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Fast and thermal Accelerator Driven Systems: Studies of secondary particle production and transport

A thesis submitted in fulfilment of the requirements for the degree of Doctor of Philosophy

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2015
Abstract

Accelerator Driven Systems (ADS) consist of a spallation neutron source coupled to a sub-critical nuclear assembly and are a proposed technology for the transmutation of nuclear waste and electricity generation. The Gamma-3 assembly of the Joint Institute for Nuclear Research (JINR), Dubna, Russia is designed to emulate the neutron spectrum of a thermal ADS. It consists of a lead spallation target surrounded by reactor grade graphite. The target was irradiated with 1.6 GeV deuterons from the Nuclotron accelerator and the neutron capture and fission rate of $^{232}$Th within the assembly were experimentally measured. $^{232}$Th is a proposed fuel for envisaged Accelerator Driven Systems and these two reactions are fundamental to the performance and feasibility of $^{232}$Th in an ADS. The spatial distribution of slow neutrons on the surface of the graphite was also measured using CR-39 track detectors coupled to LR-1152B film.

The Quinta assembly, also located at the JINR, is composed of 512 kg of natural uranium surrounded by a lead reflector and is designed for neutronic studies of fast ADS. It was irradiated with 1 and 4 GeV deuterons and the fission rate of $^{nat}$U, $^{232}$Th, $^{209}$Bi, $^{nat}$Pb and $^{197}$Au was measured using mica solid state nuclear track detectors (SSNTDs) placed around the assembly. These materials are all mooted as potential fuel and/or target materials in future ADS. $(n,\gamma)$ and $(n,xn)$ reaction rates of $^{209}$Bi and $^{197}$Au were also determined through gamma spectrometry.

Experimental reaction rates were compared to those calculated using the MCNPX 2.7 code with the INCL4/ABLA and CEM03 physics models. Evaluated cross-section libraries were used whenever available, otherwise reaction cross section data were retrieved from the literature or calculated using the TALYS and XSEX3 codes. Good agreement between the experimental and calculated results were found, although in general, the INCL4-ABLA model produced a closer agreement with the experimental results compared to CEM03. This study serves as a good validation for the computational models and cross section data used to simulate secondary particle production and transport in thermal and fast ADS. Future development of ADS will inevitably rely upon accurate and well validated simulation codes and nuclear data.

The work described in this thesis resulted in several peer-reviewed publications, as outlined in Appendix A.
Acknowledgements

Throughout the course of this PhD, I have been extremely fortunate to receive advice and assistance from many people, without which completion of this work would not have been possible. First and foremost, my supervisor A/Prof. Reza Hashemi-Nezhad has been an excellent mentor, motivator and proofreader. His patience, humorous anecdotes and tireless support when I was struggling to make sense of things will never be forgotten. Thank you for being a fantastic supervisor and all-round great person.

A special thanks also to the former students of the Applied Nuclear Science group, notably Dr Jacob Borger and Claude LeComte. I wish you guys all the best for the future.

The experimental work carried out in this thesis was performed as part of the E&T RAW (Energy and Transmutation of Radioactive Waste) collaboration, which survives on funding and administrative support from the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. I am grateful for the technical support and warm hospitality of group members Prof. Sergey Tyutyunnikov, Drs Valeri Chilap, Walter Furman, Wolfram Westmeier and the late Mikhail Kadykov. Dr Jindra Adam’s expertise in gamma spectrometry and generous allowance to use his laboratory equipment is particularly appreciated. I thank the JINR itself, especially the staff at the Nuclotron accelerator for providing access to the research facilities and Prof. Kekelidze, director of the Veksler and Baldin Laboratory of High Energies.

This PhD work was funded by an Australian Postgraduate Award (APA). The United Uranium Scholarship provided funding so I could attend an MCNPX workshop in Washington DC. A Postgraduate Research Support Scheme (PRSS) travel grant also allowed me to participate at the International Youth Nuclear Congress (IYNC), held in Burgos, Spain. I am extremely thankful for all funding received.

To my parents, Allan and Suzanne, and my boyfriend, Paul, thank you for your endless love and support over the years. Thanks also to my friends for knowing nothing about physics and providing a much needed escape from time to time.
Author’s Contribution

The review into the development of nuclear power and ADS (chapters 1 and 2) were compiled through the author’s own reasoning and understanding of the literature cited. The discussion into Monte Carlo spallation models (sections 4.1–4.5) also follows material cited from the literature and contains no original work by the author.

The description of the Gamma-3 and Quinta assemblies (chapter 3) including details of geometry and construction materials arises from a combination of technical drawings, first-hand measurements conducted by the author, and consultations with A/Prof. Reza Hashemi-Nezhad. The measurement of the beam pulse distributions (shown in Figures 3.6 and 3.7) were carried out by Nuclotron technicians at the JINR. Determination of the incident number of beam deuterons and beam shape (Tables 3.1 and 3.2) was performed by collaboration members Prof. Igor Zhuk and Dr Wolfram Westmeier.

All spectra collected from the HPGe detectors mentioned in chapter 3, including the spectra for efficiency calibrations, were done so by the author or a laboratory assistant from the JINR working under the author’s instruction. Analysis of all spectra, including reaction rate calculation, determination of detection efficiency, pile-up and true coincident summing corrections were carried out by the author using the methods described and cited in the text. The $^{232}$Th spectra discussed in Appendix B was collected by Dr Jindra Adam. However, the analysis and presentation of material in Appendix B is the author’s own. The Python script mentioned in Appendix B to match spectral data with nuclides in the ENSDF database is the work of the author.

The preparation of all experimental samples (activation foils, fission track detectors and CR-39/LR-115 2B detectors), mounting of these samples onto the Gamma-3 and Quinta assemblies, and etching, photographing, track counting and analysis of the track detectors (mica and CR-39) were performed by the author under the guidance of A/Prof. Reza Hashemi-Nezhad. The script for image post-processing and automatic track counting of the track detectors is the author’s.

The calibration of the CR-39/LR-115 2B system was performed at The University of Sydney with the assistance of A/Prof. Reza Hashemi-Nezhad and technician Marek Dollesier. The method of calibration expands on that first described in the PhD thesis of Dr Jacob Borger. However, the design of the calibration set-up, sample preparation, analysis, Monte Carlo simulations and calculation of calibration factors is the author’s own work.
The results and analysis of all MCNPX, TALYS and XSEX3 calculations presented in this thesis arise from input files written from scratch by the author. All images, graphs, diagrams and photographs are the work of the author unless stated otherwise.

I, Nicola Asquith, hereby declare that all work presented in this thesis was completed by me during my PhD candidature at The University of Sydney, Australia. To the best of my knowledge, it contains no copy or paraphrased work published by someone else, except where indicated otherwise. This work has not been submitted, either in full or in part, for a degree at The University of Sydney or any other institution.

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<tr>
<td>ABLA</td>
<td>Abrasion-ablation</td>
</tr>
<tr>
<td>ACE</td>
<td>A Compact ENDF</td>
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<tr>
<td>ADS</td>
<td>Accelerator Driven System</td>
</tr>
<tr>
<td>AMSTER</td>
<td>Actinides Molten Salt TransmutER</td>
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<tr>
<td>ATW</td>
<td>Accelerator Transmutation of Waste</td>
</tr>
<tr>
<td>AVR</td>
<td>Arbeitsgemeinschaft VersuchsReaktor (pebble bed reactor)</td>
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<tr>
<td>BRIC</td>
<td>Bruyères-le-Châtel Intranuclear Cascade (model)</td>
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<tr>
<td>BORAX</td>
<td>BOiling water ReActor eXperiment</td>
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<tr>
<td>BWR</td>
<td>Boiling Water Reactor</td>
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<tr>
<td>CANDU</td>
<td>CANada Deuterium Uranium (reactor)</td>
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<tr>
<td>CEM</td>
<td>Cascade Exciton Model</td>
</tr>
<tr>
<td>CENDL</td>
<td>Chinese Evaluated Nuclear Data Library</td>
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<tr>
<td>EN(S)DF</td>
<td>Evaluated Nuclear (Structure) Data File</td>
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<tr>
<td>EpT</td>
<td>Energy plus Transmutation</td>
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<td>Fast Breeder Reactor</td>
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<td>FEAT</td>
<td>First Energy Amplifier Test</td>
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<tr>
<td>FERFICON</td>
<td>FERtile-to-FIssile CONversion program</td>
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<tr>
<td>FLUKA</td>
<td>FLUktuierende KAskade</td>
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<td>GANEX</td>
<td>Group ActiNide EXtraction</td>
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<td>GFR</td>
<td>Gas-cooled Fast Reactor</td>
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<tr>
<td>HERMES</td>
<td>High Energy Radiation Monte carlo Elaborate System</td>
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<td>INC(L)</td>
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<td>NPT</td>
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<td>Particle and Heavy Ion Transport code System</td>
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<tr>
<td>PMA</td>
<td>Plutonium and Minor Actinides</td>
</tr>
<tr>
<td>PUREX</td>
<td>Plutonium Uranium Redox EXtraction</td>
</tr>
<tr>
<td>PWR</td>
<td>Pressurized Water Reactors</td>
</tr>
<tr>
<td>REPA</td>
<td>REprocessing-PArtitioning</td>
</tr>
<tr>
<td>RUSFOND</td>
<td>RUssian library of Files Of evaluated Nuclear Data</td>
</tr>
<tr>
<td>SSNTD</td>
<td>Solid-State Nuclear Track Detector</td>
</tr>
<tr>
<td>TARC</td>
<td>Transmutation by Adiabatic Resonance Crossing</td>
</tr>
<tr>
<td>TASSE</td>
<td>Th-fuelled ADS for both energy production and TRU-incineration</td>
</tr>
<tr>
<td>TCS</td>
<td>True Coincidence Summing</td>
</tr>
<tr>
<td>TENDL</td>
<td>TALYS-based Evaluated Nuclear Data Library</td>
</tr>
<tr>
<td>THOREX</td>
<td>THORium EXtraction</td>
</tr>
<tr>
<td>THTR</td>
<td>Thorium High Temperature Reactor</td>
</tr>
<tr>
<td>UOX</td>
<td>Uranium OXide</td>
</tr>
<tr>
<td>UREX</td>
<td>URanium EXtraction</td>
</tr>
<tr>
<td>WNA</td>
<td>World Nuclear Association</td>
</tr>
<tr>
<td>XADS</td>
<td>eXperimental Accelerator Driven System</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

By the year 2040, the world’s population is predicted to rise above 9 billion [1]. This will place an enormous strain on energy resources; in particular, net electricity generation is expected to rise 93% between 2010 and 2040 [2]. Renewable energy can not reliably meet this demand, and with increasing environmental concerns regarding CO$_2$ emissions from the burning of fossil fuels, nuclear power must be considered a viable alternative.

Currently, Light Water Reactors (LWR) consisting of both the Pressurised Water Reactor (PWR) and Boiling Water Reactor (BWR) sub-types constitute 88% of the electricity generated by nuclear power plants [3]. These reactors were first developed as a means to power naval ships and submarines [4], and while they are generally considered safe and reliable [5], better reactor designs more suited to civilian electricity generation are under development. These reactors offer improved safety, efficiency, economics, reliability, waste reduction and proliferation resistance.

Accelerator Driven Systems (ADS) have been suggested as a means for safe, economical nuclear energy production and nuclear waste incineration [6, 7]. An ADS consists of a spallation neutron source coupled to a sub-critical nuclear assembly. Neutrons available in an ADS may be used to induce fission in the assembly, breed fissile material, and/or transmute radiotoxic minor actinides and long lived fission products.

The fission spectrum in LWRs has a well established analytical function with the high energy tail extending up to $\sim 10 \text{MeV}$. By comparison, the upper energy limit of spallation neutrons in an ADS can extend up to the energy of the incident ion ($\sim \text{GeV}$), where availability of accurate nuclear data is scant. The energy and spatial distribution of these neutrons also does not have an identifiable analytical solution, and may vary considerably depending on the target material, energy and type of incident ion, presence of moderating material and geometry of the system. Therefore, Monte Carlo physics models are required for simulation of reactions and radiation transport outside the range of available evaluated nuclear data.

Various designs for an ADS have been considered. These include thermal spectrum ADS utilising graphite [8] or heavy water [6] as a moderator. Fast spectrum ADS designs involving Pb [7, 9–11] or Pb-Bi eutectic (LBE) [12] moderator-coolant have also been proposed. The
Joint Institute for Nuclear Research (JINR) in Dubna, Russia houses two small-scale assemblies suitable for experimental studies into ADS. Quinta, a uranium assembly, produces a combined fission-spallation spectra like that expected of a fast ADS. The Gamma-3 assembly, consists of a lead spallation target surrounded by a graphite moderator and is designed to emulate the neutron spectrum of a thermal ADS.

This work reports the outcome of several experimental studies performed on these two assemblies. The neutron capture and fission rate of $^{232}$Th has been measured in several locations within the Gamma-3 assembly. $^{232}$Th is a proposed fuel for ADS in order to increase fuel efficiency and reduce the radio-toxicity of waste at the back end of the fuel cycle [9]. The $^{232}$Th(n,γ) reaction rate was measured in order to study the breeding efficiency of fissile $^{233}$U in a thermal ADS. Meanwhile, $^{232}$Th is a fissionable material, which may fission by energetic neutrons. Direct fission of $^{232}$Th therefore can contribute to the neutron balance of the system. The spatial distribution of slow neutrons on the surface of the Gamma-3 was also measured using Solid State Nuclear Track Detectors (SSNTDs). Slow neutrons strongly influence the reaction rates within the sub-critical assembly, including the fission rate of fissile materials, breeding of $^{233}$U from $^{232}$Th, and nuclear waste transmutation of long lived fission products.

The fission rate of three sub-actinides ($^{209}$Bi, $^{nat}$Pb and $^{197}$Au) was measured in the Quinta assembly. These materials have shown potential for use as spallation target materials such as lead-gold eutectic [13] and the MYRHA which utilises a liquid lead-bismuth eutectic target [14]. The fission rate of $^{nat}$U and $^{232}$Th was also measured. The fission of these materials is important as it has implications for the power output, neutron balance, heat deposition, induced radioactivity and production of fission gases within the system. Additionally, the high threshold energy ($E_{th} \gtrsim 40$ MeV) of fission for sub-actinides means this reaction may be used as an integral monitor for very fast neutrons. $^{197}$Au(n,γ), $^{197}$Au(n,xn) and $^{206}$Bi(n,xn) reaction rates were also determined using activation detectors. (n,xn) threshold reactions can be used to experimentally probe narrow sections of the high energy region of the neutron spectrum, while the (n,γ) reaction is useful for the detection of slow neutrons.

The experimental conditions in the two assemblies were simulated using Monte Carlo code MCNPX 2.7. The INCL4 intranuclear cascade with the ABLA fission/evaporation models were compared to the CEM03 model and the experimentally measured data. The primary aim of this thesis was to assess the ability of the codes and physics models to correctly predict secondary particle production from spallation sources and radiation transport in both thermal and fast ADS. Efficient design of future ADS will strongly depend on accurate nuclear data and verified physics models. While ADSs are primarily envisioned for electricity production and nuclear waste transmutation, research into accelerator and spallation physics has implications over a broad range of scientific fields. This includes tritium production for fusion research, isotope production, materials science research, biological and medical research, hadron therapy and neutrino factories [15].
Chapter 2

Survey of the subject

2.1 Introduction to ADS

Accelerator Driven Systems (ADS) have been studied with renewed interest for the past two decades. The modern rendition of the ADS is generally credited to two groups led by Carlo Rubbia of CERN [7, 9] and Charles Bowman of Los Alamos National Laboratory (LANL) [6, 8]. The idea of using accelerators to breed fissile material has been around since the early 1950’s [16], however these early efforts were abandoned for technical and economic reasons.

An ADS is a type of hybrid reactor and therefore requires an external neutron source to sustain fission reactions in the sub-critical fuel assembly. This neutron source is a spallation neutron source which typically arises from a high current (∼mA), high energy (∼1 GeV) ion beam impinging on a heavy metal target. The usefulness of an ADS depends on its energy gain, $G$ and the production of neutrons which are used to maintain fission reactions, breed fissile material and transmute nuclear waste.

The energy gain, $G$ is the ratio of thermal energy produced to the energy deposited by the beam, and may be expressed as:

$$G = \frac{N_p \varphi^* k_{\text{eff}} E_f}{\nu (1 - k_{\text{eff}}) E_i}$$

where, $N_p$ is the average number of neutrons to escape the target volume per incident ion, $E_i$ is the kinetic energy of incident ion (in GeV), $\nu$ is the average number of neutrons emitted per fission, $k_{\text{eff}}$ is the effective neutron multiplication factor of the sub-critical assembly and $E_f$ is the recoverable energy per fission (∼0.2 GeV) [17]. $\varphi^*$ is a weighting factor for the effectiveness of ‘source’ neutrons, defined as the ratio of the importance of neutrons from the external source to those emitted from fission. Source neutrons leaving the target volume have very different energy and spatial distributions to neutrons emitted from fission reactions in the assembly, and $\varphi^*$ may deviate significantly from 1 [18].

The thermal power output, $(P_0)_{th}$ of an ADS (in MW) in terms of the accelerator beam current, $I$ (in mA) is

$$(P_0)_{th} = I E_i G$$

(2.2)
and the fraction of power output, \( f \) from the ADS required to power the accelerator is

\[
f = \frac{1}{G \eta_{th} \varepsilon}
\]

where, \( \eta_{th} \) is the thermodynamic efficiency of the system, and \( \varepsilon \) is the conversion efficiency from electric to beam power [17]. The excess power not required to run the accelerator may be passed on to the commercial power grid.

The introduction of a new reactor type must offer evidential improvements on existing reactors in order to justify the substantial investment in research and development. The ADS has several advantages over conventional LWRs. This includes enhancements to the safety, environmental impact and proliferation resistance.

The most characterising feature of an ADS is that it operates sub-criticality, entirely eliminating the possibility of a Chernobyl type accident. Although the chances of a criticality accident occurring in a reactor today are incredibly remote, they are a cause of great concern for the general public. All commercial reactors operating today do so critically. Criticality refers to the balance of neutrons in the system and is best defined by the effective neutron multiplication factor, \( k_{\text{eff}} \):

\[
k_{\text{eff}} = \frac{\text{Number of neutrons in generation } n+1}{\text{Number of neutrons in generation } n}
\]

If \( k_{\text{eff}} < 1 \), the reactor is sub-critical and fission chain reactions cannot be sustained without an external source of neutrons.

If \( k_{\text{eff}} = 1 \), the reactor is critical and a steady state fission rate is achieved.

If \( k_{\text{eff}} > 1 \), the reactor is supercritical. Neutron production and fission rate will continue to increase exponentially.

In an ADS, chain reactions in the core may be stopped simply by switching off or redirecting the accelerator beam, if required. The residual decay heat can then disperse through passive cooling methods e.g. natural convection of molten lead [9, 19]. The greater tolerance to reactivity swings in the reactor is also an added safety feature. An ADS can withstand large reactivity swings which would be unacceptable in a critical reactor. This allows for greater flexibility in the reactor design and operation, which is especially beneficial when trying to transmute large quantities of waste actinides or breed \(^{233}\text{U}\) from \(^{232}\text{Th}\).

Finding an appropriate solution for the long-term disposal of nuclear waste is one of the biggest challenges facing the nuclear industry. An ADS incorporating the \(^{232}\text{Th}−^{233}\text{U}\) fuel cycle would produce a significantly less radio-toxic waste stream than that of the \(^{238}\text{U}−^{239}\text{Pu}\) cycle or the once-through cycle currently embraced by most LWRs. Parasitic formation of \(^{232}\text{U}\) also makes the adoption of a \(^{232}\text{Th}−^{233}\text{U}\) fuel cycle intrinsically proliferation resistant.
2.2 Nuclear Fuel Cycles

The nuclear fuel cycle is the series of stages that nuclear fuel progresses through in its lifetime. This includes mining the ore, processing and enrichment, placement in the reactor and the final safe disposal of the spent fuel. Three options are currently considered for the disposal of spent fuel – direct disposal (open fuel cycle), a temporary storage and postponed decision or a “reprocessing and recycle” (closed fuel cycle) solution [20].

2.2.1 Open fuel cycle

The open, or once-through fuel cycle (not a cycle in itself) currently employed by most LWRs is incredibly inefficient and wasteful. There exists only one naturally occurring fissile isotope in nature, $^{235}\text{U}$ which constitutes a mere 0.72% of natural uranium (the rest is $^{238}\text{U}$, with trace quantities of $^{234}\text{U}$). The fuel for a LWR requires enrichment to levels of 3–5%, resulting in approximately 85% of the original natural uranium feed being discarded as depleted uranium [21]. Apart from possible uses as a feed for fast breeder reactors, counterweights in aircraft or for radiation shielding, depleted uranium has very few respectable uses.

As well as the inefficient use of the mined uranium, the reactors themselves do not burn fuel efficiently. Table 2.1 shows the inventory of a typical 1 GWe PWR at loading and discharge [22]. The fuel is 3.5% enriched uranium and assumes a burn up rate of 33 GWd/ton. 111 kg of the 674 kg of consumed $^{235}\text{U}$ has not undergone fission but instead converted to $^{236}\text{U}$ via neutron capture, 673 kg of $^{238}\text{U}$ has been converted to Plutonium or Minor Actinides (PMA) and of this 383 kg has undergone fission. This leaves 286 kg of highly radiotoxic PMA which due to their long half lives, the only suitable disposal options are either long term geological storage or transmutation. Each year, every once-through 1 GWe reactor requires 200 ton of fresh natural uranium [23]. At current consumption levels, known uranium reserves are estimated to last only 120 years [24].

2.2.2 Reprocessing and recycle

Throughout the 1960’s and early 1970’s, LWRs were being built with the intention of recycling the spent fuel. The unused fissile $^{235}\text{U}$, $^{239}\text{Pu}$ produced from $^{238}\text{U}$ conversion and neutron poisons would be separated and fresh fuel created. The plan was to eventually transition to a new generation of plutonium breeder reactors [4]. However, in 1974, the successful demonstration of a plutonium weapon made from reprocessed reactor fuel by India led to significant proliferation concerns. The fear that plutonium from the civilian nuclear fuel cycle could be diverted for military (or terrorism) uses, led to the 1977 indefinite ban of commercial fuel reprocessing in the United States. Despite this ban being lifted only a few years later in 1981, the lack of government subsidies convinced private start-up companies that fuel reprocessing was not commercially viable [25]. Without the ability to reprocess fuel, breeder technology became unfeasible and
TABLE 2.1: Inventory of 1 GWe PWR at loading and discharge using 3.5% enriched uranium fuel [22].

<table>
<thead>
<tr>
<th>Nuclides</th>
<th>Loading (kg)</th>
<th>Discharge (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}\text{U}$</td>
<td>954</td>
<td>280</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td>111</td>
<td></td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>26328</td>
<td>25655</td>
</tr>
<tr>
<td>U total</td>
<td>27282</td>
<td>26047</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>56</td>
<td></td>
</tr>
<tr>
<td>Pu total</td>
<td>266</td>
<td></td>
</tr>
<tr>
<td>Minor Actinides</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>$^{90}\text{Sr}$</td>
<td>13</td>
<td></td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td>Long-lived FP</td>
<td>63</td>
<td></td>
</tr>
<tr>
<td>FP total</td>
<td>946</td>
<td></td>
</tr>
<tr>
<td>Total mass</td>
<td>27282</td>
<td>27279</td>
</tr>
</tbody>
</table>

research programs were terminated. This led to further entrenchment of the open (once-through) fuel ‘cycle’, and the continued dominance of the LWR.

Reprocessing spent fuel increases the use of fissile material and reduces the volume and radiotoxicity of high level and long lived waste requiring long term disposal [20]. Currently, the only technique used for fuel reprocessing on an industrial scale is the PUREX (plutonium and uranium extraction) process for recycling of plutonium into MOX (mixed oxide) fuel. PUREX was optimised for the maximum extraction of pure plutonium for military purposes and as such, now lends itself to significant proliferation concerns. Other aqueous separation techniques being developed including the COEX, simplified PUREX, NEXT, REPA, UREX and GANEX offer increased proliferation resistance and the ability to separately extract fission products and minor actinides from the spent fuel. However, apart from the COEX process, which is showing potential for industrial scale development, none of the other technologies have progressed further than laboratory scale experiments [20].

MOX fuel is licensed to operate only in selected reactors and because of these proliferation concerns, the PUREX technology remains restricted to a select group of countries (mainly France, Japan, UK, Russia and India) [20]. The burn-up of MOX fuel is approximately the same as UOX fuel [26], and as shown in Table 2.1, LWRs do not consume all fissile material from their initial loading. Therefore, several reprocessing cycles are required for complete closure of the fuel cycle. However, after only a few of these cycles, the isotopic quality of the MOX fuel degrades significantly due to the increasing production of higher actinides and decreasing fissile content. This is compensated for by increasing the proportion of plutonium with depleted uranium support in each cycle [27]. However, in industrial PWRs there is an upper limit of 12% plutonium content, beyond that the void coefficient becomes positive if it is not mixed further with enriched uranium [28]. Therefore, even implementing fuel reprocessing and recycle in current reactors will still not remove the need for long term geological storage.
2.2.3 Conversion and breeding

As well as fuel reprocessing, fuel breeding can also be used to greatly extend the performance of the mined fuel material. Nuclear breeding involves converting fertile nuclei ($^{232}$Th or $^{238}$U) into fissile nuclei ($^{233}$U or $^{239}$Pu) via the process of neutron capture and subsequent beta decay:

$$^{232}\text{Th} + n \longrightarrow ^{233}\text{Th} \xrightarrow{\beta^-} ^{233}\text{Pa} \xrightarrow{\beta^-} ^{233}\text{U}$$

$$^{238}\text{U} + n \longrightarrow ^{239}\text{U} \xrightarrow{\beta^-} ^{239}\text{Np} \xrightarrow{\beta^-} ^{239}\text{Pu}$$

The probability that the absorption of a neutron leads to a fission is the fission probability, $P_f$. At low energies, neutron absorption consists of fission and radiative capture reactions.

$$P_f(E) = \frac{\sigma_f(E)}{\sigma_a(E)} = \frac{\sigma_f(E)}{\sigma_f(E) + \sigma_c(E)} \quad (2.5)$$

Where, $\sigma_f$, $\sigma_c$ and $\sigma_a$ are the neutron energy dependent microscopic fission, capture and absorption cross sections, respectively. The neutron reproduction factor, $\eta$ is the number of neutrons emitted per absorbed neutron. It may be calculated from the product of the fission probability and $\nu$, the mean number of neutrons released per fission, like so:

$$\eta(E) = P_f(E)\nu(E) \quad (2.6)$$

$\eta$ is a useful parameter for assessing how effective certain fuels will be for criticality or breeding fissile material. A value of $\eta > 1$ is required to achieve criticality, since at least one neutron is required to continue the chain reaction. The condition for breeding to occur is $\eta > 2$ since one of the neutrons created must go on to produce further fission and at least one must be absorbed by fertile nucleus in order to convert to fissile material. For values of $\eta$ between 1 and 2 some conversion of fuel will occur [29].

Figure 2.1 shows plots of the fission cross section, capture cross section, fission probabilities and neutron reproductive factor values, $\eta$ for three fissile nuclides – $^{233}$U, $^{235}$U and $^{239}$Pu. Integral cross data from the ENDF/BVII.1 libraries is also shown in Table 2.2. From Table 2.2 we may conclude $^{233}$U is a suitable nuclear fuel for breeding. It has a neutron reproductive factor > 2 across all regions of the neutron spectrum, therefore capable of breeding in a thermal, epithermal or fast neutron spectrum. In contrast, due to the large resonance capture cross section, $^{235}$U and $^{239}$Pu, $\eta$ falls below 2 during the resonance integral region. The unfavourable neutron economy in the resonance region is also the reason why current reactors are designed to be either thermal or fast.

In thermal region, $^{233}$U has the lowest fission cross section, as compared to $^{235}$U and $^{239}$Pu, however the relatively low capture cross section produces a high fission probability (Table 2.2). Additionally, the neutron capture cross section of $^{232}$Th is nearly three times higher than that of
CHAPTER 2. SURVEY OF THE SUBJECT

FIGURE 2.1: (a) Fission cross sections, $\sigma_f$ shown alongside the Maxwellian thermal spectrum and Watt fission spectrum (arbitrary units on y-scale) [30], (b) capture cross sections, $\sigma_c$ (c) fission probabilities, $P_f$ and (d) neutron reproductive factor, $\eta$ (neutrons emitted per neutron absorbed) for fissile isotopes – $^{233}\text{U}$, $^{235}\text{U}$ and $^{239}\text{Pu}$. Source: ENDF/B-VII.1 [31].

FIGURE 2.2: (a) Capture and (b) fission cross sections of fertile isotopes – $^{232}\text{Th}$ and $^{238}\text{U}$. Source: ENDF/B-VII.1 [31].
2.2. Nuclear Fuel Cycles

**TABLE 2.2:** Integral cross section data for $^{233}$U, $^{235}$U and $^{239}$Pu for various neutron spectra calculated using ENDF/B-VII.1 data. ‘Thermal’ refers to a Maxwellian spectrum (T=298K), ‘epithermal’ refers to neutron energies relevant for resonance integral determination and ‘fast’ refers to a fission spectrum. The energy regions are noted in Figure 2.1a.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Energy range of neutrons</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Thermal</td>
</tr>
<tr>
<td>$^{233}$U</td>
<td>$\bar{\sigma}_f$ (b)</td>
</tr>
<tr>
<td></td>
<td>$\bar{\sigma}_c$ (b)</td>
</tr>
<tr>
<td></td>
<td>$P_f$</td>
</tr>
<tr>
<td></td>
<td>$\nu$</td>
</tr>
<tr>
<td></td>
<td>$\eta$</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>$\bar{\sigma}_f$ (b)</td>
</tr>
<tr>
<td></td>
<td>$\bar{\sigma}_c$ (b)</td>
</tr>
<tr>
<td></td>
<td>$P_f$</td>
</tr>
<tr>
<td></td>
<td>$\nu$</td>
</tr>
<tr>
<td></td>
<td>$\eta$</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>$\bar{\sigma}_f$ (b)</td>
</tr>
<tr>
<td></td>
<td>$\bar{\sigma}_c$ (b)</td>
</tr>
<tr>
<td></td>
<td>$P_f$</td>
</tr>
<tr>
<td></td>
<td>$\nu$</td>
</tr>
<tr>
<td></td>
<td>$\eta$</td>
</tr>
<tr>
<td>$^{232}$Th</td>
<td>$\bar{\sigma}_f$ (b)</td>
</tr>
<tr>
<td></td>
<td>$\bar{\sigma}_c$ (b)</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>$\bar{\sigma}_f$ (b)</td>
</tr>
<tr>
<td></td>
<td>$\bar{\sigma}_c$ (b)</td>
</tr>
</tbody>
</table>

$^{238}$U (Figure 2.2). Therefore, not only does $^{233}$U burn the most efficiently, but $^{232}$Th is also the better fertile material at thermal energies.

Although the $^{232}$Th$-$$^{233}$U fuel cycle is feasible over all neutron energies, the $^{238}$U$-$$^{239}$Pu cycle clearly has superior neutron economy at fast energies. This is due to the high $\eta$ value (3.05) and the fast fission cross section of $^{238}$U being four times higher than $^{232}$Th (Table 2.2). The $\eta$ value of $^{239}$Pu has a greater increase with energy compared to $^{233}$U.

2.2.3.1 Advantages of the $^{232}$Th$-$$^{233}$U cycle

As described above, the $^{232}$Th$-$$^{233}$U fuel cycle is capable of operating in a thermal, epithermal or fast neutron spectrum. Even though the $^{238}$U$-$$^{239}$Pu fuel cycle is clearly advantageous in a fast spectrum from a neutron economy perspective, the $^{232}$Th$-$$^{233}$U fuel cycle has several practical advantages, as described below. This has led to recent renewed interest in the $^{232}$Th$-$$^{233}$U fuel cycle looking for ways to increase proliferation resistance, reduce the burden of nuclear waste,
increase fuel residence times, enable in-situ breeding of fissile material and to aid reduction in excess plutonium stockpiles [32].

Natural thorium consists of a single isotope, $^{232}$Th which is 3-4 times more abundant than uranium in the Earth’s continental crust [33]. Thorium is not yet exploited commercially, as it has very limited uses outside the nuclear industry. Examples include thorium nitrate in gas mantles, as a catalyst for synthesis of hydrocarbons, magnesium-based alloys, thoriated tungsten welding rods and refractory crucibles [32]. Therefore, the long term availability of thorium is assured and reserves may be exploitable for thousands of years [34].

Due to the parasitic formation of $^{232}$U from unavoidable (n,2n) and ($\gamma$,n) reactions with $^{233}$U and $^{233}$Pa, the $^{232}$Th–$^{233}$U fuel cycle is intrinsically proliferation resistant. $^{232}$U and its daughter products are all short-lived and one of them, $^{208}$Tl emits a 2.6 MeV $\gamma$-ray. The emission of this energetic $\gamma$-ray makes handling the material dangerous and any illicit diversions easy to trace. Increased proliferation resistance will ease the ability to deploy the technology, especially to developing countries.

The high fission probability and lower mass number of $^{233}$U compared to $^{239}$Pu leads to significantly less production of minor actinides. Moir and Teller [35] have estimated that the volume required for long term storage of waste from a thorium fuelled MSR would be 10–100 times less compared to the waste of a once-through LWR. The radioactive waste produced from mining thorium is about 100 times less than from mining uranium and so the front end of the fuel cycle is also predicted to produce much less waste from mining operations. The mill tailings are also a lot easier to manage due to the much shorter half life of thoron ($^{220}$Rn), compared to radon ($^{222}$Rn). Monazite (a major source of thorium) is open pit mined, so there are no ventilation issues from the build up of thoron as occurs from radon in underground uranium mines [32]. The short half-life of thoron (56 s) compared to radon (3.8 d), means there is not enough time for it to diffuse through rock and accumulate in unventilated areas. There is strong evidence to link radon and lung cancer, and after smoking it is believed to be the second biggest cause of lung cancer [36].

Around the world, large stockpiles of plutonium from civil reactors and military activities are continuing to grow [37]. How best to handle this material is subject to much controversy, where there are serious concerns about misuse of the plutonium or release into the environment (accidental or otherwise) [38]. One option is to incinerate it in commercial reactors where the plutonium may be combined with uranium in mixed oxide (MOX) fuel. This is currently done in a few countries (Belgium, Switzerland, Germany, France and Japan are all licensed to use MOX fuel in selected reactors [26]). However, the (U,Pu)O$_2$ performance is limited due to breeding of more plutonium and higher actinides in the fuel mix. In particular, a build up of non-fissile even mass number isotopes occurs which cannot be chemically separated from the fissile odd number isotopes. This degrades the quality of the (U,Pu)O$_2$ fuel significantly after a few cycles [27]. Calculations comparing (U,Pu)O$_2$ and (Th,Pu)O$_2$ in an LWR showed that although the (U,Pu)O$_2$ achieved higher reactivity-limited burnup by 1.3-4.6 times, the amount
2.2. Nuclear Fuel Cycles

of plutonium depleted during the cycle was more than two times greater for the (Th,Pu)O$_2$ fuel [39]. The authors of that study concluded that overall (Th,Pu)O$_2$ fuel is superior to the (U,Pu)O$_2$ MOX fuel for plutonium incineration.

The higher radiation resistance and chemical stability of ThO$_2$ compared to UO$_2$ (UO$_2$ may oxidise further to U$_3$O$_8$ or UO$_3$) greatly simplifies normal operations, accident planning and waste management [40]. ThO$_2$ also has better thermo-physical properties such as a lower coefficient of thermal expansion, higher thermal conductivity and higher melting point. The fission gas release rate for ThO$_2$-based fuels is also less than for UO$_2$ [32]. Gas release is a significant performance-limiting factor of nuclear fuels as it can lead to temperature increases caused by thermal conductivity decreases across the fuel clad gap [41]. Some authors also claim that the fission products themselves from $^{233}$U are about 25% less poisoning (neutronically) than from $^{235}$U and $^{239}$Pu [42].

2.2.3.2 Studies into the $^{232}$Th$-^{233}$U cycle

Despite all the proposed benefits from implementing a $^{232}$Th$-^{233}$U fuel cycle, unlike the $^{238}$U$-^{239}$Pu cycle it still remains unproven on a commercial scale. Research into utilising thorium as a nuclear fuel flourished during the mid 1950’s to early 1970’s, at a time when access to uranium resources seemed limited and the exceedingly optimistic nuclear industry was rapidly expanding. During this time, several experimental and prototype reactors were built and operated. Examples include five graphite moderated, helium cooled HTGRs which included two German built (AVR and THTR), two in the USA (Peach Bottom and Fort St Vrain) and one in the UK (Dragon) [32, 43]. These reactors successfully utilised ‘coated fuel particles’ of mixed thorium-uranium oxides and di-carbides in a graphite matrix. The coated particles can tolerate a high internal gas pressure and contain all fission products, while the high operating temperatures (≈850 °C) have high thermal efficiencies (45%) [43]. The generation IV GFR design is very similar to these reactor types except it is geared more towards efficient plutonium breeding and so operates a fast neutron spectrum with no moderator.

Four LWRs in the USA implemented thorium into their fuel cycle. Two BWR’s (BORAX IV and Elk River) and a PWR (Indian Point) used high density fuel pellets of (Th,U)O$_2$ mixed with a small percentage of highly enriched $^{235}$UO$_2$ [32]. The 60 MW$_e$ Shippingport LWBR (Light Water Breeder Reactor) was originally built as a PWR in 1958 as the world’s first full size nuclear power plant devoted entirely to peaceful uses [44]. After many years of successful operation as a commercial PWR, an experimental LWBR core was installed in 1977. This core consisted of 12 hexagonal shaped fuel modules surrounded by 15 reflector modules. Each fuel module contained a highly enriched $^{233}$UO$_2$–ThO$_2$ movable seed surrounded by a stationary (Th,U)O$_2$ blanket. The reflector modules were fertile ThO$_2$, designed to produce additional fissile $^{233}$U and prevent neutron leakage. The power within the core was evenly distributed by incorporating power-flattening blankets between the fuel and the reflector modules. These power-flattening blankets contained slightly more $^{233}$U than the blanket of the inner fuel modules. To minimise
the parasitic loss of neutrons, the reactivity of the core was controlled by moving the seeds and altering the geometry, rather than by using control rods or burnable poisons. After the reactor shutdown in 1982, chemical analysis of the fuel rods found that the LWBR contained 101.39% of the fissile fuel it started with. Therefore, the Shippingport LWBR not only showed it was possible to breed $^{233}$U from thorium in a light water environment, but also that existing reactors could be retrofitted with a new breeder core requiring only minimal design changes and operations of the plant [44–46]. To this day, the Shippingport LWBR is the only reactor that has successfully demonstrated the ability to breed $^{233}$U. Although, it is believed to be technically feasible to incorporate the thorium fuel cycle in all existing thermal and fast reactors including LWR, PHWR, HTGR, MSBR, LMFBR [32].

The 8 MWth experimental Molten Salt Reactor (MSRE) of Oak Ridge, operational between 1965-1969, successfully achieved criticality with $^{233}$U fuel [47]. The MSRE was graphite moderated with $^{233}$UF$_4$ in molten fluoride carrier salts, which acted as a combined fuel and coolant. The MSR is also proposed for Gen IV development and the epithermal neutron spectrum means that the $^{232}$Th$^{\rightarrow}$ $^{233}$U fuel cycle will achieve better performance than the $^{238}$U$^{\rightarrow}$ $^{239}$Pu cycle [47]. The liquid fuel allows online processing for continuous fission product removal.

Development of thorium based fuels stagnated in the mid 1970's mostly due to political reasons, rather than technical limitations. The US government suspended all thorium related research based on the success of LWRs, uranium breeder reactors being more efficient, the research being too immature, plutonium by-products being sought after for nuclear weapons, and the minimisation of nuclear waste production was not considered an important issue [23, 42, 48]. Furthermore, new uranium reserves were discovered and the electricity growth rate was much lower than expected [35].

The major exception is perhaps India, who have always had a strong motivation to pursue thorium based fuels. This was due to the presence of large thorium deposits, limited uranium sources and issues obtaining uranium for political reasons. Plans to begin construction of an advanced heavy water reactor (AWHR) by 2016 to be running by 2025 are currently underway. The 300 MWe reactor fuelled with $^{233}$UO$_2$−ThO$_2$ and PuO$_2$−ThO$_2$ pins is designed to breed $^{233}$U in-situ and employ a closed fuel cycle [49, 50]

In April 2013, pellets of thorium-MOX fuel consisting of 10% plutonium oxide were loaded into the Norwegian Halden research reactor. This irradiation test is expected to run for five years, collecting data on the physical and chemical suitability of thorium-MOX for eventual use in commercial reactors [51].

### 2.2.3.3 Disadvantages of the $^{232}$Th$^{\rightarrow}$ $^{233}$U cycle

Despite all the advantages the U-Th fuel has over the U-Pu cycle, there are still several disadvantages and technological challenges (which ironically pair with many of the advantages). The increased chemical stability of ThO$_2$ over UO$_2$, while allows more stable waste storage, also makes processing of the spent fuel more difficult. ThO$_2$ does not dissolve easily in concentrated
TABLE 2.3: The composition of minor actinides in spent nuclear fuel from a uranium fuelled 1GWe PWR 150 days after discharge [52] and their associated radiotoxicities [53].

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half life (yr)</th>
<th>kg/yr</th>
<th>Ci/yr</th>
<th>Sv/Ci</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{237}$Np</td>
<td>$2.1 \times 10^6$</td>
<td>20.4</td>
<td>14.4</td>
<td>$4.07 \times 10^3$</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>433</td>
<td>1.33</td>
<td>$4.53 \times 10^3$</td>
<td>$7.40 \times 10^5$</td>
</tr>
<tr>
<td>$^{242m}$Am</td>
<td>141</td>
<td>0.012</td>
<td>116</td>
<td>$7.03 \times 10^3$</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>$7.4 \times 10^3$</td>
<td>2.48</td>
<td>477</td>
<td>$7.40 \times 10^5$</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
<td>0.45</td>
<td>0.133</td>
<td>$4.4 \times 10^5$</td>
<td>$4.44 \times 10^5$</td>
</tr>
<tr>
<td>$^{243}$Cm</td>
<td>29</td>
<td>$1.96 \times 10^{-3}$</td>
<td>90.3</td>
<td>$5.55 \times 10^3$</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>18.1</td>
<td>0.911</td>
<td>$7.38 \times 10^4$</td>
<td>$4.44 \times 10^5$</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>$8.4 \times 10^3$</td>
<td>0.0554</td>
<td>9.79</td>
<td>$7.77 \times 10^3$</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>$4.7 \times 10^3$</td>
<td>$6.23 \times 10^{-3}$</td>
<td>1.92</td>
<td>$7.77 \times 10^3$</td>
</tr>
</tbody>
</table>

nitric acid (HNO$_3$) and so requires addition of hydrofluoric acid (HF) which corrodes the steel pipes in the reprocessing plant. ThO$_2$-based fuels require long dissolution times in the standard THOREX solution [13M HNO$_3$ + 0.05M HF + 0.1M Al(NO$_3$)$_3$] at 120°C. The higher melting point of ThO$_2$ (3350°C vs 2800°C for UO$_2$) translates to a much higher sintering temperature required to create high density fuel pellets from the oxide powder. While the production of $^{232}$U creates a barrier to proliferation, it also means the spent fuel is highly radioactive, delivering a higher radiation dose than the spent fuel from $^{239}$Pu. Reprocessing of the spent fuel requires an automated approach in heavily shielded hot cells which then increases the cost of the fuel processing [32].

The intermediary $^{233}$Pa has a long half life (27 days) as compared to $^{239}$Np (2.35 days) which requires storing the material for at least a year for it to completely decay to $^{233}$U. Finally, practical experience and information databases of the thorium fuel cycle is vastly lacking compared to UO$_2$ and (U,Pu)O$_2$-based fuels and significant improvements need to be made before commercial interests are likely to be developed [32].

2.3 Nuclear Waste Transmutation

Nuclear waste can be broadly categorised into three forms – Transuranics (PMA), fission products and reactor structural materials that have become activated via neutron capture e.g. Co-60. Waste with a half-life longer than 10 years, such as some fission products and transuranic elements are considered problematic for storage [23]. The transuranic elements, consisting of plutonium and minor actinides (neptunium, americium and curium) generally have long half lives and high radio-toxicities due to being alpha emitters. The only way to dispose of them is through long term geological disposal or incineration by fission. Table 2.3 shows the minor actinide content in the spent fuel of a 1 GWe PWR. The most desirable way to incinerate minor actinides is in a fast neutron reactor as the fission to capture ratio is much more favourable at higher energies for many minor actinides (see Figure 2.3).
CHAPTER 2. SURVEY OF THE SUBJECT

FIGURE 2.3: Fission to capture ratio for minor actinides – $^{237}\text{Np}$, $^{241}\text{Am}$, $^{243}\text{Am}$, $^{244}\text{Cm}$. Source: ENDF/B-VII.1 [31].

TABLE 2.4: The half lives and production rates of long lived fission products produced in a 1 GWe PWR reactor [23].

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half life (yr)</th>
<th>Production rate (kg/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{79}\text{Se}$</td>
<td>$3.0 \times 10^5$</td>
<td>0.11</td>
</tr>
<tr>
<td>$^{90}\text{Zr}$</td>
<td>$1.6 \times 10^6$</td>
<td>15.5</td>
</tr>
<tr>
<td>$^{99}\text{Tc}$</td>
<td>$2.1 \times 10^5$</td>
<td>17.7</td>
</tr>
<tr>
<td>$^{107}\text{Pd}$</td>
<td>$6.5 \times 10^6$</td>
<td>4.4</td>
</tr>
<tr>
<td>$^{126}\text{Sn}$</td>
<td>$2.3 \times 10^5$</td>
<td>0.44</td>
</tr>
<tr>
<td>$^{129}\text{I}$</td>
<td>$1.6 \times 10^7$</td>
<td>3.9</td>
</tr>
<tr>
<td>$^{135}\text{Cs}$</td>
<td>$2.3 \times 10^6$</td>
<td>7.7</td>
</tr>
</tbody>
</table>

There are seven long lived fission products with half-lives longer than 100,000 years (listed in Table 2.4). These elements decay by $\beta$ emission and may be transmuted into short lived or stable elements through neutron capture. As an example:

$$^{99}\text{Tc} (2.1 \times 10^5\text{y}) + n \rightarrow ^{100}\text{Tc} (15.46\text{s}) \rightarrow ^{100}\text{Mo} \text{ (stable)}$$ \hspace{1cm} (2.7)

where the neutron capture cross section of $^{99}\text{Tc}$ is shown in Figure 2.4. The medium lived fission products e.g. $^{90}\text{Sr}$ and $^{137}\text{Cs}$ have very high activities at discharge and due to low neutron absorption cross sections, it is unfeasible to attempt to transmute them. Therefore, they will require storage on the order of 300 years [23].

Finding an acceptable solution for the proper disposal of nuclear waste is one of the greatest challenges facing the nuclear industry. A detailed study examining the impact that Partitioning and Transmutation (P&T) will have on nuclear waste management strategies has been undertaken by the EU-funded RED-IMPACT project [54]. The study concluded that ‘fabrication of transmutation fuel is a significant technical and economical challenge’ and that the long
term geological disposal of HLW cannot be avoided irrespective of what waste management strategy is adopted. However, P&T could reduce the half-life of most PMA by a couple hundred years and efficient transmutation of MA could be carried out in an ADS or fast reactor [54]. Other large-scale studies have included the combined projects of EUROPART (partitioning) and EUROTRANS (transmutation). EUROTRANS focused on the transmutation of high level waste in an Accelerator Driven System and led to detailed design of XT-ADS [55].

Sub-critical reactors are capable of maintaining a much higher percentage of MA content in their fuel compared to critical reactors. The addition of MA in the fuel reduces the doppler coefficient and fraction of delayed neutrons. For liquid metal cooled reactors, particularly those with sodium coolant, the void coefficient also becomes increasingly positive [54, 56]. This introduces significant safety barriers and limits the MA content of fuel in critical reactors to a few percent [54].

