functions
- CalculatePerformanceMetrics: Engages in the computation of accuracy, precision, recall, and F1 score to assess the classification's effectiveness.
 - CalculateFunctionMatrix: Fabricates a matrix that embodies the dataset within the linear discriminants' space.
 - CalculateEllipsoids: Determines the centres and volumes of ellipsoids for the projected data points of each class.
 - CalculateConfidenceEllipsoid: Derives the centre and volume of a confidence ellipsoid for a specific dataset in three-dimensional space.

- SaveEllipsoidPropertiesAndDistances: Archives the calculated ellipsoid properties and distances into Excel files, facilitating straightforward access and subsequent analysis.

Script and functions

```

clear;
files = 'data.xlsx';
resultsDir = 'New_areadistance_FourD_LDA_results';
if ~exist(resultsDir, 'dir')
    mkdir(resultsDir); % Create results directory if it does not exist
end

[data, ~, ~] = xlsread(files);
label = repelem(1:14,4); % 14 metal ions, each with 4 replicates

% Select 4 features for LDA classification
C = nchoosek(1:22, 4); % All combinations of 4 features out of 22

for i = 1:size(C, 1)
    feature_select = C(i, :);
    feature_data = data(:, feature_select);

    lda = fittediscr(feature_data, label);
    predicted_label = predict(lda, feature_data);
    metrics = CalculatePerformanceMetrics(label, predicted_label);

    if metrics(1) == 1.0 % If accuracy is 100%
        % Create a directory for this feature combination
        featureDirName = sprintf('Features_%d_%d_%d_%d', feature_select);
        featureDirPath = fullfile(resultsDir, featureDirName);
        if ~exist(featureDirPath, 'dir')
            mkdir(featureDirPath); % Create the directory
        end

        functionMatrix = calculateFunctionMatrix(lda, feature_data);
        principalFunctions = functionMatrix(:, 1:3); % Use the first three principal functions
        [centers, volumes] = calculateEllipsoids(principalFunctions, label);
        saveEllipsoidPropertiesAndDistances(centers, volumes, label, featureDirPath); % Save in the new
    end
end

function metrics = CalculatePerformanceMetrics(true_labels, predicted_labels)
    C = confusionmat(true_labels, predicted_labels);
    TP = diag(C);
    FP = sum(C, 1)' - TP;
    FN = sum(C, 2) - TP;
    TN = sum(C(:)) - TP - FP - FN;
    Accuracy = sum(TP) / sum(C(:));
    Precision = mean(TP ./ (TP + FP));
    Recall = mean(TP ./ (TP + FN));
    F1 = 2 * (Precision * Recall) / (Precision + Recall);
    metrics = [Accuracy, Precision, Recall, F1];

```

```

end

function functionMatrix = calculateFunctionMatrix(lda, feature_data)
    numClasses = length(lda.ClassNames);
    coeffs = lda.Coeffs;
    numObservations = size(feature_data, 1);
    functionMatrix = zeros(numObservations, numClasses - 1);
    for i = 1:numClasses-1
        linearCoeffs = coeffs(i, min(i+1, numClasses)).Linear;
        const = coeffs(i, min(i+1, numClasses)).Const;
        functionMatrix(:, i) = feature_data * linearCoeffs + const;
    end
end

function [centers, volumes] = calculateEllipsoids(principalFunctions, label)
    sensingTargets = unique(label);
    numTargets = length(sensingTargets);
    centers = zeros(numTargets, 3);
    volumes = zeros(numTargets, 1);
    for i = 1:numTargets
        idx = label == sensingTargets(i);
        [volume, center] = calculateConfidenceEllipsoid(principalFunctions(idx, :));
        centers(i, :) = center;
        volumes(i) = volume;
    end
end

function [volume, center] = calculateConfidenceEllipsoid(data)
    meanData = mean(data, 1);
    covarianceMatrix = cov(data);
    [eigenvec, eigenval] = eig(covarianceMatrix);
    chiSquareVal = 6.251; % Chi-square value for 90% confidence in 3D
    axesLengths = sqrt(diag(eigenval) * chiSquareVal);
    volume = 4/3 * pi * prod(axesLengths);
    center = meanData;
end

function saveEllipsoidPropertiesAndDistances(centers, volumes, label, resultsDir)
    % Use unique labels to match volumes
    uniqueLabels = unique(label); % This assumes 'label' is the full list of labels, including duplicates
    % Convert numeric labels to string if necessary, for table variable names
    sensingTargets = cellstr(num2str(uniqueLabels)); % Adjust if your labels are already strings

    % Ensure volumes is correctly sized to match the number of unique labels/targets
    if length(volumes) ~= length(sensingTargets)
        error('Mismatch between the number of sensing targets and the number of calculated volumes.');
```

```

end

