MARGINAL LEAKAGE AND ADAPTATION
OF COMPOSITE RESIN RESTORATIONS

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INTRODUCTION

The lack of adaptation between restorative materials and tooth structure has for many years been a major problem in Dentistry. Studies during this time have emphasized that the margins of restorations are not fixed, inert and impenetrable borders, but dynamic microcrevices which contain a busy traffic of ions and molecules (Going, 1972). The microleakage which results, has been defined as "the clinically undetectable passage of bacteria, fluids and molecules or ions between a cavity wall and the restorative materials applied to it" (Kidd, 1976, b). Microleakage has been implicated in a variety of clinical conditions, including recurrent caries, tooth discolouration, hypersensitivity, pulp pathology and hastened breakdown of restorative materials (Kidd, 1976, a and b; Torney et al, 1977).

Buonocore (1955) was probably the first to bond resins to tooth structure, etched by the application of phosphoric acid to enamel surfaces. It was found that etching the enamel converted a previously hydrophobic, low energy surface to a more wettable high energy state
possessing an increased surface area with numerous retentive sites for resin tag formation.

Over the years the acid-etch technique has been used in a number of dental procedures including fissure sealing, the repair of fractured incisal edges, the repair of hypoplastic and abraded areas, the placement of orthodontic brackets and the splinting of teeth.

In recent years, studies have evaluated the ability of the acid-etch technique to improve both the adaptation and retention of composite resins to tooth structure and the marginal seal around composite restorations. However, much of the research examining the adaptation of composite resins to tooth enamel has been limited to the study of the enamel tooth surface rather than the enamel of the cavity wall. In addition, previous studies have paid little attention to the effects, on the microleakage, of different etching times and different sizes of the inorganic filler particles and to the influence of ageing the restorations on the microleakage. The review of the literature also indicated some controversy concerning the use and effectiveness of low viscosity resins.
Since the early 1970's, acid-etching of the enamel has been used increasingly in the placement of composite resin restorations, to provide retention and to minimize marginal microleakage. It was the purpose of this investigation to examine the etched enamel cavity wall, to compare the ability of composite resins, of different particle size and viscosity, to adapt to this cavity wall, and to study the influence, *in vitro*, of these and other factors, on the microleakage occurring at the margins of restorations.
REVIEW OF THE LITERATURE
CHAPTER 1

SOME STRUCTURAL ASPECTS OF ENAMEL

The outer portion of the anatomic crown of a tooth is composed of enamel, an acellular calcified material of epithelial origin, which has been described as the hardest tissue found in the body. The protein matrix, as secreted by the ameloblasts, is entirely organic and is a keratin-related material. As it becomes mineralised, hydroxyapatite precipitates, replacing the matrix until the final composition of enamel is approximately 0.5 per cent organic, 4 per cent water and 95.5 per cent mineral. In addition to the major inorganic component, hydroxyapatite, other inorganic constituents include sodium (1 per cent), magnesium (1 per cent), carbonate (3 per cent), iron, fluorine, and manganate, the last three in lower and more variable concentrations. The main organic components are two proteins, one of which is relatively insoluble and a soluble glycoprotein (Provenza, 1972; Mjor and Pindborg, 1973; Bloom and Fawcett, 1975).

The structural entity of enamel is the prism or rod. The prisms originate at the dentin-enamel junction and it seems probable that most extend through the thickness of the enamel to the tooth surface. The prism is narrowest,
approximately three micrometres in cross section, near the
dentino-enamel junction and widens gradually as it nears the
surface to approximately six micrometres. This widening is due
to the inner surface of the enamel having a smaller area than the
outer surface (Mjor and Pindborg, 1973; Osborn and Ten Cate, 1976).

In recent years it has become appropriate to describe human
enamel as consisting of "keyhole-shaped" prisms tightly locked
together. Previously, enamel prisms were described by light
microscopists as roughly hexagonal or circular prisms separated
by and cemented together with interprismatic "substance" (Poole
and Brooks, 1961; Sicher, 1966; Bloom and Fawcett, 1975), a
material thought to be less mineralised than the body of the prism.
However, electron microscopists have concluded that this "substance"
contains an identical proportion of hydroxyapatite to that of the
prism bodies and it is preferable to use the term interprismatic
"region" (Hardwick et al, 1965; Osborn and Ten Cate, 1976).