2.4 Previous studies into ADS

The idea of using accelerators to breed fissile material was first proposed in 1952 by Lewis [16] as an alternative way of breeding fissile material without relying on the existence of $^{235}$U. Concurrently, the Electronuclear (MTA) program at Livermore Research Laboratory also worked on developing accelerator based technology, primarily for producing weapons grade plutonium [57]. Upon discovery of high grade uranium ores, the concept was considered too uneconomic and abandoned. Likewise, the original early projects investigating accelerators to transmute LWR actinide and fission products were soon terminated as the beam currents required were well beyond the reach of the available technology at the time [58].

In 1977, Grand et al. proposed that an accelerator-breeder would be a practical method for breeding $^{233}$U or $^{239}$Pu, and that construction of the required accelerator should be technically possible [59, 60]. By 1985 he was claiming that the accelerator breeder was just as competitive
as the FBR and due to technological immaturity at that time, a solid fertile target would be preferably to a liquid metal target [61]. The fertile-to-fissile conversion program (FERFICON) during the 80s investigated the feasibility of producing fissile material in accelerator breeding systems for use in power reactors [62]. In the late 80s, Tolstov [63] of the JINR in Dubna, Russia proposed introducing externally produced neutrons into a sub-critical nuclear reactor in order to sustain chain reactions. The Accelerator Driven System eventually gained mainstream attention around the early 1990’s after Carlo Rubbia of CERN [7, 9] and Charles Bowman of LANL [6, 8] promoted the idea more publicly. Rubbia’s design focused on using thorium fuel for energy production and waste transmutation, while Bowman’s was aimed more towards waste transmutation and incineration.

2.4.1 Conceptual studies into ADS

Thermal systems for the Accelerator Transmutation of Waste (ATW) have been proposed by LANL [6, 8]. Bowman et al. [6] outlines a system involving a 1.6 GeV proton beam incident on a heavy water moderated flowing lead target. Inspired by the MSRE, a molten salt loop (LiF–BeF$_2$) incorporating actinide fuel is used to further enhance the neutron flux. Bowman et al. [6] proposes that the intense thermal neutron flux provided by the accelerator ($10^{16}$ n cm$^{-2}$ s$^{-1}$, approximately 100 times higher than in an LWR) makes destruction of higher actinides possible. This is because minor actinides like $^{237}$Np may capture two neutrons in succession, converting to fissionable $^{239}$Np and avoiding production of poisonous $^{238}$Pu. However, the high flux and hence high burn up rate, requires continuous flow and on-line chemistry facilities for constant removal of stable and short-lived fission products while returning the radioactive waste products. This results in enormous technical challenges. Similarly, breeding of $^{233}$U from $^{232}$Th in the high flux would be nearly impossible, as neutron capture of $^{233}$Pa would occur before it could decay to $^{233}$U.

A few years later, a once-through 750 MW$_{th}$ system capable of processing the waste from a typical 3000 MW$_{th}$ LWR at the same rate it’s produced was envisioned [8]. It involved a 1 GeV proton beam impinging on a graphite moderated lead target. Circulating NaF–ZrF$_4$ molten salt carries the actinides and fission products through narrow channels in the graphite. The average effective flux of this system was about $2 \times 10^{14}$ n cm$^{-2}$ s$^{-1}$ with $k_{\text{eff}} = 0.96$. Other ADS designs utilising the circulation of molten salts include the Russian Cascade Sub-critical Molten Salt Reactor (CSMSR) [64–66] and the French TASSE design [67].

Carlo Rubbia’s ‘Energy Amplifier’ idea was an Accelerator Driven System for nuclear energy production [7, 9]. It consisted of a 12.5 mA 1.0 GeV proton beam with a nominal unit capacity of 1500 MW$_{th}$. The main reactor vessel is 6.0 m diameter and 30 m tall, with circulating molten lead for cooling (Figure 2.5). Relying on natural convection for cooling is an important passive safety feature, because pumps may fail in the case of a major accident, such as that which occurred at Fukushima. The fuel is composed of mixed oxides, containing a large concentration of ThO$_2$ and a closed fuel cycle incorporating the THOREX liquid separation method is proposed. The
high temperature of the molten lead (600–700 °C) allows for a high thermal efficiency of 45%. A neutron multiplication coefficient, $k_{\text{eff}}$ of 0.98 would correspond an energy gain, $G$ of 120 while operating at $k_{\text{eff}}$ of 0.96 yields $G$ to be 60 [9].

Additional ADS designs operating at a fast neutron spectrum for transmutation of nuclear waste include the PHOENIX concept of Brookhaven National Laboratory [69]. More recently, designs like HYPER (Hybrid Power Extraction Reactor) from KAERI [70], NWB (Nuclear Waste Burner) from Russia [71] and one from JAERI [72] have also been proposed. All these designs recommend cooling via lead-bismuth eutectic.

### 2.4.2 Experimental studies of ADS

An industrial scale commercial ADS has never been built, however there has been several experimental studies and prototypes constructed. The First Energy Amplifier Test (FEAT) at CERN showed that an energy gain from a sub-critical assembly was possible [73]. The experiment consisted of 3.62 ton natural uranium in 270 rods immersed in a stainless steel tank. The fuel assembly had diameter 89 cm and height 107 cm, and was moderated by water with thickness 12.5–15.5 cm. It was irradiated with a proton beam of energies 0.6–2.75 GeV and found that $G$ remains essentially constant above proton energies of 1 GeV. The $k_{\text{eff}}$ of the system was found to be $0.895 \pm 0.010$, yielding an energy gain of $29 \pm 2$, which was much more than required to power the accelerator. The neutronic behaviour of FEAT was simulated using the FLUKA code, and found to be consistent with the experimental results.
The follow up to CERN’s FEAT experiment was the TARC (Transmutation by Adiabatic Resonance Crossing) experiment [10, 74, 75]. The TARC experiment demonstrated the ability to exploit Adiabatic Resonance Crossing (ARC) for transmutation of Long-Lived Fission Products in an ADS (see Figure 2.4 for $^{99}$Tc capture cross section). The experimental apparatus was a large volume of natural lead ($3.3\text{ m} \times 3.3\text{ m} \times 3.0\text{ m}$) irradiated with 2.5 and 3.5 GeV protons. It was shown to be possible to destroy large quantities of $^{99}$Tc and $^{129}$I, outside an EA core faster than it is produced, thereby making it a practical way to reduce existing stockpiles of LLFPs. Cross section data of $^{99}$Tc and $^{129}$I was found to be accurate, while they found that $^{232}$Th(n,γ), $^{186}$W(n,γ) and $^{237}$Np(n,f) reaction cross section data lacked the required precision and should be remeasured.

Preliminary Design Studies of an eXperimental ADS (PDS-XADS) were performed within the EURATOM 5th framework programme. The PDS-XADS project aimed to study the feasibility of three conceptual XADS designs for accurate portrayal of the basic features required in an ADS prototype. The three designs studied were an 80 MW$_{th}$ LBE-cooled XADS, 80 MW$_{th}$ gas-cooled XADS and LBE-cooled MYRRHA [76].

MYRRHA (Multi-purpose hYbrid Research Reactor for High-tech Applications) is a small scale eXperimental ADS (XADS) currently under construction at SCK-CEN in Mol, Belgium [14, 77, 78]. MYRRHA was designed in 1998 and aims to be fully operational before 2025. It consists of a windowless liquid lead-bismuth eutectic (LBE) spallation target coupled to a 600 MeV 2.5 mA proton accelerator. Surrounding the target is a sub-critical multiplying medium ($k_{eff} \leq 0.95$) composed of fast reactor MOX fuel (35% Pu) and cooled with LBE. The construction of MYRRHA has several goals including demonstration of the full ADS concept, performing transmutation studies of MA and obtaining operational feedback at a power level (50–100 MW$_{th}$) appropriate for an industrial scale ADS. MYRHHA may also operate in a critical mode, where it will study the Lead-cooled Fast Reactor (LFR) tagged for Gen IV development. This includes developing lead-bismuth eutectic technology and material irradiation studies in a fast neutron spectrum.

The YALINA facility operating since 1997 at the National Academy of Sciences of Belarus is a zero-power (no temperature effects), sub-critical assembly used for studying the small scale neutronics of an ADS [79]. YALINA was designed as a low-cost experiment by forgoing the spallation neutron source, instead relying on $^3\text{He}$, $^4\text{He}$ reactions or a $^{252}$Cf spontaneous-fission neutron source placed in the core. Despite the differing external neutron drivers, due to astute engineering, YALINA satisfactorily reproduces the neutron spectra of a large scale ADS. The thermal YALINA-T core consists of enriched uranium pins placed in channels of polyethylene blocks and the core is surrounded by a graphite reflector and cadmium sheet. A fully loaded core has a $k_{eff} < 0.98$. Later, the booster YALINA-B was built which extends the source region to more accurately emulate the spatial distribution and time profile of a spallation neutron source.
2.5. Spallation reactions

The MUSE program at the MASURCA facility at Cadareche was also aimed at studying the neutronics of a fast sub-critical assembly in a low-power (<5 kW) configuration. The primary motivation behind the MUSE experiments was to investigate transmutation of minor actinides and long-lived fission products in an ADS [80].

Other experimental studies into ADS include TRASCO (Italian: TRAsmutazione di SCOrie) which investigates nuclear waste transmutation in an ADS design based on Carlo Rubbia’s EA proposal [81]. The TRASCO-ADS project involved theoretical and experimental evaluation of a windowless interface between the vacuum of the accelerator and the LBE spallation target [82]. The TRADE (TRiga Accelerator Driven Experiment) project was a novel idea to couple an external proton accelerator and spallation target with the existing 1 MW TRIGA Mark II reactor at ENEA in Casaccia, Italy [83, 84].

2.5 Spallation reactions

Spallation is an inelastic nuclear collision that occurs when an energetic particle (proton, neutron, deuteron, pion etc.) interacts with a heavy nucleus leading to the ejection of several lighter particles. The concept of nuclear spallation was first devised in 1937 in the PhD thesis of Glenn Seaborg [85]. No formal definition of spallation exists and there is no clear boundary between reactions categorised as ‘spallation’ and those occurring at lower energies [15]. However, the first attempt to describe a basic reaction mechanism was by Serber [86]. He noted that at sufficiently high energies of $\gtrsim 100$ MeV, the reaction no longer proceeded via the formation of a compound nucleus. The deBroglie wavelength of the incident particle becomes similar to or smaller than the distance between nucleons in the nucleus, allowing the incident particle to interact with single nucleons rather than the nucleus as a whole. The collision time between the incident particle and a nucleon in the nucleus also becomes short compared to the time between collisions of the nucleons.

The spallation process is modelled in a series of several stages. The first stage, the Intra-Nuclear Cascade (INC) involves the incident particle colliding with individual nucleons in the nucleus which is then followed by a series of direct reactions between the nucleons. It is a very fast reaction, occurring within $10^{-22}$ s and leads to secondary particles (neutrons, protons and pions) of energies greater than 20 MeV being ejected in mainly the same direction as the incident projectile. These high energy secondary particles may also trigger further Inter-Nuclear Cascades in other nuclei, if target is thick. After the INC, the nucleus remains in a highly excited state and relaxes by isotropically ‘boiling off’ low-energy (<20 MeV) particles. This mostly includes neutrons, but also protons, deuterons, alpha particles, tritons, light heavy ions, pions, residuals etc. This evaporation stage, occurs within $10^{-18}$ s or less. For very heavy target nuclei (Z>72) e.g. lead, bismuth, tungsten, thorium and uranium, high-energy fission may also compete with evaporation. After the excitation energy of the residual nucleus falls below the neutron binding
energy (i.e. all particle decay modes have exhausted), the remaining de-excitation occurs by emission of $\gamma$-rays.

2.5.1 Studies of spallation sources

The spallation source is a fundamental component of an ADS and research efforts into ADS focuses heavily on spallation targets. This includes neutron production and transport, heat removal, material stress and fatigue, radiation damage and induced radioactivity.

Experimental studies into spallation reactions are categorised into either “thin” or “thick” target experiments. The differentiation between a thin and thick target is determined by the probability of secondary particles escaping the target. Secondary particles produced in a “thin” target do not undergo any further collisions inside the target and the energy loss of the incident beam is very small compared to the beam energy. They are best suited for investigating the intra-nuclear cascade, subsequent evaporation/fission reactions including yield of secondary particles and differential cross section data, making them ideal “code validation experiments” [15]. In “thick” targets, a significant portion of the neutron yield\(^1\) arises from hadronic cascades of nuclear reactions within the bulk of the target material – from secondary and higher-order reactions induced by the reaction products themselves. Thick targets are useful for studying properties beneficial to the application of spallation sources including particle leakage spectra, particle multiplicities\(^1\), energy deposition and induced radioactivity [15].

The first experimental efforts into the investigation of neutron yields on thick targets were carried out in 1955 by Cohen [87]. Although not technically a spallation reaction, he measured the angular distribution of neutrons from (p,n) reactions on Mg, Al, Cu, Mo, Ag, Ta, Au, Th and U induced by 23 MeV protons. Cosmic-rays have also been used as a source of protons between 250 and 900 MeV to measure low-energy neutron production in U, Pb, W and Sn [88]. Throughout the 1960s research into spallation neutron sources was dominated by groups at JINR in Dubna, BNL and ORNL [15]. Measurements collected at the Brookhaven Cosmotron accelerator in 1965 by Fraser et al. [89] remained the most comprehensive data-set with regards to beam energies and target materials for many years [15]. Data was collected for Be, Sn, Pb and $U_{\text{depleted}}$ for protons between 500 and 1500 MeV. Further measurements of neutron yield on thick targets has also been completed by Lone et al. [90], Becker et al. [91], Vasil’kov et al. [92, 93] and Yurevich et al. [94]. Double-differential cross section measurements of neutron production of 0.8, 1.2 and 1.6 GeV protons on 2 cm thick lead targets were conducted at the SATURNE accelerator in Saclay [95], whilst neutron leakage spectra from 0.5 and 1.5 GeV proton beams on lead targets were measured at KEK, Japan [96].

The first studies into a spallation target operating at the high power load expected for future ADS targets has been the carried out by the Megawatt pilot target experiment (MEGAPIE) [97, 98]. The 920 kg liquid lead-bismuth eutectic target was irradiated by a $\sim$1 MW 590 MeV proton

\(^1\)The terms neutron yield, neutron production and multiplicity are often used interchangeably. They are defined as the integral number of neutrons produced per one incident particle.
Spallation reactions

beam for 4 months (cumulative current 2.8 Ah) in 2006. Operating at the SINQ facility of the Paul Scherrer Institut (PSI), MEGAPIE demonstrated the advantages of a flowing liquid target and convectional cooling by allowing for higher power (and neutron flux) densities. MEGAPIE collected an enormous amount of data into the lifetime performance of the target including thermal hydraulics and stress analysis [99, 100], and neutron [101, 102], nuclide [103] and gas production [104].

2.5.2 Optimal parameters of spallation target

The spallation target must be optimally designed and constructed as it is such an imperative component of an ADS. In general, the target should allow easy extraction of heat and encourage the development of nuclear cascades to maximise neutron yield. Increasing the neutron yield increases the energy gain and output power of ADS (see Eq. 2.1 on page 3). The neutron yield strongly depends on the target material type and geometry as well as the projectile type and energy.

2.5.2.1 Target material type

Several potential materials for use as ADS targets have been proposed. These include lead, lead-bismuth eutectic, mercury, tantalum, tungsten, uranium and thorium and may be categorised into fissionable solids, non-fissionable solids or non-fissionable liquids. Apart from high neutron production, the ideal properties of the target material should also take into account the induced radioactivity, thermal conductivity, melting point, radiation induced material damage, life expectancy, economics and safety. Most studies into target materials has focused on natural lead given it is a popular choice based on its high nuclear density, abundance in nature, high thermal conductivity and very high neutron transparency (due to doubly magic $^{208}$Pb). One of the greatest technical challenges of high power spallation targets is the ability dissipate heat quickly. This can be more easily achieved with a flowing liquid target design. Lead-bismuth eutectic (LBE, Pb$_{0.45}$Bi$_{0.55}$) shows such promise due to its minimal neutron absorption and similar neutron yield and lower melting point as compared to elemental lead. In 2006, a demonstration experiment of an LBE target for application in an ADS as part of the MEGAPIE project showed “excellent performance of the target and ancillary systems” and “superb neutronic efficiency” [97, 98, 105]. Mercury targets have also been considered but have a much higher thermal neutron absorption cross-section and chemical toxicity compared to LBE. However, in cases where generation of high neutron flux is not important, mercury targets are preferable to LBE because they have a higher density, do not require prior heating (liquid at room temperature) and do not produce alpha emitting polonium isotopes (from transmutation of bismuth) [15]. Lead-gold eutectic targets (LGE) of composition Pb$_{0.841}$Au$_{0.159}$ are also being considered for their lower chemical toxicity compared to mercury and their lower polonium production compared to LBE targets [13]. As an
A Monte Carlo study into comparing neutron production rate of various target materials has been performed by Hashemi-Nezhad et al. [17]. Assuming a 1 GeV proton striking a target with 20 cm diameter and 170 cm length, while ignoring neutrons produced from secondary neutron induced fissions, the authors have concluded that a uranium target would provide the highest production rate and hence energy gain for an ADS. Including secondary fission reactions would increase the neutron production rate of $^{238}$U and $^{232}$Th significantly. The neutron yield calculated for these metallic target materials are shown in Figure 2.6.

Even though uranium targets show the greatest potential for producing the highest energy gain in an ADS [17], differing opinions with regards to the suitability of uranium targets are currently under debate [17, 108–110]. Targets made from fissionable materials have shown evidence of limited operational lifetimes [109] and the high fission rate results in significant heating which must be cooled using a special cooling mechanism [17]. Natural, depleted and enriched uranium targets have been used at spallation facilities at KENS (KEK), IPNS and ISIS (RAL) but were found to experience problems due to radiation damage and swelling [15]. Uranium targets also introduce proliferation concerns from plutonium production and containing several masses of critical fissionable material [109]. Thus, non-actinide liquid targets (Pb, PbBi, Hg) may be preferable for high power spallation sources [15], and some consider metallic uranium unsuitable for high power spallation sources [109] or full-scale ADS [110].

### 2.5.2.2 Projectile type

The main factor when deciding the projectile type for spallation neutron production is to maximise energy loss via secondary particle production and minimise energy loss due to heating from...
2.5. Spallation reactions

![Graph showing neutron yield per ion charge as a function of ion energy per ion charge for protons, deuterons, $^3$He, $^4$He and $^{12}$C incident on a cylindrical lead target (diameter 20 cm, length 60 cm) [93].](image)

ionisation reactions. The neutron production rate per beam energy is highest for projectiles of hydrogen isotopes [17].

Most previous studies into ADS focused on using protons as the incident projectile. However, theoretical [17, 111, 112] and experimental [93, 94, 113, 114] studies have shown that neutron yield can be increased by the use of deuterons as compared to using protons of the same energy. Barashenkov et al. [111] found that the neutron yield increased by about 15% for incident deuterons as compared to proton, while heat deposition remained constant for a 1 GeV beam on lead target. However, a significant heating increase occurred for uranium targets but this was not to the extent of the increase in neutron yield. The heating increase was caused by the increase in number of fissions which contributes to 60–70% of the heating. The experimental data of Vasil’kov et al. [93] is shown in Figure 2.7.

At incident energies above 100 MeV, the nuclear collision cross section, $\sigma_{in}$ is approximately constant with energy, increasing slowly with increasing ion and target mass [115]:

$$\sigma_{in} \propto \left( \frac{A_{ion}^{1/3} + A_{target}^{1/3}}{3} \right)^2$$  (2.8)

The stopping power i.e. rate of ionisation energy loss of an ion may be expressed in terms of the stopping power of a proton, $S_{proton}$ in the following way [15]:

$$S_{ion} = Z_{ion}^2 S_{proton} \frac{E_{ion}}{A_{ion}}$$  (2.9)

where, $S_{ion}$ is the stopping power of an ion with mass, $A_{ion}$ and kinetic energy, $E_{ion}$. The increase in neutron yield of a deuteron, as compared to the proton is because it has similar ionisation losses (Eq. 2.9) but a larger nuclear collision cross section (Eq. 2.8). The deuteron also consists of a weakly bound proton and neutron which may lead to an extra “source” of neutrons from stripping reactions [116, 117].
Increasing the mass and charge of the incident ion requires increasing the kinetic energy of the ion, to minimise ionisation losses in favour of inelastic nuclear collisions. For a given beam energy, the neutron yield per ion charge is lower for ions with higher mass, as seen in Figure 2.7.

2.5.2.3 Projectile energy

In order to produce significant numbers of neutrons, the range of the incident particle, $R_{ion}$ needs to be much larger than the mean free path of nuclear collisions, $\lambda$ (Eq. 2.11) [15]. The range of the projectile increases with kinetic energy (Eq. 2.10), and significant neutron production does not begin to occur unless beam energies are higher than several hundred MeV. This is also approximately independent of target material [15]. The purpose of using higher beam energies is to increase neutron production, rather than increase maximum neutron energy (most neutrons are emitted at a few MeV).

$$R_{ion} = \frac{A_{ion}}{Z_{ion}^2} R_{proton} \frac{E_{ion}}{A_{ion}}$$

$$\lambda = 33A^{1/3} (\text{g cm}^{-2}) \quad \text{for } E \geq 100 \text{ MeV}$$

The number of neutrons produced in a lead target (diameter 20 cm and length 170 cm) calculated by Hashemi-Nezhad et al. [17] for proton and deuterons as a function of incident ion energy is shown in Figure 2.8. The neutron yield per GeV increases sharply up to 1 GeV and then decreases slowly above 2 GeV. The maximum neutron production rate occurs at $\sim$1.5 GeV and it is noted by Hashemi-Nezhad et al. [17] that this energy is about the same for all four target materials studied in the paper (W, Pb, Th and U).
The reason for the decrease in the neutron production rate at higher beam energies is because pion production becomes significant at energies greater than 1–2 GeV. The production of pions means energy for neutron production is lost. The production of \( \pi^0 \) contributes to the so called electromagnetic drain of the hadronic cascade. This is because their mean lifetime of \( 8.3 \times 10^{-17} \text{s} \) is too short to engage in the internuclear cascade and the subsequent decay into \( 2\gamma \) leads to loss of neutron producing energy from the system. However, the charged pions, \( \pi^\pm \) mean lifetime \( (2.6 \times 10^{-8} \text{s}) \) is long enough to engage in further internuclear cascades once created. Pions are an important contributor to energy deposition in the system and a significant muon source. At energies higher than 10 GeV, other production channels become significant (e.g. kaon production), leading to further decrease in the neutron production rate.

The economy of neutron production tends to suggest that there is no advantage to beam energies greater than 1–2 GeV. However, increasing the beam energy (up to 10 GeV) decreases the required current from the accelerator and reduces radiation damage to the accelerator and target beam windows [118].

### 2.5.3 Spallation studies at the JINR

This PhD study was completed as part of the E&T RAW (Energy and Transmutation of RAdioactive Wastes) collaboration, based at the Joint Institute for Nuclear Research (JINR) located in Dubna, Russia. The E&T RAW collaboration began in the late 1990’s and now contains approximately 40 members from 13 countries – Armenia, Australia, Belarus, Bulgaria, Czech Republic, India, Germany, Greece, Mongolia, Poland, Russia, Serbia and Ukraine.

The collaboration is particularly focused on experimentally studying the energy and spatial distribution of spallation neutrons produced by GeV protons and deuterons and utilising this data for the benchmarking of computational codes, spallation physics models and cross section data libraries. Experimental studies into the spatial distribution of fission, \((n,\gamma)\) and \((n,xn)\) reaction rates, the transmutation of nuclear wastes such as \(^{239}\text{Pu}\), \(^{241}\text{Am}\), \(^{129}\text{I}\) and \(^{238}\text{Pu}\) in target set-ups, and applications of spallation neutrons in Accelerator Driven Systems are all encompassed in the main objectives. Comprehensive experimental studies into these reactions, and the neutronics of these systems in general cannot be achieved through the use of a single particle detector type. Rather, several different detector types employing many experimental techniques must be used in a coordinated manner. Examples include activation analysis, Solid State Nuclear Track Detectors (SSNTDs), He-3 detectors, ionisation chambers, nuclear emulsions and scintillation detectors.

The E&T RAW collaboration has successfully built four target systems – Gamma-2, Energy plus Transmutation (EpT), Gamma-3 and Quinta. Experiments conducted within these target systems present challenges that are unique to this environment, including:

- There is not much free space in the assemblies so the detectors must be small enough to fit in the empty spaces
CHAPTER 2. SURVEY OF THE SUBJECT

There is a wide range of neutron energies present e.g. the Gamma-3 assembly contains thermal, epithermal, and spallation neutrons that span energies of more than 11 orders of magnitude.

As well as neutrons, there are also charged particles present such as protons, deuterons, pions, tritons and alphas etc.

There is a huge background gamma radiation.

The spatial distribution of neutrons and energy spectrum changes rapidly on a small geometrical scale (<1 cm).

These factors must all be taken into account when choosing which experimental technique is most appropriate.

2.5.3.1 Gamma-2 set-up

The Gamma-2 set-up consists of a lead target (diameter of 8 cm, length of 20 cm) surrounded by 6 cm of moderating paraffin. If desired, the centre of the lead target could be replaced with uranium (Figure 2.9). Gamma-2 was irradiated with protons ranging from 0.5–7.4 GeV as well as with neutrons from a Pu-Be source placed internally. SSNTDs such as LR-115 2B [119, 120], CR-39/LR-115 2B combination [121, 122] and $^{235}$U on Makrofol [122] were used to measure slow neutrons, while recoils in CR-39 [119, 121, 123], cadmium covered CR-39 [122] and $^{232}$Th on Makrofol [122] used for fast neutron detection. Radiochemical $^{139}$La sensors were also used to detect thermal neutrons [120, 124]. Transmutation rates of long lived nuclear waste products $^{129}$I, $^{237}$Np [124, 125], and $^{239}$Pu [126] were measured. The spallation neutron spectrum has also been experimentally unfolded using $^{209}$Bi, $^{197}$Au, $^{59}$Co, $^{115}$In and $^{232}$Th threshold detectors [127].

2.5.3.2 EpT set-up

EpT consisted of a lead target encompassed by a $^{nat}$U blanket and was used for neutron production and transport benchmark studies [128]. Polyethylene surrounded the target-blanket to act as a neutron shield/moderator/reflector and the addition of cadmium sheet served to prevent thermal neutrons from re-entering the blanket region. Detailed experimental and Monte Carlo
2.5. Spallation reactions

FIGURE 2.10: The EpT set-up contains a lead target surrounded by a blanket of $^{nat}U$ rods. Cadmium sheet (pink) and polyethylene moderator (yellow) are also shown.

studies into the particle field of the EpT irradiated with 4 GeV deuterons is described in the PhD thesis of Borger [129]. The PhD thesis of Krasa [130] studied the spatial distribution of spallation neutrons produced by 0.7, 1.0, 1.5 and 2.0 GeV protons using $^{27}$Al, $^{197}$Au, $^{209}$Bi and $^{59}$Co activation threshold detectors. Likewise, the PhD thesis of Svoboda [131] employed similar experimental methodology to investigate the production and transport of high energy neutrons from 1.6–4.0 GeV deuteron irradiations.

Additional experimental studies of EpT include the transmutation of $^{129}$I, $^{237}$Np, $^{238}$Pu, $^{239}$Pu and $^{241}$Am irradiated by 0.7, 1.0, 1.5 and 2.0 GeV protons [132] and $^{129}$I, $^{237}$Np, $^{238}$Pu and $^{239}$Pu irradiated with 2.52 GeV deuterons [133]. Activation studies determining the (n,$\gamma$), (n,f) and (n,2n) reaction rates of $^{232}$Th and $^{nat}U$ [134] and measurement of $^{nat}U$ fission rate using mica SSNTDs [135] have also been performed. The neutron multiplicity of the system has been calculated using a modification of the water-bath/activation-foil method [136]. Activation measurements of the (n,xn) reactions in $^{89}$Y were used to unfold the neutron energy spectrum between 10 and 50 MeV [137] and calculations of the neutron dose rate for radiation dosimetry have been conducted [138].

Comparisons of Monte Carlo calculations and the EpT experiments have so far shown mixed agreements. In general, comparisons with fission track detectors have yielded good agreements [135, 139, 140], while activation detectors [131, 132, 134, 141] have not been so successful. Comparisons using CR-39/LR-1152B detectors [129] have achieved partial agreement – the relative spatial distribution of neutrons can be reproduced but results required normalisation. It is difficult to speculate whether the discrepancies are due to issues in the experiment or problems with the calculation, such as a fault in the nuclear data or the models themselves.

The Quinta assembly (see section 3.1.2) is the most recent experimental set-up for the E&T RAW collaboration and is similar in design to the EpT set-up. However, instead of a lead target, there is an all uranium target-blanket with no moderating polyethylene to produce a fast neutron
spectrum. Quinta was designed to provide a high energy, high flux, intense neutron source for transmutation and fission yield studies in a fast spectrum ADS.

2.5.3.3 Gamma-3 set-up

The Gamma-3 assembly (more details in section 3.1.1) was designed to study transmutation rates in the neutron field of a thermal Accelerator Driven System and is the only experimental facility for studies into graphite moderated spallation neutron sources. It was built after previous studies showed that on average, the $^{232}$Th(n,$\gamma$) reaction rate in a graphite moderated system is higher than in a lead moderated system [142]. (n,f), (n,$\gamma$) and (n,2n) reaction rates of $^{232}$Th and natU under 2.33 GeV deuteron irradiation have been measured previously [141]. However, it was found that the calculated $^{232}$Th(n,$\gamma$) reaction rate was more than two times higher than the experimentally measured rate, while the $^{232}$Th(n,f) reaction rate was less than half the experimental result [141].

2.6 Motivation for this work

The choice of projectile, as well as the geometry and material of the target, fuel, moderator, coolant and radiation shielding, covers an enormous parameter space making the execution of physical experiments to deduce the optimal design of an ADS a prohibitively complicated task. Therefore, the successful development of ADS in the future will inevitably rely upon well validated and benchmarked nuclear cross section data, spallation physics models and Monte Carlo codes.

The neutron flux, and the spatial and energy distribution of these neutrons are the most important parameters for understanding the behaviour of a nuclear system [143]. In an ADS, other secondary particles (such as protons and pions) can also contribute to the energy gain, isotope production, radiation damage, induced radioactivity of target and other structural materials, neutron multiplicity and breeding of fissile material in the system. Comprehensive knowledge of neutron, proton, pion and photon cross sections and radioisotope production data is therefore also required.

This work studied the neutronic environment of thermal and fast experimental ADS set-ups irradiated with GeV deuterons through experimental and Monte Carlo methods. Few studies into the use of deuteron projectiles for ADS have been performed previously, despite evidence to suggest they are more efficient than protons. Reliability of GeV physics models can only be improved through the collection of extensive experimental data, which is currently sparse and inconsistent.

Engineering and technical issues related to the development of ADS, such as the reliable production of a high current accelerator, development of materials suitable for use in an intense radiation field, and solutions to cooling solid targets are non-trivial problems but were beyond the scope of this study.
Chapter 3

Experimental methodology

3.1 Major Experimental Facilities used

3.1.1 The Gamma-3 assembly

The Gamma-3 assembly consists of a cylindrical lead target centred in a large block of high purity reactor grade graphite. The lead target is 8 cm in diameter, and the graphite (density \(1.7 \text{ g cm}^{-3}\)) has dimensions 110 cm \(\times\) 110 cm \(\times\) 60 cm (see Figure 3.1). There are several channels within the graphite where experimental samples may be placed e.g. nuclear waste samples for transmutation studies. The length of the target was 58.8 cm long and so fell short of the graphite length by 3 mm at the front and 9 mm at the back surface. In principle, the target should start within the moderator so as to minimise loss of backward emitted spallation neutrons. However, the current target length was chosen to provide consistency with previous Gamma-3 experiments (e.g. Adam et al. [141]).

3.1.2 The Quinta assembly

The Quinta target-blanket consists of a total of 512 kg of natural uranium in five sections. Each section is 114 mm long and separated by a 17 mm airgap, where samples mounted onto sample plates may be easily placed (Figure 3.2).

The uranium is in the form of rods, where each rod is 36 mm in diameter, 104 mm long and has a mass of 1.72 kg. These dimensions are inclusive of a 1 mm thick imperviously sealed aluminium casing, which is present for safety reasons.

Excluding the first section, 61 rods are arranged in a hexagonal lattice configuration with pitch size of 36 mm and enclosed in a hexagonal aluminium container with wall thickness of 5 mm (Figure 3.3b). The first section contains only 54 rods, the removal of the central 7 is to create a beam window. This beam window is 80 mm in diameter and serves to reduce the loss of backward emitted/scattered neutrons (Figure 3.3a). The front and back of each section are then covered by aluminium plates, 350 mm \(\times\) 350 mm \(\times\) 5 mm. These plates, which also provide structural support, are then mounted onto a single slab of 25 mm thick aluminium. The
CHAPTER 3. EXPERIMENTAL METHODOLOGY

FIGURE 3.1: Photographs of the Gamma-3 assembly: (a) front view, and (b) side view. The lead target is 8 cm in diameter and the graphite is 110 cm × 110 cm × 60 cm. The graphite consists of 25 blocks and experimental samples may be placed internally within the cylindrical channels.

FIGURE 3.2: (a) Photograph of the bare Quinta target-blanket (Image: Furman et al. [108]), and (b) 3D image with transparent first plate to allow clear viewing of the uranium rods. The positioning of the sample plates may also be seen.
3.1. Major Experimental Facilities used

The entire uranium target-blanket reaches a length of 638 mm and a mass of 538 kg including all construction materials.

The uranium target-blanket is surrounded by 100 mm thick lead bricks on all six sides (Figure 3.4). This serves as a neutron reflector and to some extent as biological shielding for $\gamma$-rays. The roof of this lead shielding is supported by 18 mm of aluminium and there is a 150 mm x 150 mm square window at the front of the lead castle. Sample plates can be inserted and removed easily into the gaps between the sections, as well as on the front and back of the target-blanket by the presence of slots and lids located on the roof of the lead castle. These plates were labelled 0–5 (as indicated in Figure 3.9 on page 39) and will be referred to accordingly throughout this manuscript.

The $k_{\text{eff}}$ of the Quinta assembly was calculated to be 0.24. Removal of the lead reflector slightly reduces it to 0.23 and flooding with water raises it to 0.56.
CHAPTER 3. EXPERIMENTAL METHODOLOGY

FIGURE 3.4: Photograph of the Quinta assembly in the F3 experimental hall of the Nuclotron. Nuclear track and activation detectors can be seen mounted on the surface of the lead castle (blue) outside the Quinta assembly. The beam extraction pipe and several independent beam monitoring devices are also visible.

3.1.3 Nuclotron Accelerator at JINR

The Gamma-3 and Quinta assemblies were both irradiated with deuteron beams extracted from the Nuclotron accelerator. The Nuclotron accelerator is located in the Veksler and Baldin Laboratory of High Energy Physics (VBLHEP) within the Joint Institute for Nuclear Research (JINR), Dubna, Russia (Figure 3.5). It was proposed in 1973, with construction beginning in 1987 and eventual commissioning in 1993. The Nuclotron was the world’s first superconducting synchrotron, designed to accelerate nuclei and heavy multi-charged ions (up to uranium) up to 6 GeV/u. It operates at a temperature of 4.5 K under a magnetic field of 2 T generated from 6 kA of current [144–146].

The Nuclotron accelerator is currently undergoing a major upgrade into an ion collider – the Nuclotron-based Ion Collider fAcility (NICA). The NICA project is part of a 7 year (2010-2016) plan, aimed at studying hot and dense baryonic matter and nucleon spin structure with polarised protons and deuterons [147].

3.2 December 2011 Irradiation Details

In December 2011, one irradiation of the Gamma-3 assembly (1.6 GeV deuterons) and three irradiations of the Quinta assembly (1, 4 and 8 GeV deuterons) were scheduled. All proceeded as planned apart from the 8 GeV deuteron irradiation which was terminated due to a power outage.
3.2. December 2011 Irradiation Details

FIGURE 3.5: (a) Layout of the Nuclotron and Synchrophasotron accelerator site. Irradiations were performed in Experimental Hall F3. In order to save space, the Nuclotron was built in a tunnel 3.7m below the Synchrophasotron (built in 1957). The Nuclotron ring has a larger circumference than the Synchrophasotron (251.5 vs 207.3 m) but is much slimmer due to the strong focusing system and use of more compact, helium cooled superconducting magnets. Image: Kovalenko [146]. (b,c) Photographs showing inside the Nuclotron accelerator.

3.2.1 Gamma-3 irradiation details

The target of the Gamma-3 assembly was irradiated with a pulsed 1.6 GeV deuteron beam extracted from the Nuclotron accelerator, over a period of approximately 17 hours in total. However, after 56 minutes, the irradiation was interrupted to allow removal of the highly sensitive CR-39 track detectors (described in section 3.6). The activation samples and fission track detectors (described in section 3.4 and 3.5, respectively) were left undisturbed for the entirety of the irradiation period. Further details of the irradiation may be seen in Table 3.1.

The distribution of the intensity of the deuteron beam pulses as extracted from the Nuclotron accelerator is shown in Figure 3.6. These pulses were measured with an ionisation chamber monitored by the Nuclotron accelerator operators. On average, the Nuclotron delivers a pulse every 8 seconds. Significant variation in intensity of the beam pulses is noted throughout the entire irradiation period, as well as extended beam stoppage in some periods. This variation in beam intensity does not affect the passive Solid State Nuclear Track Detectors but it does affect the induced activity of the activation detectors. Further corrections are required to account for the discontinuous and intermittent production and decay in the activation samples.

The total number of deuterons to hit the target was determined from the activation of aluminium via beam induced $^{27}$Al(d,x)$^{24}$Na reactions (further details in section 3.3). The total number of deuterons on target for the long (17 hours) irradiation was found to be $(1.3 \pm 0.2) \times 10^{13}$. No specific measurement for the number of deuterons on target was performed for the short
#### TABLE 3.1: Details of the Gamma-3 irradiation.

<table>
<thead>
<tr>
<th>Location</th>
<th>Nuclotron Accelerator, JINR, Russia</th>
</tr>
</thead>
<tbody>
<tr>
<td>Date</td>
<td>December 11-12\textsuperscript{th}, 2011</td>
</tr>
<tr>
<td>Incident Ion</td>
<td>Deuteron</td>
</tr>
<tr>
<td>Ion Energy</td>
<td>1.6 GeV</td>
</tr>
<tr>
<td>Irradiation Duration</td>
<td>17h 3min / 56min*</td>
</tr>
<tr>
<td>Deuterons on Target</td>
<td>(1.30 ± 0.16) × 10\textsuperscript{13} / (2.9 ± 0.4) × 10\textsuperscript{11}*</td>
</tr>
<tr>
<td>X\textsubscript{c} (cm)</td>
<td>0.14 ± 0.02</td>
</tr>
<tr>
<td>Y\textsubscript{c} (cm)</td>
<td>0.46 ± 0.01</td>
</tr>
<tr>
<td>FWHM\textsubscript{x} (cm)</td>
<td>4.50 ± 0.70</td>
</tr>
<tr>
<td>FWHM\textsubscript{y} (cm)</td>
<td>1.56 ± 0.04</td>
</tr>
</tbody>
</table>

\* Short irradiation. See section 3.2.1 for details.

![Distribution of deuteron pulses during the Gamma-3 irradiation period as measured with an ionisation chamber. The short irradiation period is shown in red and the long irradiation period shown in red+blue.](image)

(56 min) irradiation. However, the number of deuterons extracted from the Nuclotron ring was provided by the accelerator operators to be $5.26 \times 10^{11}$ and $2.37 \times 10^{13}$ for the short and long irradiation periods, respectively. These values are known to be systematically higher than the number of deuterons on target due to losses in beam transport and bending from the Nuclotron to the F3 irradiation hall (the Nuclotron ring is about 4 m below the irradiation hall). The same beam-target geometry was used for the short and long irradiation periods. Therefore the ratio of deuterons extracted from the ring to that measured on target (via Al-activation) was assumed to remain constant for the short and long irradiation periods [123]. This yielded the total number of deuterons on target for the short irradiation period to be $(2.9 \pm 0.4) \times 10^{11}$.

### 3.2.2 Quinta irradiation details

The Quinta target was irradiated three times with 1, 4 and 8 GeV deuterons. The three irradiation periods lasted approximately 21, 17.5 and 9.5 hours, respectively. Further details of the irradiation
TABLE 3.2: Details of the three Quinta irradiations.

<table>
<thead>
<tr>
<th>Location</th>
<th>Location Nuclotron Accelerator, JINR, Dubna, Russia</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incident Ion</td>
<td>Deuteron</td>
</tr>
<tr>
<td>Ion Energy</td>
<td>1 GeV 4 GeV 8 GeV</td>
</tr>
<tr>
<td>Date (Dec 2011)</td>
<td>14-15th 16-17th 18-19th</td>
</tr>
<tr>
<td>Irradiation Duration</td>
<td>21h 6min 17h 30min 9h 31min</td>
</tr>
<tr>
<td>Deuterons on Target</td>
<td>$(1.50 \pm 0.04) \times 10^{13}$ $(1.94 \pm 0.05) \times 10^{13}$ $6.3 \times 10^{10}$*</td>
</tr>
<tr>
<td>$X_c$ (cm)</td>
<td>1.3 $\pm$ 0.2 1.4 $\pm$ 0.2 $-0.5 \pm 0.2$</td>
</tr>
<tr>
<td>$Y_c$ (cm)</td>
<td>$-0.2 \pm 0.2$ $0.2 \pm 0.2$ $0.0 \pm 0.2$</td>
</tr>
<tr>
<td>FWHM$_x$ (cm)</td>
<td>2.6 $\pm$ 0.3 1.5 $\pm$ 0.3 0.6 $\pm$ 0.3</td>
</tr>
<tr>
<td>FWHM$_y$ (cm)</td>
<td>3.5 $\pm$ 0.3 1.4 $\pm$ 0.3 1.2 $\pm$ 0.3</td>
</tr>
</tbody>
</table>

* Early termination due to power outage midway through irradiation period.

FIGURE 3.7: Distribution of deuteron pulses during the 1 and 4 GeV Quinta irradiation periods as measured with an ionisation chamber.

periods may be seen in Table 3.2. In order to prevent incident beam deuterons from travelling straight through the gaps between the uranium rods, the target was rotated 2° relative to the beam axis on the $x$–$z$ plane (Figure 3.3).

The distribution of deuteron pulses extracted from the Nuclotron over the 1 and 4 GeV irradiation periods is presented in Figure 3.7. Similar to the Gamma-3 irradiation period, the variation in beam intensity is quite dramatic over the course of the irradiation. Corrections are therefore required for the activation samples due to the intermittent activity production.
3.3 Measurement of incident beam deuterons

Prior to the irradiations, polaroid films were placed on the front of the Gamma-3 and Quinta targets and several test shots of the beam were fired. This was to ensure the target axis was properly aligned and the deuteron beam was striking in the centre of the target.

The total number of deuterons to hit the target was determined from the activation of aluminium via beam induced $^{27}$Al(d,x)$^{24}$Na reactions [108]. Three independent aluminium foil monitors were placed between the beam output and the front of the target. These monitors were placed between 2 and 3m away from the front of the target to prevent high-energy backscattered neutrons contaminating the measurement. The main source of uncertainty in the number of deuterons on target is that of the $^{27}$Al(d,x)$^{24}$Na cross section as only three experimental values for the cross section exist in the GeV range – 2.33 [148], 6.0 and 7.3 GeV [149]. The cross sections at 1, 1.6, 4 and 8 GeV were estimated from interpolating the curve fitted to experimental cross section data in the energy range of 0.1–7.3 GeV. The three foils were each analysed independently and good agreement between all three measurements were reached [108].

The beam positioning and beam shape was found using an array of fission track detectors measuring beam induced $^{nat}$Pb(d,f) reactions. These were placed directly on the front of the targets (see Zhuk et al. [150] for method). The $^{nat}$Pb(d,f) reaction rates determined from the track detectors were fitted to a Gaussian to obtain the beam profile. The fitted peak centroid and FWHM along the X and Y axes are displayed in Tables 3.1 and 3.2 for Gamma-3 and Quinta irradiations, respectively.

3.4 Neutron Activation Analysis

Neutron Activation Analysis (NAA) is a highly sensitive, non-destructive technique that is typically used for trace analysis of isotopes in materials. It traditionally involves irradiating a sample containing unknown isotopic concentrations with a known flux of (usually) thermal neutrons. The sample becomes radioactive, decaying via $\alpha$, $\beta^\pm$ emission or electron capture, leaving an excited daughter nucleus that decays via internal conversion or emission of $\gamma$-rays. These $\gamma$-rays are characteristic to the nuclide that emitted them. They are mono-energetic with well known emission probabilities, and so may be used to calculate the concentrations of isotopes within the sample.

The activation analysis carried out in this study was reversed, whereby a known quantity of isotope was used to study the particle fluence in the sample locations. The overlying principle however remains the same. Activation samples were placed in both the Gamma-3 and Quinta assemblies. Thorium samples placed in the Gamma-3 assembly were used to assess the efficacy that a graphite moderated spallation neutron spectrum could breed fissile $^{233}$U via the $^{232}$Th(n,$\gamma$)

---

1 Measurement of incident beam deuterons was carried out by Dr Wolfram Westmeier of Philipps-Universität in Marburg, Germany and Professor Igor Zhuk of the Joint Institute for Power and Nuclear Research, Belarus and his team.
reaction. In the Quinta assembly, \((n,\alpha n)\) threshold reactions in bismuth and gold were used to experimentally probe the high energy region of the neutron spectrum and the \(^{197}\text{Au}(n,\gamma)\) reaction was used for the detection of slow neutrons.

For an isotope to be detectable with gamma spectrometry it must be radioactive with a half life long enough so that it does not all decay before measurements can take place, and its half life must also be short enough so that the activity builds up to sufficient levels during the irradiation period. The \(\gamma\)-rays emitted should also be of easily detectable energies and of sufficient intensity. Ideally, all reaction cross sections and channels would be well known.

### 3.4.1 Positioning of activation samples

#### 3.4.1.1 Thorium samples in Gamma-3 assembly

In total, five thorium foils, sandwiched between two pieces of muscovite mica (for fission rate measurement) were placed inside the Gamma-3 assembly. To increase the mass of the samples, several layers of the \(\sim 50 \mu\text{m}\) thick foils were folded over into an approximate \(1\text{ cm}^2\) square. The mass of the samples varied between 0.12 g and 0.22 g. The samples were then placed in aluminium tubes (diameter 27 mm and thickness 1.5 mm) and inserted into holes within the Gamma-3 assembly, arbitrarily named A and B, located 15.8 and 22.9 cm from the central target axis (see Figure 3.8). Rod A contained three samples, positioned 8.3, 25.5 and 47 cm from the front face of the graphite, while rod B contained two samples, located 27.1 and 48.4 cm from the front face. The gaps between the samples in the aluminium tubes were filled in with cylindrical pieces of graphite.

#### 3.4.1.2 Bismuth and gold samples in Quinta assembly

Five bismuth foils of thickness 250 \(\mu\text{m}\) and three gold foils of thickness 50 \(\mu\text{m}\) were placed inside the Quinta target for each of the 1 GeV and 4 GeV irradiations. Both foils were from Goodfellow with purities >99% and had approximate areas of 1 \(\text{cm}^2\). Each of these foils were sandwiched between two pieces of 100 \(\mu\text{m}\) thick muscovite mica which were used as SSNTD’s for the fission rate measurements, as described in section 3.5.

The samples were mounted onto the back of 2 mm thick aluminium plates and inserted into gaps labelled 1, 2, 3, 4 and 5 in the Quinta target (see Figure 3.9). The gold samples were placed...
only in gaps 1, 2 and 3. For the 1 GeV irradiation, the samples were positioned at \( y = -4 \text{ cm} \) from the target axis. However, for the 4 GeV irradiation, to prevent overexposing the mica with fission tracks (independent of activation measurements) due to potential overlap with the beam shadow, as experienced in the 1 GeV irradiation, the samples were lowered to \( y = -6 \text{ cm} \). The positioning of the samples on the plates is presented in Table 3.4.