```

7.3.4 Array optimisation result based on SVM and LDA analysis

Table S5.6 Summary of the occurrences of each CDs involved in the array combinations with 100% classification accuracy given by both MATLAB based SVM classifier and LDA classifier

No.	Carbon dots	SVM occurrences	LDA occurrences	Occurrences of both SVM and LDA
1	CD_1-0_6 h	29	50	13
2	CD_9-1_6 h	0	3	0
3	CD_4-1_6 h	7	0	0
4	CD_4-2_6 h	0	14	0
5	CD_4-3_6 h	1	6	0
6	CD_4-4_6 h	2	0	0
7	CD_3-4_6 h	4	2	0
8	CD_2_4_6 h	17	27	9
9	CD_1-4_6 h	10	12	2
10	CD_3-17_6 h	14	22	1
11	CD_1-9_6 h	34	68	16
12	CD_1-0_8 h	0	4	0
13	CD_9-1_8 h	10	15	6
14	CD_4-1_8 h	2	12	2
15	CD_4-2_8 h	4	7	3
16	CD_4-3_8 h	1	6	0
17	CD_4-4_8 h	2	3	1
18	CD_3-4_8 h	0	1	0
19	CD_2_4_8 h	2	3	1
20	CD_1-4_8 h	9	12	3
21	CD_3-17_8 h	7	15	3
22	CD_1-9_8 h	9	10	4

Table S5.7 The volume of each metal ions' ellipsoid of each combination of four-CDs array with 100% accuracy for both SVM and LDA

Metal Ions	Four-CDs array combinations							
	1	2	3	4	5	6	7	8
Al(III)	11624.82	109612.3	571194.2	175501.5	103379.6	396230.4	88237.04	159500.3
Ag(I)	7600.145	143266.8	135119.1	252490.1	71093.4	180124.4	42751.68	61911.02
Ca(II)	4453.858	6592.301	350126.7	31544.96	88314.33	38672.84	18078.63	14191.61
Co(II)	1228.135	66594.97	28925.65	8820.178	130832.9	87126.44	13623.58	9812.063
Cu(II)	14754.65	188277.4	294406.1	186860	184368.6	178675.8	43204.76	51242.32
Cd(II)	2849.887	108051.8	23976.76	111481	133129.6	9176.108	33645.72	379.9569
Fe(III)	21543.79	35334.66	54466.79	74038.83	4731.36	31586.26	12918.64	5783.121
Hg(II)	20505.74	191196.7	614283.2	332034	287799.2	386370.2	100677.8	90306.36
Ni(II)	5245.578	23756.84	161492.7	18053.21	23669.77	32504.8	11107.64	16230.42
Pb(II)	58101.36	393722.9	422311.6	311089.5	462624.5	441107.5	42616.38	24533.76
Zn(II)	9348.277	66202.58	13674.69	20385.39	65312.73	18610.05	1588.83	15179.71
Mg(II)	2635.726	25325.52	81329.21	35999.41	6000.757	72575.96	3164.285	30426.03
Mn(II)	214.7811	20175.68	6118.368	1769.183	2515.137	3181.101	22435.75	9032.556
Cr(III)	5544.081	36987.5	20970.96	19619.15	13709.75	7603.649	2689.974	19214.2
Metal Ions	9	10	11	12	13	14	15	16
Al(III)	322322.4	85324.03	323798	501202	34264.39	93251.98	8435.663	65142.99
Ag(I)	100867.4	46433.72	147570.1	12852.76	9182.334	26740.21	21545.75	26654.79
Ca(II)	43744.03	20900.08	98956.47	301232.2	7457.226	14179.16	527.233	7675.117
Co(II)	158460.7	21951.47	24986.39	61603.94	2383.846	5901.217	6246.471	6315.511
Cu(II)	97304.47	93111.01	64101.11	129961.5	36292.24	97023.28	6116.607	23077.87
Cd(II)	82748.02	951.9494	45254.16	122858.1	752.2185	10510.08	15834.44	20286.21
Fe(III)	17791.76	8866.182	55805.72	21781.94	276.2095	22927.22	14706.66	41882.09
Hg(II)	90949.38	104796.1	81729.43	117013.4	24714.47	96091.51	80200.01	126060.2
Ni(II)	5077.237	11719.38	84156.05	124498.8	6926.881	19320.98	7199.211	21013.47
Pb(II)	310411.8	26229.88	109141.1	76639.89	20085.18	8809.207	47885.22	159004.2
Zn(II)	31538.9	25669.22	3524.081	871.8873	5412.452	5417.953	12000.71	2999.376
Mg(II)	8503.902	9916.042	24991.83	42561.3	2839.633	4377.768	988.5599	4282.173
Mn(II)	15582.27	27485.9	11132.25	6764.078	3524.631	17094.66	854.6358	13596.81
Cr(III)	875.4488	2059.704	6449.751	2452.081	62.84968	52833.13	538.2882	30331.36