The keyhole shape of the enamel prisms allows very close
packing—the relationship of adjoining prisms is such that
between two heads is inserted a tail belonging to a neighbouring
prism. In this arrangement there is no evidence of interprismatic
"substance", because each crystallite (the microscopic unit of the
prism) can be assigned to a specific prism (Meckel et al, 1964 and
1965, b). The keyhole shape of the prisms when cut in cross
section measures approximately five micrometres in diameter across the head and approximately nine micrometres in height from head to tail. The prisms have been found to be oriented so that the heads of the prisms are directed cuspally and the tails directed cervically (Meckel et al, 1965, b).

At the ultramicroscopic level of examination, enamel is seen to be composed of a multitude of long crystallites embedded in a scant organic framework. The crystallites measure between 20 - 30 nanometres in width and 150 nanometres in length (Buonocore, 1975). The crystallites within a single prism do not all lie in the same direction. Those crystallites in the upper head region of the prism lie parallel to the long axis of the prism but, towards the tail, the crystallites increasingly tilt away from this axis. At the lower end of the tail region the long axes of the crystallites deviate away from the long axis of the prism by an angle of approximately 70 degrees.

Enamel sectioned along the prism axis and across the head-tail direction shows the crystallites of the tail region as cross-cut and those of the head region showing their long axes. However, if enamel is sectioned along the prism axis but parallel to the head-tail direction, the crystallites are depicted running along the prism axis at the upper edge of the prism and tipping down
towards the tail throughout the rest of the prism (Meckel et al, 1964 and 1965, b; Griebstein and Helmcke, 1965; Romanuik and Shroff, 1965; Scott et al, 1974; Buonocore, 1975). It is in this way that the sudden changes in crystallite orientation at the boundaries between prisms serve to define the cross-sectional keyhole shape of the individual prisms (Fig. 1.1).

Studies by Johansen (1965) indicated a great variation in crystallite size and shape. He found that those crystallites oriented parallel with the prisms showed predominantly uniform and regular outlines, but those that were angled away from the prism axis were found to display great variations in width and form, often showing protuberances and/or indentations. These findings were confirmed by Gustavson and Silness (1966), who showed that the crystallites at prism junctions have a special form and are of a different size than elsewhere in the tissue.

Enamel prisms radiate from the dentino-enamel junction toward the outer surface of the enamel. However, not all prisms reach the enamel surface (Innes and Shroff, 1966; Scott et al, 1974; Buonocore, 1975; Osborn and Ten Cate, 1976), from which it may be concluded that some ameloblasts die before the full thickness of organic enamel matrix is deposited.
Fig. 1.1. - Model illustrating the arrangement of crystallites in enamel prisms. (Courtesy Meckel et al (1965, b)).
The course of prisms from the dentino-enamel junction to the enamel surface is not straight but "S"-curved, and intertwining. Osborn (1968, b) described a basic model for enamel structure as consisting of a series of one-prism thick, interlocking, open-ended tapered cones, the bases of which constitute the surface enamel and the open ends of which lie on the dentino-enamel junction. Within each cone, the prisms wave from side to side, the bends of the prisms in one cone all being the same and slightly out of phase with the bends of prisms in adjacent cones. This leads to a decussation of rows of prisms (Osborn, 1968, a) which contributes to an optical phenomenon referred to as Hunter-Schreger bands (Buonocore, 1975).

The course of the prisms from their origin to the enamel surface can be divided into three zones (Lyon and Darling, 1957). Prisms occupying the inner zone run a wavy course, then straighten out in the central zone and run straight to the periphery in the outer zone (Romanuk and Shroff, 1965; Buonocore, 1975; Osborn and Ten Cate, 1976).

Enamel prisms have been observed to approach the surface at a variety of angles; for example, some prisms terminate at right angles to the surface, others curve towards the occlusal and others curve in an apical direction. Several textbooks on dental histology have stated that enamel prisms in the cervical two-thirds
of the permanent tooth are apically oriented (Sicher, 1966; Scott and Symons, 1971). This description has influenced the design of proximal cavity preparations. In contrast, Romanuik and Shroff (1965) and Ramsay and Ripa (1969), observed that enamel prisms of the cervical two-thirds were inclined occlusally at the surface, and the prisms of the occlusal one-third were bent apically, whilst most prisms of the incisal tips or occlusal cusps terminated at right angles to the surface.
CHAPTER 2

THE ACID-ETCHING OF ENAMEL - FUNCTIONS AND USES

2.1 The Functions of Acid-Etching the Enamel Surface

The principal purpose of acid-etching the enamel surface is to provide a means of retaining the restorative material without the need for the excessive removal of tooth structure that is frequently required in more conventional restorative techniques. As a direct result of the nature of this acid-etched retention, microscopically close adaptation of the material to the etched tooth surface is achieved. Acid-etching of enamel is, therefore, currently being used either to retain the material or to improve marginal adaptation or to achieve both improved retention and marginal adaptation.