**TABLE 3.4:** Location of \(^{209}\text{Bi}\) and \(^{197}\text{Au}\) activation samples placed in the Quinta assembly. All units are in cm relative to the origin, positioned in front and centre of the beam window on the Quinta target-blanket assembly (indicated on Figure 3.9).

<table>
<thead>
<tr>
<th>Sample</th>
<th>1 GeV</th>
<th>4 GeV</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(x)</td>
<td>(y)</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>-4</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>-4</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>-4</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>-4</td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>-4</td>
</tr>
</tbody>
</table>
3.4. Neutron Activation Analysis

3.4.2 Calculating experimental reaction rates

After irradiation, the samples were removed from the assemblies and transported to the detector labs to be analysed with gamma spectrometry. The 16 bismuth and gold samples placed in the Quinta assembly were measured using a HPGe n-type coaxial (model: GR-1819) detector manufactured by CANBERRA Industries with relative efficiency 18\% (at 1.33 MeV), as determined by the manufacturer. Meanwhile, the thorium samples placed in Gamma-3, were measured using an Ortec detector. The Ortec detector was also an HPGe n-type coaxial detector (model: GMX-23200) and had a relative efficiency of 27\%. Measurements began ∼1.5 h after the beam had stopped, continuing for up to 6 days afterwards. The spectra collection times ranged from 15 minutes to just over 3 hours. All spectra were collected using Ortec Maestro PC software and later analysed with the HYPERMET-PC program [151].

3.4.2.1 \(^{209}\text{Bi}(\text{n,}\text{xn}),\) \(^{197}\text{Au}(\text{n,}\text{xn})\) and \(^{197}\text{Au}(\text{n,}\gamma)\) reactions

The isotopes detected in the bismuth and gold samples are listed in Table 3.5. All of the isotopes listed in Table 3.5 were detected in at least one of the measured samples with the exception of \(^{195}\text{Au}.\) Due to its long half life (186 days), the activity of \(^{195}\text{Au}\) was too low to be measurable.

The \(^{209}\text{Bi}(\text{n,}\text{xn}),\) \(^{197}\text{Au}(\text{n,}\text{xn})\) and \(^{197}\text{Au}(\text{n,}\gamma)\) reactions may be represented in the following way:

\[ (1) \quad \text{Target} \quad \begin{array}{c} \text{197Au/209Bi} \rightarrow \text{Product} \\ \left(\text{n,}\gamma\right)/\left(\text{n,}\text{xn}\right) \end{array} \quad \begin{array}{c} \lambda_1 \rightarrow \text{Daughter} \end{array} \]
### TABLE 3.5: $^{209}$Bi and $^{197}$Au neutron-induced reactions, threshold energies (MeV) and half lives of product nuclei.

$E(\sigma_{\text{max}})$ refers to the neutron energy (MeV) at which the reaction cross section is maximum.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Product</th>
<th>$E_{\text{th}}$</th>
<th>$E(\sigma_{\text{max}})$</th>
<th>Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{209}$Bi(n,4n)</td>
<td>$^{206}$Bi</td>
<td>22.6</td>
<td>34</td>
<td>6.24 d</td>
</tr>
<tr>
<td>$^{209}$Bi(n,5n)</td>
<td>$^{205}$Bi</td>
<td>29.6</td>
<td>44</td>
<td>15.3 d</td>
</tr>
<tr>
<td>$^{209}$Bi(n,6n)</td>
<td>$^{204}$Bi</td>
<td>38.1</td>
<td>55</td>
<td>11.2 h</td>
</tr>
<tr>
<td>$^{209}$Bi(n,7n)</td>
<td>$^{203}$Bi</td>
<td>45.4</td>
<td>66</td>
<td>11.8 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,$\gamma$)</td>
<td>$^{198}$Au</td>
<td>0</td>
<td>2.69 d</td>
<td></td>
</tr>
<tr>
<td>$^{197}$Au(n,2n)</td>
<td>$^{196}$Au</td>
<td>8.11</td>
<td>14</td>
<td>6.17 d</td>
</tr>
<tr>
<td>$^{197}$Au(n,3n)</td>
<td>$^{196m2}$Au</td>
<td>8.71</td>
<td>15</td>
<td>9.6 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,4n)</td>
<td>$^{195}$Au</td>
<td>14.8</td>
<td>24</td>
<td>186 d</td>
</tr>
<tr>
<td>$^{197}$Au(n,5n)</td>
<td>$^{194}$Au</td>
<td>23.2</td>
<td>34</td>
<td>38.0 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,6n)</td>
<td>$^{193}$Au</td>
<td>30.2</td>
<td>44</td>
<td>17.7 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,7n)</td>
<td>$^{192}$Au</td>
<td>38.9</td>
<td>54</td>
<td>4.94 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,7n)</td>
<td>$^{191}$Au</td>
<td>46.0</td>
<td>65</td>
<td>3.18 h</td>
</tr>
<tr>
<td>$^{232}$Th(n,$\gamma$)</td>
<td>$^{233}$Th→$^{233}$Pa</td>
<td>0</td>
<td>27.0 d</td>
<td></td>
</tr>
</tbody>
</table>

The target atoms ($^{209}$Bi or $^{197}$Au) undergo reactions ((n,xn) or (n,$\gamma$)) of rate, $R_1$ transmuting to the product radionuclide. The product nuclide with decay constant $\lambda_2$ decays to an excited state of the daughter nuclide which then de-excites via the emission of $\gamma$-rays. Most of the reactions listed in Table 3.5, with the exception of $^{209}$Bi(n,4n) and $^{209}$Bi(n,5n) contain products with one or more metastable states. Apart from $^{196m2}$Au, these metastable states have half lives less than few seconds and for purposes of activity measurements, may be disregarded [152]. $^{196m2}$Au however, has a half life of 9.6 hours and is most likely still present when spectrometry measurements take place. This must be corrected for when determining the total $^{197}$Au(n,2n) reaction rate. The procedure for doing so is listed separately in section 3.4.2.4.

The experimental production rate, $R_i^\text{exp}$ of each of the measured isotopes (per target atom per incident deuteron) was determined using the following equation:

$$R_i^\text{exp} = \frac{NC}{I_\gamma \varepsilon_p T BSD \Theta m N_A V} \frac{1}{\Phi} \frac{M_m t_{irr}}{t_{irr}}$$

(3.1)

Where

- $N$: Count rate of peak of interest (counts per second)
- $C$: Correction for decay during counting time (Eq. 3.2)
- $I_\gamma$: Emission probability of measured $\gamma$-ray
- $\varepsilon_p$: Detector full-energy peak efficiency (details in section 3.4.3)
- $T$: Correction due to True Coincidence Summing (TCS) (details in section 3.4.5)
- $S$: Activity saturation factor (Eq. 3.3)
3.4. Neutron Activation Analysis

$D$: Correction for decay between end of irradiation and beginning of counting (Eq. 3.4)

$B$: Correction due to pulsed beam (Section 3.4.2.2)

$M_m$: Molar mass of activation foil

$\Theta$: Isotopic fraction of activation foil

$m$: Mass of activation foil (in grams)

$N_{AV}$: Avogadro’s number

$t_{irr}$: Deuteron irradiation time

$\Phi$: Total number of incident deuterons

\[ C = \frac{t_r \lambda_2}{1 - e^{-t_r \lambda_2}} \]  
\[ S = 1 - e^{-\lambda_2 t_{irr}} \]  
\[ D = e^{-\lambda_2 t_d} \]  

Where, $t_r$ is the real-time of measurement interval and $t_d$ is the time between end of irradiation and beginning of counting [152].

This method does not take into consideration the depletion of the target material, nor possible activation of the produced nuclide i.e. burn-up is negligible. The depletion-activation model introduced by Abdel-Rahman and Podgorsak [153] can be used to correctly account for this. However, due to the low specific activities produced and the activation factor, $m = \sigma \phi / \lambda \ll 10^{-3}$ the saturation model used here is expected to be valid.

3.4.2.2 Pulsed beam correction factor

The delivery of the deuteron beam from the Nuclotron accelerator occurs in sporadic pulses, approximately every 8 seconds. The activity induced in the samples is affected as it is produced non-uniformly in discrete bunches, rather than smoothly and continuously over the entire irradiation period. The pulsed beam correction factor, $B$ compares the activity in the sample occurring at the end of a pulsed irradiation, $A_{\text{pulse}}$ to the activity occurring at the end of a continuous irradiation, $A_{\text{cont}}$.

Detailed derivations of $B$ have been provided previously [129, 131, 154] and so will not be repeated here. However, for completeness a short explanation of the underlying concepts will be provided. Each pulse, $i$ of the irradiation is treated as a very short continuous irradiation. The activity produced in the sample from the $i^{th}$ pulse, then decays over the time from the end of the pulse to the end of the irradiation, $t_{di}$. It is assumed that the length of each pulse is short enough that no decay of the product occurs during this time. Pulse lengths from the Nuclotron were on the order of milliseconds and were interrupted by gaps of 8 seconds or more. Small variations in the length of the pulse does not make more than a negligible difference. The $B$ values may be calculated using
where, $W_i$ is the ratio of number of deuterons in the pulse to the total number of deuterons in the entire irradiation period and $t_{di}$ is the time from given deuteron pulse to end of irradiation period. The $B$ values are dependent on the half lives of the product isotopes and are unique to each irradiation period. Eq. 3.6 was solved with a short script written in Python utilising data on beam pulse timing and beam fluence provided by the accelerator operators (Figures 3.6 and 3.7).

The $B$ values calculated for the 1 and 4 GeV irradiation periods are shown in Table 3.6. It can be seen that the $B$ values for the 1 GeV irradiation are very close to unity. This implies that the deuterons were distributed approximately evenly throughout the entire irradiation period. From Figure 3.7a, we can see that most pulses delivered contained on the order of $10^{10}$ deuterons. The reason for being a little above 1 means that there were slightly more deuterons extracted in the second half of the irradiation compared to the first half. The deviation of $B$ values from unity for the 4 GeV irradiation periods are more significant because the beam instability was substantial (Figure 3.7b). The beam continued to decrease in intensity for the first 11 hours, before only beginning to pickup again at the 15 hour point. Therefore, significantly more deuterons were delivered in the first half of the irradiation compared to the second half. This uneven beam extraction particularly affects the short lived isotopes e.g. $^{191}$Au (3.18 h), because the majority of product produced in the first half has already decayed by the end of the irradiation period.

### 3.4.2.3 $^{232}$Th($n,\gamma$) reaction

The neutron capture rate of $^{232}$Th was deduced by the activity of $^{233}$Pa. Production of $^{233}$Pa from the activation of $^{232}$Th is not a direct process but occurs via the relatively short lived (22.3 min) intermediary $^{233}$Th. The production and decay of $^{233}$Th must be taken into account when determining the $^{232}$Th($n,\gamma$) rate via the detection of $^{233}$Pa [129]. The reaction processes may be represented in the form:

$$\begin{align*}
(1) \quad &^{232}\text{Th} \quad R_{1(n,\gamma)} \quad (2) \quad ^{233}\text{Th} \quad \lambda_2 \quad \beta^{-22.3m} \quad (3) \quad ^{233}\text{Pa} \quad \lambda_3 \quad \beta^{-26.97d} \quad (4) \quad ^{233}\text{U}^*
\end{align*}$$

Where, $\lambda_2$ and $\lambda_3$ are the decay constants of $^{233}$Th and $^{233}$Pa, respectively. The neutron capture rate, $R_{1,exp}$ (per deuteron per target atom) of the $^{232}$Th was determined using

$$R_{1}^{exp} = \frac{N_3 C_3}{T_{\gamma p} T} \left( S_2 D_2 B_2 + S_3 D_3 B_3 \right) \frac{M_m}{\Theta m N_{AV}} \frac{t_{irr}}{\Phi}$$

(3.7)
3.4. Neutron Activation Analysis

**TABLE 3.6:** The measured reaction products of \(^{209}\text{Bi}\), \(^{197}\text{Au}\) and \(^{232}\text{Th}\) and their half-lives. The energies of detected \(\gamma\)-rays \((E_\gamma)\), their emission probabilities \((I_\gamma)\), full-energy peak efficiencies for sample geometry \((\varepsilon_p)\), True Coincident Summing \((T)\) and beam pulse correction factors \((B)\) are also shown. \(^{209}\)Bi and \(^{197}\)Au were measured with Canberra detector and \(^{232}\)Th with Ortec detector. \(\varepsilon_p\) and \(T\) are for sample located 1.0 and 1.4 cm from the detector face for the Canberra and Ortec detector, respectively.

<table>
<thead>
<tr>
<th>Product</th>
<th>Half-life</th>
<th>(E_\gamma) (keV)</th>
<th>(I_\gamma) (%)</th>
<th>(\varepsilon_p) (%)</th>
<th>(T)</th>
<th>(B_{1\text{GeV}})</th>
<th>(B_{4\text{GeV}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{209}\text{Bi})</td>
<td>6.24 d</td>
<td>803.10</td>
<td>99</td>
<td>1.58</td>
<td>0.729</td>
<td>1.001</td>
<td>0.9851</td>
</tr>
<tr>
<td></td>
<td></td>
<td>881.01</td>
<td>66.2</td>
<td>1.46</td>
<td>0.762</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{205}\text{Bi})</td>
<td>15.3 d</td>
<td>703.44</td>
<td>32.5</td>
<td>1.72</td>
<td>0.928</td>
<td>1.000</td>
<td>0.9939</td>
</tr>
<tr>
<td>(^{204}\text{Bi})</td>
<td>11.2 h</td>
<td>374.76</td>
<td>82</td>
<td>3.12</td>
<td>0.760</td>
<td>1.012</td>
<td>0.8183</td>
</tr>
<tr>
<td></td>
<td></td>
<td>899.15</td>
<td>98</td>
<td>1.43</td>
<td>0.752</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{203}\text{Bi})</td>
<td>11.8 h</td>
<td>820.3</td>
<td>30</td>
<td>1.55</td>
<td>0.976</td>
<td>1.011</td>
<td>0.8257</td>
</tr>
<tr>
<td></td>
<td></td>
<td>825.2</td>
<td>14.6</td>
<td>1.55</td>
<td>0.999</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{198}\text{Au})</td>
<td>2.69 d</td>
<td>411.8</td>
<td>96</td>
<td>3.04</td>
<td>0.999</td>
<td>1.002</td>
<td>0.9657</td>
</tr>
<tr>
<td>(^{196\text{Au}})</td>
<td>6.17 d</td>
<td>332.98</td>
<td>22.9</td>
<td>3.83</td>
<td>0.837</td>
<td>1.001</td>
<td>0.9849</td>
</tr>
<tr>
<td></td>
<td></td>
<td>355.68</td>
<td>87</td>
<td>3.55</td>
<td>0.898</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{196\text{m2Au}})</td>
<td>9.6 h</td>
<td>147.81</td>
<td>43</td>
<td>8.07</td>
<td>0.717</td>
<td>1.014</td>
<td>0.7920</td>
</tr>
<tr>
<td></td>
<td></td>
<td>188.27</td>
<td>37.4</td>
<td>6.75</td>
<td>0.734</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{194}\text{Au})</td>
<td>38.0 h</td>
<td>328.46</td>
<td>61</td>
<td>3.88</td>
<td>0.861</td>
<td>1.003</td>
<td>0.9423</td>
</tr>
<tr>
<td>(^{193}\text{Au})</td>
<td>17.7 h</td>
<td>186.17</td>
<td>9.4</td>
<td>6.82</td>
<td>1.000</td>
<td>1.007</td>
<td>0.8797</td>
</tr>
<tr>
<td></td>
<td></td>
<td>255.57</td>
<td>6.2</td>
<td>5.08</td>
<td>1.000</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{192}\text{Au})</td>
<td>4.94 h</td>
<td>316.5</td>
<td>58</td>
<td>4.05</td>
<td>0.829</td>
<td>1.031</td>
<td>0.6560</td>
</tr>
<tr>
<td>(^{191}\text{Au})</td>
<td>3.18 h</td>
<td>586.45</td>
<td>17</td>
<td>2.15</td>
<td>1.000</td>
<td>1.052</td>
<td>0.5725</td>
</tr>
<tr>
<td>(^{233}\text{Pa})</td>
<td>27.0 d</td>
<td>311.9</td>
<td>38.5</td>
<td>4.16</td>
<td>0.995</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Where

- \(N_3\): Count rate of \(^{233}\text{Pa}\) 311.9 keV peak
- \(C_3\): Correction for decay of \(^{233}\text{Pa}\) during counting time (Eq. 3.2)
- \(S_2\): Saturation factor of \(^{233}\text{Th}\) (Eq. 3.8)
- \(S_3\): Saturation factor of \(^{233}\text{Pa}\) (Eq. 3.9)
- \(D_2\): Correction for decay of \(^{233}\text{Th}\) between end of irradiation and beginning of counting (Eq. 3.10)
- \(D_3\): Correction for decay of \(^{233}\text{Pa}\) between end of irradiation and beginning of counting (Eq. 3.11)
- \(B_2\): Correction due to pulsed beam for \(^{233}\text{Th}\) activity (Eq. 3.12)
- \(B_3\): Correction due to pulsed beam for \(^{233}\text{Pa}\) activity (Eq. 3.13)

The activity saturation factors for both \(^{233}\text{Th}\) and \(^{233}\text{Pa}\), \(S_2\) and \(S_3\) respectively, may be found by implementing the Leibniz solution of the generalised Bateman equations, as described
in De Soete et al. [152]. $S_3$ describes the growth of $^{233}\text{Pa}$ over the irradiation period.

$$S_2 = 1 - e^{-\lambda_2 t_{irr}}$$  
(3.8)

$$S_3 = \frac{\lambda_3}{\lambda_3 - \lambda_2} (1 - e^{-\lambda_2 t_{irr}}) - \frac{\lambda_2}{\lambda_3 - \lambda_2} (1 - e^{-\lambda_3 t_{irr}})$$  
(3.9)

The decay factors are used to account for the decay between the end of the irradiation period and the beginning of measurement. $D_3$ describes the decay of $^{233}\text{Pa}$ and $D_2$ describes the decay of residual $^{233}\text{Th}$, subsequent build-up of $^{233}\text{Pa}$ and then the continued decay over delay time ($t_d$). The decay factors may also be derived from the Leibniz solution of the Bateman equations [152].

$$D_2 = \frac{\lambda_3}{\lambda_3 - \lambda_2} (e^{-\lambda_2 t_d} - e^{-\lambda_3 t_d})$$  
(3.10)

$$D_3 = e^{-\lambda_3 t_d}$$  
(3.11)

The beam correction factors, $B_2$ and $B_3$, (Equations 3.12 and 3.13) were derived using the same principles described in section 3.4.2.1 [129]. The $B$ values calculated for the Gamma-3 irradiation (Figure 3.6) were 1.60 and 0.98, respectively.

$$B_2 = \frac{t_{irr} \lambda_2}{1 - e^{-\lambda_2 t_{irr}}} \sum_{i=1}^{n} W_i e^{-\lambda_2 t_{di}}$$  
(3.12)

$$B_3 = \frac{t_{irr} \lambda_2 \lambda_3}{\lambda_3 (1 - e^{-\lambda_2 t_{irr}}) - \lambda_2 (1 - e^{-\lambda_3 t_{irr}})} \sum_{i=1}^{n} W_i (e^{-\lambda_2 t_{di}} - e^{-\lambda_3 t_{di}})$$  
(3.13)

In the case that $^{233}\text{Th}$ is irradiated to saturation ($1 - e^{-\lambda_2 t_{irr}} \rightarrow 1$ and the delay time is long enough for $^{233}\text{Th}$ to completely decay to $^{233}\text{Pa}$, Eq. 3.7 may be simplified to Eq. 3.1 [152]. In our experiments, the difference between Eq. 3.7 and 3.1 was 2-3 percent.

### 3.4.2.4 $^{197}\text{Au}(\text{n},2\text{n})^{196m2,\text{g}+\text{m}1}\text{Au}$ reaction

The $^{197}\text{Au}(\text{n},2\text{n})$ reaction has the added complication of the activation branching to three possible isomers, $^{196g}\text{Au}$, $^{196m1}\text{Au}$ and $^{196m2}\text{Au}$. The $^{196m1}\text{Au}$ isomer decays quickly (8.1 s half life), and so measurements of $^{196g}\text{Au}$ include the sum of the ground state and the first isomeric state. Most activation reactions produce the $^{196g+1}\text{Au}$ state [155], however some proceed via the $^{196m2}\text{Au}$ channel. $^{196m2}\text{Au}$ has a 9.6 hour half, decaying to $^{196m1}\text{Au}$ and then to $^{196g}\text{Au}$. Determination of the total $^{196}\text{Au}$ production rate, requires the sum of the $^{196m2}\text{Au}$ and $^{196g+1}\text{Au}$ states. The reaction process may be visualised as shown in Figure 3.10.

If a transformation chain branches and the two chains rejoin together, they are treated as separate chains [152]. Therefore, the total production rate of $^{196}\text{Au}$, $R_1$ is calculated from the
3.4. Neutron Activation Analysis

3.4.1 Reaction Channels and Isomeric Ratios

**Figure 3.10:** $^{197}$Au(n,2n) reaction channels. Most reactions proceed directly to $^{196g}$Au, however some proceed indirectly via $^{196m2}$Au which has a 9.6 hour half life.

The yields of $^{196g}$Pt and $^{196g}$Hg were determined from the $147.81$ and $188.27$ keV lines and Eq. 3.1. It was found from the early spectra acquired 1.4-3 hours after irradiation that the average isomeric ratio ($R_1$) was $0.09 \pm 0.04$ and $0.12 \pm 0.04$ for the 1 and 4 GeV irradiations, respectively. According to TENDL-2012 data [156], $\sigma_{m2} / \sigma_{m1+g}$ fluctuates between 0.06 and 0.08 from 8.5 to 30 MeV neutrons. Data above 30 MeV is not available.

In the situation that the metastable isomer is shorter lived than the ground state, foils irradiated to saturation and sufficient time has passed for it to completely decay into the ground state, Eq. 3.1 may be used [152]. This requirement was satisfied for all the other reactions that produced isomeric forms. In the case of $^{197}$Au(n,2n), the difference between using the simplified solution of Eq. 3.1 and the full solution solving the transformation chains individually was about 5%.

**3.4.3 Determination of detector efficiency**

Detector efficiencies are strongly dependent on source-detector geometry. The full-energy peak efficiency, $\varepsilon_p$, is the ratio of counts in the peak area to the number of $\gamma$-rays emitted by the source at the corresponding peak energy.
CHAPTER 3. EXPERIMENTAL METHODOLOGY

\[ \varepsilon_p(E) = \frac{N_p(E)}{AI \gamma(E)} \]  

(3.14)

Where \( N_p(E) \) is the peak count rate, \( A \) is the source activity and \( I_\gamma(E) \) is the emission probability of the measured \( \gamma \)-ray. The total efficiency, \( \varepsilon_T \) is the ratio of counts recorded anywhere in the spectrum to the number of photons emitted by the source of energy \( E \). Total efficiency takes into account the full-energy peak, all incomplete absorption from Compton scattering and any secondary photons scattered into the detector from surroundings. Knowledge of detector total efficiency is required for True Coincidence Summing corrections.

\[ \varepsilon_T(E) = \frac{N_T}{AI \gamma(E)} \]  

(3.15)

Where \( N_T \) is the total count rate across the entire spectrum. It is necessary to discount the low energy components of the spectrum originating from X-rays emitted from electron capture or internal conversion \[157\]. The full-energy peak and total efficiencies of both HPGe detectors were determined using both experimental and Monte Carlo methods.

3.4.3.1 Canberra GR-1819 detector

Eleven point sources of verified activity, including \(^{241}\)Am, \(^{133}\)Ba, \(^{139}\)Ce, \(^{57}\)Co, \(^{60}\)Co, \(^{137}\)Cs, \(^{152}\)Eu, \(^{54}\)Mn, \(^{113}\)Sn, \(^{228}\)Th and \(^{88}\)Y were measured with the Canberra detector to find the full-energy peak and total efficiencies. These sources were also used for energy calibration, characterising the non-linearity of the detection system, and the FWHM-energy relationship. Four of these sources (\(^{241}\)Am, \(^{139}\)Ce, \(^{137}\)Cs and \(^{54}\)Mn) are single line (or approximately) photon emitting and hence appropriate for total efficiency calibration. \(^{60}\)Co emits two photons close in energy (\( E_1 = 1173, E_2 = 1332 \text{keV} \)) and so total efficiency may be determined for the mean of these energies (\( \bar{E} = 1253 \text{keV} \)). In the case of \(^{88}\)Y, two photons are emitted that are far apart in energy (\( E_1 = 898, E_2 = 1836 \text{keV} \)). The 898 keV total efficiency was found by interpolating existing measured values which could then be used to calculate the total efficiency at 1836 keV as described in Eq. 3.16 \[157\].

\[ \varepsilon_T(E_2) = \frac{N_{T,2} - A_1 I_{\gamma,1} \varepsilon_T(E_1)}{A_2 I_{\gamma,2}} \]  

(3.16)

To verify and interpolate the experimentally measured full-energy peak and total efficiency, Monte Carlo simulations of the detector were performed \[158–161\]. A survey of the literature provided in Helmer et al. \[160\] has noted that disagreements between Monte Carlo calculated and measured detector efficiencies are generally 5–10\%. The two ways to correct for this can involve either simply scaling the Monte Carlo results to minimise the difference between the measured and scaled result, or to adjust the physical parameters of the detector in the Monte Carlo calculations until better agreement is achieved \[160\]. In this work, the latter approach was
chosen. An example of an MCNPX input file used to determine the efficiency of the Canberra GR-1819 is provided in Appendix C on page 225.

X-ray images of detectors have shown that the technical specifications provided by detector manufacturers commonly deviate from the true detector geometry [160]. Problems have included the positioning of the detector within the housing, whether the detector is parallel to the housing axis, size and shape of sensitive volume, and inconsistencies in the thickness of dead layers [159–163].

Quality assurance data sheets and technical drawings provided by the manufacturer were used as a basis to model the Canberra detector geometry in the simulation. Without X-ray images of the detector, the optimal physical parameters were determined through trial and error. It was found that changing the crystal to end cap distance from 5 mm to 8 mm led to the most favourable outcome. Changes of 4 mm to 9 mm have been reported previously [162] and so this adjustment was not considered unreasonable. Additionally, the front face of modern detectors have rounded edges (bulletisation) in order to remove weak field regions [164]. This was not shown on technical diagram provided by the manufacturer and so was added to achieve even better agreement. It should be noted that it would be a major coincidence if this adjustment represented the true physical nature of the detector. However, with the multitude of variables and lack of X-ray images available, it was considered the most straightforward approach. It is possible that imperfections in the crystal and non-uniformities in the electric field have led to non-uniform charge collection. This effect is not accounted for in the Monte Carlo simulation.

A diagram of the Canberra GR-1819 detector is shown in Figure 3.11. The manufacturers specifications are in black, while the changes made here are indicated in red. Not shown in Figure 3.11, but important for the simulated total efficiency is to include all materials surrounding the detector, such as lead shielding. The experimental and calculated total and full-energy peak efficiency of the GR-1819 CANBERRA is shown in Figure 3.12. Measurements were performed at three distances (1.0, 3.5 and 13.2 cm) from the detector cap. All samples irradiated in the Quinta assembly had relatively low activities, therefore to ensure adequate counting statistics, they were all measured at the 1.0 cm position. Good agreement between the experimentally measured and simulated efficiencies can be seen.

3.4.3.2 Ortec GMX-23200 detector

Determination of the Ortec detector efficiency was done in the same way as it was for the Canberra detector. However, only two experimental sources – $^{241}$Am and $^{228}$Th were measured. Figure 3.13 shows the Ortec detector manufacturers dimensions (black) and adjusted dimensions (red) adopted to achieve better agreement with the experimentally measured data. Changes included increasing the crystal to end-cap distance from 3 to 4 mm and decreasing the crystal diameter from 53.6 to 52.1 mm.

The experimental and Monte Carlo full-energy peak and total efficiencies of the Ortec detector for the three measured distances (1.4, 5.5 and 13.7 cm) may be seen in Figure 3.14. $^{228}$Th is not
FIGURE 3.11: Cross section view of the Canberra GR-1819 detector. The manufacturer’s specifications are shown in black while the modifications made here are outlined in red. Changes included shifting the detector 3 mm further away from the end-cap and removal of the weak field regions in the sensitive detector volume.

FIGURE 3.12: Experimental and Monte Carlo simulated full-energy peak ($\varepsilon_p$) and total ($\varepsilon_T$) efficiency of the GR-1819 Canberra detector for three measured distances from the detector face.
3.4. Neutron Activation Analysis

3.4.1.1 Calculations of the Ortec GMX-23200 Detector Efficiency

**FIGURE 3.13:** Cross section view of the Ortec GMX-23200 detector. The manufacturers specifications are shown in black while the modifications made here are outlined in red. Changes included increasing the crystal to end-cap distance from 3 to 4 mm and decreasing the crystal diameter from 53.6 to 52.1 mm.

**FIGURE 3.14:** Experimental and Monte Carlo simulated full-energy peak ($\varepsilon_p$) and simulated total ($\varepsilon_T$) efficiency of the Ortec GMX-23200 detector for three measured distances from the detector face.

A single line gamma emitter and the energy of the major $^{241}$Am gamma line (60 keV) is too low to be useful for characterising the total efficiency of the detector. Therefore, no experimental points can verify the simulated total efficiency. However, there was good agreement between the experimental and simulated total efficiency for the Canberra detector, and as the lead castle surrounding the Ortec detector was identical to that surrounding the Canberra detector, this leads to reasonable confidence in the results. However the lack of experimental verification may contribute to an increased uncertainty in the calculation of TCS correction factors.

3.4.3.3 Efficiency corrections for sample geometry

The calibration sources used for the detector efficiency determination were all effective point sources which does not reflect the geometry of the samples placed in the Quinta and Gamma-3 assemblies. These samples were square foils (1 cm$^2$) with thickness 250 µm, 50 µm, ~150 µm for
the bismuth, gold and thorium foils respectively, and are therefore volume sources. This leads to a decrease in the solid angle subtended at the detector and a reduction in detection efficiency. In addition, bismuth, gold and thorium are all dense, high Z materials which cause significant photon absorption and scattering within the samples.

Self absorption within the sample can be easily corrected for mathematically using the following equation

\[ R_0 = \frac{R \mu t}{(1 - e^{-\mu t})} \]  

(3.17)

Where, \( R_0 \) is the true spectrum peak count rate, \( R \) is the measured spectrum peak count rate, \( t \) is the thickness of the sample and \( \mu \) is the linear attenuation coefficient of the sample material at the peak energy. Correction factors for volume sources, however cannot be calculated using simple equations [164]. Due to the success of using Monte Carlo simulations for efficiency calibration of the point sources, further Monte Carlo simulations with the specific geometries of the foils were performed to correct for variations in the individual sample geometry and self absorption of photons within these samples.

3.4.4 Random Summing (Pile-up)

Random summing (pulse pile-up) occurs when two (or more) \( \gamma \)-rays emitted in quick succession are detected within the resolving time of the detector. The detector is unable to distinguish these as separate events, the output pulse becomes misshapen and only a single event is recorded. It can be particularly problematic in samples with high count rates and close detector geometries.

Random summing may ordinarily be corrected for using pile-up rejection electronics. However as none were utilised here the pile-up losses were corrected for mathematically using

\[ A_T = A e^{2R\tau} \]  

(3.18)

where, \( A_T \) is the true peak area, \( A \) is the measured peak area, \( R \) is the count rate across the entire spectrum and \( \tau \) is the resolution time [164]. Because the summing is random, this equation may be applied to all peaks in spectrum. Without pile-up rejection electronics, \( \tau \) is best approximated as the shaping time constant of the amplifier. This was provided by the detector manufacturer and reported on quality assurance data sheets to be 4\( \mu \)s and 6\( \mu \)s for the Canberra and Ortec detectors, respectively.

The samples irradiated in Quinta and Gamma-3 had low activities and even at close detector geometries, random summing was negligible. However, the calibration sources often had high activities and thus pile-up corrections were necessary, especially at close detector geometries.

3.4.5 True Coincident Summing

True Coincident Summing (TCS) occurs when two or more photons emitted from the same cascade of a single decay process are detected simultaneously. This happens because the resolving
times of HPGe detectors are typically microseconds while the lifetimes of excited nuclear states are nanoseconds or less. TCS is independent of source activity and is a separate issue from random summing (discussed above). It is entirely dependent on the absolute detector efficiencies and decay scheme of the radionuclide being measured. TCS can lead to either summing out (peak loss), summing in (sum peak) or contributions to the continuous part of the spectrum [165].

TCS corrections must be made for absolute activity determination and detector efficiency calibrations [166] and can be particularly problematic when samples have low count rates and so are placed close to the detector. The bismuth and gold samples irradiated in Quinta and the thorium samples in Gamma-3 all had relatively low activities and so to ensure adequate counting statistics, these were all measured at the closest position to the detector. However, placing the samples this close to the detector means that TCS corrections must be made or the results will be in error. Likewise, the calibration sources used for determining the detector efficiency were also measured close to the detector where TCS cannot be avoided.

Summing out occurs when a $\gamma$-ray is detected at full energy coincidentally with another photon. In the absence of TCS, a count in the peak at the full-energy of the $\gamma$-ray would be recorded, however with TCS, the detection of the second photon means their energies will sum and a count will be recorded elsewhere in the spectrum. This causes the loss of a count to the $\gamma$-ray peak. Summing in occurs when two (or more) $\gamma$-rays are detected in coincidence at full energy. A count is recorded at the sum of their energies. If the nuclide emits a photon with the sum of detected photon energies then additional counts will be recorded in that peak. However, if the nuclide does not, then a pure sum (alias) peak will appear in the spectrum. Figure 3.16 illustrates TCS using the simple decay scheme shown in Figure 3.15.

The provided example in Figure 3.15 is very simplified and unrealistic in the context of real world decay schemes. Decay schemes can contain large numbers of levels, including metastable states. Additionally, each transition in the decay scheme is a competition between $\gamma$-ray emission and internal conversion, which emits X-rays. If the detector can detect low energy photons (n-type), then $\gamma$-X coincidences need to be considered as well as $\gamma$-$\gamma$ coincidences. The decay

![Figure 3.15: Simplest decay scheme to show True Coincident Summing (TCS).](image-url)
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FIGURE 3.16: Diagram illustrating the effects of True Coincident Summing (TCS) of decay scheme shown in Figure 3.15. 

- **a)** No coincident summing.
- **b)** Summing-out of $\gamma_{21}$. Count loss from $\gamma_{21}$ peak due to $\gamma_{10}$ depositing some energy in detector.
- **c)** Summing in of $\gamma_{10}$ and $\gamma_{21}$. Additional count recorded in $\gamma_{20}$ peak, and loss of counts from $\gamma_{10}$ and $\gamma_{21}$ peaks.

The process of the parent nuclide may also produce photons – atomic X-rays are emitted in electron capture, and annihilation of $\beta^+$ particles creates two annihilation photons.

Unlike pile-up pulses, True Coincident Summed pulses are not misshapen and so cannot be filtered out with electronic circuitry [164]. The most reliable way to correct for it is mathematically. Since the early 1970’s, a lot of work has gone into developing algorithms for accurate corrections to TCS. The first record of quantitative corrections were a recursive method published by Andreev et al. [167]. These were later revised by McCallum and Coote [168] to include annihilation photons and rewritten due to Andreev et al. work not being widely recognised at the time. Throughout the 1970’s and 80’s several groups published correction factors for specific radionuclides. This included the work of Gehrke et al. [169] containing 13 experimentally determined correction factors for common calibration sources and Schima and Hoppes [170] presenting results for 17 common nuclei based purely on first order calculations. However, these formulae were inadequate for universal application to all isotopes. Additionally, first order corrections involving combinations of only two coincident $\gamma$-rays became increasingly unsatisfactory when dealing with more complicated decay schemes.

Semkow et al. [171] introduced a matrix formalism to carve the way for a full correction for arbitrarily complicated decay schemes. The work of Semkow et al. was extended a few years later by Korun and Martinčič [165] to incorporate X-\(\gamma\) coincidences. Only K X-rays were included, while other X-rays and Bremsstrahlung radiation was neglected. Most recently, Novković et al. [172] has built on the work of Korun and Martinčič to add metastable states and allowances of the parent nuclide to decay into two or more excited daughter states.

The method used to correct for TCS was a combinatorial approach presented by Sudár [173] and made available in the “TrueCoinc” program [174]. The program requires input of the total and full-energy peak efficiencies and decay scheme data in the ENSDF format [175]. The program calculates all TCS correction factors, including X-rays (K, L, M... lines) without the time consuming data preparations and calculations of earlier programs. The calibration sources were used to determine the detector efficiency, as required for TCS corrections for the calibration sources. TCS corrections to the calibration sources were thus determined recursively until satisfactory agreement was reached. The TCS correction factors for the gold, bismuth and thorium samples are shown in Table 3.6 (page 43).
3.5 Fission track detectors

The fission rate of materials is important for ADS design as it directly contributes to the neutron balance and power output of the assembly, as well as having consequences for heat deposition, induced radioactivity and fission gas release. Fission rate measurements may also be useful for neutron detection, as most heavy metals fission when bombarded with high energy neutrons.

Different energy regions of the neutron spectrum may be probed by measuring the fission rate of different isotopes. Fissile materials \(^{233}\text{U},^{235}\text{U},^{239}\text{Pu}\) fission readily from thermal neutrons, and actinide metals \(^{232}\text{Th},^{238}\text{U}\) fission with fast neutrons >1 MeV. The threshold energy for inducing fission in sub-actinides increases with decreasing mass number of target nuclide [176].

In this study, the fission rate of materials were measured using mica Solid State Nuclear Track Detectors (SSNTDs). The SSNTDs are small enough to fit in the small gaps in the assemblies, they are economical compared to the capital costs of spectroscopic techniques, no specialised equipment is required (only chemicals and microscope), samples are easily transportable, and they can be analysed at later date when convenient. Gamma spectrometry can also be used to calculate the fission rate of samples by measuring the yields of prominent fission products e.g. \(^{97}\text{Zr},^{131}\text{I},^{133}\text{I}\) and \(^{43}\text{Ce}\). However, this requires each sample to be measured several times, and restricts the number of samples that can be used due to limited access to detectors. Also, the yields of fission products vary depending on the shape of the neutron spectra and for fast neutrons, only data for mono-energetic 14 MeV neutrons is available [31]. This method is discussed in more detail in appendix B, where fission and spallation products were measured in \(^{232}\text{Th}\). The unique conditions of the Gamma-3 and Quinta assemblies also preclude the use of miniaturised fission chambers.

3.5.1 Theory

The fission track detector technique involves placing a foil of fissionable material in between two pieces of a suitable Solid State Nuclear Track Detector (SSNTD) (Figure 3.17). Fission fragments ejected from the outer surfaces of the foil produce damage trails in the adjacent SSNTDs. Upon etching the SSNTD in a corrosive solution, the etching tracks enlarge themselves to the point that they become clearly visible and can be counted using an optical microscope.

The track density of the SSNTD in close contact with the fission foil may be expressed as

\[
\rho = n\mu \varepsilon dN_e t \int_0^\infty \sigma_f(E)\phi(E) \, dE
\]  

(3.19)
where $\rho$ is the detectable track density on the surface of the SSNTD, $n$ is the number of fission fragments emitted per fission, $\mu$ is the proportion of fissionable nuclei in the foil where fission would lead to ejection of a fragment into the adjacent SSNTD, $\varepsilon$ is the track registration efficiency, $d$ is the foil thickness, $N_v$ is the number of fissionable nuclei per unit volume of the foil, $t$ is the foil irradiation time and $\sigma_f(E)$ and $\phi_f(E)$ are the energy-dependent fission cross section and energy-dependent particle flux [177, 178].

The factor $\mu$, depends on the mean range of the fission fragment, $R$ and the thickness of the fission foil, $d$ and is given by the following relationships

$$
\mu = \begin{cases} 
\frac{1}{2} \left(1 - \frac{d}{2R}\right), & \text{for } d < R \\
\frac{1}{4}, & \text{for } d = R \\
\frac{1}{4} \frac{R}{2}, & \text{for } d > R 
\end{cases}
$$

(3.20)

The track registration efficiency, $\varepsilon$ is defined as

$$
\varepsilon = \eta \cos^2 \theta_c
$$

(3.21)

where $\eta$ is the proportion of tracks able to be observed, and $\theta_c$ is the critical etching angle [178]. Not all tracks are able to be observed due to limitations imposed by the track size, track density (being too high can cause track overlap) and observation conditions (contrast, brightness of images, dirt on sample etc.) [179]. Etching of the SSNTD removes material along the damage trail at a faster rate than the bulk of the detector. Therefore, a track may only be exposed by etching if the dip angle of the entering fragment is greater than the critical dip angle, $\theta_c$ which is defined by:

$$
\sin \theta_c = \frac{V_B}{V_T}
$$

(3.22)

where $V_B$ is the bulk etch rate of the detector and $V_T$ is the track etch rate [180].

Hashemi-Nezhad et al. [178] has defined a calibration factor, $w$ such that

$$
w = n \mu \varepsilon d N_v
$$

(3.23)

and the number of fissions per fissionable atom occurring in the foil during the irradiation time, $t$ is

$$
N_f = t \int_0^{\infty} \sigma_f(E) \phi_f(E) \, dE
$$

(3.24)

By substituting Equations 3.23 and 3.24, into Eq. 3.19 we find

$$
\rho = w N_f
$$

(3.25)

and the density of tracks in the SSNTD can be expressed in terms of only the calibration factor, $w$ and number of fission events occurring in the fission foil per fissionable atom [181].
TABLE 3.7: Calibration factors, \( w \) (tracks cm\(^{-2}\) (fissions per atom\(^{-1}\)) for fission foils as calculated by Hashemi-Nezhad et al. [181].

<table>
<thead>
<tr>
<th>Fission foil</th>
<th>Eq. 3.26</th>
<th>Eq. 3.27</th>
<th>Monte Carlo</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{natU}^* )</td>
<td>((8.99 \pm 0.22) \times 10^{18})</td>
<td>((8.99 \pm 0.22) \times 10^{18})</td>
<td>((8.51 \pm 0.32) \times 10^{18})</td>
</tr>
<tr>
<td>( ^{232}\text{Th} )</td>
<td>((9.23 \pm 0.36) \times 10^{18})</td>
<td>((8.46 \pm 0.50) \times 10^{18})</td>
<td>((8.97 \pm 0.45) \times 10^{18})</td>
</tr>
<tr>
<td>( \text{natPb} )</td>
<td>((1.03 \pm 0.05) \times 10^{19})</td>
<td>((1.10 \pm 0.12) \times 10^{19})</td>
<td>((1.00 \pm 0.05) \times 10^{19})</td>
</tr>
<tr>
<td>( ^{197}\text{Au} )</td>
<td>((1.01 \pm 0.03) \times 10^{19})</td>
<td>–</td>
<td>((1.01 \pm 0.01) \times 10^{19})</td>
</tr>
</tbody>
</table>

* \( \text{natU} \) was used as the reference calibration.

The calibration factors for thick \((d > R)\) uranium foils at differing enrichment levels for mica (natural and artificial), Lavsan and soda glass SSNTDs has been experimentally measured previously [178]. These calibration factors were then successfully utilised to determine the natural uranium fission rate in a combined spallation and fission neutron field [135]. Further studies based on Monte Carlo simulation and theoretical assumptions have determined additional calibration factors for \( ^{235}\text{U} \), \( ^{232}\text{Th} \), \( \text{natPb} \) and \( ^{197}\text{Au} \) [181]. These measurements relied on the use of the experimental measurement in Hashemi-Nezhad et al. [178] to be used as a reference foil. Two theoretical methods were then applied to find calibration factors for the foils of interest.

The calibration factor of the foil of interest can be calculated if the mean ranges of fission fragment \((R)\) in both the reference foil (subscript \( r \)) and foil of interest (subscript \( j \)) are known using

\[
w_j = w_r \frac{N_{v,j}}{N_{v,r}} \frac{R_j}{R_r} \varepsilon_j \varepsilon_r
\]  

(3.26)

If both the reference foil and foil of interest are irradiated simultaneously with the same particle beam and fluence, then the calibration factor may be found from

\[
w_j = w_r \frac{\rho_j}{\rho_r} \frac{[\sigma(E)]_j}{[\sigma(E)]_r}
\]  

(3.27)

where \( \sigma(E) \) is the spectrum averaged fission cross section of the particle field used for the irradiation. The calibration factors used in this work were a weighted average of the Monte Carlo simulations and the two theoretical methods published in Hashemi-Nezhad et al. [181] and reproduced in Table 3.7.

It may be noted that the calibration factors for the sub-actinides (\( \text{natPb} \) and \( ^{197}\text{Au} \)) are closely clustered around \(1 \times 10^{19}\). Hashemi-Nezhad et al. [181] have concluded that nuclides with \( Z \geq 79 \) without a known calibration factor may adopt the value of \(1 \times 10^{19}\) without introducing an uncertainty of more than 10%. Borger [129] took this approach and used a calibration factor of \((1.0 \pm 0.1) \times 10^{19}\) in the case of \( ^{209}\text{Bi} \) fission. As it did not introduce any systematic error, this value will also be used as the calibration factor for \( ^{209}\text{Bi} \) here.

The calibration factors are valid in particle fields of unknown energy and angular distribution, if the average density of the tracks in the detectors on both sides of the foil is used [178].
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TABLE 3.8: The weighted average of calibration factors in Table 3.7. These were the values used in this work.

<table>
<thead>
<tr>
<th>Fission foil</th>
<th>Weighted average</th>
</tr>
</thead>
<tbody>
<tr>
<td>natU</td>
<td>((8.89 \pm 0.14) \times 10^{18})</td>
</tr>
<tr>
<td>(^{232}\text{Th})</td>
<td>((8.97 \pm 0.22) \times 10^{18})</td>
</tr>
<tr>
<td>(^{209}\text{Bi})</td>
<td>((1.0 \pm 0.1) \times 10^{19})*</td>
</tr>
<tr>
<td>natPb</td>
<td>((1.02 \pm 0.02) \times 10^{19})</td>
</tr>
<tr>
<td>(^{197}\text{Au})</td>
<td>((1.01 \pm 0.01) \times 10^{19})</td>
</tr>
</tbody>
</table>

* This was an approximation. See text for details.

is because the kinetic energy (and range) of the fission fragments is not affected (or only very weakly affected) by the excitation energy of the fissioning nucleus [178].

3.5.2 Placement of fission track detectors in Gamma-3 and Quinta

3.5.2.1 Gamma-3 assembly

The Gamma-3 assembly contained five thorium fission samples, which were placed in the same location as the activation samples (see Figure 3.8 on page 38).

3.5.2.2 Quinta assembly

Fission rate measurements of \(^{nat}\text{U}\), \(^{232}\text{Th}\), \(^{209}\text{Bi}\), \(^{nat}\text{Pb}\) and \(^{197}\text{Au}\) were carried out in the Quinta assembly under 1 and 4 GeV irradiations. The chosen locations for the samples placed inside the Quinta assembly was dependent on the following restrictions – (1) the samples could only be mounted onto special plates (Figure 3.18) which were to be inserted in fixed locations within the assembly, (2) these sample plates were shared with many members of the collaboration team who also mounted their own samples onto the plates, and (3) due to the narrow gaps between the Quinta sections, the samples mounted onto the plates needed to maintain a low profile. This limited the number of samples that could be placed in each location.

For the 1 GeV irradiation, axial distribution of the fission rate of \(^{232}\text{Th}\), \(^{209}\text{Bi}\), \(^{nat}\text{Pb}\) and \(^{197}\text{Au}\) samples was determined at \(y=-4\) cm and \(y=-12\) cm (below the target axis). Additional samples were placed on plate 2 at \(y=5.5\) cm and \(y=12\) cm (above the target axis), to study the symmetry of the system. An additional two samples were placed on plate 3, which included \(^{nat}\text{U}\) samples at \(y=\pm 8\) cm. The positioning of all samples deployed for the 1 GeV irradiation is shown in Table 3.9 and also graphically in Figure 3.19a.