7.3.5 Array sensing application

7.3.5.1 The optimised final four-CDs array combination

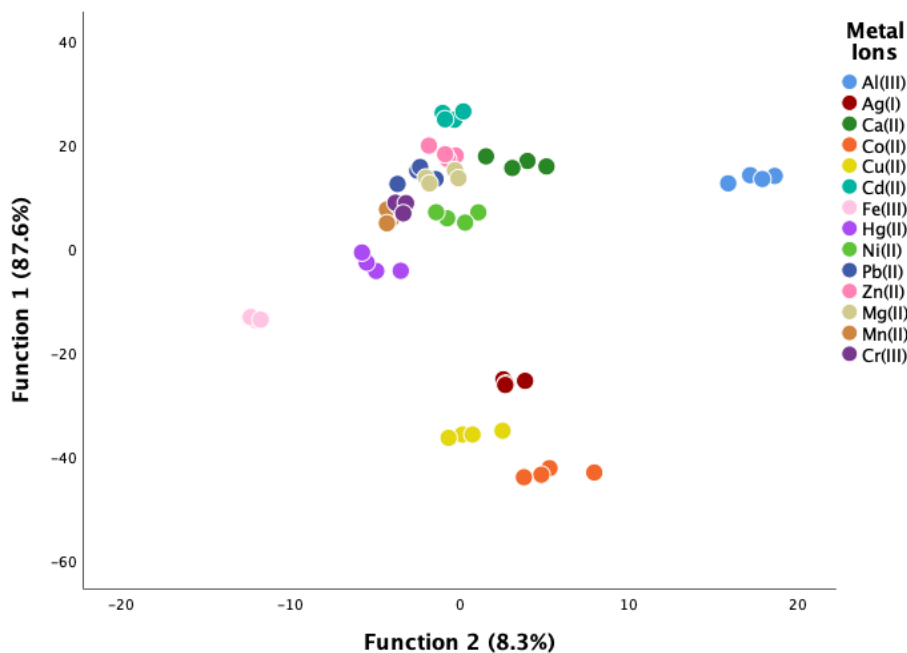


Figure S5.1 2D LDA score plots with the first two functions for the analysis of 14 metal ions ($80\ \mu\text{M}$ metal ion, 1mg/mL CDs ($\lambda_{\text{ex}} = 370\ \text{nm}$, $\lambda_{\text{em}} = 390\text{--}750\ \text{nm}$)) in buffered Milli-Q water ($20\ \text{mM}$, $\text{pH } 7.0$) by using CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h based array