Since the acid-etching technique was first described by Buonocore in 1955, many studies have investigated the effect of acid solutions on enamel (Poole and Brooks, 1961; Poole and Johnson, 1967; Sharpe, 1967; Gwinnett, 1971; Johnson et al, 1971; Hamilton et al, 1971; Jongebloed et al, 1973; Nichol et al, 1973; Retief, 1973, b; Scott et al, 1974). These studies indicated that when acid solutions were applied to enamel surfaces, a selective dissolution of the large inorganic component occurred at sites related to the cores and peripheries of enamel prisms.
The nature of the retention achieved by acid-etching enamel has usually been considered to be a simple mechanical interlocking by resin into pores created in the etched enamel surface. The term "adhesion" has frequently been applied to the state in which the material is retained to the etched surface. The continued use of this term is not appropriate since true adhesion is associated with interfacial bonding through molecular attraction—a phenomenon which is considered to play a negligible role in the retention of composite resin to an acid-etched surface.

Buonocore (1955) and Buonocore et al (1968), suggested that "adhesion" obtained intraorally on treated enamel surfaces might be due to several factors.

1. An increase in enamel surface area for attachment.
2. The exposure of the organic framework of enamel, in and about which the resins can flow.
3. The creation of spaces along "interprismatic" areas into which the resins can penetrate.
4. The removal of the old, inert enamel surface, exposing a fresh, reactive surface more favourable for adhesion.
5. The presence of an adsorbed layer of highly polar phosphate groups derived from the acid used, which may enhance
"adhesion" of the resin through increased "wettability" of the surface. Their discussion suggested the possibility of "adhesion" taking a considerable part in the retention of resins to the acid-etched surface.

The requirements for adhesion involving intermolecular attraction are several. First, only very short distances of separation (0.1 to 0.2 nanometres) should exist between adhesive and adherend, stress concentrations at the interface should be at a minimum, and thirdly, the attack on the interface by environmental factors, such as shrinkage, expansion and moisture should be limited (Gwinnett, 1965; Eick et al, 1972; Arends et al, 1975). Although true adhesive forces at the resin-enamel interface would be ideal, it is unlikely that such forces could be maintained due to the hydrophilic nature of the etched surfaces and the ability of water molecules to penetrate between adhering layers (Retief, 1970). It would appear, therefore, that mechanical bonding of resins to acid-etched enamel is the most likely mechanism of attachment and that the resins flow into irregularly shaped microspaces created by preferential dissolution of enamel prisms (Powell, 1975).
In 1974, the American Dental Association Council on Dental Materials and Devices, issued a statement regarding claims relating to "adhesion". The Council's opinion was that an "adhesive" dental material should bond to enamel "without reliance on interlocking effects". In addition, the Council also ruled that mechanical retention (rather than adhesion) was the predominant mechanism of bonding an unfilled resin to etched enamel.

There are, currently available, some materials and techniques which not only use the acid-treatment of enamel to gain retention and to remove debris from the prepared surface but also achieve some degree of true adhesion in the form of molecular bonding. These include glass ionomer cements and composite resin materials which use a tooth-restoration coupling agent.

Glass ionomer cements, currently available, are in the form of a calcium alumino-silicate glass powder and a poly (acrylic acid)-based liquid, which when combined together result in a hardening reaction between ion-leachable glasses and aqueous solutions of acrylic acid. In the fluid paste stage of the setting, the glass ionomer cements adhere to a number of substrates, including dentine and enamel, probably because of the availability of acid-COOH groups for hydrogen bonding, thereby promoting the wetting of the substrate. It is likely
that as the material hardens and set, the hydrogen bonds are replaced by more rigid metallic bonds, linking the cement firmly to the substrate. Adhesion between glass ionomer cements and substrates probably results from dipole and ionic interactions because glass ionomer cements, dentine and enamel have a polar nature. Clinical application of glass ionomer cements involves the use of a cavity cleansing solution — 50 per cent citric acid (a relatively weak acid) is used for a 30 second treatment.

The prime purpose of this "conditioner" is to provide a clean, easily wettable surface by removal of any proteinaceous debris, without causing damage to surrounding tissues. Stronger acid-etching techniques are both unnecessary and deleterious since the glass ionomer cements adhere to substrates by physico-chemical attractive forces and do not rely on mechanical retention (McLean and Wilson, 1977, a, b and c).