As can be seen in Table 3.2 on page 35 the size of the incident 1 GeV deuteron beam was large. The beam was located 0.2 cm below the target axis with a FWHM\(_y\) of 3.5 cm. This caused a small portion of the beam to impinge directly on the samples located at \(y=-4\) cm. Previous experience had shown that samples placed directly in the path of the incident beam are very difficult to analyse because the high energy deuterons induce a large number of fissions.
### 3.5. Fission track detectors

**TABLE 3.9:** Locations of fission samples for the 1 GeV Quinta irradiation. Distances given relative to the origin, positioned front and centre of the beam window on the target-blanket.

<table>
<thead>
<tr>
<th>Plate</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>z-pos (cm)</td>
<td>-0.5</td>
<td>12.6</td>
<td>25.7</td>
<td>35.8</td>
<td>51.9</td>
<td>65.0</td>
</tr>
<tr>
<td>y-pos (cm)</td>
<td>+12</td>
<td>Au,Pb</td>
<td>Bi,Th</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>+8</td>
<td>Au,Pb</td>
<td>Bi,Th,U</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>+5.5</td>
<td>Au,Pb</td>
<td>Bi,Th</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-4</td>
<td>Au,Pb</td>
<td>Bi,Th</td>
<td>Au,Pb</td>
<td>Au,Pb</td>
<td>Au,Pb</td>
</tr>
<tr>
<td></td>
<td>-6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**TABLE 3.10:** Locations of fission samples for the 4 GeV Quinta irradiation. Distances given relative to the origin, positioned front and centre of the beam window on the target-blanket.

<table>
<thead>
<tr>
<th>Plate</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>z-pos (cm)</td>
<td>-0.5</td>
<td>12.6</td>
<td>25.7</td>
<td>35.8</td>
<td>51.9</td>
<td>65.0</td>
</tr>
<tr>
<td>y-pos (cm)</td>
<td>+12</td>
<td>Au,Pb</td>
<td>Bi,Th</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>+8</td>
<td>Au,Pb</td>
<td>Bi,Th,U</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>+6</td>
<td>Au,Pb</td>
<td>Bi,Th</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
FIGURE 3.18: The Quinta sample plates showing the samples mounted on them. Samples belonging to the author and also other collaboration members are shown. Other members of the collaboration have studied characteristics of the ADS neutronics, other than those reported in this thesis.

TABLE 3.11: Location of external thorium samples for 4GeV irradiation. Position relative to origin located at the front and centre of beam window.

<table>
<thead>
<tr>
<th>Sample</th>
<th>x (cm)</th>
<th>y (cm)</th>
<th>z (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>30</td>
<td>-2.5</td>
<td>1.9</td>
</tr>
<tr>
<td>2</td>
<td>30</td>
<td>-2.5</td>
<td>11.9</td>
</tr>
<tr>
<td>3</td>
<td>30</td>
<td>-2.5</td>
<td>21.9</td>
</tr>
<tr>
<td>4</td>
<td>30</td>
<td>-2.5</td>
<td>31.9</td>
</tr>
<tr>
<td>5</td>
<td>30</td>
<td>-2.5</td>
<td>41.9</td>
</tr>
</tbody>
</table>

in the sample, resulting in overlapping of tracks in the mica detector. Therefore, for the 4GeV irradiation, samples were lowered to the \( y = -6 \) cm position from the \( y = -4 \) cm position. Of course, as can be seen from Table 3.2, this change was redundant given the relatively small size of the 4 GeV deuteron beam. The positioning of samples in the 4 GeV irradiation is shown in Table 3.10 and also graphically in Figure 3.19b.

In order to measure the fast neutron leakage from the assembly, five of the six thorium samples at \( y = -6 \) cm were relocated to the outside of the Quinta assembly. This was also done for fear the samples would be overexposed leaving them with an uncountable track density. The accurate determination of neutron leakage from nuclear assemblies has important implications for dose rate calculations, design of external shielding, addition of any blanket or neutron multipliers and any applications that the escaping neutrons will be used for e.g. material studies or nuclear waste transmutation. The precise coordinates of these samples are shown in Table 3.11 and indicated graphically in Figure 3.19c.
3.6 CR-39/LR-1152B track detector technique

Graphite is a very effective neutron moderator causing neutrons to quickly thermalise. The majority of neutrons on the surface of the Gamma-3 assembly are slow neutrons, but have an energy range spanning more than 11 orders of magnitude. Measuring position-dependent energy spectra across this range with a single measurement technique is not practical. Slow neutrons strongly influence reaction rates within the assembly, including fission rates of fissile materials, breeding of $^{233}$U from $^{232}$Th and nuclear waste transmutation of long lived fission products. In previous works, nuclear track detectors have been used to study the neutron field around a paraffin moderated lead target (Gamma-2 set-up) [143, 182]. In this work, the spatial

3.5.3 Analysis of fission track detectors

After the irradiation, the mica track detectors were separated from the fission foils and etched in 7% hydrofluoric acid at 60°C. The time of etching depended on the expected track density of the samples. Samples with very high track densities (e.g. uranium samples irradiated with 4 GeV) were etched for 20 minutes, while samples with the lowest track densities were etched for several hours. Etching the samples for longer periods enlarges the tracks making them visible with lower magnifications of the microscope.

Several images of the tracks in each of the mica detectors were taken using an Olympus CX41 microscope in conjunction with a Canon EOS 550D Digital SLR camera. Unlike the track counting procedure of the CR-39 detectors (discussed on page 62) which could be performed automatically using a macro written in the ImageJ software, track counting of the mica detectors needed to be performed manually. This was because the mica crystal had many internal fractures, which interfered with the automatic counting. These anomalies in the image led to many false positive results in the automatic track counting procedure. Instead, the inbuilt ‘cell counter’ function of ImageJ was used after post-processing the images (background subtraction, brightness/contrast adjusted etc.). The statistical uncertainty of the track density for each mica detector was kept below 5% in all cases except for those samples with very low track densities.

3.6 CR-39/LR-1152B track detector technique

Graphite is a very effective neutron moderator causing neutrons to quickly thermalise. The majority of neutrons on the surface of the Gamma-3 assembly are slow neutrons, but have an energy range spanning more than 11 orders of magnitude. Measuring position-dependent energy spectra across this range with a single measurement technique is not practical. Slow neutrons strongly influence reaction rates within the assembly, including fission rates of fissile materials, breeding of $^{233}$U from $^{232}$Th and nuclear waste transmutation of long lived fission products. In previous works, nuclear track detectors have been used to study the neutron field around a paraffin moderated lead target (Gamma-2 set-up) [143, 182]. In this work, the spatial
distribution of slow neutrons on the Gamma-3 assembly was measured using TASTRAK CR-39 track detectors in conjunction with Kodak-Pathé LR-115 Type 2B film.

CR-39 was first synthesised in 1940 by the Pittsburgh Plate Glass Company [183] finding widespread usage in the manufacturing of eyeglass lenses. It is a colourless, transparent, scratch resistant plastic formed by the polymerisation of allyl diglycol carbonate (PADC - \((\text{C}_{12}\text{H}_{18}\text{O}_7)\)) and was first used as a SSNTD in 1978 at the Lawrence Livermore Laboratory [184]. Etchable damage tracks can form in CR-39 by the interaction of charged particles (protons, deuterons, tritons, alpha) with energies greater than 100 keV. \(\beta\)-particles, \(\gamma\)-rays and thermal neutrons leave no recordable tracks. Fast neutrons may be detected indirectly via the production of recoil ions which in turn produce etchable tracks. Bare CR-39 detectors have become effective fast neutron dosimeters for this reason [185].

The LR-115 Type 2B film developed and manufactured by Kodak-Pathé, consists of a solid layer of compressed powdered lithium tetraborate (\(\text{Li}_2\text{B}_4\text{O}_7\)) on a 15 \(\mu\text{m}\) cellulose nitrate (CN) substrate mounted on 100 \(\mu\text{m}\) polyester (PET) [186] (see Figure 3.22).

CR-39 itself is unresponsive to slow neutrons, but can detect the alpha particles produced by slow neutron induced \(^{10}\text{B}(n,\alpha)^7\text{Li}\) and \(^6\text{Li}(n,t)^4\text{He}\) reactions occurring in the \(\text{Li}_2\text{B}_4\text{O}_7\) layer of the LR-115 2B film. Boron-10 (19.9 \% of natural boron) and lithium-6 (7.5 \% of natural lithium) are attractive isotopes for slow neutron detection due to the large cross sections and the following of \(1/v \left(1/\sqrt{E}\right)\) relationship over a wide range of energies. The cross sections of the \(^{10}\text{B}(n,\alpha)^7\text{Li}\) and \(^6\text{Li}(n,t)^4\text{He}\) are shown in Figure 3.20.

### 3.6.1 Placement of CR-39/LR-115 2B on Gamma-3

The CR-39/LR-115 2B detectors were placed in 27 locations on the external surfaces of the Gamma-3 assembly. This included 17 samples on the top, 5 samples on the front and 5 samples
3.6. CR-39/LR-1152B track detector technique

Sample locations

110 cm
60 cm
110 cm
110 cm

a) Top view
b) Front/Back view

Sample
A
Graphite
Textolite strips
Sample
B

FIGURE 3.21: Locations of CR-39/LR-1152B track detectors placed on the Gamma-3 assembly. Samples were placed on the top, front and back surface. Each sample location contains a bare CR-39/LR-1152B sample (Figure 3.22) and a cadmium covered CR-39/LR-1152B sample (Figure 3.23). Figures are not to the same scale.


LR115 Type 2B
Lithium tetraborate
Cellulose Nitrate
Polyester (PET)

FIGURE 3.23: Cadmium covered CR-39/LR-1152B used to filter out thermal neutrons. The broken lines on the edges are used to signify the sealed and unsealed versions of the cadmium cover that were simulated with Monte Carlo (see section 3.6.4.2 for details).

3.6.2 Analysis of CR-39 detectors

After the irradiation, the samples were removed and transported back to Sydney. The CR-39 was separated from the LR-1152B film and etched in 6N NaOH at 70°C for 90 min. This etching time is enough to resolve alpha tracks but not proton recoils produced by high energy neutrons. The required number of images were taken using an Olympus CX41 microscope coupled to a Canon EOS 550D Digital SLR camera. A macro was written for the image processing software
CHAPTER 3. EXPERIMENTAL METHODOLOGY

FIGURE 3.24: (a) Sample photo of tracks on the CR-39 surface as viewed through the microscope and (b) the same image with the background subtracted ready for automated track counting.

ImageJ [187] to automatically post process the photographs and count the number of tracks in each image. A sample image of the tracks on the CR-39 surface is shown in Figure 3.24.

Fast neutrons may produce etchable tracks directly in the CR-39 from recoil scattering events. These tracks, along with background tracks, are produced irrespective of whether the LR-115 2B is present or not. Correction for these tracks were determined from the back side and front side of the CR-39, which was not in contact with LR-115 2B film. This was then subtracted from the track density in the region covered by the LR-115 2B film to ensure the measured track density was exclusively due to slow neutron induced \((n,\alpha)\) reactions in \(^{10}\)B and \(^6\)Li.

3.6.3 Theoretical considerations

The neutron spectrum emerging from a moderator is assumed to be the superposition of a Maxwell-Boltzmann distribution and a continuous slowing down spectrum [188]. The Maxwell-Boltzmann distribution portrays neutrons that have reached thermal equilibrium with the moderating environment, while the slowing down spectrum describes neutrons that have not yet reached thermal equilibrium. These epithermal neutrons follow an approximate \(1/E\) distribution which arises from the constant fractional energy loss after many successive elastic collisions.

The energy distribution of the neutrons may therefore be expressed as:

\[
\Phi(E) = \phi_{th} \frac{E}{(kT)^2} \exp\left(-\frac{E}{kT}\right) + \phi_{epi} \frac{\Delta(E/kT)}{E^{1+\alpha}}
\]

where \(\phi_{th}\) and \(\phi_{epi}\) are the thermal and epithermal neutron flux constants, \(T\) is the effective neutron temperature, \(k\) is Boltzmann’s constant, \(\Delta(E/kT)\) is the joining function and \(\alpha\) is the epithermal neutron flux shape factor [189, 190].

The thermal flux constant, \(\phi_{th}\) is equivalent to the total thermal neutron flux and \(\phi_{epi}\) is the integrated epithermal neutron flux per unit lethargy. The effective neutron temperature, \(T\) is always higher than the moderator temperature because of preferential absorption at thermal energies which removes lower energy neutrons before they can thermalise completely. The
3.6. CR-39/LR-1152B track detector technique

spectrum becomes ‘absorption hardened’ and as the absorption and hardness of the spectrum increases, the effective temperature does too [29]. The epithermal neutron flux shape factor, $\alpha$ is used to adjust for the fact that the epithermal spectrum often deviates from the ideal $1/E$ law. It can be positive or negative, is less than an absolute value of 1, and can vary at different irradiation sites within the same neutron field [191]. Moderator material, geometry of the irradiation site and configuration of the neutron source all serve to impact on the value $\alpha$ in a complicated manner [192]. It has also been shown using simple age theory arguments that for graphite moderators $\alpha$ varies with distance from the source in a very complex way [190].

3.6.3.1 Joining Function

The purpose of the joining function, $\Delta(E/kT)$ is to smoothly merge the Maxwellian distribution of the thermal neutrons to the $1/E$ tail of the epithermal neutrons. The joining function (also known as the Westcott cut-off function) is dependent on temperature and the molecular properties of the moderator [193].

The Gamma-3 assembly was composed of a graphite moderator while the calibration of the track samples (discussed further in section 3.6.4) took place using a polyethylene moderator. The joining function of graphite, as measured by Coates [194] and polyethylene, as measured by Mildner et al. [195] can be viewed in Figure 3.25. Clear differences in the joining function can be attributed to the different chemical make-up and molecular bonding of the two moderators.

The joining function can only be determined empirically. Coates [194] evaluated the joining function of graphite at four different temperatures and found it is essentially independent of temperature. Mildner et al. [195] measured the joining function of polyethylene at 300 K and 77 K which revealed a large difference between the two. The empirical formula for the joining

![Graph showing the joining function of graphite and polyethylene](image)

**FIGURE 3.25:** The joining function of graphite [194] and polyethylene [195]. The energy scale corresponds to a neutron temperature of 300 K.
function of polyethylene at 300 K as measured by Mildner et al. [195] and shown in Figure 3.25 is

$$\Delta(E/kT) = \frac{1}{1 + \exp\left(\frac{29.287}{\sqrt{E/kT} - 11.052}\right)}$$  \hspace{1cm} (3.29)

### 3.6.3.2 Cadmium Difference method

The track density (tracks cm\(^{-2}\)) on the surface of the CR-39 detectors is directly proportional to the number of \(^6\)Li(n,t) and \(^{10}\)B(n,\(\alpha\)) reactions occurring in the LR-115 2B film. This track density may be divided into tracks originating from thermal neutrons and tracks originating from epithermal neutrons so

$$\rho = \rho_{th} + \rho_{epi}$$  \hspace{1cm} (3.30)

where \(\rho\) is the total track density, and \(\rho_{th}\) and \(\rho_{epi}\) is the track density caused by thermal and epithermal neutrons, respectively [129]. The track density due to the thermal (\(\rho_{th}\)) and epithermal (\(\rho_{epi}\)) neutrons are directly proportional to the thermal (\(\phi_{th}\)) and epithermal (\(\phi_{epi}\)) flux constants so we can write

$$\rho = w_{th}\phi_{th} + w_{epi}\phi_{epi}$$  \hspace{1cm} (3.31)

where \(w_{th}\) and \(w_{epi}\) are the thermal and epithermal detection efficiencies of the track detectors (in tracks per neutron) [129].

In order to separate the thermal from the epithermal neutrons incident on the detectors, the cadmium difference method was used [189]. This method involves simultaneous irradiation of two detectors, one is covered with a cadmium cover 0.5–1.5 mm thick while the other remains bare. The absorption cross section of cadmium (Figure 3.26) is very high at low energies (2500 b at thermal energies combined with a large absorption resonance at 0.172 eV) but drops very sharply at higher energies (0.06 b at 10 eV). As a very rough first approximation it may be assumed that the cadmium cover absorbs all the thermal neutrons while letting the epithermal neutrons pass through. Therefore, tracks on the cadmium covered CR-39 would be entirely due to epithermal neutrons and tracks on the bare CR-39 would be due to both thermal and epithermal neutrons.

A perfect filter with an ideal step function for a cross section, would have a neutron cut-off energy such that all neutrons with energies below the cut-off would be absorbed and all neutrons above would pass through the filter. However, cadmium is not a perfect filter and the low energy section of the neutron spectrum incident on the detector becomes distorted. The effective cut-off energy, \(E_{Cd}\) has therefore been defined as the energy that allows the same total number of absorptions in the sample as in the cadmium filter. It is a function of cadmium thickness, neutron flux spectrum, angular distribution of neutrons, and sample material size and shape [196].

The effective cadmium cut-off energy has been measured and calculated under a variety of conditions. This includes slab, spherical and cylindrical cadmium cover geometry, different thicknesses of cadmium, isotropic and monodirectional flux and different thermal to epithermal flux ratios (\(\phi_{th}/\phi_{epi}\)) [196–198]. From all these values we chose the value for the effective cut-off
energy that most resembled the conditions of our experiment. Dayton and Pettus [196] reported that a 1 mm thick cadmium in slab geometry exposed to monodirectional neutrons has an effective cut-off energy of $(0.43 \pm 0.01)$ eV (uncertainty arises from reading the data off a graph). Results using this $E_{Cd}$ value were published in our earlier publication [199]. However, we have since improved the accuracy of this value because in our case the neutrons were not strictly monodirectional. Less monodirectional neutrons increase the cut-off energy due to the effective thickness of the cadmium being higher. Further Monte Carlo simulations found that the average angle of neutrons leaving the top surface of the Gamma-3 was $35^\circ$ relative to the surface normal. This increases the effective thickness of the cadmium from 1 mm to 1.2 mm, in turn changing $E_{Cd}$ to $(0.48 \pm 0.03)$ eV. The increased uncertainty accounts for the variation in emission angles of neutrons passing the surface of the Gamma-3. This updated value had negligible impact on the final results presented in Figure 5.4 on page 115, compared to those published in our earlier publication [199].

This cadmium cut-off energy is higher than the lower energy cut-off ($\mu kT$) of the 1/$E$ epithermal spectrum which usually lies around $\sim 0.1$ eV ($\mu$ defined below). It would be desirable to have $E_{Cd}$ as close to or equal to $\mu kT$, however this requires the use of very thin cadmium filters which are very sensitive to the flux ratio ($\phi_{th}/\phi_{epi}$) [197]. This would cause $E_{Cd}$ to fluctuate depending on the hardness of the spectrum. Stoughton et al. [197] have reported that the best compromise between stability over a wide range of flux ratios ($\phi_{th}/\phi_{epi}$) and a low effective cut-off energy is with filters of a ‘40 mil’ (1 mm) thickness, as used here.

The cadmium cover not only absorbs all of the thermal neutrons, but some of the lower energy epithermal neutrons as well. The cadmium correction factor, $F_{cd}$ [189] is the ratio of the track density due to all epithermal neutrons ($\rho_{epi}$) and the measured track density from the cadmium covered sample ($\rho_{cd}$)

$$F_{Cd} = \frac{\rho_{epi}}{\rho_{Cd}}$$ (3.32)
The total track density, $\rho$ is directly proportional to the total number of $(n,\alpha)$ reactions occurring in the lithium tetraborate volume, so

$$\rho = \varepsilon \eta t \int_0^\infty \sigma(E)\phi(E)\,dE \quad (3.33)$$

where $\varepsilon$ is the track detection efficiency which includes critical angle limitations, minimum detectable track size and track observation conditions [178], $\eta$ is the probability of ions ($\alpha$, $t$ and Li and B recoils) being ejected from the $\mathrm{Li}_2\mathrm{B}_4\mathrm{O}_7$ layer, $t$ is the irradiation time and $\sigma(E)$ and $\phi(E)$ are the energy dependent reaction cross section and neutron flux, respectively.

Combining Equations 3.28 and 3.33, the track density due to only epithermal neutrons for a $1/v$ absorber becomes

$$\rho_{epi} = \varepsilon \eta t \int_0^\infty \sigma_0 \sqrt{\frac{E_0}{E}} \frac{\Delta(E/kT)}{E^{1+\alpha}}\,dE \quad (3.34)$$

$$= \varepsilon \eta t \int_{\mu kT}^\infty \sigma_0 \sqrt{\frac{E_0}{E}} \frac{\Delta(E/kT)}{E^{1+\alpha}}\,dE \quad (3.35)$$

$$\rho_{Cd} = \varepsilon \eta t \int_{E_{Cd}}^\infty \sigma_0 \sqrt{\frac{E_0}{E}} \frac{\Delta(E/kT)}{E^{1+\alpha}}\,dE \quad (3.36)$$

where $\sigma_0$ is the cross section at neutron energy $E_0$ (typically 0.025 eV). Substituting Eq. 3.35 and 3.36 into Eq. 3.32 and integrating we may calculate $F_{Cd}$ in terms of $E_{Cd}$, $\mu kT$ and $\alpha$:

$$F_{Cd} = \left(\frac{E_{Cd}}{\mu kT}\right)^{\alpha+\frac{1}{2}} \quad (3.37)$$

The value $\mu$ is determined from the joining function and is therefore a property of the moderator. Coates [194] have calculated the value of $\mu$ for a graphite moderator to be $3.6 \pm 0.1$, while Mildner et al. [195] have estimated the value of $\mu$ for polyethylene to be somewhere between 4 and 5. $\mu$ may also be obtained by solving the integral

$$\mu = \frac{4}{\left(\int_0^\infty \Delta(x) x^{-\frac{3}{2}}\,dx\right)^2} \quad (3.38)$$

where $x = E/kT$ [189]. Substituting Eq. 3.29 into this reveals $\mu$ to be 7 which is unreasonably high for polyethylene and outside the range of Mildner et al. estimate. As a more precise value of $\mu$ for polyethylene could not be found in the literature, the value of $\mu$ used in this work was $4.5 \pm 0.5$, corresponding to the mid-value of the 4–5 range estimated by Mildner et al. [195]. For an ideal epithermal spectrum (i.e. $\alpha = 0$), an $E_{cd}$ value of $(0.48 \pm 0.03)$ eV and $T$ value of 300 K we find from Eq. 3.37 that $F_{Cd}$ is $2.26 \pm 0.08$ and $2.02 \pm 0.13$ for the graphite and polyethylene moderators, respectively.
3.6. CR-39/LR-1152B track detector technique

![Diagram of the X3 capsule](image)

**FIGURE 3.27:** (a) Amersham International Plc X3 capsule (Image: Lorch [201]). (b) High resolution measurement of the neutron energy spectrum emitted from the $^{241}$Am–Be X3 capsule [200].

### 3.6.4 Determination of $w_{th}$ and $w_{epi}$

#### 3.6.4.1 Experimental Procedure

Calibration of the CR-39/LR-1152B system (determination of $w_{th}$ and $w_{epi}$ from Eq. 3.31) was performed in a reference neutron field consisting of a polyethylene moderated $^{241}$Am–Be neutron source. The neutron source consisted of 4.6 g beryllium and 0.37 g of AmO$_2$ (1 Ci) pressed into a homogenised pellet and placed into an Amersham International Plc X3 capsule. The X3 capsule is a small steel cylinder, 22.4 mm in diameter, 31 mm in height with 2.4 mm wall thickness (Figure 3.27a). A high resolution measurement of the neutron energy spectrum emerging from the capsule, as measured using a $^3$He sandwich spectrometer, is shown in Figure 3.27b [200]. The total number of neutrons emitted by the Amersham X3 source was specified by the manufacturer to be $(2.2 \pm 0.2) \times 10^6$ s$^{-1}$.

This neutron source capsule has previously been used to calibrate track detectors by being placed in the centre of a block of $30 \times 30 \times 31$ cm paraffin moderator. Hashemi-Nezhad et al. [120] successfully used it to calibrate isolated LR-1152B track detectors. Excellent agreement between MCNP simulations, neutron diffusion calculations and $^3$He detector measurements were found. Borger [129] used it to calibrate a different geometry of CR-39/LR-1152B detector but did no $^3$He detector measurements and did not achieve good agreement with his experimental results.

In this case, the X3 neutron source was placed in the centre of the back of a block of high density polyethylene ($0.95$ g cm$^{-3}$ HDPE PE300) with dimensions $50 \times 50 \times 8$ cm. The thickness of the polyethylene was chosen to be 8 cm as Monte Carlo calculations revealed this thickness to produce the best balance between an ideal ($\alpha=0$) epithermal neutron spectrum and high neutron flux. The height and width was 50 cm so as to be the same length as the $^3$He detector that was used for the absolute neutron measurements. Five cadmium covered CR-39/LR-1152B samples (Figure 3.23) and five bare CR-39/LR-1152B samples (Figure 3.22) were placed horizontally

along the front of the polyethylene, 10 cm apart (see Figure 3.28) and irradiated for a period of three weeks.

The track densities of the cadmium covered, $\rho_{\text{Cd}}$ and bare CR-39/LR-115 2B, $\rho$ were determined for the central three sample locations under the exact same etching and counting procedures as for the Gamma-3 samples. The two samples on the edges had track densities that were too low to enable good counting statistics and so were not included in the calibration.

The thermal neutron flux on the surface of the polyethylene was verified using a $^3\text{He}$ detector with known efficiency for thermal neutrons. The cylindrical $^3\text{He}$ detector was 50 cm in length, 2.54 cm in diameter with 1 mm thick stainless steel wall. The operating voltage was 1000 V and pulses were analysed using a Davidson multichannel analyser (model 1056 C). Due to the large detector size, and the small area of where the neutrons were measured, the entire detector was covered in cadmium with thickness of 1 mm, apart from a window of area 1 cm$^2$. This window was to be located over the position where the samples were placed. The cross section of the $^3\text{He}(n,p)^3\text{H}$ reaction is shown in Figure 3.20 (page 60) and it can be seen that it has very similar response $(1/v)$ as the $^6\text{Li}(n,t)^4\text{He}$ and $^{10}\text{B}(n,\alpha)^7\text{Li}$ reactions.

Measurements of the neutron flux on the surface of the polyethylene were performed with the cadmium window both open and closed. The difference between the count rates with the window open and closed corresponds to all neutrons below the cadmium cut-off energy (0.43 eV) entering the detector via the 1 cm$^2$ window. A pulse height distribution with the cadmium window both open and closed as measured with the $^3\text{He}$ detector is shown in Figure 3.29. The efficiency of this specific detector is known to be 1 count per 5.36 thermal neutrons [120]. The $^3\text{He}$ detector measurements combined with the Monte Carlo simulations revealed the source to have a total neutron emission rate of $(2.16 \pm 0.09) \times 10^6$ s$^{-1}$ which is well within the manufacturers provided value of $(2.2 \pm 0.2) \times 10^6$ s$^{-1}$.
3.6. CR-39/LR-1152B track detector technique

3.6.4.2 Calculations and Monte Carlo simulations of calibration source

The values of $\rho_{epi}$ and $\rho_{th}$ were found from counting the track densities of the cadmium covered ($\rho_{Cd}$) and bare samples ($\rho$).

$$\rho_{epi} = F_{Cd} \rho_{Cd}$$

$$\rho_{th} = \rho - F_{Cd} \rho_{Cd}$$

The neutron spectrum on the surface of the polyethylene at each of the three sample locations was simulated using MCNPX, implementing the Am-Be neutron source energy spectrum given by Marsh et al. [200] (Figure 3.27b). The resulting spectrum was then fitted in the region $<1$ keV to Eq. 3.28 [129] with $\phi_{th}$, $\phi_{epi}$, $T$ and $\alpha$ as fitting parameters. All spectra were fitted in Python using the Levenberg-Marquadt least-squares fitting algorithm. Neutrons with energies $>1$ keV have very low interaction probability with the lithium-borate due to the $1/v$ cross section dependence and the number of neutrons $>1$ keV in the sample locations being several orders of magnitude lower than at thermal energies. Therefore, the number of fast neutrons contributing to $(n,\alpha)$ reactions in the LR-1152B film was deemed to be negligible. $w_{th}$ and $w_{epi}$ were then calculated from $\phi_{th}$, $\phi_{epi}$, $\rho_{th}$ and $\rho_{epi}$ using

$$w_{th} = \frac{\rho_{th}}{\phi_{th} S t}$$  \hspace{1cm} (3.41)

$$w_{epi} = \frac{\rho_{epi}}{\phi_{epi} S t}$$  \hspace{1cm} (3.42)

where $t$ is the irradiation time and $S$ is the strength of the neutron source i.e. $(2.16 \pm 0.09) \times 10^6$ s$^{-1}$ as determined from $^3$He measurements.

Equations 3.41 and 3.42 are only valid in cases where a completely sealed cadmium cover is used to shield the detector from thermal neutrons. As can be seen from Figure 3.23, in our

![Pulse height distribution measured with the $^3$He detector measured over the centre sample with the cadmium window open and closed.](image)

**Figure 3.29:** Pulse height distribution measured with the $^3$He detector measured over the centre sample with the cadmium window open and closed.
experiments the edges of the cadmium cover were open. A small number of thermal neutrons leaked through the sides of the cadmium covered samples which needed to be corrected for when calculating $\rho_{\text{epi}}$. MCNPX calculations comparing the effect of sealing the edges of the cadmium cover showed that while the unsealed cover stopped $>99.2\%$ of thermal neutrons, the sealed cover stopped $100\%$ of thermal neutrons. The sealed cadmium cover involved sealing the edges with 1 mm of cadmium, as indicated by the dotted lines in Figure 3.23 (page 61). The comparison of neutron spectra between the uncovered, sealed and unsealed cadmium covers is shown in Figure 3.30, with a comparison to the Gamma-3 spectrum.

The corrected $\rho_{\text{epi}}$ was found by fitting the thermal component of the unsealed spectrum (refer to Figure 3.30) with the Maxwellian distribution described in the first part of Eq. 3.28. Replacing $\phi_{\text{th}}$ with $\phi_{\text{th}}^{Cd}$ to obtain the thermal neutron flux leaking through the sides of the cadmium, the corrected $\rho_{\text{epi}}$ could then be found using

$$\rho_{\text{epi}} = F_{Cd}(\rho_{Cd} - w_{\text{th}}^{Cd} \phi_{\text{th}}^{Cd})$$

(3.43)

where $w_{\text{th}}^{Cd}$ is now determined recursively from Equations 3.44 and 3.45 (n is iteration number). The initial guess for $w_{\text{th}}^{Cd}$ is the equation for zero leakage through the sides of the detector. Convergence to a stable solution was reached after approximately five iterations.

$$w_{\text{th}}^{n} = \frac{\rho - F_{Cd}(\rho_{Cd} + w_{\text{th}}^{n-1} \phi_{\text{th}}^{Cd})}{\phi_{\text{th}} S t}$$

(3.44)

$$w_{\text{th}}^{0} = \frac{\rho - F_{Cd}\rho_{Cd}}{\phi_{\text{th}} S t}$$

(3.45)
TABLE 3.12: Track densities and fitting parameters for the neutron spectra of the samples used for the CR-39/LR-1152B calibration. The spectrum of the central sample (0 cm) is shown in Figure 3.30. The parameters have the following units: \( \rho \) (tracks cm\(^{-2}\) neutron\(^{-1}\)), \( \phi \) (cm\(^{-2}\) neutron\(^{-1}\)) and \( w \) (tracks neutron\(^{-1}\)). The anisotropy between the \(-10\) cm and \(+10\) cm sample is due to the asymmetry of the \(^{241}\)Am–Be source and its positioning.

<table>
<thead>
<tr>
<th></th>
<th>-10 cm</th>
<th>0 cm</th>
<th>+10 cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho )</td>
<td>((5.52 \pm 0.33) \times 10^{-8})</td>
<td>((1.93 \pm 0.93) \times 10^{-7})</td>
<td>((5.20 \pm 0.36) \times 10^{-8})</td>
</tr>
<tr>
<td>( \rho_{Cd} )</td>
<td>((2.14 \pm 0.39) \times 10^{-9})</td>
<td>((6.6 \pm 1.2) \times 10^{-9})</td>
<td>((1.70 \pm 0.28) \times 10^{-9})</td>
</tr>
<tr>
<td>( \rho_{th} )</td>
<td>((5.28 \pm 0.42) \times 10^{-8})</td>
<td>((1.84 \pm 0.12) \times 10^{-7})</td>
<td>((5.04 \pm 0.42) \times 10^{-8})</td>
</tr>
<tr>
<td>( \rho_{epi} )</td>
<td>((2.4 \pm 0.8) \times 10^{-9})</td>
<td>((9.4 \pm 2.5) \times 10^{-9})</td>
<td>((1.6 \pm 0.6) \times 10^{-9})</td>
</tr>
<tr>
<td>( \phi_{th} )</td>
<td>((4.61 \pm 0.04) \times 10^{-5})</td>
<td>((1.72 \pm 0.01) \times 10^{-4})</td>
<td>((4.48 \pm 0.03) \times 10^{-5})</td>
</tr>
<tr>
<td>( \phi_{epi} )</td>
<td>((3.43 \pm 0.19) \times 10^{-6})</td>
<td>((1.31 \pm 0.04) \times 10^{-5})</td>
<td>((2.91 \pm 0.14) \times 10^{-6})</td>
</tr>
<tr>
<td>( T(K) )</td>
<td>(381 \pm 3)</td>
<td>(378 \pm 2)</td>
<td>(375 \pm 2)</td>
</tr>
<tr>
<td>( T(K) )</td>
<td>(381 \pm 3)</td>
<td>(378 \pm 2)</td>
<td>(375 \pm 2)</td>
</tr>
<tr>
<td>( \alpha )</td>
<td>(-0.019 \pm 0.005)</td>
<td>(-0.011 \pm 0.003)</td>
<td>(-0.003 \pm 0.004)</td>
</tr>
<tr>
<td>( F_{Cd} )</td>
<td>(1.76 \pm 0.11)</td>
<td>(1.79 \pm 0.11)</td>
<td>(1.81 \pm 0.11)</td>
</tr>
<tr>
<td>( \phi_{th}^{Cd} )</td>
<td>((7.26 \pm 0.14) \times 10^{-7})</td>
<td>((1.36 \pm 0.02) \times 10^{-6})</td>
<td>((7.23 \pm 0.14) \times 10^{-7})</td>
</tr>
<tr>
<td>( w_{th} )</td>
<td>((1.14 \pm 0.09) \times 10^{-3})</td>
<td>((1.07 \pm 0.07) \times 10^{-3})</td>
<td>((1.13 \pm 0.09) \times 10^{-3})</td>
</tr>
<tr>
<td>( w_{epi} )</td>
<td>((6.9 \pm 2.5) \times 10^{-4})</td>
<td>((7.2 \pm 1.9) \times 10^{-4})</td>
<td>((5.5 \pm 2.1) \times 10^{-4})</td>
</tr>
<tr>
<td>( \bar{w}_{th} )</td>
<td>((1.11 \pm 0.05) \times 10^{-3})</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \bar{w}_{epi} )</td>
<td>((6.5 \pm 1.3) \times 10^{-4})</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The weighted average of \( w_{th} \) and \( w_{epi} \) from the three calibrations samples were therefore found to be \((1.11 \pm 0.05) \times 10^{-3}\) and \((6.5 \pm 1.3) \times 10^{-4}\) tracks neutron\(^{-1}\), respectively for etching conditions and counting procedures described above. A complete list of results for the three samples is shown in Table 3.12. Without correction for the thermal neutrons leaking into the sides of the cadmium covered samples, the value of \( w_{th} \) would be lower by about 2%, while \( w_{epi} \) would be about 60% higher. It is noted that in future, any further calibration of the track detectors will be better achieved using a completely sealed cadmium cover removing the need to solve for \( w_{th} \) recursively.

The requirement of Monte Carlo simulations to calibrate the detectors may seem contradictory. However, it is noted that only cross section data tables (ENDF/B-VII.0) were used because the maximum neutron energy was less than 20 MeV and so no physics models were required. The secondary objective of this work was to assess the spallation neutron production calculated by the high energy physics models CEM03 and INCL4-ABLA and so we do not feel this is compromised.
Chapter 4

Monte Carlo Simulations

The Monte Carlo method involves solving physical or mathematical problems through the repeated generation of random numbers. It is particularly useful in cases where there are no analytical solutions and deterministic methods are cumbersome or inaccurate. Although the process of using random numbers to solve numerical problems has been around for centuries [202], the term ‘Monte Carlo’ was not coined until the 1940’s [203]. The name is a reference to the uncle of Los Alamos mathematician, Stanislas Ulam who would regularly borrow money from relatives to visit the Monte Carlo casino [203]. Monte Carlo simulations are popular for modelling stochastic processes such as radiation transport, economics, traffic flow, weather forecasting and population changes.

4.1 Radiation Transport Codes

The Monte Carlo method has widespread usage in radiation transport codes. Particles are transported through time and space in a material volume and possible interactions are sampled from Probability Distribution Functions based on reaction cross section data. Several radiation transport codes developed in a range of countries are currently available. Popular general purpose codes commonly in use today include MCNP5/MCNPX/MCNP6 [204], Geant4 [205, 206], PHITS [207] and FLUKA [208, 209].

An extensive comparison between the Geant4 and MCNPX codes for spallation target modelling was undertaken in the PhD study of Borger [129]. It was found that MCNPX better reproduced experimental results, as Geant4 tended to overestimate the neutron yield from spallation targets. In simulations of a 1 GeV proton on lead target using the Bertini INC model (described in section 4.4.1.1), Geant4 was shown to overestimate the neutron yield by 35%. This divergence was attributed to differences in the fission-evaporation models of the two codes. Although Geant4 is an excellent code for particle physics studies, charged particle transport and detector simulation, in its current form it remains less suitable for neutron transport and ADS studies [129]. Therefore, MCNPX remains the preferred option for simulation of spallation
targets at this stage. Additionally, our research group has a long history of using MCNP/MCNPX providing an extensive knowledge and support base. Therefore, MCNPX was also the code of choice for this study.

4.2 MCNPX

MCNPX (Monte Carlo N-Particle eXtended) is a general purpose radiation transport and nuclear reaction simulation code. It is capable of transporting 34 particle types (+2205 heavy ions) over a broad range of energies (up to $\sim$1 TeV) in a completely 3-dimensional and time dependent fashion. It finds applications in many areas of studies including medical physics, space physics, radiation shielding and health physics, accelerators, radiation detection, nuclear fuel cycle, and criticality, transmutation, activation and burnup in nuclear reactors.

Motivated by the Accelerator Production of Tritium (APT) project, MCNPX began in 1994 as a seamless merger of the MCNP4B and LAHET 2.8 codes [210]. The LAHET (Los Alamos High Energy Transport) code [211], itself based on Oak Ridge’s HETC (High Energy Transport Code) [212] was for the transport of nucleons (neutrons $>20\text{MeV}$), pions and muons. Neutrons below the cut-off energy (typically $20\text{MeV}$) had their kinematic parameters stored to an external file (NEUTP), which was manually passed to MCNP as an input source for continued particle transport. MCNP makes use of cross section data tables and is thus limited by the upper energy limits of these tables. The first public release of MCNPX (version 2.1.5, November 1999) [213] finally allowed uninterrupted tracking of all neutrons from $10^{-11}\text{MeV}$ to $>\text{GeV}$, through the use of a single input and output file. The version of MCNPX used in this study was 2.7.0 (released in April 2011 [210]) which will be the last release of MCNPX before merging with MCNP5 to become MCNP6.

4.3 Low energy physics: Cross section data

Low energy reactions below a specific cut-off energy can be calculated by use of cross section data tables and the MCNP side of MCNPX. Evaluated Nuclear Data Files (ENDF), containing sets of cross section data tables form the foundation of radiation transport codes. These databases contain a wealth of nuclear interaction information, often covering several hundred isotopes across a broad range of incident projectile energies (up to an upper energy limit), and the energy and angular distribution of all outgoing particles. The development of complete evaluated libraries is an impressive undertaking, relying on a combination of experimental data, nuclear theory and modelling, statistical analysis, radiation transport physics, and fundamental computer code and database improvements [214]. For extra precision and accuracy, these libraries are then validated against further experiments. Several well known general purpose evaluated libraries originating from all over the world are available today. These include ENDF/B-VII.1 (USA [31]),
Jeff-3.2 (Europe [215]), JENDL-4.0 (Japan [216]), CENDL-3.1 (China [217]), RUSFOND-2010 (Russia [218]) and BROND-2.2 (Russia).

Up until the mid 1990’s, evaluated data only extended up to neutron energies of 20 MeV (e.g. ENDF/VI). However, with increasing interest in nuclear fusion and accelerator/spallation research (particularly ADS), there became an increased need to extend this upper energy limit. The LA150n and LA150h libraries extended over 40 target isotopes deemed important for spallation targets, structural materials and shielding for ADS from the ENDF/B-VI library up to 150 MeV [219]. These improvements were based on measured data and calculations using the nuclear model GNASH code [220]. The upper limit of 150 MeV was merely dictated by the limitations of GNASH, which did not take pion production into account. The LA150n and LA150h libraries were later incorporated into the ENDF/B-VI.6 evaluation. The IAEA also compiled a test library (ADS-LIB/V1.0) based on JEFF-3.1 specifically for analysis of ADS systems and benchmarking of experimental results. This library was then updated to ADS-LIB/V2.0 which extended the number of materials from 30 to 156, borrowing data from ENDF/B-VII.0 and JENDL/AC libraries. Extensions up to 200 MeV came with the TENDL [221] libraries, formed entirely from TALYS-based calculations. A special purpose high energy JENDL library (JENDL/HE-2007 [222]) has created neutron and proton libraries up to 3 GeV for 132 nuclides.

The arrival of ENDF/B-VII.0 [214] vastly improved on the previous ENDF/B-VI.8 evaluation and was used extensively in this study. It contains 14 sublibraries including brand new photomuclear cross section data for 163 isotopes up to 140 MeV. Other improvements include expanding radioactive decay data from 979 to 3838 isotopes and neutron reaction cross section data from 328 to 393 materials (390 isotopes and 3 elements). The other 11 sublibraries are photo-atomic, spontaneous fission yields, atomic relaxation, neutron fission yields, thermal neutron scattering, standards, electro-atomic and non-neutron data including proton, deuteron, triton and $^3$He-induced reactions. The ENDF/B-VII.0 evaluation relied on several reaction physics codes. GNASH, COH by Kawano [LANL, unpublished] and EMPIRE code [223] were primarily used in modelling medium and heavy nucleus reactions (i.e. actinides) and calculating fission product yields, Los Alamos EDA [224] and Oak Ridge SAMMY [225] codes were used for light nucleus reactions and lower energy reactions on heavy targets and Atlas [226] was used for resolved and unresolved resonance calculations based on multi-level Breit-Wigner formalism. Five years later, the ENDF/B-VII.1 [31] evaluation contained new additions to the minor actinides borrowed from JENDL-4.0 [216].

MCNPX requires evaluated data to be processed using the NJOY code [227] into the ACE format. MCNPX 2.7.0 comes packaged with the complete ACE formatted ENDF/B-VII.0 evaluation. Therefore, all simulations carried out in this study utilised cross section data from the ENDF/B-VII.0 library.

The continued development and improvements to nuclear data is an ongoing process. Experimental projects around the world have invested greatly into improving the accuracy and precision.
of nuclear data. Recent efforts have included the HINDAS (High and Intermediate energy Nuclear Data for Accelerator-driven Systems) project which involved a number of European organisations working to provide nuclear data and models relevant to ADS in the 20–2000 MeV range. HINDAS focused on three selected elements deemed integral for ADS development – Pb as target material, Fe for shielding and U representative for the actinides [228, 229]. Since 2001, CERN’s n_TOF (neutron Time-Of-Flight) facility’s major purpose has been to measure the neutron induced reaction cross sections of isotopes relevant to incineration of radioactive nuclear waste and ADS development up to 1 GeV [230–232].

4.4 Intermediate energy physics: Physics models

Above the upper energy limits of the evaluated libraries, or in situations where a nuclide or particle type is missing in the reaction cross section data, physics models must be relied upon to simulate nuclear reactions and particle transport. At sufficiently high energies (>100 MeV), nuclear interactions no longer proceed via the formation of a compound nucleus and the incident projectile interacts with individual nucleons rather than the nucleus as a whole. These spallation reactions are generally modelled in three stages.

The first is the Intranuclear Cascade (INC) stage. The incident particle collides with nucleons in the nucleus leading to further nucleon-nucleon collisions. Some of these nucleons, as well as pions may be ejected from the nucleus with high energies. The INC affects only a small number of nucleons in a small volume of the nucleus. After the INC, a pre-equilibrium stage may occur where the energy is redistributed evenly among all remaining nucleons to form a classically equivalent compound nucleus. In this stage low-energy particles (low MeV energy range) may also be ejected. After pre-equilibrium, the nucleus is left in a highly excited state and de-excitation occurs via the evaporation of neutrons, protons and light ions. Once the excitation energy is below the threshold for particle emission, the nucleus may be left radioactive and undergo gamma decay. For heavy target nuclei, evaporation may compete with high energy fission.

MCNPX provides a seamless transition between table physics and model physics by ‘energy matching’ i.e. the code automatically uses tables up to the maximum energy and above that will switch to a model. It offers several physics model options which may simulate only a single stage of the spallation reaction or combine several of the stages into a single model. Three INC models (Bertini [233, 234], ISABEL [235, 236] and INCL4 [237]), a fission-evaporation model (ABLA [238]) and an evaporation model (Dresner [239]) are available to use in MCNPX. Dresner may be used in combination with either the ORNL [240] or RAL [241] fission models. CEM03 [242] is an entirely self-contained package consisting of INC, pre-equilibrium and evaporation/fission models. For interactions above INC energies, MCNPX also provides two very high energy models – FLUKA (FLUktuierende KAskade) and LAQGSM (Los Alamos Quark-Gluon String Model) [243]. The version of FLUKA (FLUKE89 [244]) within MCNPX is kept primarily for historical reasons and is not recommended for use [210].
4.4. Intermediate energy physics: Physics models

4.4.1 Intranuclear Cascade Models

The first attempt at qualitatively describing a basic reaction mechanism for the Internuclear Cascade is attributed to Serber [86] in 1947. His model introduced several fundamental assumptions which were adapted for implementation into later INC models. He proposed that at sufficiently high energies (\(\sim 100 \text{MeV}\)), the mean free path of the incident projectile will be comparable to nuclear radii (\(4 \times 10^{-13} \text{cm}\)) and the reaction will no longer proceed via the formation of a compound nucleus. Additionally, the collision time between a high energy incident particle and a nucleon in the nucleus is much shorter than the nucleon-nucleon collision time. The nucleon-nucleon scattering cross sections are approximately the same as the free particle cross sections but modified by Pauli exclusion effects owing to the degeneracy of the nucleus. As the nucleon-nucleon scattering cross section is inversely proportional to the energy of the incident particle, the mean free path of nucleons within the nucleus also increases with energy [86].

Equations detailing the energy transfer from high energy neutrons to heavy nuclei were devised by Goldberger [245]. Goldberger combined the assumptions of Serber [86] with the statistical model of Bethe and Bacher [246] to describe the nuclear degeneracy. The equations were solved graphically using the primitive Monte Carlo method of Ulam and Neumann [247] and experimentally measured neutron-proton cross section [248]. Calculations of 100 histories for incident particles of energy 86.6 MeV took two people working full-time for two weeks.

The construction of the Los Alamos MANIAC computer allowed the first large-scale Monte Carlo calculations using computer to occur [15]. Metropolis et al. [249] reported numerous results from simulations for incident particles up to 365 MeV. These were later extended above the pion threshold, up to 1.8 GeV [250]. Pion production (single and double), pion-nucleon scattering, pion absorption, and charge exchange were all taken into account. These calculations were completely 3-dimensional with relativistic kinematics, utilising the latest double differential (wrt. energy and angle) p-p and n-p scattering cross sections [250].

4.4.1.1 The Bertini model

Together with the early works of Goldberger [245] and Metropolis et al. [249, 250], the Bertini INC model still forms the basis of all existing INC models [15]. The traditional Bertini model [233, 234] has been successfully implemented into many codes including HETC [212], HERMES [251], LAHET [252] and MCNPX [210]. A more advanced Bertini model, based off the INUCL code [253] is available in Geant4 [254].