Table S5.8 LDA classification results using CD_1-0_6 h, CD_2-4_6 h, CD_1-4_6 h, and CD_1-9_6 h based array

Classification Results ^{a,c}																	
		Predicted Group Membership															
		Metal ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4

Cross-validate d ^b		Mg(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	100	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	100	100
	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	4
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	4	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	4	4
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	100

		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	0	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	0	0	0	0	0	0	0	0	0	100	0	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	100	0	100
		Cr(III)	0	0	0	0	0	0	0	0	0	0	0	0	0	100	100
a. 100.0% of original grouped cases correctly classified.																	
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																	
c. 100.0% of cross-validated grouped cases correctly classified.																	

7.3.5.2 Array application on the metal classification in lack environmental sample

Table S5.9 LDA classification results using 4-CDs-based array for environmental application with lake sample

Classification Results ^{a,c}																	
		Predicted Group Membership															
		Metal Ions	Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Pb(II)	Zn(II)	Mg(II)	Mn(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	2	0	0	0	0	0	0	0	0	2	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	3	0	0	0	1	0	0	0	0	0	4
		Cd(II)	0	0	0	0	0	4	0	0	0	0	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	0	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	3	0	0	1	0	0	0	4
		Ni(II)	0	0	0	0	0	0	0	0	4	0	0	0	0	0	4
		Pb(II)	0	0	0	0	0	0	0	0	0	4	0	0	0	0	4
		Zn(II)	0	0	0	0	0	0	0	0	0	0	4	0	0	0	4
		Mg(II)	0	0	0	0	0	0	0	1	0	0	0	3	0	0	4
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	4	0	4
		Cr(III)	0	0	0	0	0	1	0	0	0	0	0	0	0	3	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	50	0	0	0	0	0	0	0	0	50	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	75	0	0	0	25	0	0	0	0	0	100
		Cd(II)	0	0	0	0	0	100	0	0	0	0	0	0	0	0	100

		Fe(III)	0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	75	0	0	25	0	0	0	100
		Ni(II)	0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
		Pb(II)	0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
		Zn(II)	0	0	0	0	0	0	0	0	0	0	100	0	0	0	100
		Mg(II)	0	0	0	0	0	0	0	25	0	0	0	75	0	0	100
		Mn(II)	0	0	0	0	0	0	0	0	0	0	0	0	100	0	100
		Cr(III)	0	0	0	0	0	25	0	0	0	0	0	0	0	75	100
		Cross-validated ^b	Count	Al(III)	3	0	0	0	0	0	0	0	0	0	0	0	0
Ag(I)	0			4	0	0	0	0	0	0	0	0	0	0	0	0	4
Ca(II)	0			0	1	0	0	0	0	0	0	0	1	2	0	0	4
Co(II)	0			0	0	4	0	0	0	0	0	0	0	0	0	0	4
Cu(II)	0			0	0	0	3	0	0	0	1	0	0	0	0	0	4
Cd(II)	0			0	0	0	0	4	0	0	0	0	0	0	0	0	4
Fe(III)	0			0	0	0	0	0	4	0	0	0	0	0	0	0	4
Hg(II)	0			0	1	0	0	1	0	0	0	0	1	1	0	0	4
Ni(II)	0			0	0	0	0	0	0	0	4	0	0	0	0	0	4
Pb(II)	0			0	0	0	0	0	0	0	0	4	0	0	0	0	4
Zn(II)	0			0	1	0	0	0	0	2	0	0	1	0	0	0	4
Mg(II)	0			0	1	0	0	0	0	1	0	1	0	1	0	0	4
Mn(II)	0			0	0	0	0	0	0	0	0	0	0	0	4	0	4
Cr(III)	0			0	0	0	0	1	0	0	0	0	0	0	1	2	4
%	Al(III)		75	0	0	0	0	0	0	0	0	0	0	0	0	25	100
	Ag(I)		0	100	0	0	0	0	0	0	0	0	0	0	0	0	100
	Ca(II)		0	0	25	0	0	0	0	0	0	0	25	50	0	0	100
	Co(II)		0	0	0	100	0	0	0	0	0	0	0	0	0	0	100
	Cu(II)		0	0	0	0	75	0	0	0	25	0	0	0	0	0	100
	Cd(II)		0	0	0	0	0	100	0	0	0	0	0	0	0	0	100
	Fe(III)		0	0	0	0	0	0	100	0	0	0	0	0	0	0	100
	Hg(II)		0	0	25	0	0	25	0	0	0	0	25	25	0	0	100
	Ni(II)		0	0	0	0	0	0	0	0	100	0	0	0	0	0	100
	Pb(II)		0	0	0	0	0	0	0	0	0	100	0	0	0	0	100
	Zn(II)		0	0	25	0	0	0	0	50	0	0	25	0	0	0	100
	Mg(II)		0	0	25	0	0	0	0	25	0	25	0	25	0	0	100
	Mn(II)		0	0	0	0	0	0	0	0	0	0	0	0	100	0	100
	Cr(III)		0	0	0	0	0	25	0	0	0	0	0	0	25	50	100
a. 89.3% of original grouped cases correctly classified.																	
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.																	
c. 69.6% of cross-validated grouped cases correctly classified.																	