Some techniques use a coupling agent or primer applied to the etched enamel surface to increase the retention and improve the marginal seal of the completed composite resin restoration. The tooth-restoration coupling agents are described as multifunctional molecules that promote adhesion of the solid substrate to the restorative material by absorbing onto and altering the surface of the tooth to facilitate interaction
with the material by either chemical or physical processes. The portion of the molecule that is not absorbed presents a surface that can be more easily wetted by the composite resin (McLean and Wilson, 1977, c). The coupling agent used in the Cosmic* composite resin system, is a surface active comonomer, the reaction product of N-phenylglycine and glycidylmethacrylate (NPG-GMA). It was postulated that the coupling agent, containing polar groups, would be capable of chelating with surface calcium atoms in mineralised, dental tissue and that additional groups would be capable of copolymerising with the resin system of the restorative material (Chandler et al, 1974; Retief, 1975, b). Another example of a coupling agent, is the primer used in the Simulate** composite resin system, which is described as an alcohol solution of an acrylic acid-butyl acrylate (Gritz et al, 1979).

* Cosmic - Amalgamated Dental, De Trey, Wiesbaden, Zurich.

2.2 Clinical Uses of Acid-Etching the Enamel Surface

Since first described in 1955 by Buonocore, acid-pretreatment techniques have been used for a number of dental procedures:-

(i) Fissure Sealing

An increasing emphasis on preventive techniques in Dentistry has resulted in the development of a method of sealing pits and fissures against caries. The simple technique is particularly appropriate for recently erupted teeth and involves the application of a liquid resin onto acid-treated occlusal enamel surfaces. The resin flows into the fissures and the etched micropores, forming intimate contact with the enamel and conferring a high degree of resistance to caries by covering the susceptible fissures (Buonocore, 1971; Lee and Swartz, 1971; Rock, 1973).

(ii) Fractured Incisal Edges

A combination of a filled composite resin and an unfilled resin layer used in conjunction with acid-etching of the enamel, has been used to restore fractured incisal edges. This conservative, if somewhat temporary, procedure allows the attachment of an aesthetic restorative material to tooth structure with minimal tooth preparation (Buonocore and Davila, 1973; Roberts and Moffa, 1973).
(iii) **Abraded or Hypoplastic Areas**

The treatment of non-carious lesions resulting from toothbrush abrasion or hypoplastic defects can be accomplished without conventional cavity preparation by using the acid-etch technique to obtain a mechanical bond between the resin restoration and that part of the margin which is in enamel (Friedman and Retief, 1973; Harris et al, 1974). The effect of acid-etching either a part of the margin which is in dentine or the dentine surface of a prepared cavity is discussed subsequently (Chapter 4).

(iv) **Sealing of Cavity Margins**

Pretreatment of enamel cavity walls with phosphoric acid prior to the placement of the composite resin, has been shown to substantially improve the retention of the restoration and the seal at its margins. This is discussed subsequently (Chapter 6).

(v) **Orthodontic Brackets**

Aesthetically pleasing, tooth coloured, plastic brackets can be bonded directly onto the labial surfaces of teeth, by means of a resin, after acid-pretreatment. It has been found that these brackets can withstand the forces of mastication, the stresses from an arch wire and the degrading effects of the humid, aqueous oral environment. They can be readily removed without permanently affecting the integrity of the tooth enamel (Newman, 1976).
(vi) **Splinting Teeth**

The acid-etching technique used with composite resins can provide temporary stabilisation of mobile teeth, which may be the result of occlusal trauma associated with periodontal disease (Feldstein and Teitel, 1975; Goodman and Lester, 1977).

In summary, the function of acid-etching of enamel in restorative dentistry is to provide a surface suitable for the penetration of resin so that the subsequent restoration is retained by mechanical interlocking into the etched surface. Currently, the major use of acid-etching tooth enamel is to assist in the mechanical retention of composite resin restorations and, thereby, assist in sealing the margins against leakage. The vast majority of composite resin restorative materials using the acid-etch technique rely, for retention of the material, either completely on acid-etching of the enamel or on acid-etching in combination with the retention form of the cavity preparation. The investigation, described subsequently, examined the degree of marginal leakage and adaptation of certain composite resins after application to the acid-etched walls of prepared cavities.
CHAPTER 3

THE EFFECT OF ACID SOLUTIONS ON ENAMEL

3.1 Introduction

The application of an etchant, usually phosphoric acid, to the enamel surface causes a reaction with the inorganic calcium salts (Smith et al, 1976). Silverstone (1974), using both transmitted and polarised light, observed that the result of this reaction on the enamel surface was, superficially, a loss of surface contour relative to adjacent unetched enamel as the enamel was dissolved. At the base of the etched region, the enamel was modified by the etchant and rendered "porous" due to the partial loss of mineral.