The Bertini model [233] attempted to address the discrepancies of the Metropolis et al. [249, 250] theoretical models with experimental data. Bertini attributed these discrepancies to the oversimplification of the nuclear model, namely that the nucleon density within the spherical nucleus was assumed to remain constant. The Bertini model allows simulations of incident protons, neutrons and pions on all target nuclei. It is basically parameter free, only requiring knowledge of free particle-particle cross sections which is currently well established. Additionally,
the results do not require normalisation and provide reasonable agreement with experimental data [15].

The idea of the Bertini model is to portray the nucleus as three concentric spheres, each having a uniform nucleon density. The zone boundaries for protons and neutrons are equivalent and the proton-to-neutron ratio is constant across all three zones and at the nuclear surface. The nucleon density within each region is a discretized approximation of the continuous non-zero Fermi distribution obtained from Hofstadter [255] experimental electron scattering data. The radial dependence of nucleon density in the nucleus is approximated as the following Fermi function

$$\rho(r) = \frac{\rho_0}{1 + \exp\left(\frac{r - r_0}{\alpha}\right)} \quad (4.1)$$

where $\rho_0$ is the central nucleon density, $r_i$ ($i = 1, 2, 3$) corresponds to the outer radii of the three zones in the nucleus, $r_0$ is $1.07 A^{1/3} \times 10^{-13}$ cm, $\alpha$ is $0.545 \times 10^{-13}$ cm and $A$ is the mass number of the nucleus [15]. The outer radii of the zones are positioned so that they lie at 90, 20 and 1% of the central nucleon density [233], although some other sources position the boundaries at 90, 20 and 10% [15]. The nucleon density therefore decreases with increasing distance from the centre.

In each zone, the momentum distribution of neutrons and protons are approximated as a degenerate (zero-temperature) Fermi energy distribution [233]. The Fermi momentum, $P_{F_i}(r_i)$ in each zone is dependent on the nucleon density and can be found using

$$P_{F_i}(r_i) = \hbar \left(\frac{3\pi^2 \rho(r_i)}{2}\right)^{1/3} \quad (4.2)$$

which corresponds to the Fermi energy as

$$E_{F_i}(r_i) = \frac{P_{F_i}(r_i)^2}{2m_N} \quad (4.3)$$

where $m_N$ is the nucleon mass [15]. As the Fermi momentum depends on the nucleon density, which differs for each zone, the composite momentum across the entire nuclear volume cannot be modelled as a zero-temperature Fermi distribution. Rather, it is represented as a Gaussian distribution with $kT$ value of 15 MeV [233] obtained from experimental data [256].

For a degenerate Fermi gas, all energy states below the Fermi energy are occupied and all energy states above this are unoccupied. The Fermi energy is therefore the kinetic energy of the highest occupied state and the nuclear potential is the nucleon binding energy plus the Fermi energy. Bertini [233] has assigned a nucleon binding energy of 7 MeV for all three regions in every nuclide. The pion potential is assumed equivalent to the nuclear potential of the nucleon it is interacting with.

The INC model is a semi-classical model where the only quantum mechanical effect considered is the Pauli exclusion principle. In the Bertini model, only protons and neutrons obey the exclusion
principle as the densities of other fermions is considered negligibly small. It is assumed that as
the collision time is very short ($\leq 10^{-22}$ s), the nucleus will remain in the ground state. Therefore,
the nucleons are treated as a completely degenerate Fermi gas where all states below the Fermi
energy are occupied. Consequently, all secondary neutrons (and protons) produced in the cascade
must have an energy greater than the Fermi energy, forbidding any collisions involving very large
or small energy transfers. In Monte Carlo calculations, if an interaction results in any secondary
nucleon with momentum less than the Fermi momentum it is automatically rejected. This is
called Pauli blocking.

The Bertini model requires free particle cross section data for nucleon-nucleon elastic and
inelastic, pion-nucleon elastic scattering, charge exchange and pion production which is well
characterised by experimental data [15]. Pion production from nucleon-nucleon or pion-nucleon
collisions is calculated using the isobaric nucleon model [257, 258]. This model assumes pions
are produced from the decay of isobars ($\Delta$-resonances) of excited nucleons. The original Bertini
model only considers single and double pion production, creating an upper energy limit of 3.5 GeV
for nucleon-nucleon interactions and 2.5 GeV for pion-nucleon interactions. Above these energies,
higher order pion and other meson production channels start becoming relevant. An extrapolation
procedure for finding the energy, angle, and multiplicity of products from inelastic collisions
for 3–10 GeV using scaling relations based on scant experimental data has been developed [259].
MCNPX switches to this scaling procedure above 3.495 GeV [210].

The Bertini model is executed in Monte Carlo simulations as a series of basic steps [15]:

1. The collision impact parameter is determined from uniform sampling across the cross sectional
area of nucleus.
2. The path length of the incident particle before experiencing a collision is calculated from total
particle-particle cross section and zone dependent nucleon densities.
3. When incident particle experiences a collision, the type of reaction, momentum transfer
to struck nucleon, and the energy and direction of reaction products are calculated from
statistical sampling and free particle cross section data. Relativistic kinematics is in effect at
all times.
4. If collision is not Pauli blocked, then reaction products are transported as per step 2 until they
are either emitted from the nucleus or fall below a predefined cut-off energy. In general, the
cut-off energy is taken to be half the coulomb barrier at the nuclear surface [260]. Secondary
particles are transported no different to primary particles with the exception that they begin
their trajectory inside the nucleus.
5. The INC is terminated when all reaction products have either become absorbed or escaped
the nucleus. The excitation energy $E^*$, and the mass and charge, $A^*$ and $Z^*$, of the residual
nucleus can have a large range of values because of the high energies involved in the interactions.
These properties are evaluated from a sampling of energy and particle balances, respectively.
The output (A*, Z* and E*) of the residual nuclei is then used as input for the subsequent evaporation calculation, and the double differential particle spectra (wrt. energy and angle) are used to continue the particle transport.

4.4.1.2 The INCL model

Despite the widespread success and implementation of the Bertini model in many code systems, it has been surpassed by more recent advanced models. The INCL (INC-Liège) model was originally conceived at the University of Liège, Belgium in the early 1980’s [261]. Today, the model is still being actively developed and is jointly maintained by Liège and CEA-Saclay in France. INCL was originally dedicated to heavy ion collisions in the GeV range [261] but INCL4 is now used for simulating nucleon and light-ion (A ≤ 4, ≤2AGeV) induced reactions. INCL4 was developed as part of the European HINDAS project [229].

INCL4.2 [237] was released in 2002 and is considered to be the ‘standard version’ of INCL. INCL4.2 has been successfully integrated into LAHET, MCNPX and the recent MCNP6 [262]. As it is included in MCNPX 2.7, this is the version used in this work. The INCL code was originally written in Fortran, however a C++ edition (INCL++ [263]) has been reprogrammed from scratch for use in Geant4. This version is physically equivalent to INCL4.6 [264] with the exception it can handle incident light ions up to mass 18.

A brief description of the fundamental principles behind INCL4.2 [237] model is provided below, and henceforth the INCL4.2 model will be simply referred to as INCL4. The model adopts many of the same fundamental semi-classical assumptions of the Bertini model, eg:

- The particle cascade consists of a series of two body point-like collisions
- Collisions are considered instantaneous and follow the laws of classical mechanics
- Relativistic kinematics is in effect
- The only quantum mechanical effect enforced is the Pauli exclusion principle. However, INCL4 applies the more sophisticated dynamic Pauli blocking, which takes into account depletion of the Fermi sphere from prior reactions.

The INCL4 model makes accommodation for a diffuse nuclear surface giving a more realistic nucleon density distribution than the sharp boundary of Bertini. The nuclear density is defined by a Woods-Saxon density distribution of the form:

$$
\rho(r) = \begin{cases} 
\rho_0 & \text{for } r < R_{\text{max}} \\
\frac{\rho_0}{1 + \exp \left( \frac{r-R_0}{a} \right)} & \text{for } r > R_{\text{max}} 
\end{cases}
$$

(4.4)

$R_{\text{max}}$ is defined as $R_0 + 8a$ where $a$ is $0.510 + 1.63 \times 10^{-4} A$ fm and $R_0$ is parametrised from electron scattering measurements as $(2.745 \times 10^{-4} A + 1.063)A^{1/3}$ fm. $\rho_0$ normalises the distribution to the target mass, $A$ [237].
INCL4 tracks all particles as a function of time and determines a “self-consistent” stopping time for the cascade i.e. the point where the INC model stops and hands over to a pre-equilibrium or evaporation model. This is in contrast to Bertini which tracks particles one by one until they either escape or fall below the cut-off energy. The stopping time is a parametrised equation depending only on the mass of the target nuclide; it is largely independent of the impact parameter and incident projectile energy [237].

4.4.2 Pre-equilibrium models

After the conclusion of the INC, the energy in the residual nucleus is concentrated to a small area of the nuclear volume. The pre-equilibrium stage acts to distribute this energy evenly among all remaining nucleons in the nucleus. The Bertini model utilises the multistage Multistep Pre-equilibrium exciton Model (MPM) [252], originally developed by Griffin [265], which may be toggled on/off within MCNPX according to the preferences of the user. An exciton is either a nucleon above the Fermi energy, or an unoccupied state below it. The MPM model requires the excitation energy and particle-hole configuration from the outcome of the INC. At each stage of the MPM, a neutron, proton, deuteron, \(^3\)H, \(^3\)He or \(^4\)He may be emitted until the equilibrium exciton number is reached and evaporation model invoked [252]. CEM03 employs the integrated Modified Exciton Model (MEM) [266], which also incorporates the evaporation stage of the reaction. INCL4 does not use a pre-equilibrium model.

4.4.3 Evaporation and High-Energy Fission

After the INC (or pre-equilibrium) stage, the residual nucleus is left in a highly excited state. Most nuclear evaporation models are based on the statistical theory of evaporation from a compound nucleus originally developed by Weisskopf [267]. The compound nucleus is described only in terms of its mass, charge and excitation energy and has no “memory” of the formation mechanism. Therefore, the method of de-excitation is independent of the formation process. The Dresner EVAP Monte Carlo code [239], based on the Dostrovsky et al. [268] model has been added into LAHET using the level density parameter formulation of Ignatyuk et al. [269]. The original EVAP model was revised several times by Guthrie [270, 271] and Cloth et al. [251] to take into account updated atomic mass tables [272] and level density parameters with shell corrections [273] before being implemented into LAHET [15].

The Dresner EVAP model is restricted to neutrons and light nuclei up to \(^4\)He as ejectiles but the more recent Generalised Evaporation Model (GEM) allows 66 nuclides up to Mg as ejectiles [274]. For heavy nuclei, high energy fission is a competing decay mechanism to evaporation at every stage of the nuclear de-excitation. Therefore, evaporation models are often coupled to high energy fission models. An improved GEM has been successfully joined with the RAL model [241] to produce fission fragments and subsequent evaporation from them [275]. MCNPX
CHAPTER 4. MONTE CARLO SIMULATIONS

offers two high energy physics models that can be invoked when using the Dresner evaporation model – RAL and ORNL [240].

While RAL and ORNL are standalone fission models, the ABLA model developed at GSI, Darmstadt [238, 276] encompasses a combined fission-evaporation model. The ABLA model has the notable advantage of being a dynamical code which takes into account the dynamical nature of fission. The coupling of INCL4 with ABLA has so far shown good agreement with a large range of experimental observables [15].

After all particle decay channels have been depleted (usually at some predefined cut-off like 7 MeV [15]), the remainder of the nuclear de-excitation occurs via emission of gamma photons. Gamma emission does not compete with particle emission at any stage during the spallation process. The photons are emitted with discrete energies, which may be obtained from ENSDF libraries [175] or calculated from photon evaporation models such as the Los Alamos PHT model [211]. The PHT model is integrated into the LAHET and MCNPX codes.

For low mass nuclides (A ≤ 13 at all excitation energies, or 14 ≤ A ≤ 20 with excitation energies < 44 MeV [210]), MCNPX employs a Fermi breakup model [277, 278] instead of the pre-equilibrium and evaporation models. The Fermi breakup model is much more computationally efficient yielding similar results to the more complicated evaporation models [242].

4.4.4 Cascade-Exciton Model (CEM)

The Cascade-Exciton Model (CEM) was originally devised in the early 1980’s at the JINR [279]. CEM is different to other INC models in that it is an amalgamation of many other models describing the different stages of the spallation reaction. It considers the spallation process occurring in three stages – cascade, pre-equilibrium and equilibrium (compound nucleus evaporation and fission).

The cascade stage is based on the standard Dubna INC model [280] and pre-equilibrium is calculated using the Modified Exciton Model (MEM) [266]. The most recent version, CEM03 [242] incorporates the GEM2 evaporation/fission code [281] for the equilibrium stage. CEM03 also includes a Fermi breakup model and the LAQGSM extension [282], allowing simulations of both particle-nucleus and nucleus-nucleus collisions up to 1 TeV per nucleon [242]. CEM03 uses up-to-date experimental data and the latest systematics developed by other codes (e.g. from INCL [237] and BRIC [283]) so as not to rely on the 30 year old data of the original Dubna INC code.

4.5 The TALYS code

Jointly developed by NRG Petten, the Netherlands and CEA Bruyères-le-Châtel, France, TALYS is a deterministic code for simulating nuclear reactions [284]. TALYS was completed as part of the HINDAS project for simulating low energy (1 keV–250 MeV) nuclear reactions (while INCL4 was developed for higher energies). TALYS combines several nuclear models – optical, direct,
4.6 Simulations of experimental set-ups

The entire geometries of the Gamma-3 and Quinta assemblies were programmed into an MCNPX input file and ‘irradiated’ with the same beam positions and shapes as experimentally measured (shown in Tables 3.1 and 3.2 on pages 34 and 35, respectively). Deuterons, neutrons, protons, pions (charged and neutral), muons and photons were all transported. Electrons were not transported as they have negligible effect on the overall results and only slow the simulation down significantly [135]. Neutron transport cross section libraries were from the ENDF/B-VII.0 evaluation [214] and the S(α,β) thermal tables were also used to correctly represent thermal neutron scattering in the graphite. Examples of MCNPX input files for the simulations of the Gamma-3 and Quinta assemblies are provided in appendix C.

4.6.1 Gamma-3 assembly

4.6.1.1 Neutron leakage from target

The neutron leakage from the Gamma-3 lead target irradiated with protons and deuterons is shown in Figure 4.1 as a function of incident projectile energy. These results were calculated using INCL4-ABLA and CEM03 assuming a symmetrical centred beam with a FWHM of 2 cm. For reasons described in chapter 2 (section 2.5.2.3), substantial neutron production does not occur until the projectile energy becomes greater than several hundred MeV. Neutron production for deuteron projectile is also much greater than for proton (explained in section 2.5.2.2). The total neutron multiplicity of deuteron-induced reactions is less than the sum of individual proton and neutron-induced reactions. This is because if a stripping reaction occurs, one of the nucleons may not interact at all, or one nucleon may ‘clear the space’ for the other [286]. The neutron economy increases sharply up to 1 GeV and then decreases slowly above 1.5 GeV (explained in section 2.5.2.3). For 1.6 GeV deuterons, the total neutron leakage is 31 and 32 neutrons per deuteron for INCL4-ABLA and CEM03, respectively.
CHAPTER 4. MONTE CARLO SIMULATIONS

![Figure 4.1](image)

**FIGURE 4.1:** Total neutrons escaping the Gamma-3 lead target. *(a)* per incident ion and *(b)* per GeV for the Gamma-3 under proton and deuteron irradiation up to 4 GeV. Results calculated using MCNPX 2.7 code with INCL4-ABLA and CEM03 physics models.

4.6.1.2 Particle leakage spectra

The energy spectra of particles – neutrons, photons, protons and pions escaping from the bare lead target and graphite moderated lead target of the Gamma-3 set-up is shown in Figure 4.2. These spectra were generated using the CEM03 model emulating the experimental conditions i.e. 1.6 GeV deuteron irradiation with the beam position and shape shown in Table 3.1.

Figure 4.2a shows a typical spallation neutron spectrum. Neutrons escaping the bare lead target span in energies from $\sim 0.1\text{keV}$ all the way up to about the incident deuteron energy (1.6 GeV). The neutron flux drops off significantly above 800 MeV due to the breakup of the incident beam deuteron [287]. The neutron and proton constituting the deuteron are weakly bound and easily separated in "stripping" reactions. The neutrons arise with an energy around $\frac{1}{2}E_d$ and half width of $1.5(E_d\epsilon_d)^{\frac{1}{2}}$, where $E_d$ is the kinetic energy and $\epsilon_d$ is the binding energy of the incident deuteron (2.2 MeV) [116]. The stripping process is the most dominant reaction involved in the first interaction of the high energy deuteron [113].

Most neutrons have energies 1–4 MeV originating from the evaporation stage of the spallation reaction. The presence of the graphite around the target strongly moderates the neutrons producing a significant Maxwellian thermal peak and $1/E$ slowing down epithermal region in the spectrum (4.2a).

The spectra of photons emitted from the bare lead target and the graphite moderated target are shown in Figure 4.2b. The majority of photon production occurs during the last stage of the evaporation process. Several sharp peaks corresponding to characteristic X-rays from the lead target are visible. This includes the $K_\alpha$ and $K_\beta$ lines at 74 and 85 keV respectively, and the L-series lines ($\sim 12$ keV). Another peak at 511 keV is from electron-positron annihilation. Even though electrons are not being transported in these simulations, all photo-electron physics is still calculated. The high energy photons $>50$ MeV mostly originate from the decay of neutral pions.
4.6. Simulations of experimental set-ups

In MCNPX, these pions decay at the place where they are created (half life of \(10^{-16}\) s) and are not transported. The presence of the graphite causes significant inelastic scattering of photons and an extra peak at 4.9 MeV corresponding to the neutron capture of $^{12}$C is present.

The spectra of leakage protons are shown in Figure 4.2c. In both cases, the energy of the protons extend from about 1 MeV to 1 GeV. The presence of the graphite does not perturb the shape of the spectrum too much but there is a reduction in the number of protons due to absorption in the graphite. Similar to the neutrons the flux of protons rapidly decreases above 800 MeV from the breakup of the deuteron. These break-up protons have a very strong forward momentum and are thus not strongly affected by the graphite.

The spectra of charged pions are shown in Figure 4.2d. MCNPX transports negative and positively charged pions together. Charged pions are much longer lived than neutral pions ($10^{-8}$ s) and so may contribute to continuation of the hadronic cascades or decay into a muon and muon neutrino.

\[ \begin{align*}
\text{Neutron Leakage} & \quad \text{Graphite Moderator} \\
\text{Photon Leakage} & \quad \text{Bare Target} \\
\text{Proton Leakage} & \\
\text{Pion Leakage} & \end{align*} \]

**FIGURE 4.2**: Neutron, photon, proton and pion leakage spectra from the bare and graphite moderated Gamma-3 lead target irradiated with 1.6 GeV deuterons. Spectra calculated using CEM03 and equal logarithmic binning with 20 bins per decade is used.
4.6.1.3 Spatial distribution of particles in Gamma-3 set-up

Figure 4.3 shows the $y - z$ cross section (cf. Figure 3.8 on page 38) of the spatial distribution of particles around the bare and graphite moderated lead target at incident deuteron energies of 1.6 GeV. The graphite homogenises the spatial distribution of neutrons within the assembly and reflects neutrons back into the target region, increasing the neutron flux in the vicinity of the target (Figures 4.3a and 4.3b).

The graphite also scatters photons and homogenises the photon flux within the graphite (Figures 4.3c and 4.3d). The protons (Figures 4.3e and 4.3f) and pions (Figures 4.3g and 4.3h) are emitted in a strong forward direction. The graphite moderator does not strongly affect the flux of protons and pions in the immediate vicinity of the target region. This implies that protons and pions are not significantly scattered but continue travelling close to their original trajectory until their energy falls below the specified cut-off energy within MCNPX (default is 0.14875 MeV).

4.6.1.4 Neutron spectra in the thorium sample locations

Five thorium samples were placed within the graphite of the Gamma-3. The locations of these samples is given in Table 3.3 on page 37. The neutron spectra at these five sample locations are shown in Figure 4.4. It can be seen that samples closer to the target (Rod A) and towards the front of the assembly (Th1) have harder neutron spectra than those in Rod B. CEM03 simulates more neutrons with energies less than 20 MeV, while INCL4-ABLA simulates more neutrons with energies greater than 20 MeV.

4.6.2 Quinta assembly

4.6.2.1 Particle spectra outside Quinta

The neutron yield as a function of incident deuteron energy for the Quinta assembly calculated with CEM03 and INCL4-ABLA is shown in Figure 4.5. As can be seen, the two models predict almost the same number of total neutrons.

The spectra of neutrons emitted from the Quinta assembly irradiated with 1 and 4 GeV deuterons are shown in Figure 4.6a. The shape of the 1 and 4 GeV spectra is the same up to 500 MeV (the energy of resulting neutrons from the breakup of the 1 GeV deuteron). The neutron spectrum from Quinta is a combined fission-spallation spectrum, while for a lead target it is a pure spallation spectrum.

FIGURE 4.3 (facing page): Mesh tally plots showing a $y - z$ cross section (cf. 3.8 on page 38) of the spatial distribution of the neutron, photon, proton and pion flux along the central target axis of the Gamma-3 assembly irradiated with 1.6 GeV deuterons. The left hand column shows the bare lead target, while the right column shows the graphite moderated target. In the case of the bare target, the space occupied by the graphite was set to void material but the geometry is still shown for reference.
4.6. Simulations of experimental set-ups

(a) Neutron Flux
(cm$^2$ deuteron$^{-1}$)

(b) Photon Flux
(cm$^2$ deuteron$^{-1}$)

(c) Proton Flux
(cm$^2$ deuteron$^{-1}$)

(d) $\pi^0$ Flux
(cm$^2$ deuteron$^{-1}$)
FIGURE 4.4: Neutron spectrum in each of the five thorium sample locations calculated using MCNPX with the INCL4-ABLA (solid line) and CEM03 (dotted line) models. Equal logarithmic binning with 20 intervals per decade is used.

FIGURE 4.5: Neutron yield of Quinta target as a function of incident deuteron energy.

Notable features in the spectra include the troughs at ~35, 88 and 150 keV corresponding to capture resonances of $^{27}$Al which is a major construction material in the Quinta assembly. A spike at ~2 GeV is from the breakup of the 4 GeV deuteron but a similar spike does not occur for the 1 GeV irradiation as the range of the deuteron is much smaller than the length of the assembly. The range of a 1 GeV deuteron in uranium and lead is 25 cm and 40 cm respectively, while for a 4 GeV deuteron it is 162 cm and 262 cm respectively.

$K_{\alpha}$ X-rays of lead (72 keV) and uranium (94 keV) can be seen in the photon spectra in Figure 4.6b. The proportion of high energy ($E_\gamma > 10$ MeV) photons is much greater at 4 GeV due to the increased production and decay of neutral pions. The proton and pion spectra are shown in Figures 4.6c and 4.6d, respectively. Similar to the neutron spectra, we see the proton flux dramatically drop off at energies above half the incident deuteron energy. However, it is less than half the deuteron energy due to ionisation losses that occur from travelling through...
4.6. Simulations of experimental set-ups

**FIGURE 4.6:** Spectra of particles (neutrons, photons, protons and charged pions) leaving the Quinta assembly irradiated with 1 and 4 GeV deuterons. Spectra calculated using INCL4-ABLA and binned logarithmically with 20 intervals per decade.

The vast majority of experimental samples were placed inside the Quinta assembly. An example of the particle spectra inside Quinta is displayed in Figure 4.7. These spectra were calculated at sample positions on plate 2, \( z = 25.7 \text{ cm} \) (cf. Fig 3.9 on page 39) at \( y = -4 \) and \( y = -6 \) cm for 1 and 4 GeV deuteron irradiation, respectively. Neutron, proton, pion, deuteron and photon spectra, as calculated using INCL4-ABLA and CEM03 are shown.

Important points to note include the significant amount of high energy deuterons for the 1 GeV sample location which is not present in the 4 GeV sample locations. This is because the 4 GeV samples were placed 2 cm lower than the 1 GeV samples, and the 1 GeV beam was much broader than the 4 GeV beam (refer to Table 3.2 on page 35). Consequently, the axial samples along the \( y = -4 \) cm line were directly hit by some of the incident beam deuterons. It
FIGURE 4.7: Neutron, proton, pion, photon and deuteron particle spectra inside the Quinta assembly calculated with MCNPX. Location of spectra is at the sample position on plate 2 indicated by the figure inset. INCL4-ABLA shown as the darker line and CEM03 shown as the lighter line. Equal logarithmic energy binning with 20 intervals per decade is used.
4.7. Calculating reaction rates

The Monte Carlo calculated reaction rates were determined by folding the particle spectrum with the corresponding reaction cross section in the following way

\[ R_{\text{calc}} = \int_{E_{\text{th}}}^{\infty} \sigma(E) \Phi(E) \, dE \]  \hspace{1cm} (4.5)

FIGURE 4.8: Neutron spectra at sample positions in the Quinta assembly. Spectra calculated using INCL4-ABLA on the six sample plates at \( y = 4\) cm and \( y = 6\) cm for the 1 and 4 GeV deuteron irradiation, respectively.

also causes the 1 GeV neutron spectrum around the target to be much harder than the 4 GeV. Pion production is also much more significant at 4 GeV compared to 1 GeV.

For the 1 and 4 GeV neutron spectra, the INCL4-ABLA model produces more neutrons with energies \( > 20 \text{ MeV} \) but less neutrons with energies \( < 20 \text{ MeV} \) compared to CEM03. Neutrons with energies \( > 20 \text{ MeV} \) arise from the high energy nuclear cascades or from dissociation of the incident beam deuteron.

Activation and fission samples were placed axially along the target axis at \( y = 4\) cm and \( y = 6\) cm for the 1 and 4 GeV deuteron irradiation, respectively. To demonstrate the change in neutron spectra along the assembly, Figure 4.8 shows the neutron spectra on each of the six sample plates at these locations.

4.6.2.3 Spatial distribution of particles within Quinta

Figure 4.9 shows the spatial distribution of neutrons, photons, protons and pions within the Quinta assembly under 1 and 4 GeV irradiation. It can be seen that protons and pions are emitted in a very strong forward direction compared to the neutrons. Neutrons are much more isotropically dispersed and homogeneously spread in the assembly. The difference in the ranges and number of secondary particles released by the different deuteron energies is also apparent.

4.7 Calculating reaction rates

The Monte Carlo calculated reaction rates were determined by folding the particle spectrum with the corresponding reaction cross section in the following way

\[ R_{\text{calc}} = \int_{E_{\text{th}}}^{\infty} \sigma(E) \Phi(E) \, dE \]  \hspace{1cm} (4.5)
CHAPTER 4. MONTE CARLO SIMULATIONS

FIGURE 4.9: Mesh tally plots showing a cross section of the $y-z$ plane (cf. 3.3 on page 31) of the spatial distribution of the neutron, photon, proton and pion flux along the central target axis of the Quinta assembly under deuteron irradiation. The left hand column shows the 1 GeV irradiation, while the right column shows the 4 GeV irradiation.
where $\sigma(E)$ is the energy dependent reaction cross section (cm$^2$), $\Phi(E)$ is the particle flux within the sample volume (cm$^{-2}$ MeV$^{-1}$ incident deuteron$^{-1}$) and $E_{th}$ is the threshold energy of the reaction.

Gamma spectrometry only reveals information about the final product yield, and not which reaction channels occurred to produce that final product. The reactions listed in Table 3.5 on page 40 have included only the neutron induced reactions, however reactions due to other particles (protons and deuterons), leading to the same final product are also possible. Therefore, reactions due to neutrons, protons and deuterons must all be calculated. Likewise, the fission track detectors only measure the total number of fission events in the foil, which could have been induced by neutrons, protons, deuterons, photons and charged pions. The total reaction rate was then found by summing across all particle types, using:

$$ R_{\text{calc}} = \sum_{i=n,p,d,\gamma,\pi} \int_{E_{th}}^{\infty} \sigma_i(E) \Phi_i(E) \, dE $$

(4.6)

Particle spectra in each of the sample locations were calculated using MCNPX e.g. Figures 4.4, 4.7 and 4.8. For reactions that have evaluated cross section data, such as $^{197}$Au(n,$\gamma$) and $^{232}$Th(n,$\gamma$), Eq. 4.5 may be solved automatically in MCNPX by combining an F4 particle flux tally with an FM tally multiplier. For cross sections calculated using the TALYS code or retrieved from the literature, these cross sections may be input into MCNPX as a user supplied dose function using the DE/DF cards. Further details of reaction cross sections used in this work are provided in sections 4.8 and 4.9.

4.8 Cross sections for activation analysis calculations

4.8.1 $^{209}$Bi(n,$xn$) reactions

The neutron induced $^{209}$Bi(n,$xn$), $x=4,5,6,7$ reaction cross sections are shown in Figure 4.10. Cross section data from the TENDL-2009 evaluation [288] was used for the $^{209}$Bi(n,4n) reaction. As no evaluated data were available for $^{209}$Bi(n,$xn$), $x=5,6,7$ reactions, these were calculated using the TALYS code and compared to experimental data from Kim et al. [289], Svoboda et al. [290] and Vrzalová et al. [291].

Good agreement is found between the TENDL-2009 and experimental data for the $^{209}$Bi(n,4n) reaction. Agreement is also seen between the TALYS calculation and experimental data for the $^{209}$Bi(n,5n) reaction. However, for the $^{209}$Bi(n,6n) and $^{209}$Bi(n,7n) reactions, the results of the TALYS calculations are higher than the experimentally measured data. This is most prominent where the cross section is maximum. For the $^{209}$Bi(n,6n) reaction, reasonable agreement is reached for neutron energies less than 50 MeV and greater than 70 MeV, however at the peak energy of 55 MeV the ratio $\frac{\sigma_{\text{TALYS}}}{\sigma_{\text{Kim}}}$ is approximately 1.5. For the $^{209}$Bi(n,7n) cross section, the ratio $\frac{\sigma_{\text{TALYS}}}{\sigma_{\text{Kim}}}$ at peak energy 66 MeV is approximately 1.6.
Due to the inconsistency and lack of continuity in the experimental data, TENDL-2009 evaluation and TALYS calculated cross sections were used for the reaction rate calculations. However, the discrepancy between the experimentally measured and TALYS calculated data is noted, and was the major contributor to the overall uncertainty in the final results, especially for the $^{209}$Bi(n,6n) and $^{209}$Bi(n,7n) reactions. The cross sections have a long, high energy tail and remain approximately constant above 100 MeV. As no experimental or calculated data exists beyond 250 MeV (the highest TALYS calculations can go), the cross sections were assumed to remain constant above this range.

Experimental cross section data for proton induced $^{209}$Bi(p,x) reactions are available up to 2.6 GeV which covers the entire range of proton energies expected in the sample locations of the Quinta assembly [292–295] (Figure 4.11). Sotnikov et al. [296] have measured $^{209}$Bi(d,x) cross sections at deuteron energies of 1.6 and 4 GeV (Table 4.1). Most deuteron induced reactions occur directly from the incident beam (1 and 4 GeV). On the basis that the (p,x) cross sections become approximately linear at GeV energies, the data of Sotnikov et al. [296] was linearly extrapolated to cover this range.
4.8. Cross sections for activation analysis calculations

FIGURE 4.11: Reaction cross section data for $^{209}$Bi(p,x)$^{206}$Bi, $^{209}$Bi(p,x)$^{205}$Bi, $^{209}$Bi(p,x)$^{204}$Bi and $^{209}$Bi(p,x)$^{203}$Bi reactions. Data shown includes experimental cross sections obtained from Bell and Skarsgard [292], Kuhnhenn et al. [293], Michel et al. [294] and Titarenko et al. [295].

TABLE 4.1: Cross sections for $^{209}$Bi(d,X)Y reactions for 1.6 and 4 GeV deuterons [296]. Extrapolated data for 1 GeV deuterons also shown.

<table>
<thead>
<tr>
<th>Y</th>
<th>Cross section (mb)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 GeV *</td>
</tr>
<tr>
<td>$^{206}$Bi</td>
<td>84 ± 9</td>
</tr>
<tr>
<td>$^{205}$Bi</td>
<td>81 ± 10</td>
</tr>
<tr>
<td>$^{204}$Bi</td>
<td>52 ± 6</td>
</tr>
<tr>
<td>$^{203}$Bi</td>
<td>41 ± 5</td>
</tr>
</tbody>
</table>

* Extrapolated value
4.8.2 $^{197}$Au(n,xn) and $^{197}$Au(n,$\gamma$) reactions

The neutron induced $^{197}$Au(n,$\gamma$) and (n,xn), $x=2,4,5,6,7$ reaction cross sections can be seen in Figure 4.12. Evaluated data from ENDF/B-VII is available for the $^{197}$Au(n,$\gamma$) reaction up to 30 MeV. This is less than the maximum neutron energy present in the Quinta sample locations, which extends all the way up to about half that of the incident deuteron energy. The $^{197}$Au(n,$\gamma$) reaction rate is dominated at thermal and in the resonance region for neutrons of energies less than 0.01 MeV. The cross section at 0.01 MeV is more than five orders of magnitude higher than at 30 MeV and the number of neutrons above 30 MeV in the sample positions is also at most 4% of the total neutrons. Therefore, the lack of evaluated data at higher energies is not expected to be a significant disadvantage.

TALYS calculated data were used for the $^{197}$Au(n,2n) and (n,4n) reactions as it showed the best agreement between the experimental data [290, 291, 297–299], compared to the ENDF/B-VII and TENDL-2009 data. There is no evaluated data for the $^{197}$Au(n,xn), $x=5,6,7$ reactions and only very limited experimental sources of data exist. Svoboda et al. [290] and Vrzalová et al. [291] have produced a few scattered data points, which in isolation is not particularly helpful but may be used to compare to the TALYS calculations. As with the $^{209}$Bi(n,xn) cross sections, the $^{197}$Au(n,xn) cross sections were assumed to remain constant above 250 MeV.

Experimental proton cross section data were available for $^{197}$Au(p,x)$^{196}$Au and $^{197}$Au(p,x)$^{194}$Au reactions only [300–302] (Figure 4.13). No deuteron-induced reaction cross section data for reactions of interest could be found in the EXFOR database or literature. TALYS was not used to calculate proton and deuteron induced cross sections because in our calculations, there were large unexplained discrepancies with the experimentally measured cross sections.

Due to the lack of cross section data for $^{197}$Au(p,x)$^{193–191}$Au and $^{197}$Au(d,x) reactions, contributions due to proton and deuteron-induced reactions had to be estimated. It was assumed that the percentage of proton and deuteron-induced reactions for the $^{197}$Au samples were the same as for the $^{209}$Bi samples. This was done on the basis that $^{209}$Bi(n,xn) and $^{197}$Au(n,xn) have closely resembling cross sections (see Figure 4.14) and are both sub-actinides with odd-Z, even-N nuclei. Further details are described in section 6.1.2 on page 119.

4.8.3 $^{232}$Th(n,$\gamma$) reaction

The $^{232}$Th(n,$\gamma$) cross section, shown in Figure 4.15 is available up to 60 MeV from the ENDF/B-VII library. Neutron energies in the Gamma-3 assembly extend from thermal all the way to GeV energies. In a similar case to the $^{197}$Au(n,$\gamma$) reaction, the lack of cross section data at higher energies is not considered a disadvantage. This is because the $^{232}$Th(n,$\gamma$) cross section drops nearly 5 orders of magnitude from 1 MeV to 60 MeV, and the proportion of neutrons in the sample positions with energies greater than 60 MeV is no more than 2% of the total number of neutrons.
4.8. Cross sections for activation analysis calculations

**FIGURE 4.12**: Reaction cross section data for $^{197}$Au($n, \gamma$) and $^{197}$Au($n,xn$) reactions. Evaluated data is from the ENDF/B-VII.1 and TENDL-2009 libraries and experimental data is from Svoboda et al. [290], Vrazilová et al. [291], Tewes et al. [297], Uwamino et al. [298] and Philis and Bersillon [299]. Cross sections calculated with TALYS are also shown.
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FIGURE 4.13: Available proton induced reaction cross section data for $^{197}$Au(p,x) reactions. TENDL-2009 data contains the sum of $^{197}$Au(p,d)$^{196}$Au and $^{197}$Au(p,np)$^{196}$Au cross sections. Experimental data obtained from Kavanagh and Bell [300], Michel et al. [301] and Szelecsenyi et al. [302].

FIGURE 4.14: (n,xn) reaction cross for $^{209}$Bi and $^{197}$Au. The (n,4n) data is from the TENDL-2009 evaluation, while the rest is calculated using the TALYS code.

4.9 Fission Cross sections

Fission of materials in the Quinta assembly is not exclusively caused by secondary neutrons but also secondary protons, photons and charged pions. For samples located close to the beam, fission induced by primary deuterons is also possible. Therefore, to accurately calculate the total fission rate we must consider reactions due to neutrons, protons, photons, deuterons and charged pions. Energy-dependent fission cross sections were collated from the literature or in the case of pion-induced fission, calculated using the XSEX3 code (in MCNPX). In cases where sufficient data wasn’t available, it was necessary to extrapolate or scale existing data to make reasonable approximations.
4.9. Fission Cross sections

4.9.1 Neutron-induced fission cross sections

The neutron-induced fission cross sections for $^{nat}$Pb, $^{209}$Bi, $^{197}$Au, $^{232}$Th and $^{nat}$U is presented in Figure 4.16. Evaluated data for $^{nat}$Pb(n,f) and $^{209}$Bi(n,f) reactions exist in the JENDL/HE-2007 library up to $E_n = 3$ GeV. However, it appears inconsistent with the experimental data [303] and so was not used.

4.9.1.1 Sub-actinide fission cross sections

Smirnov et al. [176] have developed an empirical parametrisation formula for neutron-induced fission which takes the form

$$\sigma_{nf}(E_n) = P_1 \exp \left[ - \left( \frac{P_2}{E_n} \right)^{P_3} \right]$$

where $\sigma_{nf}$ is the neutron induced fission cross in mb, $E_n$ is the incident neutron energy in MeV, and $P_1$, $P_2$, $P_3$ are fitting parameters which are dependent on the target nuclide. The parametrisation formula by Smirnov et al. [176] was based on experimental data available at the time, which extended up to only 180 MeV. More recently, experimental data up to 1 GeV for $^{nat}$Pb(n,f) and $^{209}$Bi(n,f) reactions have been made available by the n-TOF collaboration [230, 231, 303] and refit to Eq. 4.7. The data of Tarrio et al. extends up to 1 GeV, while neutron energies in Quinta may reach few GeV. It was deemed unrealistic to extrapolate Eq. 4.7 beyond 1 GeV as the last few experimental data points of Tarrio et al. [303] are seen to taper off at around 800 MeV. The cross section was therefore assumed to remain constant above 800 MeV.

Only a single experimental data point for the $^{197}$Au(n,f) cross section exists above 180 MeV (Figure 4.16c). This data point at 350 MeV [307] agrees well with the extrapolation of the Smirnov et al. parametrisation. However, as seen from the $^{209}$Bi(n,f) and $^{nat}$Pb(n,f) cross sections (Figure 4.16a and 4.16b), the data of Goldanskiy and Tarumo [307] and the parametrisation of Smirnov
FIGURE 4.16: Neutron-induced fission cross sections for a) natPb b) 209Bi c) 197Au d) 232Th e) low-energy natU and f) high-energy natU. Evaluated data is from the ENDF/B-VII and JENDL/HE-2007 libraries, parametrised data from Smirnov et al. [176] and Tarrio et al. [303] and experimental data from Shcherbakov et al. [304], Paradela et al. [305] and Vorotnikov and Larionov [306]. The data points of Tarrio et al. (2011) in subfigures e and f is the average ratio of 235U between natPb and 209Bi multiplied by parametrised data of Tarrio et al. shown in subfigures a and b, respectively.
4.9. Fission Cross sections

TABLE 4.2: Fitting parameters for $^{nat}\text{Pb}$, $^{209}\text{Bi}$ and $^{197}\text{Au}$ (n,f) cross section parametrisations (Eq. 4.7). Parameters for $^{197}\text{Au}$ in Tarrio et al. [303] column are estimates from this work. See text for details.

<table>
<thead>
<tr>
<th>Target</th>
<th>Parameter</th>
<th>Smirnov et al.</th>
<th>Tarrio et al.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{nat}\text{Pb}$</td>
<td>$P_1$</td>
<td>70.4</td>
<td>198.9</td>
</tr>
<tr>
<td></td>
<td>$P_2$</td>
<td>171.9</td>
<td>379.2</td>
</tr>
<tr>
<td></td>
<td>$P_3$</td>
<td>1.27</td>
<td>0.839</td>
</tr>
<tr>
<td>$^{209}\text{Bi}$</td>
<td>$P_1$</td>
<td>109.2</td>
<td>250</td>
</tr>
<tr>
<td></td>
<td>$P_2$</td>
<td>131.5</td>
<td>259</td>
</tr>
<tr>
<td></td>
<td>$P_3$</td>
<td>1.32</td>
<td>0.895</td>
</tr>
<tr>
<td>$^{197}\text{Au}$</td>
<td>$P_1$</td>
<td>35</td>
<td>$89 \pm 9$</td>
</tr>
<tr>
<td></td>
<td>$P_2$</td>
<td>207.2</td>
<td>$432 \pm 24$</td>
</tr>
<tr>
<td></td>
<td>$P_3$</td>
<td>1.18</td>
<td>$0.79 \pm 0.01$</td>
</tr>
</tbody>
</table>

et al. [176] deviates significantly from the experimental data of Tarrio et al. [303] above 200 MeV. Therefore, an estimation of the $^{197}\text{Au}$ (n,f) cross section from 0.2–1 GeV was required. The ratio of $\sigma^{Au}_{f}/\sigma^{Pb}_{f}$ and $\sigma^{Au}_{f}/\sigma^{Bi}_{f}$ from Smirnov et al. [176] parametrisation was used as a basis to estimate $\sigma^{Au}_{f}/\sigma^{Pb}_{f}$ and $\sigma^{Au}_{f}/\sigma^{Bi}_{f}$ for Tarrio et al. [303] parametrisation, in the following way

$$P^T_{Au} = \frac{1}{2} \sum_{i=Pb,Bi} \frac{P^T_{i}}{P^S_{i}} P^S_{Au}$$  \hspace{1cm} (4.8)

where, $P$ refers to the fitting parameters $P_1$–$P_3$, the subscripts refer to the target nuclide (Au, Pb or Bi) and superscripts refer to parametrisations of Tarrio et al. ($T$) and Smirnov et al. ($S$). The estimates of the fitting parameters ($P_1, P_2, P_3$) adjusted to Tarrio et al. parametrisation for the $^{197}\text{Au}$ (n,f) cross section are shown in Table 4.2 and Figure 4.16c. The validity of this approach will only be confirmed after high energy (up to 1 GeV) experimental cross section data for $^{197}\text{Au}$ (n,f) is measured.

4.9.1.2 Actinide fission cross sections

Evaluated ENDF/B-VII cross section data for $^{232}\text{Th}$ (n,f) is available up to 60 MeV and $^{nat}\text{U}$ (n,f) up to 30 and 20 MeV for $^{238}\text{U}$ and $^{235}\text{U}$, respectively. JENDL/HE-2007 cross section data exists up to 3 GeV for $^{nat}\text{U}$ (n,f) reaction but was not immediately relied upon because of the inconsistencies with experimental data for the $^{nat}\text{Pb}$ (n,f) and $^{209}\text{Bi}$ (n,f) reactions [303].

Experimental data up to 200 MeV for $^{232}\text{Th}$ (n,f) has been measured by Shcherbakov et al. [304] and agrees extremely well with the ENDF/B-VII data below 60 MeV. Above 200 MeV, the only experimental data involves highly scattered data points [305]. Therefore, the region 200–1000 MeV was least-squares fitted with a 2nd order polynomial and normalised to the 200 MeV data point of Shcherbakov et al. as described in Borger et al. [139]. Above 1 GeV, the cross section was assumed to remain constant, maintaining the end value of the fitted polynomial.
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Shcherbakov et al. [304] also provide $^{\text{nat}}\text{U}(n,f)$ cross section data up to 200 MeV. Above this range, there are no absolute cross section measurements. Tarrio et al. [303] have measured the $^{\text{nat}}\text{U}(n,f)$ cross section ratio with $^{\text{nat}}\text{Pb}$ ($\sigma_{\text{Pb}}^{nf}/\sigma_{\text{U}}^{nf}$) and $^{209}\text{Bi}$ ($\sigma_{\text{Bi}}^{nf}/\sigma_{\text{U}}^{nf}$) up to 1 GeV. Figure 4.16e and f shows the average of these two ratios multiplied by the parametrised cross sections of Tarrio et al. [303]. It can be seen that the uncertainty is large, but in agreement with JENDL/HE-2007. ENDF/B-VII was used below 20 MeV, and JENDL/HE data used above 20 MeV.

4.9.2 Proton-induced fission cross sections

The proton-induced fission cross sections for $^{\text{nat}}\text{Pb}$, $^{209}\text{Bi}$, $^{197}\text{Au}$, $^{232}\text{Th}$ and $^{\text{nat}}\text{U}$ are presented in Figure 4.17. These cross sections were all based on the parametrisations of Prokofiev [308] who collated a large amount of data from the EXFOR database and fitted it with a modified version of the Fukahori and Pearlstein [309] equation:

$$\sigma_{pf} = P_1 \{1 - \exp[-P_3(E_p - P_2)]\} \times (1 - P_4 \ln E_p)$$  \hspace{1cm} (4.9)

where, $\sigma_{pf}$ is the proton induced fission cross section in mb, $E_p$ is the incident proton energy in MeV, and $P_1$, $P_2$, $P_3$, $P_4$ are fitting parameters. The parameter $P_4$ was introduced by Prokofiev to account for the apparent decrease in cross section at high energies, particularly for the actinide nuclei. The parametrisation formula is deemed valid from 70–30 000 MeV for the sub-actinides ($^{\text{nat}}\text{Pb}$, $^{209}\text{Bi}$, $^{197}\text{Au}$) and 20–30 000 MeV for the actinides ($^{232}\text{Th}$, $^{\text{nat}}\text{U}$).

In the case of the sub-actinides, the parametrisations were refit to accommodate the experimental data of Flerov et al. [312], Yuvevich et al. [315], Smirnov [316], Kotov et al. [317] and Vaishnene et al. [318]. Updated fitting parameters are shown in Table 4.3.

The parametrisation as calculated by Prokofiev [308] was used for $^{232}\text{Th}(p,f)$ and $^{\text{nat}}\text{U}(p,f)$. As the parametrisation is only valid for proton energies above 20 MeV, the experimental data of Kudo et al. [319] and Calboreanu et al. [320] were used for energies below this.

4.9.3 Photofission cross sections

Photofission cross sections for $^{\text{nat}}\text{Pb}$, $^{209}\text{Bi}$, $^{197}\text{Au}$, $^{232}\text{Th}$ and $^{\text{nat}}\text{U}$ are shown in Figure 4.18. The $\Delta$-resonance of the sub-actinides may be fitted to a modified Breit-Wigner formula:

$$\sigma_{\gamma f} = a + \frac{bE^c}{(E - d)^2 + e}$$  \hspace{1cm} (4.10)

where, $\sigma_{\gamma f}$ is the photofission cross section in mb, $E$ is the photon energy in MeV, and $a$, $b$, $c$, $d$ and $e$ are fitting parameters [321].