Table S5.10 Summary of canonical discriminant functions of 4-CDs-based array for environmental application with lake sample

Summary of Canonical Discriminant Functions									
Eigenvalues					Wilks' Lambda				
Function	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation	Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1	50.990 ^a	67.9	67.9	0.99	1 through 4	0	373.786	5	<.001
2	22.627 ^a	30.1	98	0.979	2 through 4	0.015	192.038	3	<.001
3	1.319 ^a	1.8	99.8	0.754	3 through 4	0.363	46.568	2	0.002
4	.187 ^a	0.2	100	0.396	4	0.843	7.866	1	0.042
a. First 4 canonical discriminant functions were used in the analysis.									

Table S5.11 Standardized canonical discriminant function coefficients of 4-CDs-based array for environmental application with lake sample

Standardized Canonical Discriminant Function Coefficients				
CDs	Functions			
	1	2	3	4
CD_1-0_6 h	0.151	0.207	0.971	-0.045
CD_2-4_6 h	0.361	0.951	-0.265	0.066
CD_1-4_6 h	0.859	-0.197	-0.075	-0.53
CD_1-9_6 h	0.567	-0.542	0.167	0.71

Table S5.12 Structure matrix of 4-CDs-based array for environmental application with lake sample

Structure Matrix				
CDs	Functions			
	1	2	3	4
CD_1-0_6 h	0.392	.817*	-0.218	0.362
CD_2-4_6 h	0.078	0.246	.961*	-0.101
CD_1-4_6 h	0.472	-0.234	0.016	.850*
CD_1-9_6 h	0.674	-0.232	-0.088	-.695*
Pooled within-groups correlations between discriminating variables and standardized canonical discriminant functions				
Variables ordered by absolute size of correlation within function.				
*. Largest absolute correlation between each variable and any discriminant function				

7.3.5.3 Array application on the metal classification in urine biological sample

Table S5.13 LDA classification results using 4-CDs-based array for biological application with urine sample