Hamilton et al (1972) suggested that acid attack started at the prism periphery where the enamel crystallites changed orientation rapidly and were less tightly packed, thereby providing diffusion channels. The initial reaction to the acid attack was found to be the opening of a narrow gap in the prism boundary zone (Johnson et al, 1971; Simmelink et al, 1972).

Subsequently, Nichol et al (1973) proposed a two-stage model to explain the variations observed in the patterns of demineralisation and found that the site of initiation of attack was always the prism boundary, consistent with the work of
previous researchers. The attack could then follow several directions, either towards the prism core or along and around the prism boundaries; the primary direction of attack depended on the type of acid used. The direction of spread after treatment with hydrochloric acid was perpendicular to the prism, both towards the centre of the same prism and towards the tail region of adjacent prisms. After treatments with EDTA (ethylene diamine tetra-acetic acid) the spread was parallel to the prism axis and, after lactic acid treatments, it was parallel to the prism axis as well as perpendicular to the prism boundary.

The spread of attack, following initiation, could also be aided by the molecular sieve behaviour of enamel (Darling et al, 1961; Poole et al, 1961). Enamel contains a system of micropores and intercrystalline spaces of various sizes which act as a molecular sieve. The volume of pores is approximately 0.2 per cent of the enamel volume (Osborn and Ten Cate, 1976).

A number of factors have been found to influence the depth and pattern of etching of the enamel by phosphoric acid solutions:
(i) Instrumentation during cavity preparation (3.2).
(ii) Differential dissolution of enamel (3.3).
(iii) Acid concentration and duration of application (3.4).
(iv) Fluoride concentration of the surface enamel (3.5).
(v) Prismless enamel (3.6).
(vi) Formation of reaction products (3.7).

3.2 Instrumentation During Cavity Preparation

It has been suggested (Barnes, 1977, d) that the way in which a composite resin adapts to tooth substance depends more upon the microscopic contour of the tooth surface, which may be influenced by instrumentation during cavity preparation and etching, than on the properties of the composite resin.

The cut enamel surface varies depending upon the type of instruments that have been used during cavity preparation. One variation in the cut enamel surface takes the form of enamel smears (Eick et al, 1970; Boyde, 1976; Evans and Kasloff, 1976; Barnes, 1977, d) which are sheets of amorphous, fused enamel which differ in size, pattern and probably the ease with which they can be dislodged from the underlying tooth. Because the smeared layer of enamel might be resistant to acid attack in proportion to its thickness or degree of compaction, the methods used to finish the cavity, prior to etching, might be of clinical significance.
(Barnes, 1977, d). The size of enamel smears have been found to be reduced by the use of a tungsten-carbide bur, a tungsten-carbide "stone" and a "Jet" fine finishing bur, the last producing the least amount of smearing.

Various studies have shown that steel and diamond burs produce cavity margins with defects up to to 20 micrometres wide and deep grooves in the cavity walls (Provenza and Sardana, 1966; Boyde and Knight, 1969; Eick et al, 1970; Boyde, 1976). Tungsten-carbide burs have been found to produce the smoothest cavity margins, with defects greater than 0.5 micrometres being uncommon (Boyde, 1969; Eick et al, 1970). In contrast with these investigations, a study by Kasloff et al (1962) indicated that, at high speeds, diamond burs were less likely than tungsten-carbide burs to produce cracking and crazing of enamel during cavity preparation.

The clinical relevance of the microscopic finish produced on the cavity walls has, however, been questioned by Barnes (1977,d) who found that in all teeth where the enamel cavity walls had been etched it was no longer possible to distinguish the particular method of instrumentation of the surface. This indicated that the application of 30 per cent phosphoric acid for 60 seconds was sufficient to dissolve enamel smears and scratches of different degrees of severity and to dissolve differentially the underlying
enamel prisms. In contrast, work by Ramirez De Salazar (1972) and Dennison and Craig (1978) showed that etching with 50 per cent phosphoric acid for 60 seconds only reduced the depth of irregularities created during cavity preparation.