This formula was fitted to the $^{\text{nat}}\text{Pb}(\gamma,f)$ cross section and the resulting parameters shown in Table 4.4. This fit was then scaled to the data of the $^{209}\text{Bi}$ and $^{197}\text{Au}$ cross sections (Terranova et al. [322] and Lucherini et al. [323]), by normalising fitting parameter $b$. Photon energies in Quinta extend up to a maximum of about 300 MeV (see Figure 4.7), therefore any variations in
FIGURE 4.17: Proton-induced fission cross sections for a) $^{209}$Pb b) $^{209}$Bi c) $^{197}$Au d) $^{232}$Th and $^{nat}$U. Evaluated data is from JENDL/HE-2007 and parametrised data from Prokofiev [308]. Experimental data obtained from Konshin et al. [310], Brandt et al. [311], Flerov et al. [312], Hudis and Katcoff [313], Khan and Khan [314], Yurevich et al. [315], Smirnov [316], Kotov et al. [317], Vaishnene et al. [318], Kudo et al. [319] and Calboreanu et al. [320]. Figure does not show all experimental data used for Prokofiev parametrisation.
TABLE 4.3: Fitting parameters of $^{nat}$Pb, $^{209}$Bi and $^{197}$Au \(\text{(p,f)}\) parametrisations (Eq. 4.9) as determined by Prokofiev [308] and refit to the latest experimental data.

<table>
<thead>
<tr>
<th>Target</th>
<th>Parameter</th>
<th>Prokofiev</th>
<th>Refit (This work)</th>
</tr>
</thead>
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<tr>
<td>$^{nat}$Pb</td>
<td>P1</td>
<td>156</td>
<td>214 ± 38</td>
</tr>
<tr>
<td></td>
<td>P2</td>
<td>51.0</td>
<td>44 ± 5</td>
</tr>
<tr>
<td></td>
<td>P3</td>
<td>0.00725</td>
<td>0.0030 ± 0.0004</td>
</tr>
<tr>
<td></td>
<td>P4</td>
<td>0.0206</td>
<td>0.040 ± 0.015</td>
</tr>
<tr>
<td>$^{209}$Bi</td>
<td>P1</td>
<td>262</td>
<td>421 ± 24</td>
</tr>
<tr>
<td></td>
<td>P2</td>
<td>41.9</td>
<td>42.1 ± 0.5</td>
</tr>
<tr>
<td></td>
<td>P3</td>
<td>0.0103</td>
<td>0.0059 ± 0.0004</td>
</tr>
<tr>
<td></td>
<td>P4</td>
<td>0.046</td>
<td>0.065 ± 0.004</td>
</tr>
<tr>
<td>$^{197}$Au</td>
<td>P1</td>
<td>76.8</td>
<td>91.6 ± 3.7</td>
</tr>
<tr>
<td></td>
<td>P2</td>
<td>50.9</td>
<td>47 ± 2</td>
</tr>
<tr>
<td></td>
<td>P3</td>
<td>0.00431</td>
<td>0.0029 ± 0.0004</td>
</tr>
<tr>
<td></td>
<td>P4</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

TABLE 4.4: Photofission fitting parameters for Eq. 4.10. $^{nat}$Pb was fitted to data of Cetina et al. [321], while $^{209}$Bi and $^{197}$Au were scaled to the data of Terranova et al. [322] and Lucherini et al. [323], respectively.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$^{nat}$Pb</th>
<th>$^{209}$Bi</th>
<th>$^{197}$Au</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$b$</td>
<td>$0.54 \pm 0.13$</td>
<td>$0.99 \pm 0.36$</td>
<td>$0.18 \pm 0.05$</td>
</tr>
<tr>
<td>$c$</td>
<td>$2.11 \pm 0.03$</td>
<td>$2.11 \pm 0.03$</td>
<td>$2.11 \pm 0.03$</td>
</tr>
<tr>
<td>$d$</td>
<td>$290 \pm 4$</td>
<td>$290 \pm 4$</td>
<td>$290 \pm 4$</td>
</tr>
<tr>
<td>$e$</td>
<td>$(2.28 \pm 0.14) \times 10^4$</td>
<td>$(2.28 \pm 0.14) \times 10^4$</td>
<td>$(2.28 \pm 0.14) \times 10^4$</td>
</tr>
</tbody>
</table>

the position and shape of the $\Delta$-resonance or inaccuracy in the photofission cross section above 300 MeV will not introduce additional error in this work. Furthermore, the photofission cross section is considerably lower than the neutron or proton-induced fission cross section so we do not expect the amount of photofission in Quinta to be significant.

Photofission data for $^{232}$Th and $^{nat}$U is available up to 20 MeV from ENDF/B-VII and up to 3.8 GeV from experimental data [321, 324–326, 328, 330]. Features of the photoisotopic cross section includes the $\Delta$-resonance (above 140 MeV), the pygmy resonance (5–9 MeV), the giant dipole resonance (10–30 MeV) and the “quasi-deuteron” region (30–140 MeV). In the “quasi-deuteron” region, the photon interacts with a neutron-proton pair in the nucleus, rather than the nucleus as a whole.

4.9.4 Deuteron-induced fission cross sections

The deuteron-induced fission cross sections for $^{nat}$Pb, $^{209}$Bi, $^{197}$Au and $^{232}$Th are presented in Figure 4.19. The $^{nat}$U samples were placed further away from the target axis where the deuteron
4.9. Fission Cross sections

**Figure 4.18:** Photo-fission cross sections for a) $^{nat}$Pb, $^{209}$Bi, and $^{197}$Au b) $^{232}$Th and c) $^{nat}$U. Experimental data obtained from Cetina et al. [321], Terranova et al. [322], Lucherini et al. [323], Bowman et al. [324], Caldwell et al. [325], Leprêtre et al. [326], Martins et al. [327], Frommhold et al. [328], Terranova et al. [329] and Sanabria et al. [330].
flux is expected to be negligible. Unfortunately there is very limited experimental data and the data that is available has large uncertainties. Therefore, the parametrised proton-induced fission cross sections were normalised to the experimental data of the deuteron. The proton-deuteron fission cross section ratio is shown in Table 4.5, and it is apparent that the uncertainties are quite large (16–36%).

4.9.5 Pion-induced fission cross sections

The available experimental data of pion-induced fission is either highly scattered (such as the case of $^{209}$Bi and $^{197}$Au) or inadequate (case of $^{nat}$Pb and $^{232}$Th), as shown in Figure 4.20. Pion-induced fission cross sections were also calculated using the XSEX3 code (part of MCNPX). XSEX3 processes the history tape (HISTP) files produced by LAHET to generate double-differential cross section data. For calculating cross sections, only the initial nuclear interaction of the primary source particle is simulated – elastic scattering, particle transport of secondary
4.10 Calculating CR-39 track density

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\sigma_f \ (d/p)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$Th</td>
<td>$1.35 \pm 0.15$</td>
</tr>
<tr>
<td>$^{209}$Bi</td>
<td>$1.25 \pm 0.40$</td>
</tr>
<tr>
<td>nat Pb</td>
<td>$1.22 \pm 0.20$</td>
</tr>
<tr>
<td>$^{197}$Au</td>
<td>$1.4 \pm 0.5$</td>
</tr>
</tbody>
</table>

TABLE 4.5: Ratio of deuteron to proton-induced fission cross sections.

particles and slowing down are all switched off. Only CEM03 and Bertini allow pions to be primary source particles, and as Bertini has a suggested minimum energy of 20 MeV compared to 100 MeV for CEM03 [213], the Bertini model was used.

MCNPX does not discriminate between $\pi^+$ and $\pi^-$, and transports them together. This makes it more difficult to directly compare with the available experimental data, where $\pi^+$ and $\pi^-$ fission are measured separately. Peterson [338] has measured both $\pi^+$ and $\pi^-$ fission cross sections for $^{209}$Bi and $^{197}$Au and it can be seen that the calculated XSEX3 results lie in between the values for $\pi^+$ and $\pi^-$-induced fission. In absence of reliable ($\pi,f$) cross section we used the results obtained via the XSEX3 code.

4.10 Calculating CR-39 track density

To calculate the expected track density in each of the CR-39 detectors located on the surface of the Gamma-3, the neutron spectra in all sample locations were determined from MCNPX simulation. As an example, the neutron spectra in the positions of A and B (Figure 4.21) is shown in Figure 4.22.

The simulated neutron spectra were then fitted to Eq. 3.28, ie:

$$\Phi(E) = \phi_{th} \frac{E}{(kT)^2} \exp\left(\frac{-E}{kT}\right) + \phi_{epi} \frac{\Delta(E/kT)}{E^{1+\alpha}}$$

in the region $< 1$ keV [129] with $\phi_{th}$, $\phi_{epi}$, $T$ and $\alpha$ as fitting parameters and utilising the joining function, $\Delta(E/kT)$ from Coates [194]. The spectra were fitted in Python using the Levenberg-Marquadt least-squares fitting algorithm as described in section 3.6.4.2 on page 69. The values of $\phi_{th}$, $\phi_{epi}$, $T$ and $\alpha$ in the positions of A and B are shown in Table 4.6 for reference.

The Monte Carlo track density was then calculated using Eq. 3.31, ie:

$$\rho = w_{th}\phi_{th} + w_{epi}\phi_{epi}$$

where $\phi_{th}$ and $\phi_{epi}$ were found from the spectrum fitting and $w_{th}$ and $w_{epi}$ were found to be $(1.11 \pm 0.05) \times 10^{-3}$ and $(6.5 \pm 1.3) \times 10^{-4}$ tracks neutron$^{-1}$, respectively from the calibration procedure described in section 3.6.4 on page 67.
CHAPTER 4. MONTE CARLO SIMULATIONS

FIGURE 4.20: Pion-induced fission cross sections for a) $^{nat}$Pb b) $^{209}$Bi c) $^{197}$Au and d) $^{232}$Th. Data calculated with the XSEX3 code and compared to experimental data [336–339].

FIGURE 4.21: Locations of CR-39/LR-115 2B track detectors placed on the Gamma-3 assembly. The two samples labelled A and B correspond to the positions where the spectra shown in Figure 4.22 were calculated.
4.10. Calculating CR-39 track density

FIGURE 4.22: Neutron spectra in two sample locations – A and B (Figure 4.21) on the Gamma-3 assembly. The fitting parameters are listed in Table 4.6.

TABLE 4.6: Fitting parameters for the neutron spectra shown in Figure 4.22

<table>
<thead>
<tr>
<th></th>
<th>Sample A</th>
<th>Sample B</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\phi_{th}$ (cm$^{-2}$ deuteron$^{-1}$)</td>
<td>$(4.57 \pm 0.08) \times 10^{-4}$</td>
<td>$(1.06 \pm 0.02) \times 10^{-3}$</td>
</tr>
<tr>
<td>$\phi_{epi}$ (cm$^{-2}$ deuteron$^{-1}$)</td>
<td>$(1.18 \pm 0.04) \times 10^{-5}$</td>
<td>$(7.66 \pm 0.25) \times 10^{-4}$</td>
</tr>
<tr>
<td>$T$ (K)</td>
<td>346 ± 5</td>
<td>372 ± 5</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>0.046 ± 0.003</td>
<td>−0.140 ± 0.003</td>
</tr>
</tbody>
</table>
Chapter 5

Results I: Gamma-3 assembly

5.1 $^{232}\text{Th}(n,\gamma)$ and $^{232}\text{Th}(n,f)$ reaction rates

The experimental and Monte Carlo calculated neutron capture rate of $^{232}\text{Th}$ in the Gamma-3 set-up is shown in Figure 5.1 and Table 5.1. This shows the spatial distribution of the $^{232}\text{Th}(n,\gamma)$ reaction rate (per thorium atom per incident deuteron) along the entire $z$-axis at the two nominated locations, A and B which were situated 15.8 and 22.9 cm radially from the target axis. The reaction rate is minimum at the front and back end of the target, rising to a maximum at 24–25 cm along the target. The reaction rate is also $\sim$30\% higher along rod A due to the fact the neutron flux decreases with increasing distance from the target. Within the experimental uncertainties, there is good agreement between the experimental and both Monte Carlo results of INCL4-ABLA and CEM03, but better agreement with INCL4-ABLA is seen.

The experimentally measured and Monte Carlo calculated $^{232}\text{Th}$ fission rate is presented in Figure 5.2 and Table 5.1. Similar to the neutron capture rate, the fission rate is lowest towards the front and back of the target, with the maximum INCL4-ABLA fission rate occurring $\sim$16 cm and $\sim$18 cm along rod A and B, respectively. Meanwhile, CEM03 calculates the maximum fission rate to occur at $\sim$18 cm and $\sim$22 cm. Averaged over the entire length of the rod, the fission rate along rod A is higher by a factor of about two as compared with rod B. Good agreement between the experimental and Monte Carlo results can once again be seen. However, better agreement is achieved by using the CEM03 model.

The $^{232}\text{Th}(n,\gamma)$ reaction rate shows a more centralised peak compared to $^{232}\text{Th}(n,f)$ reaction rate, which peaks closer to the front of the assembly. The $^{232}\text{Th}(n,\gamma)$ reaction is most sensitive to thermal and resonance neutrons, while $^{232}\text{Th}(n,f)$ is only sensitive to fast neutrons $>1$ MeV. Thermal neutrons become centralised due to many scatterings and reflections in the graphite moderator. The relative decrease in reaction rate from rod B to rod A is also much less for the $(n,\gamma)$ compared to the $(n,f)$ reaction due to the fast neutrons becoming increasingly thermalised away from the target.
FIGURE 5.1: The spatial distribution of the $^{232}$Th(n,γ) reaction rate in the Gamma-3 assembly along rods A and B. Results normalised to per thorium atom per incident deuteron. Experimental data is shown as points and the Monte Carlo calculations shown as solid lines. Uncertainties in the Monte Carlo simulations are signified by the shaded area and the width of the experimental data points corresponds to the size of the X-error bars.

FIGURE 5.2: The spatial distribution of the $^{232}$Th(n,f) reaction rate in the Gamma-3 assembly along rods A and B. Results normalised to per thorium atom per incident deuteron. Experimental data is shown as points and the Monte Carlo calculations shown as solid lines. Uncertainties in the Monte Carlo simulations are signified by the shaded area and the width of the experimental data points corresponds to the size of the X-error bars.
5.1. $^{232}\text{Th}(n,\gamma)$ and $^{232}\text{Th}(n,f)$ reaction rates

![Graph showing $^{232}\text{Th}(n,\gamma)$ and $^{232}\text{Th}(n,f)$ reaction rates for rods A and B.](image)

**FIGURE 5.3:** The spatial distribution of the neutron capture-to-fission ratio ($\alpha$) for $^{232}\text{Th}$ along rods A and B. Experimental data is shown as points and the Monte Carlo calculations shown as solid lines. Uncertainties in the Monte Carlo simulations are signified by the shaded area and the width of the experimental data points corresponds to the size of the X-error bars.

**TABLE 5.1:** Experimental and Monte Carlo calculated ($n,\gamma$), ($n,f$) and capture-to-fission ratios ($\alpha$) in the five thorium sample locations within the Gamma-3 set-up. ($n,\gamma$) and ($n,f$) reaction rates are normalised per thorium atom per incident deuteron and $\alpha$ is dimensionless.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Exp. ($n,\gamma$) $10^{-25}$</th>
<th>MC ($n,\gamma$) $10^{-25}$</th>
<th>Exp. ($n,f$) $10^{-28}$</th>
<th>MC ($n,f$) $10^{-28}$</th>
<th>$\alpha \left( \frac{n,\gamma}{n,f} \right)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th1</td>
<td>1.10±0.14</td>
<td>1.17±0.04</td>
<td>10.1±1.3</td>
<td>11.8±0.39</td>
<td>109±17</td>
</tr>
<tr>
<td>Th2</td>
<td>1.76±0.23</td>
<td>1.90±0.06</td>
<td>10.7±1.4</td>
<td>11.8±0.43</td>
<td>165±26</td>
</tr>
<tr>
<td>Th3</td>
<td>0.95±0.12</td>
<td>1.00±0.03</td>
<td>5.88±0.81</td>
<td>5.46±0.22</td>
<td>162±26</td>
</tr>
<tr>
<td>Th4</td>
<td>1.43±0.19</td>
<td>1.42±0.04</td>
<td>5.11±0.71</td>
<td>5.53±0.21</td>
<td>279±46</td>
</tr>
<tr>
<td>Th5</td>
<td>0.66±0.09</td>
<td>0.70±0.02</td>
<td>3.32±0.49</td>
<td>3.15±0.13</td>
<td>198±35</td>
</tr>
</tbody>
</table>

The neutron capture-to-fission ratio ($\alpha$) for $^{232}\text{Th}$ within the Gamma-3 assembly is shown in Figure 5.3 and Table 5.1. $\alpha$ is useful for determining where fissile $^{233}\text{U}$ is produced at the highest rate and as an approximate measure of the slow-to-fast neutron flux ratio. Neutron capture is strongly favoured over fission in all locations within the Gamma-3 assembly due to the highly thermalised neutron spectrum. Fission reactions are most probable towards the front and back of the assembly where the neutron spectrum is harder.

$\alpha$ is on average ~50% higher in rod B compared to rod A and it peaks at ~43 cm and ~40 cm along rod A and B, respectively. Production of $^{233}\text{U}$ is thus more likely to occur further from the target and towards the centre of the assembly where the proportion of slow neutrons is highest. Overall, the experimental results achieve good agreement with those calculated using MCNPX.

As described in section 4.9.1.2 on page 101, calculation of the $^{232}\text{Th}$ fission rate occurred in two stages – reactions induced by neutrons below 60 MeV and those above 60 MeV. 60 MeV is the upper energy limit of the $^{232}\text{Th}(n,\gamma)$ reaction in the ENDF/B-VII data library. Table 5.2
CHAPTER 5. RESULTS I: GAMMA-3 ASSEMBLY

TABLE 5.2: Monte Carlo calculated $^{232}$Th fission rate, $R_f^{MC} (10^{-28} \text{ atom}^{-1} \text{ deuteron}^{-1})$ induced by neutrons below 60 MeV and above 60 MeV. z-pos indicates the distance along target. Note that the uncertainties specified include only the statistical uncertainty from the simulation and experimental uncertainty in the cross section data. It does not include the systematic uncertainties arising from the beam shape or sample locations.

<table>
<thead>
<tr>
<th>Rod</th>
<th>Sample</th>
<th>z-pos (cm)</th>
<th>$E_n \leq 60 \text{ MeV}$</th>
<th>$E_n &gt; 60 \text{ MeV}$</th>
<th>$% (E_n &gt; 60 \text{ MeV})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Th1</td>
<td>8.3</td>
<td>9.24±0.04</td>
<td>2.52±0.13</td>
<td>22±1</td>
</tr>
<tr>
<td></td>
<td>Th2</td>
<td>25.5</td>
<td>7.90±0.03</td>
<td>3.89±0.20</td>
<td>33±2</td>
</tr>
<tr>
<td></td>
<td>Th3</td>
<td>47.0</td>
<td>3.00±0.02</td>
<td>2.46±0.13</td>
<td>45±3</td>
</tr>
<tr>
<td>B</td>
<td>Th4</td>
<td>27.1</td>
<td>3.61±0.02</td>
<td>1.92±0.10</td>
<td>35±2</td>
</tr>
<tr>
<td></td>
<td>Th5</td>
<td>48.4</td>
<td>1.69±0.01</td>
<td>1.46±0.08</td>
<td>46±3</td>
</tr>
</tbody>
</table>

shows the relative contribution of the two neutron energy groups to the overall fission rate. It can be seen that the contribution from neutrons above 60 MeV is sizeable and increasing with distance along the target (z-axis). The significant contribution from neutrons above 60 MeV highlights the importance of the availability of accurate high energy (up to a few GeV) cross section data for future ADS development. The reliability of the simulated results remain limited by the availability of accurate cross section data.

These experimental results do not provide enough evidence to conclusively decide which spallation model is best suited to simulation of a thermal ADS. INCL4-ABLA provides better agreement for the thermal/resonance region, while CEM03 agrees best for the fast region of the neutron spectrum. However, it should be noted that all data points do agree within the measurement uncertainties for both reactions.

5.2 Spatial distribution of slow neutrons on the Gamma-3

The experimentally measured and Monte Carlo calculated track densities in the CR-39/LR-1152B samples are presented in Figure 5.4. Monte Carlo track densities were calculated using the INCL4-ABLA models, as this provided the best agreement for the $^{232}$Th(n,$\gamma$) reaction. The track densities on the top surface of the Gamma-3 (Figure 5.4a and b) are maximum in the centre of the surface of the graphite, in both the X and Z-directions, falling off with increasing distance from the centre. As expected, the track densities on the front and back surface (Figure 5.4c and d) are highest closest to the lead target. The track densities then decrease with increasing distance from the centre.

In general, good agreement between the experimental and calculated track densities was observed apart from the X-direction on the top surface (Figure 5.4b) where the two samples on both edges do not agree within the measurement uncertainties. The experimental track density is consistently higher than the calculated track density. At the $x=\pm 55 \text{ cm}$ position, the experimental value is approximately 85% higher than the calculated, while for the $x=\pm 40 \text{ cm}$
5.2. Spatial distribution of slow neutrons on the Gamma-3

The Monte Carlo calculated thermal and epithermal flux constants on the surface of the Gamma-3 is shown in Figure 5.5. These calculated flux constants were used to determine the Monte Carlo calculated track densities presented in Figure 5.4. As expected from the measured track densities, the thermal flux constants are at a maximum closest to the centre of the top surface of the Gamma-3. Along the Z-direction, the epithermal neutrons peak 5–10 cm closer to the front of the assembly compared to the thermal neutrons (Figure 5.5a).

The thermal to epithermal flux ratios, $\frac{\phi_{\text{th}}}{\phi_{\text{epi}}}$ increases with increasing distance from the centre. On the front surface (Figure 5.5c) the epithermal flux drops off at a much faster rate than the thermal flux, compared to the back surface (Figure 5.5d). The thermal to epithermal flux ratios, $\frac{\phi_{\text{th}}}{\phi_{\text{epi}}}$ on the top surface are quite high, ranging from approximately 40 at the centre to >90 at the edges. This corresponds to the proportion of epithermal tracks in the detectors to being no more than 1.5% of the total measured track density. The experimental uncertainty in the total track density for samples placed on the top surface is $\sim$13% (including $\sim$12% from the incident number of deuterons) and so accounting for the additional 1.5% of tracks from epithermal position the deviation is about 40%. The possible reason for this discrepancy is discussed in section 5.2.1.
FIGURE 5.5: Thermal and epithermal flux constants on the surface of the Gamma-3, as calculated using MCNPX. Lines connecting the points are there to guide the eye only.

neutrons will not increase the overall precision of these measurements. We may therefore infer that the track density on the top surface is directly proportional to the thermal flux constant. A contour plot of the thermal flux constant on the top surface of Gamma-3 is shown in Figure 5.6.

The total number of neutrons escaping from the top surface (and the other three symmetrical equivalent surfaces) calculated with Monte Carlo is 1.37 neutrons per 1.6 GeV deuteron. Of these, 48% are thermal neutrons i.e. have energies less than $\mu kT$ (0.11 eV). There is 16.8 and 7.45 neutrons per incident deuteron escaping from the front and back surfaces, of which 17% and 34% are thermal, respectively.

5.2.1 Discussion of results

The reason for the disagreement between the experimental and measured track density for the two samples nearest the edge on the top surface (Figure 5.4b) is possibly attributed to the roughness of the graphite on the top surface. The graphite moderator was built from individual graphite bricks, rather than being a single block. The top surface was modelled as being completely flat and horizontal in the simulation, however, in reality this was not the case.
 FIGURE 5.6: Contour plot showing thermal flux constants on the top surface of Gamma-3. Also shown for reference are positions of the experimental samples. The track density measured on these samples is primarily due to thermal neutrons as contribution from epithermal neutrons is insignificant.

It can be seen in Figure 5.4b that the experimentally measured track density in the centre sample \((x=0\,\text{cm})\) is lower than in the adjacent samples at \((x=\pm10\,\text{cm})\). This is in contrast to the calculated results where the maximum track density is in the centre. The reason for the dip in the centre of the experimental results is likely due to the overlapping textolite strips that the samples were mounted on.

The good agreement between the experimental and calculated track density, shows promising potential for the INCL4-ABLA physics models to model the spallation process and provides validation of the experimental procedure to measure the slow neutron flux. However, there are still limitations with this experimental technique; the most significant being the requirement of Monte Carlo simulations to separate the thermal and epithermal neutrons. Cadmium covered CR-39/LR-115 2B samples were irradiated simultaneously with the bare CR-39/LR-115 2B samples in an attempt to experimentally separate the thermal and epithermal neutrons. However, the external surfaces of the Gamma-3, particularly the top surface has very high thermal to epithermal flux ratios, \(\frac{\phi_{\text{th}}}{\phi_{\text{epi}}}\). This means that there were very few epithermal neutrons, and the track densities on the cadmium covered samples were extremely low. These low track densities were not statistically viable and thus were omitted from the final results.

5.3 Sources of measurement uncertainties

Sources of uncertainty in the experimental data arise from the 12\% uncertainty in the measured number of incident deuterons. For the \(^{232}\text{Th}(n,\gamma)\) reaction evaluated from gamma spectrometry,
an additional $\sim 6.5\%$ arises from the combination of the full-energy peak efficiency ($4\%$), measured peak area ($\sim 1\%$), known intensity of 311.9 keV $\gamma$-ray ($\sim 1\%$) [175] and pile-up and TCS correction factors. Uncertainties for the $^{232}$Th(n,f) reaction rate, determined using the fission track detector technique also include statistical uncertainties from track counting ($< 5\%$) and the calibration factor, $w_{Th}$ (2.5\%). An estimated uncertainty of 5 mm is attributed to the horizontal positioning of samples within the tubes, as it was not possible to verify the precise location of samples once inserted into the graphite. In the worst case, a sample shift of 5 mm would not cause more than a 5\% variation to the reaction rate. This uncertainty from sample shift is not included in the overall uncertainty but rather indicated by the X-error bars in Figures 5.1, 5.2 and 5.3.

The statistical uncertainties for the $^{232}$Th(n,$\gamma$) and $^{232}$Th(n,f) Monte Carlo simulations were kept below 3\% in all cases (most were 1–2\%). Uncertainty in the $^{232}$Th(n,f) cross section above 60 MeV was inferred from the uncertainty of the experimental measurements [304, 305]. An additional 3\% was attributed to the uncertainty in the experimentally measured beam position and shape whereby the FWHM$_x$ value (4.5±0.7 cm, Table 3.1) is quite large and it is expected that some of the incident deuterons will miss the lead target. This reduces secondary neutron production and the overall neutron yield in the assembly.

Statistical uncertainties from CR-39 track counting were below 5\% in all cases. The uncertainty in the calculated CR-39 track densities involved 5\% and 19\% from $w_{th}$ and $w_{epi}$, respectively. Another 2\% and 5\% arose due to the spectrum fitting of $\phi_{th}$ and $\phi_{epi}$, respectively. The uncertainty of $w_{epi}$ is quite high (19\%), arising from a combination of the uncertainty from $F_{Cd}$ (6\%), intensity of source neutrons (4\%), $\phi_{th}^{Cd}$ (2\%), $w_{th}$ (5\%) and statistical uncertainties from track counting. The total combined weighted uncertainties amounted to 12.3–15.4\% for experimental results and 4.5–6.5\% for the Monte Carlo results.
Chapter 6

Results II: Quinta assembly

6.1 Results of activation analysis

6.1.1 $^{209}$Bi activation

The experimental and Monte Carlo calculated reaction rates of the $^{209}$Bi sample for the 1 and 4 GeV deuteron irradiation of Quinta are shown in Figures 6.1 and 6.2, respectively. The production rates of $^{206}$Bi, $^{205}$Bi, $^{204}$Bi and $^{203}$Bi calculated by both CEM03 and INCL4-ABLA are shown. These reactions are of particular interest for validation of spallation models with experimental data, as they are most sensitive to neutrons in the energy range 20–80 MeV (Figure 4.14), where local discrepancies in this region have been reported to be as large as a factor of 2 in extreme cases [340]. In general, good agreement between the experimental and the Monte Carlo simulations is observed. All points except the $^{203}$Bi measurement of the 1 GeV sample on plate 1 agree within the limits of measurement uncertainties with either CEM03 or INCL4-ABLA.

The proportion of $^{206}$Bi, $^{205}$Bi, $^{204}$Bi and $^{203}$Bi produced in $^{209}$Bi from neutron, proton and deuteron-induced reactions is presented in Table 6.1. The results from this table were used to estimate $^{197}$Au(p,x)$^{192–191}$Au and $^{197}$Au(d,x) reactions as described below in section 6.1.2. It is shown that neutron-induced reactions were most dominant, particularly for the 4 GeV irradiation. The 4 GeV irradiation samples were placed out of the way of the incident deuteron beam, thus deuteron-induced reactions were negligible, and proton-induced reactions minimal. In comparison, the 1 GeV irradiated samples show a significant proportion of deuteron-induced reactions, up to 31% of the total reaction rate. Deuteron-induced reactions are also much more significant for samples on plate 1.

6.1.2 $^{197}$Au activation

The experimental and Monte Carlo calculated production rates of $^{198}$Au, $^{196}$Au, $^{194}$Au, $^{193}$Au, $^{192}$Au and $^{191}$Au from $^{197}$Au for the 1 and 4 GeV irradiations of Quinta are shown in Figures 6.3 and 6.4, respectively. Contributions of $^{197}$Au(p,x)$^{192–191}$Au and $^{197}$Au(d,x) reactions to the
FIGURE 6.1: The experimental and Monte Carlo calculated axial distribution at $-4 \text{ cm}$ of $^{209}\text{Bi}$ activation in the Quinta assembly irradiated with 1 GeV deuterons. Production rate of $^{206}\text{Bi}$, $^{205}\text{Bi}$, $^{204}\text{Bi}$ and $^{203}\text{Bi}$ due to neutron, proton and deuteron-induced reactions is shown.
6.1. Results of activation analysis

**FIGURE 6.2:** The experimental and Monte Carlo calculated axial distribution at −6 cm of $^{209}$Bi activation in the Quinta assembly irradiated with 4 GeV deuterons. Production rate of $^{206}$Bi, $^{205}$Bi, $^{204}$Bi and $^{203}$Bi due to neutron, proton and deuteron-induced reactions is shown.
TABLE 6.1: Proportion of total $^{206}$Bi, $^{205}$Bi, $^{204}$Bi and $^{203}$Bi produced due to neutron, proton and deuteron induced reactions in the 1 and 4 GeV irradiations as calculated with MCNPX from the INCL4-ABLA models. In the case of the 4 GeV irradiation, the contribution of deuteron induced reactions is negligible. Sample coordinates are listed in Table 3.4 on page 38.

<table>
<thead>
<tr>
<th>Product</th>
<th>Sample</th>
<th>1 GeV (% reactions)</th>
<th>4 GeV (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Neutrons</td>
<td>Protons</td>
</tr>
<tr>
<td>$^{206}$Bi</td>
<td>1</td>
<td>75</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>85</td>
<td>6</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>86</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>85</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>88</td>
<td>4</td>
</tr>
<tr>
<td>$^{205}$Bi</td>
<td>1</td>
<td>57</td>
<td>12</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>75</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>76</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>77</td>
<td>12</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>79</td>
<td>12</td>
</tr>
<tr>
<td>$^{204}$Bi</td>
<td>1</td>
<td>54</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>70</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>72</td>
<td>19</td>
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<tr>
<td></td>
<td>4</td>
<td>74</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>73</td>
<td>18</td>
</tr>
<tr>
<td>$^{203}$Bi</td>
<td>1</td>
<td>58</td>
<td>18</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>71</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>75</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>76</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>78</td>
<td>17</td>
</tr>
</tbody>
</table>
6.1. Results of activation analysis

The total reaction rate were estimated from the results shown in Table 6.1. It was assumed that the percentage of proton and deuteron-induced reactions to the $^{197}\text{Au}$ reactions were the same as for the $^{209}\text{Bi}$ reactions. This was done on the basis that $^{209}\text{Bi}(n,xn)$ and $^{197}\text{Au}(n,xn)$ reactions have closely resembling cross sections (see Figure 4.14 on page 98) and $^{209}\text{Bi}$ and $^{197}\text{Au}$ are both sub-actinides with odd-Z, even-N nuclei. Without this correction, the Monte Carlo results of $^{197}\text{Au}$ would be lower by 4 to 38\%, depending on the sample position in Quinta and the reaction involved. As an example, the 38\% adjustment occurs for sample 1 of $^{192}\text{Au}$ (produced from $^{197}\text{Au}(n,6n)$) because for $^{204}\text{Bi}$ (produced from $^{209}\text{Bi}(n,6n)$), 16\% is attributed to proton and 22\% is attributed to deuteron induced reactions. This is not the most ideal solution, but with the lack of further information and cross section data it is considered a reasonable first approximation.

The sample located on plate 1 of the 1 GeV measurement has a systematically higher experimental result compared to the calculation. This disagreement applies only to the high energy (>20 MeV) threshold reactions and not to the lower energy threshold reactions – (n,γ) and (n,2n). We suspect this was caused by an unintended ~0.5 cm positioning error of the sample within the Quinta (details in section 6.4 on page 141).

The experimental (n,γ) reaction rate is about 25\% higher than those calculated by both CEM03 and INCL4-ABLA and outside the measurement uncertainties for all INCL4-ABLA results. Other reaction channels that lead to the production of $^{198}\text{Au}$ such as $^{197}\text{Au}(d,p)^{198}\text{Au}$ were also considered. This reaction has a relatively low cross section – maximum 200 mb at 13 MeV, dropping rapidly to 0.2 mb at 200 MeV [288], the maximum energy that cross section data is available. As the vast majority of deuterons in the sample locations are the incident deuterons, this reaction was deemed negligible for both 1 and 4 GeV irradiations.

The experimental reaction rate of $^{197}\text{Au}(a,b)^{192}\text{Au}$ shows a peculiarly large disagreement between the experimental and calculated results for both 1 and 4 GeV irradiations. The experimental result is roughly two times higher than the INCL4-ABLA calculations. This large disagreement is unlikely to be due to an error in the cross section calculation because for there to be agreement, the $^{197}\text{Au}(n,6n)$ cross section would need to be larger than the (n,4n) cross section and also deviate significantly from the $^{209}\text{Bi}(n,6n)$ cross section (Figure 4.14 on page 98). This was considered illogical. Another considered possibility was the presence of $^{192}\text{Ir}$ from $^{197}\text{Au}(n,2p4n)$ reaction which emits the same signature γ-rays upon $\beta^-$ decaying\(^1\). However, the relatively long half life of $^{192}\text{Ir}$ (74 days) would not allow significant activity build up in the short irradiation times experienced here. Additionally, multiple spectra of each sample were taken over a period of time, and peaks attributed to $^{192}\text{Au}$ were absent from all but the first spectra of each sample, implying a short half life. The possibility of any “interfering nuclear reactions” [152] is being considered and the cause of this large discrepancy is still unknown and currently under investigation.

\(^1\) $^{192}\text{Au}^{\text{EC}+\beta^+}_{4.9\text{h}}^{192}\text{Pt}^*$ or $^{192}\text{Ir}^{\beta^-}_{74\text{d}}^{192}\text{Pt}^*$
FIGURE 6.3: The experimental and Monte Carlo calculated axial distribution at $-4$ cm of $^{197}$Au activation in the Quinta assembly irradiated with 1 GeV deuterons. Production rate of $^{198}$Au, $^{196}$Au, $^{194}$Au, $^{193}$Au, $^{192}$Au and $^{191}$Au due to neutron, proton and deuteron-induced reactions is shown.
FIGURE 6.4: The experimental and Monte Carlo calculated axial distribution at −6 cm of $^{197}$Au activation in the Quinta assembly irradiated with 4 GeV deuterons. Production rate of $^{198}$Au, $^{196}$Au, $^{194}$Au, $^{193}$Au, $^{192}$Au and $^{191}$Au due to neutron, proton and deuteron-induced reactions is shown.
In general, the experimental results of the gold samples agree better with INCL4-ABLA calculations than CEM03 (apart from the \((n,\gamma)\) reaction). This is in contrast to bismuth where the experimental results show a much more mixed agreement between the two models. We suspect this contradiction is caused by uncertainties in the cross section data. A more detailed comparison between the two models is supplied in section 6.3.

6.2 Results of fission track detectors

6.2.1 Sub-actinide fission rates

6.2.1.1 Axial distribution

The axial distribution of the \(^{209}\text{Bi}\), \(^{nat}\text{Pb}\) and \(^{197}\text{Au}\) fission rate at \(y = -4\) cm and \(y = -12\) cm for the 1 GeV irradiation, and \(y = -6\) cm and \(y = -12\) cm for the 4 GeV irradiation are shown in Figures 6.5 and 6.6, respectively. These plots show both the experimental and Monte Carlo calculated (INCL4-ABLA and CEM03) fission rate in terms of ‘fissions per sample atom per incident deuteron’. Experimental samples for the three materials were placed in all locations indicated in the graphs, however the 1 GeV irradiation samples on plate 0 at \(y = 12\) cm had extremely low track densities \(<10\) tracks cm\(^{-2}\) which were not statistically reliable and therefore excluded from the final results.

For all 1 GeV samples positioned at \(y = -4\) cm, the experimentally measured fission rate is systematically higher than the Monte Carlo calculated rate. We attribute this discrepancy to the samples being hit directly with high energy primary deuterons as these samples were placed in the beam shadow (explained further in section 6.4). The samples at \(y = -12\) cm were well clear of the incident beam and there is much better agreement between the experiment and calculations.

The experimental result on plate 1 at \(y = -4\) cm is also anomalously high. This is consistent with results of the activation analysis of the \((n,xn)\) reactions which were performed on the same samples. As described in section 6.4, the discrepancy was likely caused by a \(~0.5\) cm vertical displacement of the sample which likely occurred either when mounting the sample onto the plate or when lowering the plate into the gap between the Quinta sections.

Activation samples were placed at only one position on plate 1 (\(y = -4\) cm), while fission samples were placed at two positions (\(y = -4\) cm and \(y = -12\) cm). The sample at \(y = -12\) cm also shows an elevated experimental reaction rate, so this may suggest that the cause of the positioning error may be due to the sample plate not being lowered completely into position.

However, the results for the 4 GeV samples also show experimental rates that are systematically higher on plates 0 and 1 at both the \(y = -6\) cm and \(y = -12\) cm positions. It is possible that the Monte Carlo code underestimates the number of high-energy backward emitted neutrons in thick targets [96, 130], but further investigation is required to reach a final conclusion.

On plate 4 and 5 at \(y = -6\) cm, the experimentally measured fission rate is higher than calculated by both models. At \(y = -6\) cm, the maximum fission rate occurs on plate 2, while at
6.2. Results of fission track detectors

FIGURE 6.5: Experimental and Monte Carlo calculated axial distribution of bismuth, lead and gold fission rates in Quinta irradiated with 1 GeV deuterons. For z-coordinates of the plates refer to Table 3.9 on page 57.
CHAPTER 6. RESULTS II: QUINTA ASSEMBLY

FIGURE 6.6: Experimental and Monte Carlo calculated axial distribution of bismuth, lead and gold fission rates in Quinta irradiated with 4 GeV deuterons. For z-coordinates of the plates refer to Table 3.10 on page 57.
6.2. Results of fission track detectors

When $y = -12$ cm it occurs on plate 3. This is due to the emission angles of the high energy secondary particles. As fission of sub-actinides is only induced by neutrons of $E_n > 40$ MeV, these secondary particles have undergone minimal scattering. For the samples located at $y = -12$ cm, the fission rate at 4 GeV is higher by a factor of 5–10 as compared to 1 GeV.

6.2.1.2 Radial distribution

The radial distribution of the fission rate along plates 2 and 3 for the 1 and 4 GeV irradiation is shown in Figures 6.7 and 6.8, respectively. The experimental measurements agree much better with the calculated results at 4 GeV compared to 1 GeV. We suspect this is due to the issues regarding the 1 GeV deuteron beam described in section 6.4. The radial distribution of these two plates clearly show that CEM03 calculates higher fission rates in the path of the incident beam, while INCL4-ABLA calculates higher away from the beam. The deuteron inelastic cross sections seems to differ between the two models, being higher for INCL4. This causes more deuterons for CEM03 and more high energy neutrons for INCL4 in the beam region (see section 6.3). The differences between the two physics models highlights the importance of experimental benchmarking.

The shapes of the peaks vastly differ between the 1 and 4 GeV irradiations. The 1 GeV results show a much broader peak, owing to the much wider deuteron beam. The width of the flat part of the peak on plate 2 that occurs for the 1 GeV irradiation corresponds approximately to the FWHM of the beam. On plate 3, the fission rate distribution along the $y$-axis shows an unusual shape at the $y = \pm 2$ cm locations. Figure 6.9 shows that these spikes were predominantly caused by deuteron-induced fissions, and not neutrons. This implies that the deuteron flux was much higher in these locations. At $x = 0$ cm, $y = \pm 2$ cm there is an air gap above and below the central rod in each section in Quinta. Despite rotating the Quinta assembly $2^\circ$ on the $x$-$z$ plane to prevent incident deuterons passing straight through these gaps, the gaps were large enough that it was still possible for a small number of incident deuterons to pass through two sections of the assembly without colliding with the uranium rods. Calculations show that a minimum gap between two rods along the $x$-axis of 0.82 cm would be enough for deuterons to travel through two sections unimpeded. The maximum width of the gap along the $x$-axis between two adjacent rods in Quinta sections is 1.14 cm.

6.2.1.3 Contribution of different particle types to Bismuth fission rate

The relative contribution of each particle type to the overall bismuth fission rate in samples positioned axially along the Quinta set-up at $y = -4$, $-6$ and $-12$ cm is displayed in Table 6.2. The numbers in this table do not include uncertainties from the Monte Carlo simulations or cross section values and are indicative only.

Deuteron-induced fission is dominant when samples are located in the path of the incident beam, but is otherwise negligible for samples away from the beam. As expected, photofission...
FIGURE 6.7: Experimental and Monte Carlo calculated radial distribution of bismuth, lead and gold fission rates in Quinta irradiated with 1 GeV deuterons.
FIGURE 6.8: Experimental and Monte Carlo calculated radial distribution of bismuth, lead and gold fission rates in Quinta irradiated with 4 GeV deuterons
FIGURE 6.9: Breakdown of the radial $^{209}$Bi fission rate on plate 3 of the 1 GeV irradiation (cf. Figure 6.7b) comparing the neutron, proton, deuteron and total fission rate. Results calculated with INCL4-ABLA model and contributions from pion-induced and photofission are minimal.
**TABLE 6.2:** Percentage of $^{209}$Bi fissions attributed to neutrons, photons, protons, pions and deuterons calculated with INCL4-ABLA. $z$-pos is the distance in cm along target axis from Quinta origin.

<table>
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<th>Plate</th>
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<th>Proton</th>
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is insignificant due to the small photofission cross sections and the extremely low photon flux above $\sim 100$ MeV. The relative proportion of neutrons contributing to fission reactions increases further away from the centre of the target due to the high electronic energy losses and shorter range of charged particles travelling through the dense uranium. As mentioned in section 4.9.5, no reliable pion-induced fission cross section data is available for the energy range required in this work. Therefore, the accuracy of the calculated pion-induced fission rates at 4 GeV will be limited by the accuracy of the calculated fission cross section. The absence of high energy pion cross sections will become a potential complication in future ADS utilising beam energies greater than 1–2 GeV, as pion production starts to become more significant.
6.2.2 Actinide fission rates

The fission rate of the $^{232}\text{Th}$ samples is shown in Figures 6.10 and 6.11 for the 1 and 4 GeV deuteron irradiations, respectively. As was the case for the sub-actinide fission, the 1 GeV sample on plate 1 at $y = -4$ cm has an experimental result abnormally higher than the Monte Carlo calculated result (reasons discussed in section 6.4). The samples on plate 0 for the 1 GeV irradiation also show higher experimental results compared to the calculation. The 1 GeV radial distributions all show good agreement between experiment and calculation.

For unknown reasons, the experimental samples placed in the 4 GeV irradiation shows a systematically lower fission rate than the Monte Carlo calculations predict. This is especially noticeable in the radial distribution (Figures 6.11c and d). In contrast, the samples placed on the external surface do not show this systematic discrepancy. The samples placed inside the...
6.2. Results of fission track detectors

FIGURE 6.11: Experimental and Monte Carlo calculated thorium fission rate in Quinta irradiated with 4 GeV deuterons.

a) Axial distribution at $y = -12$ cm
b) Axial distribution on the external surface of the Quinta assembly
c) Radial distribution on plate 2
d) Radial distribution on plate 3
TABLE 6.3: Percentage of $^{232}$Th fissions attributed to high energy ($E_n > 60$ MeV) neutrons, low energy ($E_n \leq 60$ MeV) neutrons, protons, pions and deuterons calculated with INCL4-ABLA. Contribution from photons were negligible (<0.1%).

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<td>26.3</td>
<td>66.0</td>
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</table>

* No experimental sample were placed at these locations

Quinta also agree better with CEM03 (similar to Gamma-3), while the samples placed on the outside agree best with INCL4-ABLA, where the average energy of neutrons is lower.

The breakdown of thorium fissions attributed to high energy ($E_n > 60$ MeV), low energy ($E_n \leq 60$ MeV), protons, pions and deuterons is shown in Table 6.3. As expected, deuterons are a significant contributor to fissions in the beam region. However, pion fissions are much less significant than in the sub-actinides as the much higher neutron cross section dominates the total fission rate. Similar to $^{232}$Th fission in the Gamma-3 assembly (section 5.1), the contribution of high energy neutrons to the overall fission rate is sizeable and highlights the need for more accurate cross section measurements in this energy region.
6.3. Comparison between CEM03 and INCL4-ABLA

The fission rate of natU was measured in two locations within the Quinta assembly for each of the 1 and 4 GeV irradiations. The results are shown in Table 6.4. It can be seen that the experimental results agree best with INCL4-ABLA.

6.2.2.1 Fission and spallation products in $^{232}$Th

In addition to the $^{232}$Th fission rate being measured using mica track detectors, the yield of fission and spallation products produced in a $^{232}$Th sample placed in the Quinta assembly under 2 GeV deuteron irradiation has also been measured with gamma spectrometry. This sample was intentionally placed in the location of Quartz where the neutron flux is highest i.e. centre of plate 2 (at $x=0$, $y=0$, $z=25.7$ cm). This irradiation period was entirely separate from the irradiation periods mentioned in chapter 3. Therefore, further details of this irradiation and results are reported in the self-contained appendix chapter B.

6.3 Comparison between CEM03 and INCL4-ABLA

The Monte Carlo calculations of the activation analysis and fission rate measurements show there is a clear variation between the INCL4-ABLA and CEM03 physics models. These differences arise from deviations between particle multiplicity, particle spectra and emission angle of secondary

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**TABLE 6.4:** Experimental and Monte Carlo calculated thorium and uranium fission rate on plate 3 at ±8 cm for the 1 and 4 GeV irradiations. Results shown in units of ‘fissions per 10$^{27}$ atoms per incident deuteron’. The $^{natU}/^{232}$Th fission rate and 4/1 GeV ratios are also shown. The systematic uncertainty for the incident number of deuterons in $^{natU}/^{232}$Th cancels out.

| Deuteron $y$-pos Sample Exp. CEM03 INCL4-ABLA |
|-----------------|-----------------|-----------------|-----------------|
| $^{232}$Th      |                 |                 |                 |
|                  |                 |                 |                 |
| 1 GeV +8 cm     |                  | 1.8 ± 0.2       | 2.08 ± 0.06     | 1.98 ± 0.07     |
| natU            | 5.3 ± 0.6       | 6.9 ± 0.1       | 6.3 ± 0.1       |
| U/Th            | 2.9 ± 0.5       | 3.3 ± 0.1       | 3.2 ± 0.1       |
|                 |                 |                 |                 |
| 1 GeV -8 cm     |                  | 1.9 ± 0.2       | 2.22 ± 0.06     | 2.10 ± 0.07     |
| natU            | 6.4 ± 0.7       | 7.3 ± 0.1       | 6.6 ± 0.1       |
| U/Th            | 3.4 ± 0.6       | 3.3 ± 0.1       | 3.2 ± 0.1       |
|                 |                 |                 |                 |
| 4 GeV +8 cm     |                  | 9 ± 1           | 10.0 ± 0.3      | 10.5 ± 0.3      |
| natU            | 30 ± 3          | 31.5 ± 0.3      | 31.2 ± 0.3      |
| U/Th            | 3.3 ± 0.3       | 3.14 ± 0.09     | 2.98 ± 0.08     |
|                 |                 |                 |                 |
| 4 GeV -8 cm     |                  | 8 ± 1           | 9.3 ± 0.3       | 9.6 ± 0.3       |
| natU            | 29 ± 3          | 29.7 ± 0.2      | 29.1 ± 0.3      |
| U/Th            | 3.6 ± 0.3       | 3.2 ± 0.1       | 3.0 ± 0.1       |
|                 |                 |                 |                 |
| 4/1 ratio +8 cm |                  | 4.2 ± 0.5       | 5.1 ± 0.2       | 5.8 ± 0.7       |
| natU            | 4.3 ± 0.5       | 5.0 ± 0.1       | 5.9 ± 0.7       |
|                 |                 |                 |                 |
| 4/1 ratio -8 cm |                  | 3.6 ± 0.4       | 4.4 ± 0.2       | 5.1 ± 0.6       |
| natU            | 4.0 ± 0.4       | 4.5 ± 0.1       | 4.5 ± 0.5       |
particles calculated by both models. Figures 6.12 and 6.14 show a comparison of the neutron spectra calculated by the two models with an overlay of reaction cross sections for reference. In general, for the 1 GeV irradiation, CEM03 calculates a total neutron production rate 1.2% higher than INCL4-ABLA. For comparison, calculations performed by Hashemi-Nezhad et al. [17] of a 1 GeV proton beam on a uranium target found that the CEM03 model has a total neutron yield 10% higher than calculations using the INCL4-ABLA models. The major difference between these two calculations is the incident projectile, where Hashemi-Nezhad et al. [17] compares incident protons and in our study we have compared incident deuterons. This 10% difference for the incident proton is much higher than the 1.2% difference for incident deuteron. However, this discrepancy is consistent with the data shown in Figure 4.1a on page 84 for lead target, which shows the difference in neutron production between CEM03 and INCL4-ABLA models is much higher for incident proton than it is for incident deuteron.