Classification Results ^{a,c}													
Predicted Group Membership													
			Al(III)	Ag(I)	Ca(II)	Co(II)	Cu(II)	Cd(II)	Fe(III)	Hg(II)	Ni(II)	Cr(III)	Total
Original	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	4	0	0	0	0	0	0	0	0	4
		Ca(II)	0	0	4	0	0	0	0	0	0	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	4
		Cd(II)	0	0	1	0	0	3	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	4	0	0	4
		Ni(II)	0	0	1	0	0	0	0	0	3	0	4
		Cr(III)	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	100
		Ag(I)	0	100	0	0	0	0	0	0	0	0	100
		Ca(II)	0	0	100	0	0	0	0	0	0	0	100
		Co(II)	0	0	0	100	0	0	0	0	0	0	100
		Cu(II)	0	0	0	0	100	0	0	0	0	0	100
		Cd(II)	0	0	25	0	0	75	0	0	0	0	100
		Fe(III)	0	0	0	0	0	0	100	0	0	0	100
		Hg(II)	0	0	0	0	0	0	0	100	0	0	100
		Ni(II)	0	0	25	0	0	0	0	0	75	0	100
		Cr(III)	0	0	0	0	0	0	0	0	0	100	100
Cross-validated ^b	Count	Al(III)	4	0	0	0	0	0	0	0	0	0	4
		Ag(I)	0	3	1	0	0	0	0	0	0	0	4
		Ca(II)	0	0	3	0	0	0	0	0	1	0	4
		Co(II)	0	0	0	4	0	0	0	0	0	0	4
		Cu(II)	0	0	0	0	4	0	0	0	0	0	4
		Cd(II)	0	0	1	0	0	3	0	0	0	0	4
		Fe(III)	0	0	0	0	0	0	4	0	0	0	4
		Hg(II)	0	0	0	0	0	0	0	3	1	0	4
		Ni(II)	0	0	1	0	0	0	0	0	3	0	4
		Cr(III)	0	0	0	0	0	0	0	0	0	4	4
	%	Al(III)	100	0	0	0	0	0	0	0	0	0	100

	Ag(I)	0	75	25	0	0	0	0	0	0	0	100
	Ca(II)	0	0	75	0	0	0	0	0	25	0	100
	Co(II)	0	0	0	100	0	0	0	0	0	0	100
	Cu(II)	0	0	0	0	100	0	0	0	0	0	100
	Cd(II)	0	0	25	0	0	75	0	0	0	0	100
	Fe(III)	0	0	0	0	0	0	100	0	0	0	100
	Hg(II)	0	0	0	0	0	0	0	75	25	0	100
	Ni(II)	0	0	25	0	0	0	0	0	75	0	100
	Cr(III)	0	0	0	0	0	0	0	0	0	100	100
a. 95.0% of original grouped cases correctly classified.												
b. Cross validation is done only for those cases in the analysis. In cross validation, each case is classified by the functions derived from all cases other than that case.												
c. 87.5% of cross-validated grouped cases correctly classified.												

Table S5.14 Summary of canonical discriminant Functions of 4-CDs-based array for biological application with urine sample

Summary of Canonical Discriminant Functions									
Function	Eigenvalues				Wilks' Lambda				
	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation	Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1	27.381 ^a	86.4	86.4	0.982	1 through 4	0.003	260.219	5	0
2	3.158 ^a	10	96.4	0.871	2 through 4	0.099	106.316	3	0
3	.811 ^a	2.6	98.9	0.669	3 through 4	0.412	40.769	2	0.00
4	.340 ^a	1.1	100	0.504	4	0.746	13.457	1	0.19
a. First 4 canonical discriminant functions were used in the analysis.									

Table S5.15 Standardized canonical discriminant function coefficients of 4-CDs-based array for biological application with urine sample

Standardized canonical discriminant function coefficients				
CDs	Functions			
	1	2	3	4
CD_1-0_6 h	0.693	0.194	0.909	0.092
CD_2-4_6 h	1.092	-0.315	-0.062	-0.196
CD_1-4_6 h	0.375	0.047	-0.21	0.928
CD_1-9_6 h	0.515	0.891	-0.104	-0.29

Table S5.16 Structure matrix of 4-CDs-based array for biological application with urine sample

Structure matrix				
CDs	Functions			
	1	2	3	4
CD_1-0_6 h	.679*	-0.543	-0.368	-0.33
CD_2-4_6 h	0.205	.900*	-0.328	-0.202
CD_1-4_6 h	0.115	0.102	.974*	0.168
CD_1-9_6 h	0.196	0.156	-0.277	.928*
Pooled within-groups correlations between discriminating variables and standardized canonical discriminant functions				
Variables ordered by absolute size of correlation within function.				
*. Largest absolute correlation between each variable and any discriminant function				