3.3 **Differential Dissolution of the Enamel**

Depending on a number of factors, the enamel crystallites have been found to exhibit a difference in reactivity when subjected to acid attack. Microscopically, the appearance of the etched enamel surface reflects this variation in solubility. Factors related to the differential dissolution of the enamel during acid-etching include:

(i) Prism direction and crystallite orientation relative to the direction of acid attack (3.31).

(ii) Lattice imperfections in the enamel crystallites (3.32).

(iii) The distribution of organic material (3.33).

(iv) The presence of trace elements (3.34).

(v) Variations between teeth and between different parts of one tooth (3.35).

(vi) The reprecipitation of mineral at prism junctions (3.36).

3.31 The pattern on the enamel surface produced by etching is related to the orientation of the prisms exposed to acid attack. Early studies concentrated largely on examining the etched tooth
surface, where the direction of acid attack is parallel to prism
direction, producing an etched surface with a structure resembling
a honeycomb (Sharpe, 1967; Poole and Johnson, 1967) (Fig. 3.1(a)).
In contrast, relatively little emphasis has been given to prepared
cavity walls which present enamel prisms exposed in a variety of
planes — the prisms have most commonly been observed in longitudinal
section and less often in oblique or transverse section (Gwinnett,
1973, a). The most frequent direction of acid attack on these
surfaces is therefore perpendicular to prism direction, producing
an etched pattern that has been described as a fibrous structure
(Sharpe, 1967) (Fig. 3.1(b)).

The extent of acid attack has been found to be dependent on
the particular cavity wall or tooth surface being etched. The
crystallites that comprise the exposed enamel prisms are selectively
attacked according to their orientation (Sharpe, 1967; Nygaard
and Simmelink, 1972). Based on a knowledge of the orientation of
the crystallites within enamel it is possible to account for the
variations in solubility patterns exhibited (Gwinnett, 1973, a).

The work of Johnson (1966) and Sharpe (1967) suggested that
an enamel crystallite dissolved in acid more rapidly along its
c-axis (the long axis of the crystallite) than perpendicular to
this axis. Research by Poole and Johnson (1967) and Sharpe (1967)
showed that the head and tail portions of prisms exhibited different
Fig. 3.1. - Photographs showing, (a) Etched enamel of the tooth surface, x1000; (b) Etched enamel of the cavity wall, x1000.
reactivities to acid. When the attack was parallel to the prisms, as is the case on the enamel surface, the heads or centres of the prisms were most reactive because in this region the crystallites were parallel to the prisms and would dissolve more rapidly along their c-axes. However, when the acid attack was perpendicular to the long axes of the prisms, as is often the case with enamel cavity walls, the tails or borders of the prisms were more reactive and the heads were left moderately intact, because the crystallites in the centre of the prism were also oriented perpendicular to the acid attack and would not dissolve as rapidly as those of the tail portion which were inclined away from the long axis of the prism (Sharpe, 1967) (Fig. 3.2).

There is also a considerable difference in the overall rate of penetration of the enamel by acid (Sharpe, 1967). Parallel attack is rapid along the prism heads and differentiation of the head and tail region is very noticeable. Perpendicular attack, however, is much slower and far less selective. The head portions are easily penetrated and dissolved along their length but are more resistant in other directions, especially during perpendicular attack.
Fig. 3.2. - Model illustrating the arrangement of crystallites in enamel and their behaviour during parallel and perpendicular acid attack.  (Courtesy of Sharpe (1967).)
Various explanations for this directional reactivity have been suggested. Arends (1973) indicated that this phenomenon might be caused by the combined effects of greater chemical reactivity along the c-axis and the "coating" of the external crystallite surface with a protective layer of organic material.

3.32 An alternative suggestion by Arends (1973), related to the presence of dislocations, which are lattice imperfections on an atomic scale. Dislocations have been found in the central regions of some enamel crystallites, are intimately associated with the twist of the apatite needle and are possibly created in the early stages of enamel formation (Jongebloed et al, 1973). Around the point where a dislocation emerges at the crystallite surface, a few ions are shifted from their original positions of minimum energy, thus increasing their susceptibility to chemical attack (Arends, 1973). It has been demonstrated by Cabrera, as reported by Arends (1973), that dislocations are regions of undersaturation and that preferential dissolution takes place at dislocations rather than in the perfect lattice. A consequence of the presence of a dislocation is that transport and diffusion along a dislocation line is much faster and easier than in the surrounding lattice.