Figure 6.12 shows separate neutron spectra for the neutrons emitted during the cascade and evaporation stages for the two models. This is averaged over the entire Quinta assembly under 1 GeV irradiation and so is not entirely indicative of the individual sample positions. Nevertheless, it is immediately obvious that INCL4 produces considerably more neutrons from the INC stage above 20 MeV.

The (n,xn) reaction cross sections are overlaid in Figure 6.12a. The threshold energy of the $^{197}$Au(n,2n) reaction (8.11 MeV) is above the energy of fission neutrons and is a measure of both high energy evaporation and cascade neutrons. In this energy region, CEM03 produces more neutrons than INCL4-ABLA and the experimental results agree best with INCL4-ABLA. The (n,xn), for $x \geq 4$, reactions have a high threshold energy that exclusively detect cascade neutrons of a narrow energy range. INCL4 produces significantly more (~2X) cascade neutrons in this region compared to CEM03. Therefore, the INCL4-ABLA calculated reaction rates are greater than the CEM03 reaction rates (Figures 6.1–6.4). The $^{197}$Au(n,xn) experimental measurements agree best with INCL4-ABLA, while $^{209}$Bi(n,xn) produces a more varied agreement between the two models.

The (n,f) reaction cross sections are overlaid in Figure 6.12b. The sub-actinide fission reactions can only be induced by the highest energy cascade neutrons. INCL4 produces more of these neutrons compared to CEM03. This is why for samples not directly hit by the incident beam, INCL4 calculates a higher fission rate compared to CEM03. In general, the experimental results agree better with INCL4-ABLA compared to CEM03.

As is evident from the samples placed at $y=-4$ cm for the 1 GeV irradiation, and also seen in Figures 6.7 and 6.8, CEM03 calculates a higher fission rate for samples placed in the path of the incident beam, while INCL4 calculates a higher fission rate for samples not in the direct path of the beam. This is due to CEM03 calculating a higher deuteron flux in the beam region (see e.g. Figure 4.7). Figure 6.13 illustrates the differences of the deuteron flux within the beam region calculated using INCL4 and CEM03. The histograms represent the flux of the deuterons that have survived inelastic nuclear interactions. The total number of survived deuterons in the case
FIGURE 6.12: Comparison between cascade and evaporation spectra for the CEM03 and INCL4-ABLA model averaged over entire Quinta assembly under 1 GeV deuteron irradiation. Logarithmic binning at 20 intervals per decade is used. Also shown for reference are the (a) (n,xn) cross sections and (b) (n,f) cross sections.
CHAPTER 6. RESULTS II: QUINTA ASSEMBLY

FIGURE 6.13: Comparison of the deuteron flux along the Quinta target axis for the 1 GeV deuteron irradiation as calculated using INCL4 and CEM03. Deuteron flux was tallied in discs of radius 5 cm and thickness 1 cm.

of INCL4 is significantly less than in the case of CEM03. This implies that the deuteron inelastic interaction cross section in INCL4 is higher than in CEM03, and also provides explanation for the higher number of cascade neutrons produced with INCL4. Deuterons are the dominant contributor to fission for the samples at \( y = -4 \) cm, while neutrons are the largest contributor to fission at \( y = -12 \) cm (refer to Table 6.2). For samples where neutron-induced fission is dominant (outside of the beam), INCL4-ABLA calculations are higher and produce the best agreement with experimental data.

Figure 6.14 shows a comparison of the total neutron spectra as calculated with the two models for the 1 GeV irradiation on plate 2 at \( y = -4 \) cm. Also shown is an overlay of the relevant part of the \(^{197}\text{Au}(n,\gamma)\) cross sections demonstrating that the reaction is most sensitive to slow neutrons. It is again noticeable that INCL4-ABLA produces more neutrons above 20 MeV (25% more in that sample position), despite the total number of neutrons produced being 1.2% less than CEM03. Therefore, CEM03 has more neutrons than INCL4-ABLA for neutron energies less than 20 MeV which is consistent with the \(^{197}\text{Au}(n,\gamma)\) calculated reaction rate. The experimental \(^{197}\text{Au}(n,\gamma)\) reaction rate agrees better with CEM03 in all cases apart from sample 1 of the 1 GeV irradiation.

The clear differences between the INCL4-ABLA and CEM03 models, serves to highlight the importance of experimentally validating each of the physics models. Although, these results are unable to conclusively agree on which model is best to reproduce the entire particle spectra of a fast ADS. There is strong evidence from the \(^{209}\text{Bi}, \text{nat}\text{Pb}\) and \(^{197}\text{Au}\) fission rate measurements that INCL4-ABLA better reproduces the very high energy region of the neutron spectrum. The \(^{209}\text{Bi}(n,xn)\) and \(^{197}\text{Au}(n,xn)\) reactions have produced more arbitrary agreements even though they probe the same region of the neutron spectrum. This ambiguity is most likely caused by the cross sections that were used to calculate the reaction rate. Accurate cross sections are critical for experimentally validating these spallation models. The \(^{197}\text{Au}(n,2n)\) and \(^{197}\text{Au}(n,\gamma)\) reactions

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6.4 Effect of sample/beam position on reaction rate

Two entirely independent sources of discrepancies between the experimental and calculated results were realised. The uncharacteristically high experimental result of the 1 GeV sample on plate 1 at \( y = -4 \) cm was attributed to a problem with the positioning of the individual sample. Additionally, all sub-actinide fission results at \( y = -4 \) cm for the 1 GeV irradiation showed an experimentally higher fission rate than was predicted by the calculation. We suspect this discrepancy arose due to the true deuteron beam size being wider than measured. Further details of both cases are provided below.

**Displacement of 1 GeV sample on plate 1 at \( y = -4 \) cm**

In comparison to the other sample locations, reactions measured in the 1 GeV irradiation samples located on plate 1 at \(-4\) cm have shown an experimental result that is uncharacteristically higher than the calculated result. Fission rate in \( ^{232}\text{Th}, ^{209}\text{Bi}, ^{nat}\text{Pb} \) and \( ^{197}\text{Au} \), \( ^{197}\text{Au} \) activation reactions with \( E_{th} > 20 \text{ MeV} \) and the \( ^{209}\text{Bi}(n,7n) \) reaction experimental results do not agree within the measurement uncertainties. The reaction rates of \( ^{209}\text{Bi}(n,4n), ^{209}\text{Bi}(n,6n) \) and \( ^{197}\text{Au}(n,2n) \) agree within the measurement uncertainties but still appear higher than is expected. The only reaction that does not appear uncharacteristically high in this location is \( ^{197}\text{Au}(n,\gamma) \).

The 1 GeV samples (at \( y = -4 \) cm) were placed much closer to the centre of the incident deuteron beam than the 4 GeV samples (\( y = -6 \) cm). The FWHM\(_y\) of the 1 GeV beam was also more than twice the width of the FWHM\(_y\) of the 4 GeV beam (Table 3.2 on page 35). Therefore, the samples in the 1 GeV irradiation at \( y = -4 \) cm are being directly hit by the incident
deuteron beam and exposed to a much harder neutron spectrum. This leads to a lot more direct deuteron-induced reactions and an increased high energy neutron flux.

From Table 6.1 it is clear that the percentage of deuteron-induced reactions in the activation samples on plate 1 is much larger than for any of the other samples for 1 GeV irradiation. However, inclusion of this contribution is still not enough to make a good agreement. Therefore, it was suspected that the discrepancy of the sample on plate 1 was either due to an error in the beam location on the target and/or the positioning of the sample itself.

In order to investigate the effect of beam position and/or sample placement on the reaction rates of sample 1 in the 1 GeV irradiation, further Monte Carlo simulations were carried out. Two methods were tested, firstly, the beam was shifted down by up to 1.0 cm and secondly, the sample was shifted up by up to 1.0 cm. If the elevated experimental reaction rate in sample 1 of the 1 GeV irradiation is caused by an error in the beam position and/or sample placement it is unlikely to be more than 1 cm. Analysis of the results suggested that the discrepancy is most likely due to a ~0.5 cm vertical displacement of the sample. This probably occurred when mounting the sample onto the plate and/or when lowering the plate into the slot between the Quinta sections. Error in the positioning of the beam was deemed unlikely to be the cause as this led to further inconsistencies of the reaction rates in the other activation samples.

The $^{197}$Au(n,$\gamma$) reaction is unaffected by this shift because it is only sensitive to slow neutrons, while the $^{197}$Au(n,2n) reaction ($E_{\text{Th}}=8.11$ MeV) is only moderately affected. Neutrons with energies greater than 20 MeV arise from the cascade stages [15] or from the disintegration of the incident beam deuterons [287] and are emitted with a strong forward direction. Neutrons with energies $<20$ MeV originate from low energy pre-equilibrium emission (if applicable), evaporation, (n,$x$n) and fission reactions, the majority of which are emitted isotropically [15]. These lower energy reactions would therefore be less affected by small variations in the relative position of the incident beam.

Fission samples were placed at two positions on each sample plate ($y=-4$ cm and $y=-12$ cm) and the sample at $y=-12$ cm also shows an elevated experimental reaction rate. The difference to the fission rate at $y=-4$ cm is much more significant than at $y=-12$ cm as changes to the fission rate are much more sensitive to vertical positioning closer to the beam centre (refer to Figures 6.7 and 6.8). So, at first look, this suggested that the cause of the positioning error may be due to the sample plate not being lowered completely into position. However, the results for the 4 GeV sub-actinide fission (Figure 6.6), also show experimental results that are systematically higher on plates 0 and 1 at both the $y=-6$ and $y=-12$ cm positions. Previous studies have shown that Monte Carlo simulations tend to underestimate the number of backward emitted high energy neutrons in thick targets [96, 130]. Therefore, it is not possible to tell if the 1 GeV sample at $y=-12$ cm was also out of position. We suspect that it is most likely an error in the individual sample positioning of the $y=-4$ cm sample.
6.5 Uncertainties and possible sources of error

Discrepancy of 1 GeV sub-actinide fission samples at $y=-4$ cm

In addition to the isolated discrepancy of the 1 GeV sample located on plate 1 at $y=-4$ cm, sub-actinide fission samples on all plates at $y=-4$ cm show an experimentally measured fission rate higher than predicted by the calculations. The sub-actinide fission samples are only sensitive to the highest energy cascade neutrons/protons and incident deuterons, all of which are emitted with a very strong forward direction. Therefore, these samples would be much more sensitive to small variations to the beam flux in those sample locations. The $(n,xn)$ reactions only probe a very narrow region of the neutron spectrum, and the $^{232}$Th(n,f) reaction is also sensitive to fission and evaporation neutrons, making them less affected by small variations to the incident deuteron beam flux.

This disagreement is therefore likely caused by an error in the deuteron-induced fission cross section or simulated deuteron flux (or both) at these locations. On average, the experimental results are $\sim 3$ times higher than the calculated ones, and $\sim 50\%$ of total fissions are induced by primary deuterons (Table 6.2). Thus in order to reach agreement, the deuteron-induced fission cross section will require an increase by a factor of five. Despite the deuteron cross section not being accurately known (uncertainties of 16–36\%), it was deemed unrealistic to believe it could be erroneous by a factor of five. We therefore attribute this disagreement to an error in the simulated particle flux stemming from an imprecise measurement in the shape of the incident deuteron beam.

The disagreement occurs for samples located both above and below the beam axis (See Figure 6.7) and so it’s likely that the width of the deuteron beam in the $y$-direction is broader than what was measured (and presented in Table 3.2 on page 35). A wider incident beam would increase the deuteron (and high energy neutron/proton) flux in the samples placed close to the beam axis while leaving samples located further away relatively unaffected. Further simulations incorporating the uncertainties of the beam parameters in Table 3.2 was not enough to establish an agreement. The size of the beam was determined using passive track detectors (to detect beam induced $^{nat}$Pb(d,f) reactions) which provides an average beam profile over the entire irradiation period. It is also possible that the beam may have shifted around over the course of 21 hour irradiation period affecting fission rates of samples close to the beam due to beam induced fission. Previous studies have reported beam drifting during the irradiation period [129], although we do not have strong evidence to support this possibility.

6.5 Uncertainties and possible sources of error

Sources of uncertainty in the experimental data arise from the 11\% uncertainty in the measured number of incident deuterons. For the activation samples, statistical uncertainties from counting statistics ranged between 1 and 10\% and in rare cases were as high as 20\%, depending on the reaction and sample involved. An additional 6\% is attributed to the full-energy peak efficiency, TCS and the emission probability of detected $\gamma$-ray. For the fission samples, systematic
uncertainties for the calibration factors (2.5, 10, 5 and 2\% for $^{232}$Th, $^{209}$Bi, $^{nat}$Pb and $^{197}$Au) were also included.

Two sources of uncertainty were considered for the Monte Carlo results. The statistical uncertainties of the simulations were kept below 5\% in all cases. Uncertainties in the cross sections were estimated from the goodness of the fits in the parametrisations or from the experimentally measured data. The precision of the calculated reaction rates is therefore limited by the uncertainties of the experimentally measured cross sections, which unfortunately in some cases were quite large. The results shown in Figures 6.1–6.11 and Table 6.4 do not include the systematic uncertainty from the measurement of the deuteron beam position and shape.
Chapter 7

Conclusion

Accelerator Driven Systems (ADS) have been studied with broad interest over the past couple of decades. They consist of a spallation neutron source linked to a sub-critical nuclear assembly, and are one proposed solution for transmutation of nuclear waste and energy production. They are predicted to offer improvements over current generation light water reactors in areas of safety, efficiency, waste reduction and proliferation resistance. The development of ADS has surged forward with the advancement of INC models and Monte Carlo codes such as MCNPX. There now exists a need to validate these models and codes with small-scale experiments. This is particularly the case with investigations into GeV deuteron projectiles, which is currently a very sparse area of study. This is despite strong evidence to suggest they will be the most efficient incident projectile for operation of an ADS.

The E&T RAW (Energy and Transmutation of RAdioactive Waste) collaboration, based at the Joint Institute for Nuclear Research (JINR) in Dubna, Russia aims to design and construct experimental set-ups for studies into the energy and spatial distribution of spallation neutrons, and their ability to transmute radiotoxic nuclear waste. Two of these experimental assemblies – Gamma-3 and Quinta, were designed to emulate the particle field of thermal and fast ADS, respectively. Small-scale experiments performed with these two assemblies are suitable for the benchmarking of nuclear models and Monte Carlo codes, when combined with detailed and thorough simulations.

The Gamma-3 assembly consists of a cylindrical lead target centred within a graphite moderator and is the only facility for experimentally studying graphite moderated spallation neutrons. In our study, it was irradiated with 1.6 GeV deuterons and the neutron capture and fission rate of thorium within the moderator was measured. Simulations performed using the MCNPX 2.7 Monte Carlo code with the INCL4-ABLA and CEM03 physics models found good agreement with the experimental measurements. This is the first report of an agreement being found between experimental and calculated $^{232}$Th(n,$\gamma$) and $^{232}$Th(n,f) reaction rates in the Gamma-3 assembly. It was also found that INCL4-ABLA provided the better agreement with experimental data for the neutron capture rate, while CEM03 agreed better for the fission rate
calculations. Accurate $^{232}\text{Th}(n,f)$ cross section data is currently only available up to 60 MeV. However, we have shown that a significant number of fissions were induced by neutrons with energies greater than 60 MeV. This highlights the need for improved cross section data at higher energies.

The spatial distribution of slow neutrons on the surface of the Gamma-3 were measured using CR-39 track detectors paired with a lithium tetraborate converting layer of LR-115 2B film. These CR-39/LR-115 2B detectors needed calibration as previous attempts to calibrate them had been unsuccessful. The detection efficiencies of the CR-39/LR-115 2B detectors were determined using a custom-built calibration set-up and found to be $(1.11 \pm 0.05) \times 10^{-3}$ and $(6.5 \pm 1.3) \times 10^{-4}$ tracks neutron$^{-1}$, for thermal and epithermal neutrons, respectively. A novel method including the development of a recursive algorithm was required to correct for thermal neutrons leaking into the detector. Special attention to the joining functions was also required to account for the different moderators of the calibration set-up and Gamma-3 assembly. The successful calibration of the CR-39/LR-115 2B detector combination has for the first time allowed these detectors to be used for absolute slow neutron measurement (i.e. no normalisation of results required). Monte Carlo simulations were used to distinguish between the proportion of tracks caused by thermal and epithermal neutrons and to calculate the expected track density on the surface of the Gamma-3. The calculations using the INCL4-ABLA physics model yielded good agreement with the experimental results. This agreement suggests that the use of Monte Carlo simulations to determine $w_{th}$ and $w_{epi}$ in the calibration procedure was justified.

The Quinta assembly consists of 512 kg natural uranium surrounded by a lead reflector. In this work, Quinta was irradiated with 1 and 4 GeV deuterons to produce the combined fission-spallation neutron spectrum expected in a fast ADS. We have reported the first comparative results of experimental measurements and Monte Carlo simulations of the Quinta assembly.

Gamma spectrometry was used to measure the rate of $^{209}\text{Bi}(n,xn)$, $^{197}\text{Au}(n,xn)$ and $^{197}\text{Au}(n,\gamma)$ reactions inside the Quinta assembly. This required the generation of high resolution efficiency curves of the HPGe detectors through experimental and Monte Carlo methods. However, complications arose with suspicions that the true detector geometry differed from the manufacturers specifications, confirming reports in the literature that this is a widespread occurrence. Where experimental and evaluated nuclear data were unavailable, neutron-induced reaction cross sections were calculated using the TALYS code. Proton and deuteron-induced reactions were found to be significant for samples placed in the path of the incident deuteron beam. As TALYS calculations were not deemed reliable for proton and deuteron-induced cross sections, very careful approximations based on existing experimental data had to be made. In general, good agreement between the experimental and Monte Carlo calculations was found. Small variations in sample positioning relative to the incident deuteron beam had a dramatic effect on reactions with high threshold energies ($>20$ MeV), while reactions occurring with lower energy thresholds ($<20$ MeV) were less affected.
The fission rate of \( ^{232}\text{Th} \), \( ^{209}\text{Bi} \), \( ^{233}\text{Pa} \) and \( ^{197}\text{Au} \) samples were measured using mica track detectors placed in various locations around the Quinta assembly. The calculation of this fission rate required the collation of vast amounts of cross section data as fissions induced by neutrons, protons, deuterons, charged pions and photons were all calculated individually. An extensive search of the literature was performed, and very carefully scrutinised to ensure only the most accurate and up-to-date cross section values were used. A method was devised to extrapolate the \( ^{197}\text{Au}(n,f) \) cross section from 200 MeV up to GeV energies. Improvements to Prokofiev’s [308] parametrised (p,f) cross section data for the sub-actinides, based on the most recent experimental data was also presented. Previous works have often overlooked charged pion-induced fission due to the lack of cross section data, and the assumption it was insignificant. However, using cross sections calculated with the XSEX3 code, we have shown here that pion-induced fission of sub-actinides in spallation neutron fields induced by deuterons of 4 GeV can be significant. Therefore, if future ADS are to utilise incident projectile energies \( \gtrsim 1\text{–}2\text{ GeV} \), then significant work measuring pion-induced fission cross sections will need to be carried out.

Overall, there was good agreement between the experimental measurements and Monte Carlo calculations of the fission rate in the Quinta assembly. The exception was in samples placed in the path of the incident deuteron beam, in the case of the 1 GeV irradiation. We speculate the cause of this disagreement was due to the true beam size being larger than specified. It was also found that the sub-actinide and \( ^{nat}\text{U} \) fission rate agreed better with INCL4-ABLA, while the \( ^{232}\text{Th}(n,f) \) reaction rate agreed better with CEM03 (like in Gamma-3). Significant differences in the deuteron inelastic cross section between the two models were observed, whereby it is much higher for INCL4. This has implications for the high energy neutron flux in the beam region. Evidence to suggest that Monte Carlo codes underestimate the number of backward emitted high energy neutrons was also provided.

Residual nuclei produced in \( ^{232}\text{Th} \) located in the bare Quinta assembly (no lead shielding) and irradiated with 2 GeV deuterons were measured using gamma spectrometry (appendix B). 46 fission products, 18 spallation products and \( ^{233}\text{Pa} \) were identified. The fission product distribution was found to be a combination of symmetric fission induced by high energy particles, and asymmetric fission induced by low energy particles. The mass distribution of residual isotopes were compared to the mass distribution of isotopes calculated with MCNPX and INCL4-ABLA. Due to limitations of the experimental technique, it was not possible to identify all residual nuclei. Therefore, the experimental data were only able to follow the general trend of the Monte Carlo results.

The overall good agreement between the experimental and calculated results provides positive validation for Monte Carlo codes to accurately predict secondary particle production, transport, energy spectra and moderation in fast and thermal Accelerator Driven Systems. Additionally, verification of the cross sections used here is also provided to some extent. Cross sections were collected from the literature, or calculated using the TALYS and XSEX3 codes, and in some instances required approximations. The generally good agreement between experimental and
CHAPTER 7. CONCLUSION

simulation results shows the use of these cross sections were appropriate. However, due to the fact that reactions may be induced by many particle types, it is not possible to validate individual cross section data. The inadequate availability of cross section data, is a major limiting factor to the accuracy of Monte Carlo simulations.

Even though the results presented here show great potential for Monte Carlo codes to simulate ADSs, there still remains many issues that hamper the accuracy of these calculations which can only be solved through further research. The improvement of high energy cross section data, specifically $^{232}$Th(n,f) above 60 MeV, charged pion induced fission for incident beam energies above 2 GeV, $^{197}$Au(n,f) above 180 MeV and (d,f) at all energies should be treated as a priority. Additionally, the tendency for INC models to underestimate the number of backward emitted neutrons requires further investigation.
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Appendices
Appendix A

List of publications

Several publications resulted from the work completed during the candidature for this thesis. The following is a list of peer reviewed papers that have been published, or have been submitted for publication at the time of thesis submission:


A further publication about the thorium fission measurements in the Quinta assembly detailed in section 6.2.2 is also expected soon after the date of submission.
The following conference presentations have also been made:


Appendix B

Production of fission and spallation products in $^{232}$Th

B.1 Introduction

This appendix chapter describes the analysis of $\gamma$-ray spectra obtained from a sample of thorium placed in the Quinta assembly irradiated with 2 GeV deuterons in March, 2011\(^1\). Unlike the thorium spectra analysed in section 3.4, which served only to identify the rate of the $^{232}$Th(n,\(\gamma\)) reaction, the aim of this work was to identify and measure the production rate of as many fission and spallation products as possible. Similar work is described in publications by Adam et al. [134] and Adam et al. [344].

B.2 Experimental details

B.2.1 Sample irradiation

The thorium sample (0.1268 g), was placed in the centre (at $x=0$, $y=0$ cm) of plate 2 ($z=25.7$ cm) and inserted into the Quinta assembly (Figure B.1). The sample was intentionally placed in this location as this is where the neutron flux is highest (cf. Figure 4.8 on page 91). The Quinta assembly was rotated 2° around the $y$-axis, just like that described in section 3.2.2. The set-up was irradiated for 17.5 hours in total with $(1.54 \pm 0.07) \times 10^{13}$ deuterons. This irradiation of Quinta did not include the lead castle, unlike that described in section 3.1.2 on page 29.

B.2.2 Spectral analysis of the sample

Shortly after irradiation, acquisition of $\gamma$-ray spectra started. All together, nine spectra of the sample were measured using the Canberra HPGe detector (Figure 3.11 on page 48) followed by

\(^1\)Thorium sample irradiation and spectral collection was performed by Dr Jindra Adam of the JINR. Analysis of these spectra were performed by the Author.
APPENDIX B. PRODUCTION OF FISSION AND SPALLATION PRODUCTS IN $^{232}$Th

FIGURE B.1: The Quinta assembly without the lead castle showing the location of the $^{232}$Th sample. The sample itself is not shown to scale. The yellow represents the natural uranium target-blanket and positions of sample plates are signified by labels 0–5. Refer to Figure 3.9 on page 39 for dimensions of the Quinta assembly.

Five spectra taken using a similar Ortec n-type coaxial detector (model: GMX-30). Details of the spectra acquired are shown in Tables B.1 and B.2, where the ‘delay time’ refers to the time interval from the end of irradiation to the start of spectral acquisition.

<table>
<thead>
<tr>
<th>Filename</th>
<th>Live time (min)</th>
<th>Real time (min)</th>
<th>Delay time (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a2Th1P2</td>
<td>10.3</td>
<td>10.4</td>
<td>1.32</td>
</tr>
<tr>
<td>a2Th2P2</td>
<td>10.2</td>
<td>10.3</td>
<td>2.89</td>
</tr>
<tr>
<td>a2Th3P2</td>
<td>21.0</td>
<td>21.1</td>
<td>4.38</td>
</tr>
<tr>
<td>a2Th4P2</td>
<td>35.0</td>
<td>35.1</td>
<td>6.91</td>
</tr>
<tr>
<td>a2Th5P2</td>
<td>120</td>
<td>120</td>
<td>20.8</td>
</tr>
<tr>
<td>a2Th6P2</td>
<td>121</td>
<td>121</td>
<td>37.4</td>
</tr>
<tr>
<td>a2Th7P2</td>
<td>179</td>
<td>180</td>
<td>48.4</td>
</tr>
<tr>
<td>a2Th8P2</td>
<td>244</td>
<td>245</td>
<td>98.4</td>
</tr>
<tr>
<td>a2Th9P2</td>
<td>438</td>
<td>439</td>
<td>228</td>
</tr>
</tbody>
</table>

The two sets of spectra were analysed separately but the method of analysis was identical for both. The spectra were analysed using the Hypermet-PC program [151] to extract information about peak area, peak position and peak FWHM. Efficiency of the detectors were obtained using the EFFCOR program. Additional calibrations regarding the non-linearity and FWHM-energy dependence of the detectors were also required, to ensure the energy of peaks were accurately identified and doublets noted. The non-linearity of the Canberra detection system was found to be minimal with no variations greater than 0.5 channels. However, the non-linearity of the Ortec detection system was up to 3.5 channels (1.3 keV).

\[^{2}\text{EFFCOR program is an in-house software program developed by the Academy of Sciences of the Czech Republic. The calibration spectra were collected by Dr Jindra Adam of the JINR.}\]
TABLE B.2: Properties of $^{232}$Th spectra collected using the Ortec detector.

<table>
<thead>
<tr>
<th>Filename</th>
<th>Live time (h)</th>
<th>Real time (h)</th>
<th>Delay time (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>c2Th1P2</td>
<td>23</td>
<td>24</td>
<td>19</td>
</tr>
<tr>
<td>c2Th2P2</td>
<td>47</td>
<td>48</td>
<td>28</td>
</tr>
<tr>
<td>c2Th3P2</td>
<td>115</td>
<td>122</td>
<td>52</td>
</tr>
<tr>
<td>c2Th4p2</td>
<td>6.6</td>
<td>7.1</td>
<td>86</td>
</tr>
<tr>
<td>c2Th5p2</td>
<td>137</td>
<td>146</td>
<td>88</td>
</tr>
</tbody>
</table>

FIGURE B.2: Example of gamma-ray spectrum taken of natural thorium.

A script was written using Python to match all peaks with identical energies from all spectra. Peaks originating from the natural thorium spectrum (Figure B.2) were automatically discarded. The half life of the produced residual nuclide responsible for the peak of interest was found from the slope of a log count-rate vs. delay time plot. A positive identification of an isotope occurred when the peak energy, peak intensity and half life matched data provided in ENSDF data libraries [175].

Once the produced nuclide had been identified, the production rate, $R$ was calculated in the same way as described in section 3.4.2.1 on page 39 (Eq. B.1). For peaks appearing in multiple spectra, the average reaction rate ($<R>$) across all spectra was also found.

$$ R = \frac{NC}{I_\gamma \varepsilon_p T BSD M_m \frac{t_{irr}}{m N_{AV} \Phi}} $$

(B.1)

Where

$N$: Count rate of peak of interest
$C$: Correction for decay during counting time (Eq. B.2)
$I_\gamma$: Emission probability of measured $\gamma$-ray
$\varepsilon_p$: Detector full-energy peak efficiency
APPENDIX B. PRODUCTION OF FISSION AND SPALLATION PRODUCTS IN $^{232}\text{Th}$

FIGURE B.3: (a) Distribution of 2 GeV deuteron pulses during the irradiation of Quinta assembly. (b) Relationship between the pulsed beam correction factor, $B$, and product half-life for the irradiation period shown in subfigure a.

$T$: Correction due to True Coincidence Summing (TCS)
$S$: Activity saturation factor (Eq. B.3)
$D$: Correction for decay between end of irradiation and beginning of counting (Eq. 3.4)
$B$: Correction due to pulsed beam (details in section 3.4.2.2)
$M_m$: Molar mass of activation foil
$m$: Mass of activation foil (in grams)
$N_{AV}$: Avogadro’s number
$t_{irr}$: Deuteron irradiation time
$\Phi$: Total number of incident deuterons

\[
C = \frac{t_r \lambda_2}{1 - e^{-t_r \lambda_2}} \tag{B.2}
\]
\[
S = 1 - e^{-\lambda_2 t_{irr}} \tag{B.3}
\]
\[
D = e^{-\lambda_2 t_d} \tag{B.4}
\]

Where, $t_r$ is the real-time of measurement interval and $t_d$ is the delay time, i.e. the time between end of irradiation and beginning of counting.

The pulsed beam correction factor, $B$ is dependent on the half lives of the produced isotopes and is unique to each irradiation period. The beam pulse distribution of this irradiation period is shown in Figure B.3a, and the relationship between $B$ and product half life shown in Figure B.3b.

A few peaks consisted of a doublet i.e. two unresolvable peaks originating from two different nuclides. This was evident when the count rate was plotted against delay time and a second order exponential was revealed. In this case, two peaks were fitted and the peak area of the longer lived isotope was subtracted from the shorter lived one. The reaction rate for the shorter
lived isotope could then be found from the same method as above. Only the shorter lived isotope was analysed as there was not enough data to reliably analyse the longer lived isotope.

B.3 Monte Carlo simulations

The irradiation of the bare Quinta assembly with 2 GeV deuterons was simulated using MCNPX 2.7 in the same way as described in chapter 4. The particle spectra calculated using INCL4-ABLA in the location of the thorium sample is shown in Figure B.4. Fission products and spallation nuclei were identified by using the FT8 RES special tally option within MCNPX. The neutron capture and total fission rate of the $^{232}\text{Th}$ sample were also calculated using the methods described in section 4.7 on page 91.

![Neutron, proton, pion, photon and deuteron particle spectra present in the $^{232}\text{Th}$ sample inside the bare Quinta assembly (no lead shielding) irradiated with 2 GeV deuterons. INCL4-ABLA physics models and equal logarithmic energy binning with 20 intervals per decade was used.](image)

**FIGURE B.4:** Neutron, proton, pion, photon and deuteron particle spectra present in the $^{232}\text{Th}$ sample inside the bare Quinta assembly (no lead shielding) irradiated with 2 GeV deuterons. INCL4-ABLA physics models and equal logarithmic energy binning with 20 intervals per decade was used.

B.4 Results and discussion

A list of all residual nuclides including fission and spallation products in the $^{232}\text{Th}$ sample, and their associated $R$ values are shown in Table B.3. Figure B.5 shows the distribution of produced isotopes by mass number, and Figures B.6 and B.7 show the detected fission and spallation products, respectively. Also shown in Table B.3 is the half lives of these nuclides, both the library value and those calculated from measurements.
TABLE B.3: Production rate of fission and spallation products in the $^{232}$Th sample irradiated with 2 GeV deuterons in the bare Quinta assembly. The uncertainty in the calculated half life is an underestimate, as the uncertainty calculation did not take into account the uncertainty of individual data points. Altogether, nine spectra of the sample were taken. These are indicated in the ‘spectra’ column as boxes in order of delay time, $t_d$. The presence of an ‘X’ in place of a box indicates that the specified peak was able to be identified in that spectra. A ‘C’ in the ‘spectra’ column indicates that peaks were also present in spectra collected using the Ortec detector.

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**APPENDIX B. PRODUCTION OF FISSION AND SPALLATION PRODUCTS IN $^{232}$Th**

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<td>374.72</td>
<td>82</td>
<td>16.0(16)</td>
<td>2.52(20)E-28</td>
</tr>
<tr>
<td>899.15</td>
<td>98</td>
<td>12.5(7)</td>
<td>3.59(25)E-28</td>
</tr>
<tr>
<td>984.02</td>
<td>59</td>
<td>13.9(12)</td>
<td>2.47(19)E-28</td>
</tr>
<tr>
<td>$^{205}$Bi</td>
<td>367.44 h</td>
<td>3.31(33)E-27</td>
<td>□□□□□□□□</td>
</tr>
<tr>
<td>1764.36</td>
<td>32.5</td>
<td>1317(1834)</td>
<td>3.31(33)E-27</td>
</tr>
<tr>
<td>$^{206}$Bi</td>
<td>6.24d</td>
<td>1.79(25)E-27</td>
<td>□□□□□□□□</td>
</tr>
<tr>
<td>537.45</td>
<td>30.5</td>
<td>15.6(29)</td>
<td>4.59(52)E-27</td>
</tr>
<tr>
<td>803.1</td>
<td>99</td>
<td></td>
<td>3.32(82)E-28</td>
</tr>
<tr>
<td>803.1</td>
<td>99</td>
<td>14.7(30)</td>
<td>1.53(17)E-27</td>
</tr>
<tr>
<td>881.01</td>
<td>66</td>
<td>9.6(22)</td>
<td>1.67(19)E-27</td>
</tr>
<tr>
<td>1718.7</td>
<td>31.8</td>
<td></td>
<td>8.1(31)E-28</td>
</tr>
<tr>
<td>$^{207}$Po</td>
<td>5.80 h</td>
<td>4.08(26)E-28</td>
<td>□□□□□□□□</td>
</tr>
<tr>
<td>992.33</td>
<td>59.3</td>
<td>7.3(5)</td>
<td>4.08(26)E-28</td>
</tr>
<tr>
<td>$^{209}$At</td>
<td>5.41 h</td>
<td>4.05(32)E-28</td>
<td>□□□□□□□□</td>
</tr>
<tr>
<td>545</td>
<td>91</td>
<td>4.5(2)</td>
<td>5.27(33)E-28</td>
</tr>
<tr>
<td>781.9</td>
<td>83.5</td>
<td>9.1(12)</td>
<td>2.82(31)E-28</td>
</tr>
<tr>
<td>$^{210}$At</td>
<td>8.10 h</td>
<td>3.49(58)E-28</td>
<td>□□□□□□□□</td>
</tr>
<tr>
<td>245.31</td>
<td>79</td>
<td>7.7(10)</td>
<td>3.68(75)E-28</td>
</tr>
</tbody>
</table>

continues on next page
TABLE B.3 – continued from previous page

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>$I_\gamma$</th>
<th>$T1/2$ Library</th>
<th>$&lt; R &gt;$</th>
<th>Spectra</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>T1/2 Experiment</td>
<td>$R$ (atom$^{-1}$ deuteron$^{-1}$)</td>
<td></td>
</tr>
<tr>
<td>$^{224}$Ac</td>
<td>1181.39</td>
<td>99.3</td>
<td>7.6(4)</td>
<td>3.40(21)E-28</td>
<td>X□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>1483.39</td>
<td>46.5</td>
<td>3.40(80)E-28</td>
<td>□□□□□□□□□□</td>
<td></td>
</tr>
<tr>
<td>$^{226}$Ac</td>
<td>131.613</td>
<td>26.9</td>
<td>2.7(3)</td>
<td>7.67(50)E-28</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>215.983</td>
<td>52.3</td>
<td>5.5(7)</td>
<td>4.71(40)E-28</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td>$^{226}$Pa</td>
<td>158.18</td>
<td>17.5</td>
<td>33.7(61)</td>
<td>1.06(9)E-27</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>230.37</td>
<td>27</td>
<td>23.9(23)</td>
<td>1.20(8)E-27</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td>$^{228}$Pa</td>
<td>129.065</td>
<td>4.26</td>
<td>29.3(14)</td>
<td>4.42(46)E-27</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>951.95</td>
<td>29.1</td>
<td>1.94(38)E-27</td>
<td>□□□□□□□□□□</td>
<td></td>
</tr>
<tr>
<td>$^{230}$Pa</td>
<td>300.34</td>
<td>6.62</td>
<td>13.1(30)</td>
<td>2.8(14)E-25</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>312.17</td>
<td>38.6</td>
<td>28.2(29)</td>
<td>4.68(23)E-26</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>312.17</td>
<td>38.6</td>
<td>26.2(3)</td>
<td>7.57(37)E-26</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>340.8</td>
<td>4.47</td>
<td>80.5(52)</td>
<td>9.55(49)E-26</td>
<td>□□□□□□□□□□</td>
</tr>
<tr>
<td></td>
<td>415.76</td>
<td>1.745</td>
<td>60.5(45)</td>
<td>8.98(49)E-26</td>
<td>□□□□□□□□□□</td>
</tr>
</tbody>
</table>

* Value not used when calculating $<R>$

** Peak was a doublet

Cumulative yield data of fission products for monoenergetic 14 MeV neutrons from the ENDF/B-VII.1 data library [31] was used to calculate the total fission rate in the $^{232}$Th sample. The total fission rate was found by averaging over the 46 identified fission products and determined to be $(6.3 \pm 1.4) \times 10^{-26}$ atom$^{-1}$ deuteron$^{-1}$. The Monte Carlo fission rate was calculated to be $(7.82 \pm 0.51) \times 10^{-26}$ atom$^{-1}$ deuteron$^{-1}$, which is within the measurement uncertainties of the experimental result. The other major reactions including $^{232}$Th($n,\gamma$) are shown in Table B.4.

As described in section 4.7, fissions may be induced by many particle types - neutrons, protons, deuterons, photons and charged pions. The energy spectra of these particle types, as calculated using MCNPX with the INCL4-ABLA model is shown in Figure B.4. The breakdown of fission events by fission-inducing particle is shown in Table B.5.

The experimental and Monte Carlo calculated mass distribution of residual nuclei in the $^{232}$Th sample in the Quinta assembly is shown in Figure B.8. Also shown for comparison is the mass distribution of residual nuclei from direct irradiation of $^{232}$Th with 2 GeV deuterons. Only
FIGURE B.5: Mass number of produced nuclides in the $^{232}$Th sample.

FIGURE B.6: Production rate of detected fission products in the $^{232}$Th sample.

FIGURE B.7: Production rate of detected spallation products in the $^{232}$Th sample.
B.4. Results and discussion

**TABLE B.4:** Reaction rates of major reactions occurring from activation of $^{232}$Th sample (cf. Figure B.1) in 2GeV deuteron irradiation of Quinta.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$R$ (atom$^{-1}$ deuteron$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$Th(n,$\gamma$)$^{233}$Th*</td>
<td>$(7.69 \pm 0.39) \times 10^{-26}$</td>
</tr>
<tr>
<td>$^{232}$Th(n,f)</td>
<td>$(6.3 \pm 1.4) \times 10^{-26}$</td>
</tr>
<tr>
<td>$^{232}$Th(n,p6n)$^{226}$Ac</td>
<td>$(1.13 \pm 0.08) \times 10^{-27}$</td>
</tr>
<tr>
<td>$^{232}$Th(n,p8n)$^{224}$Ac</td>
<td>$(0.62 \pm 0.05) \times 10^{-27}$</td>
</tr>
</tbody>
</table>

* Measured via $^{233}$Pa peak. The decay product of $^{233}$Th.

**TABLE B.5:** Breakdown of $^{232}$Th fissions by fission-inducing particle type. Uncertainty shown is only the statistical uncertainty from Monte Carlo calculation and systematic uncertainties from cross sections is not included.

<table>
<thead>
<tr>
<th>Particle</th>
<th>$R$ (atom$^{-1}$ deuteron$^{-1}$)</th>
<th>% of reactions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron ($E_n \leq 60$ MeV)</td>
<td>$(2.10 \pm 0.05) \times 10^{-26}$</td>
<td>26.9</td>
</tr>
<tr>
<td>Neutron ($E_n &gt; 60$ MeV)</td>
<td>$(1.52 \pm 0.07) \times 10^{-26}$</td>
<td>19.4</td>
</tr>
<tr>
<td>Proton</td>
<td>$(1.09 \pm 0.06) \times 10^{-26}$</td>
<td>14.0</td>
</tr>
<tr>
<td>Photon</td>
<td>$(7.84 \pm 0.92) \times 10^{-29}$</td>
<td>0.1</td>
</tr>
<tr>
<td>Deuteron</td>
<td>$(2.85 \pm 0.17) \times 10^{-26}$</td>
<td>36.5</td>
</tr>
<tr>
<td>Pion</td>
<td>$(2.49 \pm 0.35) \times 10^{-27}$</td>
<td>3.2</td>
</tr>
</tbody>
</table>

the outline of the experimental results of $^{205}$Bi and $^{206}$Bi are shown in Figure B.8 due to their anomalously high $R$ values.

From Figure B.8 we can see that there is a clear separation of fission and spallation products based on their mass numbers. The spallation products themselves also consist of two groups. For the direct irradiation of $^{232}$Th the first group peaks at $A=232$, however for the $^{232}$Th in Quinta this group peaks at $A=233$ owing to a significant number of $^{232}$Th(n,$\gamma$) reactions that occur. The second group lies around an intermediate lower mass of $A \approx 200$. This intermediate mass peak is much more obvious for direct deuteron irradiation of $^{232}$Th. This is because the average energy of particles initiating spallation reactions is much lower than 2GeV in the sample location. The higher energy of the incident particle leads to a higher excitation energy of the reaction, producing more lower mass fragments where decay by neutron emission is more probable than decay by fission [15].

The mass distribution of fission products also differs significantly between the two calculated cases. This is again caused by the higher excitation energy of the 2GeV deuteron reaction. The more symmetric mass curve for higher excitation energies tends to ‘wash out’ any shell effects [15]. As excitation energy decreases, the mass curve becomes increasingly asymmetric. The experimental fission mass distribution in Figure B.8 is a combination of symmetric fission induced by high energy particles (neutrons, protons and deuterons) and asymmetric fission induced by low energy particles.
APPENDIX B. PRODUCTION OF FISSION AND SPALLATION PRODUCTS IN $^{232}$Th

The production rate of fission and spallation residual nuclei were measured in a sample of $^{232}$Th placed in the bare Quinta assembly and irradiated with 2 GeV deuterons. Residual nuclei were detected using gamma spectrometry and spectral analysis performed using the HYPERMET-PC and EFFCOR programs. A Python program written by the author was used to correlate peak data from separate spectra and calculate the half-lives of detected nuclides. An automatic search of the ENSDF database would then be performed to identify the appropriate isotope based on peak energy, peak intensity and half-life. The mass distribution of isotopes in the $^{232}$Th were also calculated using MCNPX and the INCL4-ABLA models. The experimental results were able to identify the major features and follow the general trend of the calculated mass distribution curve.

There is good agreement between the general trends of the calculated and experimental data. However, it is apparent that this technique of measuring spallation and fission fragments has many limitations. The major limitation is that many product isotopes are not detectable due to having half lives that are too long or too short for gamma spectrometry. Additionally, fission and spallation events are induced by many particle types containing a wide range of energies. Therefore, individual reactions can not be uniquely identified to a given particle and make it impossible to calculate individual particle induced reaction cross sections.

B.5 Conclusion

The production rate of fission and spallation residual nuclei were measured in a sample of $^{232}$Th placed in the bare Quinta assembly and irradiated with 2 GeV deuterons. Residual nuclei were detected using gamma spectrometry and spectral analysis performed using the HYPERMET-PC and EFFCOR programs. A Python program written by the author was used to correlate peak data from separate spectra and calculate the half-lives of detected nuclides. An automatic search of the ENSDF database would then be performed to identify the appropriate isotope based on peak energy, peak intensity and half-life. The mass distribution of isotopes in the $^{232}$Th were also calculated using MCNPX and the INCL4-ABLA models. The experimental results were able to identify the major features and follow the general trend of the calculated mass distribution curve.
however due to half-life limitations of the experimental technique, not all reaction products could be identified.
Appendix C

Example of MCNPX input files

The following chapter contains example input files for the Gamma-3 and Quinta irradiations. Comments in MCNPX are signified by a ‘c’ in the first column of the line or a ‘$’ mid-line. Further details of syntax and commands are outlined in the MCNPX manual [210].

C.1 Gamma-3 assembly

Gamma-3 assembly irradiated with 1.6 GeV deuterons
c MCNPX input file prepared by Nicola Asquith

| Cells |
|------------------|------------------|
| Lead target and graphite moderator |
| 10 1 -11.34 -10 21 -22 | $ Lead target |
| 20 3 -1.7 10 31 32 -100 | $ Graphite block |

| Cells |
|------------------|------------------|
| Rods where thorium activation samples were placed |
| 60 2 -2.7 -31 311 -100 | $ Rod A - Aluminium tubing |
| 61 3 -1.7 -311 -100 61 62 63 | $ Rod A - Graphite filling |
| 70 2 -2.7 -32 321 -100 | $ Rod B - Aluminium tubing |
| 71 3 -1.7 -321 -100 64 65 | $ Rod A - Graphite filling |

| Cells |
|------------------|------------------|
| Thorium sample cells |
| 81 3 -1.7 -61 | $ Th1 |
| 82 3 -1.7 -62 | $ Th2 |
| 83 3 -1.7 -63 | $ Th3/U |
| 84 3 -1.7 -64 | $ Th4 |
| 85 3 -1.7 -65 | $ Th5 |

| Cells |
|------------------|------------------|
| Space outside the graphite |
| 90 9 -0.0012 -99 (100):(-100 -10 22):(-100 -10 -21) | $ Air outside |
| 99 0 99 | $ Outside universe |
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

Surfaces

10 cz 4 $ Lead target
21 pz 0.3 $ Space in front of target
22 pz 59.1 $ Space at back of target

31 c/z -5 -15 1.35 $ Rod A - outer
311 c/z -5 -15 1.15 $ Rod A - inner
32 c/z 4.5 -22.5 1.35 $ Rod B - outer
321 c/z 4.5 -22.5 1.15 $ Rod B - inner

61 rpp -5.5 -4.5 -15.5 -14.5 46.8 47 $ Th1 47
62 rpp -5.5 -4.5 -15.5 -14.5 25.3 25.5 $ Th2 26.5
63 rpp -5.5 -4.5 -15.5 -14.5 8.1 8.3 $ Th3 8.3
64 rpp 4 5 -23 -22 48.2 48.4 $ Th4 5.2
65 rpp 4 5 -23 -22 26.9 27.1 $ Th5 26.5

99 sph 0 0 30 100 $ Outside area
100 rpp -55 +55 -55 +55 0 60 $ Graphite moderator

Materials

Natural lead target

m1 82204.70c 0.014 $ Pb-204 1.4%
  82206.70c 0.241 $ Pb-206 24.1%
  82207.70c 0.221 $ Pb-207 22.1%
  82208.70c 0.524 $ Pb-208 52.4%
  cond=1

Aluminium for rods

m2 13027.70c 1. cond=1

Graphite moderator with thermal scattering card

m3 6000.70c 1. cond=1
  mx3:h 6012.70h
  mx3:p 6012.70u
  mt3 grph.10t

Thorium

m4 90232.70c 1.