3.33 Crystallite orientation relative to the direction of attack is not the only factor involved in the differential dissolution of
enamel. An additional factor may be associated with quantitative or qualitative differences in the distribution of organic material. Prism junctions are sites where there is an abrupt change in crystallite orientation and where more intercrystalline space exists which could be filled with hydrated organic matrix. It has been suggested by Johnson et al (1971), Nichol et al (1973) and Scott et al (1974) that the higher organic content of this region could account for the reduced solubility in acid, because the crystal surfaces are protected.

3.34 The slow accumulation of trace elements, for example, fluorine, at sites adjacent to diffusion pathways hinders the movement of ions and causes variation in the dissolution pattern (Poole and Johnson, 1967). As a result, clinically, it has been found necessary to increase the etching time.

3.35 Different teeth and different portions of one tooth exhibit different patterns of dissolution when subjected to acid attack. Jorgensen (1975, a) studied phosphoric acid-etched surfaces of human teeth with a scanning electron microscope and showed that the etched pattern could vary significantly from tooth to tooth and between different areas of the same tooth. A similar etched pattern could only be found along the midline of pairs of contralateral teeth.
3.36 Tyler (1976) suggested that another important factor influencing differential dissolution of enamel might be the reprecipitation of minerals in the prism junctions. On acid-etched surfaces, there appeared to be an actual elevation of the prism borders above the original level of the cut surface and foreign crystals, noticed in electron photomicrographs (Johnson, 1967), appeared to persist until the final stages of enamel dissolution. This study described the result of enamel dissolution using static, unstirred, demineralising systems in which the reaction products accumulated at the surface of the specimens being etched. Tyler (1976) experimented with stirred demineralising systems to discover the effect on etched patterns of the removal of reaction products.

The dissolution of ionic crystals was considered by Napper and Smythe (1966) to occur in two consecutive stages. The initial process involved the disengagement of solute ions from steps in the crystal surface, followed by a second stage associated with the transfer of ions into the bulk of the dissolution medium by diffusion and convection. They suggested that mass transport processes were rate-limiting and therefore, in unstirred systems where the dissolution products accumulated, the dissolution rate became diffusion controlled. However, in stirred systems which
facilitated the mass transfer of dissolution products, ionic disengagement from crystal surfaces became the rate-limiting process and, as a result, the dissolution rate was controlled by surface area availability.

The application of these findings, to different etching systems, suggests that etching agents applied to the enamel tooth surface preferentially penetrate the prism peripheries where ionic disengagement begins. With stirred systems the dissolution products are continuously removed, diffusion barriers are never created and the etch patterns are determined solely by the relative areas of enamel exposed to the etchant at different sites. As the surface area of the crystallites at the prism borders is greater than the surface area of the crystallites at the etched ends of the prisms, preferential dissolution continues at the prism borders leaving the axial parts of the prism cores less affected, in contrast to the accepted honeycomb pattern where the central regions of the prisms are extensively etched compared to the peripheries (Tyler, 1976).
In contrast, a study carried out by Silverstone et al (1975) on smooth enamel surfaces produced results which suggested that there is no one specific etching pattern produced by acid action on human dental enamel. Their study, which utilised an unstirred dissolution system, established three types of etching patterns. The first and most common pattern involved preferential removal of the prism core, leaving the prism peripheries relatively intact. The second pattern was the reverse involving preferential removal of the prism periphery (Fig. 3.3). The third pattern was a random pattern containing areas corresponding to the first and second patterns together with regions where the etching pattern could not be related to prism morphology. These findings differed slightly from those presented by Tyler (1976) but differed markedly from those presented by Poole and Johnson (1967) who considered that the first pattern was the result of acid-etching while the second pattern was the result of the action of chelators.

As a result of these findings, it may be preferable, clinically, to use a small cotton pellet to apply the acid in a stirring motion rather than painting it on, to ensure that the enamel is constantly wet. However, it may not always be possible to use this preferred stirring action in all parts of cavities, for example, at the gingival margin of small Class III cavities.
Fig. 3.3  -  (a) Transverse section through enamel, etched for two minutes, showing types 1 and 2 etching patterns, x300; (b) Same area, x1,000.
3.4 Acid Concentration and Duration of Application

The use of an acid solution to etch the enamel surface is an essential prerequisite for the successful mechanical bonding of resins to the hard tissue at the microscopic level. Buonocore (1955) used 85 per cent phosphoric acid solution to etch or "condition" enamel prior to the use of acrylic restorative materials in order to improve the edge adaptation. Since then, many workers have used a solution of 50 per cent phosphoric acid, buffered with 7 per cent zinc oxide by weight as an etching agent (Buonocore, 1955 and 1970; Gwinnett and Buonocore, 1965; Retief, 1974, b; Ferreira, 1976).