Natural uranium

m5 92234.70c -5.7e-005
  92235.70c -0.007204
  92238.70c -0.992739

Dry air at sea level

200
C.1. Gamma-3 assembly

m9  7014.70c  -0.755636  $ N-14
8016.70c  -0.231475  $ O-16
18036.70c  -3.9e-005  $ Ar-36
18038.70c  -8e-006  $ Ar-38
18040.70c  -0.012842  $ Ar-40

Source definition

Transporting neutron, proton, deuteron, charged pion, neutral pion, muon, photon, kaon
mode n h d / z | p k
imp:n,h,d,/,z,|,p,k 1 11r 0

Cut-off energies for particles
phys:n 1610
phys:h 1610
phys:d 1610
phys:/ 1610
phys:z 1610
phys:| 1610
phys:p 1610 2j -1  $ Photonuclear physics turned on
phys:k 1610

c Physics models

c lca 8j 1 1  $ CEM/LAQGSM
lca 6j 1 j 2  $ INCL4
lea 1 5j 2 j  $ ABLA

c Tallies (for producing required output)
Thorium and uranium neutron capture and fission tallies
fc14 Thorium neutron capture  $ Tally comment
f14:n 81 82 83 84 85  $ Cells for tallying
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

```
fm14 (1e3 4 102) $ Flux multiplier card
fq14 F E $ Print hierarchy card
e14 60 1610 $ Energy binning card
c
fc24 Uranium neutron capture
f24:n 81 82 83 84 85
fm24 (1e3 5 102)
fq24 F E
e24 30 1610
c
fc34 Thorium fission low energy (En<60MeV)
f34:n 81 82 83 84 85
fm34 (1e3 4 18)
fq34 F E
e34 60 1610
c
fc44 Thorium fission high energy (En>60MeV)
f44:n 81 82 83 84 85
c File containing energy-dependent cross section data (DE/DF card)
read file=Th_nf.d noecho
fq44 F E
e44 60 1610
c
fc54 Uranium fission low energy (En<30MeV)
f54:n 81 82 83 84 85
fm54 (1e3 5 18)
fq54 F E
e54 30 1610
c
fc64 Uranium fission high energy (En>30MeV)
f64:n 81 82 83 84 85
read file=U_nf.d noecho
fq64 F E
e64 30 1610
c
-------------------------------------------------------------------------------
c Point detector tallies for surface spectra (CR-39 track detectors)
c Maximum of 20 detectors allowed
fc105 Top surface - Z-direction
f105:n 0 55.2 1 0.2
  0 55.2 11 0.2
  0 55.2 21 0.2
  0 55.2 30 0.2
  0 55.2 39 0.2
  0 55.2 49 0.2
  0 55.2 59 0.2 ND
fq105 E F
e105 1e-9 119log 1000
```
C.1. Gamma-3 assembly

cfc115 Top surface - X-direction
f115:n -54 55.2 30 0.2
-39 55.2 30 0.2
-29 55.2 30 0.2
-19 55.2 30 0.2
-9 55.2 30 0.2
0 55.2 30 0.2
9 55.2 30 0.2
19 55.2 30 0.2
29 55.2 30 0.2
39 55.2 30 0.2
54 55.2 30 0.2 ND
cfc125 Front surface
cf125:n -5 0 -0.2 0.2
c -15 0 -0.2 0.2
c -25 0 -0.2 0.2
c -35 0 -0.2 0.2
c -45 0 -0.2 0.2 ND
cfc135 Back surface
cf135:n -5 0 60.2 0.2
c -15 0 60.2 0.2
c -25 0 60.2 0.2
c -35 0 60.2 0.2
c -45 0 60.2 0.2 ND
cfq115 E F
e115 1e-9 119log 1000
c
cfc125 Front surface
cf125:n -5 0 -0.2 0.2
c -15 0 -0.2 0.2
c -25 0 -0.2 0.2
c -35 0 -0.2 0.2
c -45 0 -0.2 0.2 ND
cfq125 E F
c e125 1e-9 119log 1000
c
cfc135 Back surface
cf135:n -5 0 60.2 0.2
c -15 0 60.2 0.2
c -25 0 60.2 0.2
c -35 0 60.2 0.2
c -45 0 60.2 0.2 ND
cfq135 E F
c e135 1e-9 119log 1000
c
---
c Particle spectra
cfc4 Neutron spectra in thorium sample locations
f4:n 81 82 83 84 85
cfq4 E F
e4 1e-9 259log 1e4
c
cfc11 Neutron leakage spectrum
f11:n 99
e11 1e-9 259log 1e4
c
cfc21 Proton leakage spectrum
f21:h 99
e21 1e0 79log 1e4

---

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APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

```
c fc31 Charged pion leakage spectrum
 f31: 99
e31 1e-3 139log 1e4
c
fc41 Photon leakage spectrum
 f41:p 99
e41 1e-3 139log 1e4
c  
-----------------------------------------------------------------------
c Mesh tallies
 TMESH
c Neutron mesh tally
 RMESH51:n flux
 CORA51 -2 2
 CORB51 -70 139i 70
 CORC51 -15 89i 75
 c Proton mesh tally
 RMESH61:h flux
 CORA61 -2 2
 CORB61 -70 139i 70
 CORC61 -15 89i 75
 c Pion mesh tally
 RMESH71:/ flux
 CORA71 -2 2
 CORB71 -70 139i 70
 CORC71 -15 89i 75
 c Photon mesh tally
 RMESH81:p flux
 CORA81 -2 2
 CORB81 -70 139i 70
 CORC81 -15 89i 75
 c Deuteron cylindrical mesh tally
 CMESH91:d flux
 CORA91 0 7i 4
 CORB91 0 119i 60
 CORC91 360
 c Energy deposition in target
 CMESH3 TOTAL
 CORA3 0 7i 4
 CORB3 0 119i 60
 CORC3 360
 ENDM
```
C.2 Quinta assembly

Quinta assembly irradiated with 1 GeV deuterons

MCNPX input file prepared by Nicola Asquith

Cells

Individual uranium rods inside hexagonal lattice

<table>
<thead>
<tr>
<th>Rod ID</th>
<th>X</th>
<th>Y</th>
<th>Z</th>
<th>Rod ID</th>
<th>X</th>
<th>Y</th>
<th>Z</th>
<th>Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>11</td>
<td>2</td>
<td>-2.7</td>
<td>-11</td>
<td>12</td>
<td>u=2</td>
<td>$ Aluminum cladding</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>1</td>
<td>-18.9</td>
<td>-12</td>
<td>u=2</td>
<td>$ Uranium rod</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>4</td>
<td>-0.00129</td>
<td>#11</td>
<td>#12</td>
<td>u=2</td>
<td>$ Space outside rod</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

SECTION 1

Hexagonal lattice placement of rods in section 1 with hole for beam window

16 | 4 | -0.00129 | -15 | lat=2 | u=1 | fill=-5:5 | -5:5 | 0:0 |

Construction materials of section 1

<table>
<thead>
<tr>
<th>Material ID</th>
<th>X</th>
<th>Y</th>
<th>Z</th>
<th>Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>110</td>
<td>2</td>
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<td>111</td>
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<td>111</td>
<td>4</td>
<td>-0.00129</td>
<td>-111</td>
<td>$ The hole in the front plate</td>
</tr>
<tr>
<td>112</td>
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<td>-112</td>
<td>$ Aluminium back plate</td>
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<td>2</td>
<td>-2.7</td>
<td>-120</td>
<td>121</td>
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</tr>
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<td>4</td>
<td>-0.00129</td>
<td>-131</td>
<td>$ Beam window in first section</td>
</tr>
<tr>
<td>140</td>
<td>2</td>
<td>-2.7</td>
<td>-140</td>
<td>$ Cylinder 1 joining Al plates</td>
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<tr>
<td>141</td>
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<td>-2.7</td>
<td>-141</td>
<td>$ Cylinder 2 joining Al plates</td>
</tr>
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<td>-2.7</td>
<td>-142</td>
<td>$ Cylinder 3 joining Al plates</td>
</tr>
<tr>
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<td>-2.7</td>
<td>-143</td>
<td>$ Cylinder 4 joining Al plates</td>
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<td>-0.00129</td>
<td>-150</td>
<td>$ Hole in front sample holder</td>
</tr>
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<td>151</td>
<td>2</td>
<td>-2.7</td>
<td>150</td>
<td>-151</td>
</tr>
<tr>
<td>152</td>
<td>2</td>
<td>-2.7</td>
<td>-152</td>
<td>$ Back sample Al plate</td>
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SECTION 2

Different to section 1 as no beam window

<table>
<thead>
<tr>
<th>Rod ID</th>
<th>X</th>
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<th>Z</th>
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<td>21</td>
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<td>-2.7</td>
<td>-21</td>
<td>22</td>
</tr>
<tr>
<td>22</td>
<td>1</td>
<td>-18.9</td>
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<td>u=5</td>
</tr>
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</table>
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

23 4 -0.00129  #21 #22 u=5 $ Region Outside rod
26 4 -0.00129 -25 lat=2 u=4 fill=-5:5 -5:5 0:0
  4 4 4 4 4 4 4 4 4 4 4
  4 4 4 4 5 5 5 5 5 5 4
  4 4 4 5 5 5 5 5 5 5 4
  4 4 5 5 5 5 5 5 5 5 4
  4 5 5 5 5 5 5 5 5 5 4
  4 5 5 5 5 5 5 4 4 4 4
  4 5 5 5 5 4 4 4 4 4 4
  4 4 4 4 4 4 4 4 4 4 4
c Construction materials of section 2 - Translations of section 1
210 like 112 but trcl=(0 0 2.2)
212 like 112 but trcl=(0 0 13.1)
220 2 -2.7 -220 221 $ Hexagonal aluminum container
221 0 -221 fill=4 $ Inner hexagon filled with u-rods
240 like 140 but trcl=(0 0 13.1) $ Cylinder 1 joining Al plates
241 like 141 but trcl=(0 0 13.1) $ Cylinder 2 joining Al plates
242 like 142 but trcl=(0 0 13.1) $ Cylinder 3 joining Al plates
243 like 143 but trcl=(0 0 13.1) $ Cylinder 4 joining Al plates
252 like 152 but trcl=(0 0 13.1) $ Al sample plate
c-------------------------------------------------------------------------------
c SECTION 3
c Identical to section 2 but translated along Z-axis
310 like 112 but trcl=(0 0 15.3)
312 like 112 but trcl=(0 0 26.2)
320 like 220 but trcl=(0 0 13.1)
321 like 221 but trcl=(0 0 13.1)
340 like 140 but trcl=(0 0 26.2)
341 like 141 but trcl=(0 0 26.2)
342 like 142 but trcl=(0 0 26.2)
343 like 143 but trcl=(0 0 26.2)
352 like 152 but trcl=(0 0 26.2)
c-------------------------------------------------------------------------------
c SECTION 4
c Identical to section 2 but translated along Z-axis
410 like 112 but trcl=(0 0 28.4)
412 like 112 but trcl=(0 0 39.3)
420 like 220 but trcl=(0 0 26.2)
421 like 221 but trcl=(0 0 26.2)
440 like 140 but trcl=(0 0 39.3)
441 like 141 but trcl=(0 0 39.3)
442 like 142 but trcl=(0 0 39.3)
443 like 143 but trcl=(0 0 39.3)
452 like 152 but trcl=(0 0 39.3)
C.2. Quinta assembly

c SECTION 5
c Identical to section 2 but translated along Z-axis
510 like 112 but trcl=(0 0 41.5)
512 like 112 but trcl=(0 0 52.4)
520 like 220 but trcl=(0 0 39.3)
521 like 221 but trcl=(0 0 39.3)
540 like 140 but trcl=(0 0 52.4)
541 like 141 but trcl=(0 0 52.4)
542 like 142 but trcl=(0 0 52.4)
543 like 143 but trcl=(0 0 52.4)
552 like 152 but trcl=(0 0 52.4)

c Aluminium plates above and below Quinta
601 2 -2.7 -601 $ Bottom plate
602 2 -2.7 -602 $ Top plate

c Lead castle
700 3 -11.34 -700 701 702 $ Castle
702 4 -0.00129 -702 $ Beam window
710 3 -11.34 -710 $ Top lead
711 3 -11.34 -711 $ Block 1 on top
712 3 -11.34 -712 $ Block 2 on top
713 3 -11.34 -713 $ Block 3 on top
714 3 -11.34 -714 $ Block 4 on top
715 3 -11.34 -715 $ Block 5 on top
716 3 -11.34 -716 $ Block 6 on top

c Outside setup

c Air between the five sections and inside the lead castle
c Excludes all cells in this region inc. sample cells
990 4 -0.00129 110 112 120 140 141 142 143 151 152 220 -701
#210 #212 #240 #241 #242 #243 #252
#310 #312 #320 #321 #340 #341 #342 #343 #352
#410 #412 #420 #421 #440 #441 #442 #443 #452
#510 #512 #520 #521 #540 #541 #542 #543 #552
#601
#1010 #1002 #1110 #1102 #1200 #1201 #1202 #1203 #1204 #1205
#1206 #1207 #1208 #1209 #1210 #1211 #1212 #1213 #1214 #1215
#1216 #1217 #1218 #1219 #1220 #1221 #1222 #1223 #1224 #1225
#1226 #1227 #1228 #1300 #1301 #1302 #1303 #1304 #1305 #1306
#1307 #1308 #1309 #1310 #1311 #1312 #1313 #1314 #1315 #1316
#1317 #1318 #1319 #1320 #1321 #1322 #1323 #1324 #1325 #1326
#1327 #1328 #1410 #1402 #1510 #1502

c Air outside lead castle
991 4 -0.00129 700 602 710 711 712 713 714 715 716 -999

c Outside universe

207
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

999 0 999

c

c

--- SAMPLE CELLS ---

c Sample plate 0
1010 4 -0.00129 -1010 $ Y = -4 \text{ cm}$
1002 4 -0.00129 -1002 $ Y = -12 \text{ cm}$

c Sample plate 1
1110 4 -0.00129 -1110 $ Y = -4 \text{ cm}$
1102 4 -0.00129 -1102 $ Y = -12 \text{ cm}$

c Sample plate 2 - Cells placed along entire Y-axis
1200 4 -0.00129 -1200 $ Y = -14 \text{ cm}$
1201 4 -0.00129 -1201 $ Y = -13 \text{ cm}$
1202 4 -0.00129 -1202 $ Y = -12 \text{ cm}$
1203 4 -0.00129 -1203 $ Y = -11 \text{ cm}$
1204 4 -0.00129 -1204 $ Y = -10 \text{ cm}$
1205 4 -0.00129 -1205 $ Y = -9 \text{ cm}$
1206 4 -0.00129 -1206 $ Y = -8 \text{ cm}$
1207 4 -0.00129 -1207 $ Y = -7 \text{ cm}$
1208 4 -0.00129 -1208 $ Y = -6 \text{ cm}$
1209 4 -0.00129 -1209 $ Y = -5 \text{ cm}$
1210 4 -0.00129 -1210 $ Y = -4 \text{ cm}$
1211 4 -0.00129 -1211 $ Y = -3 \text{ cm}$
1212 4 -0.00129 -1212 $ Y = -2 \text{ cm}$
1213 4 -0.00129 -1213 $ Y = -1 \text{ cm}$
1214 4 -0.00129 -1214 $ Y = 0 \text{ cm}$
1215 4 -0.00129 -1215 $ Y = 1 \text{ cm}$
1216 4 -0.00129 -1216 $ Y = 2 \text{ cm}$
1217 4 -0.00129 -1217 $ Y = 3 \text{ cm}$
1218 4 -0.00129 -1218 $ Y = 4 \text{ cm}$
1219 4 -0.00129 -1219 $ Y = 5 \text{ cm}$
1220 4 -0.00129 -1220 $ Y = 6 \text{ cm}$
1221 4 -0.00129 -1221 $ Y = 7 \text{ cm}$
1222 4 -0.00129 -1222 $ Y = 8 \text{ cm}$
1223 4 -0.00129 -1223 $ Y = 9 \text{ cm}$
1224 4 -0.00129 -1224 $ Y = 10 \text{ cm}$
1225 4 -0.00129 -1225 $ Y = 11 \text{ cm}$
1226 4 -0.00129 -1226 $ Y = 12 \text{ cm}$
1227 4 -0.00129 -1227 $ Y = 13 \text{ cm}$
1228 4 -0.00129 -1228 $ Y = 14 \text{ cm}$

c Sample plate 3 - Cells placed along entire Y-axis
1300 4 -0.00129 -1300 $ Y = -14 \text{ cm}$
1301 4 -0.00129 -1301 $ Y = -13 \text{ cm}$
1302 4 -0.00129 -1302 $ Y = -12 \text{ cm}$
1303 4 -0.00129 -1303 $ Y = -11 \text{ cm}$
1304 4 -0.00129 -1304 $ Y = -10 \text{ cm}$
1305 4 -0.00129 -1305 $ Y = -9 \text{ cm}$
### C.2. Quinta assembly

<table>
<thead>
<tr>
<th>Layer</th>
<th>Type</th>
<th>Height</th>
<th>Width</th>
<th>$Y$</th>
<th>Notes</th>
</tr>
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<tbody>
<tr>
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<td>1308</td>
<td>4</td>
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<td>$Y =  0$ cm</td>
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</tr>
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<td>4</td>
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<td>-1315</td>
<td>$Y =  1$ cm</td>
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<td>-1316</td>
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</tr>
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<td>$Y = 11$ cm</td>
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<td>$Y = 12$ cm</td>
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<tr>
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<td>-1327</td>
<td>$Y = 13$ cm</td>
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#### Sample plate 4
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<td>-1402</td>
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#### Sample plate 5
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<tr>
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<td>-1502</td>
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#### Outside samples
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<th>Notes</th>
</tr>
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<td>-2012</td>
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<td>2013</td>
<td>4</td>
<td>-0.00129</td>
<td>-2013</td>
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<td>2014</td>
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#### Surfaces

**SECTION 1 - Beam along Z-axis, z=0 will be front of section 1**

<table>
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<tr>
<th>Layer</th>
<th>Type</th>
<th>Height</th>
<th>Width</th>
<th>$Y$</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>11</td>
<td>RCC</td>
<td>0 0 0 0.5 0 0 10.4 1.8</td>
<td>Al cladding around U rod</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>RCC</td>
<td>0 0 0.63 0 0 10.14 1.67</td>
<td>Uranium rods</td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>HEX</td>
<td>0 0 0.5 0 0 10.4 1.8 0 0</td>
<td>Lattice hexagon - U rod pitch</td>
<td></td>
<td></td>
</tr>
<tr>
<td>110</td>
<td>RPP</td>
<td>-17.5 17.5 -17.5 17.5 0 0.5</td>
<td>Aluminium front plate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>111</td>
<td>RCC</td>
<td>0 0 0 0 0 0.5 4</td>
<td>Hole in first plate for target</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

112 RPP -17.5 17.5 -17.5 17.5 10.9 11.4 $ Aluminium back plate
120 HEX 0 0 0.5 0 10.4 0 14.77 0 $ Outer aluminium hexagon
121 HEX 0 0 0.5 0 10.4 0 14.27 0 $ Inner aluminium hexagon
130 RCC 0 0 0.5 0 10.4 4.43 $ Outer cylinder for beam window
131 RCC 0 0 0.5 0 10.4 4.00 $ Inner cylinder for beam window
140 RCC 15.5 15.5 0.5 0 10.4 0.5 $ Cylinder 1 joining Al plates
141 RCC -15.5 15.5 0.5 0 10.4 0.5 $ Cylinder 2 joining Al plates
142 RCC -15.5 -15.5 0.5 0 10.4 0.5 $ Cylinder 2 joining Al plates
143 RCC 15.5 -15.5 0.5 0 10.4 0.5 $ Cylinder 2 joining Al plates
150 RCC 0 0 -0.2 0 0 0.2 3.8 $ Hole in first sample plate
151 RPP -4.2 4.2 -17.5 19.0 -0.2 0 $ Front plate
152 RPP -4.2 4.2 -17.5 19.0 11.5 11.7 $ Back plate

c ---------------------------------------------------------------
c SECTION 2,3,4,5
21 RCC 0 0 13.6 0 0 10.4 1.8 $ Al cladding around rod
22 RCC 0 0 13.74 0 0 10.12 1.66 $ Uranium rods
25 HEX 0 0 13.6 0 0 10.4 1.8 0 0 $ Lattice hexagon
220 HEX 0 0 13.6 0 0 10.4 0 14.77 0 $ Outer aluminium hexagon
221 HEX 0 0 13.6 0 0 10.4 0 14.27 0 $ Inner aluminium hexagon
c ---------------------------------------------------------------
c Aluminium plates top and bottom
601 RPP -20 20 -20 -17.5 -3.1 66.9 $ Bottom plate
602 RPP -30 30 21.6 -17.1 80.9 $ Top plate
c Lead castle
700 RPP -30 30 -30 20 -13.1 76.9 $ Outer wall
701 RPP -20 20 -20 20 -3.1 66.9 $ Inner wall
702 RPP -7.5 7.5 -7.5 7.5 -13.1 -3.1 $ Hole for beam window
710 RPP -30 30 21.6 31.6 -13.1 76.9 $ Top plate
711 RPP -5 5 31.6 36.6 -2.5 2.5 $ Block 1 lid
712 RPP -5 5 31.6 41.6 10.6 15.6 $ Block 2 lid
713 RPP -5 5 31.6 41.6 23.7 28.7 $ Block 3 lid
714 RPP -5 5 31.6 41.6 36.8 41.8 $ Block 4 lid
715 RPP -5 5 31.6 41.6 49.9 54.9 $ Block 5 lid
716 RPP -5 5 31.6 36.6 63.0 68.0 $ Block 6 lid
c Outside box to cease particle transport
999 RPP -34 34 -39 49 -19 84
c
c c SAMPLE CELLS

c Sample plate 0
1010 RPP -0.5 0.5 -4.5 -3.5 -1.2 -0.2 $ Y = -4 cm
1002 RPP -0.5 0.5 -12.5 -11.5 -1.2 -0.2 $ Y = -12 cm
c Sample plate 1
1110 RPP -0.5 0.5 -4.5 -3.5 11.9 12.9 $ Y = -4 cm
1102 RPP -0.5 0.5 -12.5 -11.5 11.9 12.9 $ Y = -12 cm
c Sample plate 2 - Cells placed along entire Y-axis
1200 RPP -0.5 0.5 -14.5 -13.5 25.0 26.0 $ Y = -14 cm

210
<table>
<thead>
<tr>
<th>RPP</th>
<th>X1</th>
<th>X2</th>
<th>Y1</th>
<th>Y2</th>
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</tr>
</thead>
<tbody>
<tr>
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<td>-13.5</td>
<td>-12.5</td>
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</tr>
<tr>
<td>1202</td>
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<td>0.5</td>
<td>-12.5</td>
<td>-11.5</td>
<td>25.0</td>
<td>26.0</td>
</tr>
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<td>0.5</td>
<td>-11.5</td>
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**Sample plate 3 - Cells placed along entire Y-axis**

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</table>

*Y coordinates are given in cm.*

**C.2. Quinta assembly**
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

1318 RPP -0.5 0.5 3.5 4.5 38.1 39.1 $ Y = 4 cm
1319 RPP -0.5 0.5 4.5 5.5 38.1 39.1 $ Y = 5 cm
1320 RPP -0.5 0.5 5.5 6.5 38.1 39.1 $ Y = 6 cm
1321 RPP -0.5 0.5 6.5 7.5 38.1 39.1 $ Y = 7 cm
1322 RPP -0.5 0.5 7.5 8.5 38.1 39.1 $ Y = 8 cm
1323 RPP -0.5 0.5 8.5 9.5 38.1 39.1 $ Y = 9 cm
1324 RPP -0.5 0.5 9.5 10.5 38.1 39.1 $ Y = 10 cm
1325 RPP -0.5 0.5 10.5 11.5 38.1 39.1 $ Y = 11 cm
1326 RPP -0.5 0.5 11.5 12.5 38.1 39.1 $ Y = 12 cm
1327 RPP -0.5 0.5 12.5 13.5 38.1 39.1 $ Y = 13 cm
1328 RPP -0.5 0.5 13.5 14.5 38.1 39.1 $ Y = 14 cm
c Sample plate 4
1410 RPP -0.5 0.5 -4.5 -3.5 51.2 52.2 $ Y = -4 cm
1402 RPP -0.5 0.5 -12.5 -11.5 51.2 52.2 $ Y = -12 cm
c Sample plate 5
1510 RPP -0.5 0.5 -4.5 -3.5 64.3 65.3 $ Y = -4 cm
1502 RPP -0.5 0.5 -12.5 -11.5 64.3 65.3 $ Y = -12 cm
c Outside samples
2011 RPP 30.0 31.0 -3.0 -2.0 1.4 2.4
2012 RPP 30.0 31.0 -3.0 -2.0 11.4 12.4
2013 RPP 30.0 31.0 -3.0 -2.0 21.4 22.4
2014 RPP 30.0 31.0 -3.0 -2.0 31.4 32.4
2015 RPP 30.0 31.0 -3.0 -2.0 41.4 42.4
2016 RPP 30.0 31.0 -3.0 -2.0 51.4 52.4
2017 RPP 30.0 31.0 -3.0 -2.0 61.4 62.4
c Materials
c Natural uranium
m1 92234.70c 0.0055
   92235.70c 0.72
   92238.70c 99.2745
   cond=1
c Aluminium
m2 13027.70c 1.
   cond=1
c Natural lead
m3 82204.70c 1.4
   82206.70c 24.1
   82207.70c 22.1
   82208.70c 52.4
   cond=1
c Air
m4 7014.70c 78.084
   8016.70c 20.946
   18038.70c 0.467

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C.2. Quinta assembly

gas=1

c Materials for detectors - Not used in construction
m5 90232.70c 1 $ Thorium
m6 79197.70c 1. $ Gold
m7 83209.00c 1. $ Bismuth

c Source definition

c 1 GeV deuteron beam irradiated 2 degrees from target axis
c Gaussian with centre X = 1.3, Y = -0.2 and shape FWHM_X = 2.6, FWHM_Y = 3.5
sdef par=d erg=1000 x=d1 y=d2 z=-13.1 sur=700.6 vec=-0.0349 0 1 dir=1
sp1 -41 2.6 1.3
sp2 -41 3.5 -0.2
nps 100000 $ Required number of particle histories
c dbcn 7j 1786904 $ For debugging

c Physics Options

c Transporting neutron, proton, deuteron, charged pion, neutral pion, muon
c photon, and kaon
mode n h d / z | p k
imp:n,h,p,/,z,|,k 2 57r 0.5 0.5 0.25 0.5 0.125 0.0625 5r 1 0.125 0 4 72r
imp:d 1 70r 0 1 72r
c ACT fission=all nonfiss=all dn=both dg=mg $ For creating delayed gammas
c DXT:n 0 -12 64.7 2 4 e-4 1e-4
c DD1 -1e-4

c Particle cut-off energies
phys:n 1010
phys:h 1010
phys:p 1010 2j -1 $ Turn on photonuclear physics
phys:/ 1010
phys:d 1010
phys:z 1010
phys:| 1010
phys:k 1010

cut:n 2j 0 0 $ Force analog capture
cut:h j 0.001 0 0 $ Set cut-offs to 1 keV
cut:d j 0.001 0 0
cut:/ j 0.001 0 0
cut:| j 0.001 0 0
cut:k j 0.001 0 0

c Physics models

c lca 8j 1 1 $CEM03/LAQGSM
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

lca 6j 1 j 2 $INCL4
lea 1 5j 2 j $ABLA
c histp -500000000 1010 1002 1110 1102 1210 1202 1310 1302 1410 1402 1510 1502

c Tallies (for controlling contents of output file)

c Calculating bismuth reaction rates for activation samples

fc4 Bismuth n4n reaction - TENDL library $ Tally name card
f4:n 1010 1110 1210 1310 1410 1510 $ Cells for tallying

read file=dosefunctions/tendl_bi_n4n.d noecho
e4 200 1010 $ Energy binning
fq4 F E $ Print hierarchy

c
c14 Bismuth n4n reaction - TALYS calculation
f14:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_bi_n4n.d noecho
e14 250 1010
fq14 F E

c
c24 Bismuth n5n reaction - TALYS calculation
f24:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_bi_n5n.d noecho
e24 250 1010
fq24 F E

c
c34 Bismuth n6n reaction - TALYS calculation
f34:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_bi_n6n.d noecho
e34 250 1010
fq34 F E

c
c44 Bismuth n7n reaction - TALYS calculation
f44:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_bi_n7n.d noecho
e44 250 1010
fq44 F E

c
c54 Bismuth (p,X) Bi206 reaction
f54:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_px_206.d noecho
e54 250 1010
fq54 F E

c
c64 Bismuth (p,X) Bi205 reaction
f64:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_px_205.d noecho
C.2. Quinta assembly

```plaintext
e64 250 1010
f64 F E

fc74 Bismuth (p,X) Bi204 reaction
f74:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_px_204.d noecho
e74 250 1010
f74 F E

fc84 Bismuth (p,X) Bi203 reaction
f84:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_px_203.d noecho
e84 250 1010
f84 F E

fc94 Bismuth (d,X) Bi206 reaction
f94:d 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_dx_206.d noecho
e94 250 1010
f94 F E

fc104 Bismuth (d,X) Bi205 reaction
f104:d 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_dx_205.d noecho
e104 250 1010
f104 F E

fc114 Bismuth (d,X) Bi204 reaction
f114:d 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_dx_204.d noecho
e114 250 1010
f114 F E

fc124 Bismuth (d,X) Bi203 reaction
f124:d 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_bi_dx_203.d noecho
e124 250 1010
f124 F E

Calculating gold reaction rates for activation samples
fc204 Gold (n,g) - ENDF/B-VII.0 data and FM card
f204:n 1010 1110 1210 1310 1410 1510
fm204 (1e3 6 102) $ Flux multiplier card
e204 30 1010
f204 F E

fc214 Gold n2n reaction - TALYS data
```

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APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

```
f214:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_n2n.d noecho
e214 250 1010
fq214 F E
c
fc224 Gold n3n reaction - TENDL data
f224:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/tendl_au_n3n.d noecho
e224 200 1010
fq224 F E
c
fc234 Gold n4n reaction - TALYS data
f234:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_n4n.d noecho
e234 250 1010
fq234 F E
c
fc244 Gold n5n reaction - TALYS data
f244:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_n5n.d noecho
e244 250 1010
fq244 F E
c
fc254 Gold n6n reaction - TALYS data
f254:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_n6n.d noecho
e254 250 1010
fq254 F E
c
fc264 Gold n7n reaction - TALYS data
f264:n 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_n7n.d noecho
e264 250 1010
fq264 F E
c
fc274 Gold (p,X) Au196 reaction - TENDL data
f274:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/tendl_au_px_196.d noecho
e274 200 1010
fq274 F E
c
fc284 Gold (p,X) Au195 reaction - TALYS data
f284:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_px_195.d noecho
e284 250 1010
fq284 F E
c
```
C.2 Quinta assembly

fc294 Gold (p, X) Au194 reaction
f294:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_au_px_194.d noecho
e294 250 1010
fq294 F E
c
fc304 Gold (p, X) Au193 reaction
f304:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/exfor_au_px_193.d noecho
e304 250 1010
fq304 F E
c
fc314 Gold (p, X) Au192 reaction
f314:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_px_192.d noecho
e314 250 1010
fq314 F E
c
fc324 Gold (p, X) Au191 reaction
f324:h 1010 1110 1210 1310 1410 1510
read file=dosefunctions/talys_au_px_191.d noecho
e324 250 1010
fq324 F E
c
 Lead Fission calculations
fc404 Lead Neutron Fission
f404:n 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
  1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
  1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
  1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
  1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Pb_nf.d noecho
fq404 F E
c
fc504 Lead photofission
f504:p 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
  1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
  1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
  1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
  1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Pb_gf.d noecho
fq504 F E
c
fc604 Lead proton fission
f604:h 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
  1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
  1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
  1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
  1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Pb_pf.d noecho
fq604 F E
217
read file=XS/Pb_pf.d noecho
fq604 F E
c
fc704 Lead pion+ fission
f704:/ 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Pb+_f.d noecho
fq704 F E
c
fc804 Lead deuteron fission
f804:d 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Pb_df.d noecho
fq804 F E
c
f414 Bismuth Neutron Fission
fc414 Bismuth Fission calculations
f414:n 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Bi_nf.d noecho
fq414 F E
c
fc514 Bismuth photofission
f514:p 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Bi_gf.d noecho
fq514 F E
c
fc614 Bismuth proton Fission
f614:h 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
218
C.2. Quinta assembly

read file=XS/Bi_pf.d noecho

fc714 Bismuth pion+ Fission

f714:/ 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209 1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223 1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308 1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322 1323 1324 1325 1326 1327 1328 1410 1402 1510 1502

read file=XS/Bi_+f.d noecho

fc814 Bismuth deuteron Fission

f814:d 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209 1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223 1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308 1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322 1323 1324 1325 1326 1327 1328 1410 1402 1510 1502

read file=XS/Bi_df.d noecho

fc914 Gold Fission calculations

fc424 Gold Neutron Fission

f424:n 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209 1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223 1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308 1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322 1323 1324 1325 1326 1327 1328 1410 1402 1510 1502

read file=XS/Au_nf.d noecho

fc524 Gold photofission

f524:p 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209 1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223 1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308 1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322 1323 1324 1325 1326 1327 1328 1410 1402 1510 1502

read file=XS/Au_gf.d noecho

fc624 Gold proton Fission

f624:h 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209 1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223 1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308 1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322 1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

read file=XS/Au_pf.d noecho
fq624 F E
c
fc724 Gold pion+ Fission
f724:/ 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
       1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
       1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
       1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
       1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Au+_f.d noecho
fq724 F E
c
fc824 Gold deuteron Fission
f824:d 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
       1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
       1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
       1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
       1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Au_df.d noecho
fq824 F E

---

c Thorium Fission calculations

 fc434 High Energy Thorium Fission
f434:n 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
       1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
       1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
       1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
       1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Th_nf.d noecho
fq434 F E
e434 60 1010
c
fc454 Low Energy Thorium Fission
f454:n 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
       1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
       1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
       1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
       1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
fm454 (1e3 5 18)
e454 60 1010
fq454 F E
c
fc534 Thorium photofission
f534:p 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
       1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
read file=XS/Th_gf.d noecho
fq534 F E
c fc634 Thorium proton fission
f634:h 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Th_pf.d noecho
fq634 F E
c fc734 Thorium pion+ fission
f734:/ 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Th_+f.d noecho
fq734 F E
c fc834 Thorium deuteron fission
f834:d 1010 1002 1110 1102 1200 1201 1202 1203 1204 1205 1206 1207 1208 1209
1210 1211 1212 1213 1214 1215 1216 1217 1218 1219 1220 1221 1222 1223
1224 1225 1226 1227 1228 1300 1301 1302 1303 1304 1305 1306 1307 1308
1309 1310 1311 1312 1313 1314 1315 1316 1317 1318 1319 1320 1321 1322
1323 1324 1325 1326 1327 1328 1410 1402 1510 1502
read file=XS/Th_df.d noecho
fq834 F E
c Uranium Fission calculations
fc444 High Energy uranium Fission
f444:n 1206 1223 1306 1323
read file=XS/Us_nf.d noecho
e444 30 4010
fq444 F E
c fc464 Low Energy uranium Fission
f464:n 1206 1223 1306 1323
fm464 (1e3 1 18)
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

e464  30 4010
fq464 F E

c
fc544 Uranium photofission
f544:p 1206 1223 1306 1323
read  file=XS/U_gf.d noecho
fq544 F E

c
fc644 Uranium proton Fission
f644:h 1206 1223 1306 1323
read  file=XS/U_pf.d noecho
fq644 F E

-- Particle spectra

-- Particle spectra in sample locations
fc914 Neutron Energy Spectrum in sample locations
f914:n 1010 1002 1110 1102 1210 1202 1310 1302 1410 1402 1510 1502
fq914 E F
e914 1e-3 119log 1e3

c
fc924 Photon Energy Spectrum in sample locations
f924:p 1010 1002 1110 1102 1210 1202 1310 1302 1410 1402 1510 1502
fq924 E F
e924 1e-3 119log 1e3

c
fc934 Proton Energy Spectrum in sample locations
f934:h 1010 1002 1110 1102 1210 1202 1310 1302 1410 1402 1510 1502
e934 1e-3 119log 1e3
fq934 E F

c
fc944 P+/− Energy Spectrum in sample locations
f944:/ 1010 1002 1110 1102 1210 1202 1310 1302 1410 1402 1510 1502
e944 1e-3 119log 1e3
fq944 E F

c
fc954 Deuteron Energy Spectrum in sample locations
f954:d 1010 1002 1110 1102 1210 1202 1310 1302 1410 1402 1510 1502
e954 1e-3 119log 1e3
fq954 E F

-- Particle spectra leakage

fc911 Neutron Energy Spectrum leakage
f911:n (999.1 999.2 999.3 999.4 999.5 999.6)
fq911 E F
e911 1e-3 119log 1e3

c

222
fc921 Photon Energy Spectrum leakage  
f921:p (999.1 999.2 999.3 999.4 999.5 999.6)  
e921 1e-3 119log 1e3  

c fc931 Proton Energy Spectrum leakage  
f931:h (999.1 999.2 999.3 999.4 999.5 999.6)  
e931 1e-3 119log 1e3  

c fc941 P+/- Energy Spectrum leakage  
f941:/ (999.1 999.2 999.3 999.4 999.5 999.6)  
e941 1e-3 119log 1e3  

c fc951 Deuteron Energy Spectrum leakage  
f951:d (999.1 999.2 999.3 999.4 999.5 999.6)  
e951 1e-3 119log 1e3  

prdm 2j -1 $ Creates MCTAL file containing mesh tally data  

C.2. Quinta assembly
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

CORB61 -100 99i 100
CORC61 -70 99i 130
ENDMD
C.3 HPGe detector efficiency

Efficiency of A Canberra Detector at P2 (326 kev)

MCNPX Input file prepared by Nicola Asquith

Cells

<table>
<thead>
<tr>
<th>Number</th>
<th>X</th>
<th>Y</th>
<th>Z</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>600</td>
<td>-5.323</td>
<td>-11.22</td>
<td>(-35 : -36 : -33 ) $ Sensitive volume of detector</td>
</tr>
<tr>
<td>11</td>
<td>408</td>
<td>-0.534</td>
<td>(-23 : 21 ):(-22 : 21 ) $ Lithium electrode</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(-12.2 : 11.2 : -36 ):(35 : 36 : -34 : 33 )</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>601</td>
<td>-1.397</td>
<td>-31</td>
<td>$ Mylar layer</td>
</tr>
<tr>
<td>14</td>
<td>603</td>
<td>-1.43</td>
<td>-32</td>
<td>$ Kapton layer</td>
</tr>
<tr>
<td>30</td>
<td>208</td>
<td>-2.699</td>
<td>(-60 : 62 ):(-61 : 62 ) $ Aluminium base</td>
<td></td>
</tr>
<tr>
<td>31</td>
<td>464</td>
<td>-2.25</td>
<td>-62 : 70</td>
<td>$ Teflon core holder</td>
</tr>
<tr>
<td>40</td>
<td>208</td>
<td>-2.699</td>
<td>-70</td>
<td>$ Core signal contact</td>
</tr>
<tr>
<td>50</td>
<td>208</td>
<td>-2.699</td>
<td>80 : -81 82</td>
<td>$ Endcap</td>
</tr>
<tr>
<td>51</td>
<td>212</td>
<td>-1.85</td>
<td>-82</td>
<td>$ Be cryostat window</td>
</tr>
<tr>
<td>60</td>
<td>0</td>
<td>-12.2 : -41 : 57 #10 #11 #12 #40</td>
<td>$ Vacuum gap</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>0</td>
<td>(-80 : 57 : (51 : 42 #20 )):(-80 : -57 : 60 : 61 )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>71</td>
<td>456</td>
<td>-0.93</td>
<td>-90 : 81</td>
<td>$ Endcap protector</td>
</tr>
<tr>
<td>101</td>
<td>468</td>
<td>-1.406</td>
<td>101 : -102</td>
<td>$ Sample rack</td>
</tr>
<tr>
<td>102</td>
<td>468</td>
<td>-1.406</td>
<td>-110</td>
<td>$ Sample rack plate</td>
</tr>
<tr>
<td>110</td>
<td>208</td>
<td>-2.699</td>
<td>-120</td>
<td>$ Aluminium plate</td>
</tr>
<tr>
<td>111</td>
<td>602</td>
<td>-8.94</td>
<td>-121</td>
<td>$ Copper plate on top</td>
</tr>
<tr>
<td>112</td>
<td>602</td>
<td>-8.94</td>
<td>-122</td>
<td>$ Copper plate on bottom</td>
</tr>
<tr>
<td>120</td>
<td>406</td>
<td>-11.35</td>
<td>-130</td>
<td>$ Upper lead house</td>
</tr>
<tr>
<td>121</td>
<td>406</td>
<td>-11.35</td>
<td>-131 : 132 : 133 : 134</td>
<td>$ Lower lead house</td>
</tr>
<tr>
<td>130</td>
<td>318</td>
<td>-8.65</td>
<td>-132 : 140 : 142</td>
<td>$ Cadmium box</td>
</tr>
<tr>
<td>900</td>
<td>456</td>
<td>-0.93</td>
<td>-900</td>
<td>$ Circular Sample</td>
</tr>
<tr>
<td>901</td>
<td>604</td>
<td>-1.17</td>
<td>900 : -901</td>
<td>$ Plexiglass sample plate</td>
</tr>
<tr>
<td>910</td>
<td>204</td>
<td>-0.001225</td>
<td>81 : -131 #71 #101 #102 #111 #121 #130 #900 #901 #980</td>
<td>$ Outside end cap</td>
</tr>
<tr>
<td>911</td>
<td>204</td>
<td>-0.001225</td>
<td>-999 : 120 : 130 : 131 #101</td>
<td></td>
</tr>
<tr>
<td>980</td>
<td>204</td>
<td>-0.001225</td>
<td>-101 : 901.6 : 81 : #71 #900</td>
<td></td>
</tr>
<tr>
<td>999</td>
<td>0</td>
<td>999</td>
<td></td>
<td>$ Outside universe</td>
</tr>
</tbody>
</table>

Surfaces

Detector and dead layers

<table>
<thead>
<tr>
<th>Number</th>
<th>Surface Type</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>11</td>
<td>rcc</td>
<td>0 0 0.05 0 0 4.85 2.4 $ Main detector cylinder</td>
</tr>
<tr>
<td>12</td>
<td>rcc</td>
<td>0 0 0 0 0 4.90003 $ Boron dead layer</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.40003</td>
</tr>
</tbody>
</table>
APPENDIX C. EXAMPLE OF MCNPX INPUT FILES

21  rcc 0 0 0 0 0 3.45 0.45 $ Inner Hole (Empty)
22  rcc 0 0 0.05 0 0 3.45 0.5 $ Outer Hole (Lithium)
23  rcc 0 0 0 0 0.05 2.40003 $ Dead layer at back
31  rcc 0 0 4.90003 0 0 0.00085 2.45 $ Mylar layer
32  rcc 0 0 4.90088 0 0 0.01 2.45 $ Kapton layer
33  tz 0 0 4.3 1.8 0.6 0.6 $ Detector donut
34  tz 0 0 4.3 1.8 0.60003 0.60003 $ Detector donut
35  pz 4.3 $ Detector plane
36  cz 1.8 $ Plane for top of donut

c Aluminium casing
41  cz 2.45
42  cz 2.53
43  cz 2.72
51  pz 4.91088
52  pz 4.82
53  pz 3.65
54  pz 2.79
55  pz 1.62
56  pz 0.76
57  pz -1.8

c Electronics
60  rcc 0 0 -1.8 0 0 -0.32 2.53 $ Aluminium base plate
61  rcc 0 0 -2.12 0 0 -2.9 1.25 $ Aluminium base cylinder
62  rcc 0 0 -1.8 0 0 -2.22 0.75 $ Teflon core holder
70  rcc 0 0 -3.4 0 0 6.7 0.3 $ Core signal contact

c End cap
80  rcc 0 0 -7.65 0 0 13.45 3.6
81  rcc 0 0 -7.80 0 0 13.75 3.75
82  rcc 0 0 5.95 0 0 -0.05 3.4

c Plastic cover on end cap
90  rcc 0 0 6.15 0 0 -0.5 3.95 $ Outer cylinder
91  rcc 0 0 6.15 0 0 -0.3 3.75 $ Inner cylinder

c Sample Rack
101  rpp -4.25 4.25 -4 3 -5 50 $ Inner rack
102  rpp -5.25 5.25 -7 3 -5 50 $ Outer rack
110  rpp -18.75 18.75 -8.8 -8.8 -31.75 19.75 $ Rack plate

c Detector housing
120  rpp -28 29 19 20.8 -40.25 28.25 $ Aluminium top plate
121  rpp -19.4 20.4 18.8 19.9 -31.65 19.65 $ Copper plate on top
122  rpp -19.4 20.4 -8.9 -8.8 -31.65 19.65 $ Copper plate on bottom
130  rpp -27 28 20.8 28.3 -39.25 27.25 $ Upper lead box
131  rpp -27 28 -16.5 19 -39.25 27.25 $ Lower lead box (outer)
132  rpp -19.5 20.5 -9 19 -31.75 19.75 $ Lower lead box (inner)
133  rpp -2 3 -9 19 -39.25 -31.75 $ Lower lead box (exit)
134  rpp -10 8 -9 19 19.75 27.25 $ Lower lead box (entrance)
140  rpp -19.4 20.4 -8.9 18.9 -31.65 19.65 $ Cadmium box inner
142  rpp -10 8 -9 19 19.65 19.75 $ Cadmium box entrance gap
C.3. HPGe detector efficiency

Outside cells:
- 900 rcc 0 0 7.275 0 0 0.15 1 $ Polyethylene sample holder
- 901 rpp -4.25 4.25 -4 4.5 7.35 7.85 $ PVC sample holder
- 999 sph 0 5 5 70

Materials:
- m204 7000 -0.755636 $ Air
- m220 5000 1 $ Boron (natural)
- m408 3000 1 $ Lithium
- m600 32000 1 $ Germanium
- m208 13000 1 $ Aluminum
- m468 1000 -0.048382 $ Polyvinyl Chloride
- m456 1000 -0.143716 $ Polyethylene
- m464 6000 -0.240183 $ Teflon
- m212 4000 1 $ Beryllium metal
- m601 1000 8 $ Aluminised Mylar
- m603 1000 10 $ Kapton
- m406 82000 1 $ Lead
- m318 48000 1 $ Cadmium
- m602 29000 1 $ Copper
- m604 1000 6 $ Plexiglass

Physics Options:
- mode p e
- imp:p 4 3 1 3 2 1r $ 10, 20
  1 1r 2 1 1r 2 1 1r $ 30, 71
  0.5 1r 0.2 1 0.3 0.2 1r $ 101, 121
  0.3 1 0.5 1r 0 1 $ 130, 980
  0 $ 999
- imp:e 1 13r 0 2r 1 0 3r 1 1r $ 10, 901
  0 1r 1 0 $ 910, 999
cut:p 2j 0 0 $ Analog capture
fcl:p 1 0 26r $ Forced collision

c Source definition

sdef pos=0 0 7.35 par=p axs=0 0 1 rad=d1 vec=0 0 -1 dir=d2 erg=0.326
si1 0 0.05
sp1 -21 1
si2 -1 0 1
sp2 0 0.5 0.5
sb2 0 0 1

c Pulse height tally

f8:p 10
e8 0 12.5e-3 0.3259 0.3261 3.1
stop f8 0.01