A series of different acid solutions, including phosphoric acid in the concentration range 20-70 per cent, were investigated by Silverstone (1974) for their effect on human enamel. The results showed that an unbuffered solution of 30 per cent phosphoric acid produced a surface considered to offer the most favourable conditions for bonding. Exposure of a region of enamel to an etching solution resulted in both the loss of surface contour and the creation of a residual porous surface. Use of acid concentrations of 5-15 per cent and 70-80 per cent produced only minimal etching changes. Over the concentration range 20-60 per cent, enamel surfaces demonstrated the three basic etched patterns described by Silverstone et al (1975)
and referred to, in detail, previously in this chapter.

A 30 per cent unbuffered solution of phosphoric acid used with a 60 second exposure period produced the most consistent and evenly distributed etch over an enamel surface (Silverstone, 1975) and resulted in a lost thickness of surface enamel of approximately 10 micrometres. The remaining enamel comprised of a porous zone measuring approximately 20 micrometres and, deep to this, a zone of approximately 20 micrometres, described as a slightly porous region, which had a lower observed birefringence, but was histologically indistinguishable from sound enamel.

In comparison, Buonocore (1975) considered a 40-50 per cent concentration of phosphoric acid to be well suited for tooth etching purposes and found that a 60 second etch time using 50 per cent phosphoric acid provided both a reasonable and safe working time as well as ensuring an adequate etch.

In somewhat marked contrast, Wickwire and Rentz (1973), concluded from their studies that a four minute etch with 50 per cent phosphoric acid exposed the best surface for wetting and surface area contact, and produced a level of uniform roughness that could not be obtained with a two minute etch. They examined cut sections of enamel, treated with 50 per cent phosphoric acid solution, under the light microscope and found that the amount of enamel dissolved after the two minute acid-
treatment of the surface enamel ranged from 5-10 micrometres. After four minutes of acid-treatment approximately 5-15 micrometres was lost from the surface and after six minutes, the loss was between 20 and 30 micrometres.

Brannstrom and Nordenvall (1977), using 37 per cent phosphoric acid, claimed that there was no appreciable difference in appearance of cavity surfaces and surrounding enamel when etching for 15 seconds or two minutes.

Although it is apparent from the preceding discussion that the ideal concentration is disputed, it is generally agreed that phosphoric acid is the best acid for the etching of enamel. In addition, there is undoubtedly an ideal depth of etching which should be deep enough to provide resin tags of adequate length. However, if the tags become too long they may fracture when subjected to stress, resulting in a loss of retention. Furthermore, exaggerated etching could weaken tooth structure between the tags causing the enamel itself to fracture under stress (Phillips, 1976).
3.5 Fluoride Concentration of the Surface Enamel

The possibility that etching enamel surfaces with concentrated acid solutions may predispose the enamel to the development of caries has been considered. In response, the treatment of enamel with fluoride solutions, before, as well as after etching, has been advocated in order to reduce the solubility of enamel (Kochavi et al, 1975).

Observations using the scanning electron microscope have revealed that the presence of a significant fluoride content inhibited enamel surface alterations during acid-etching (Hoffman et al, 1969). This was supported by Lee et al (1972) and Buonocore (1975) who showed that teeth from areas of high natural fluoridation were highly resistant to phosphoric acid.

Several investigators have found that fluoride treatment of etched enamel surfaces significantly reduced the bond strengths of applied resins (Lee et al, 1972; Sheykholeslam et al, 1972). It appeared that fluoride application detrimentally affected the bond strengths whether applied to the enamel before or after acid-etching procedures. Lee et al (1972) found that when enamel was treated with a fluoride solution prior to etching, little or no bonding was obtained.
and Hoffman et al (1969) reported that enamel pre-treatment with sodium or stannous fluorides reduced the etching action of phosphoric acid, probably because of a decrease in enamel crystallite solubility. They also noted that surface changes following etching were considerably greater in enamel samples that were not pre-treated with fluoride solutions than in those that were. However, when the enamel was treated with fluoride solutions after etching, bond strengths were marginally better, but significantly less than teeth which had not been treated with fluoride solutions (Lee et al, 1972).

Sheykholeslam et al (1972) and Gwinnett et al (1972) suggested that the reduced bond strengths could be related to changes produced on the enamel surface. They showed by scanning electron microscopy that fluoride application after etching produced reaction products or precipitates, namely Ca₅(PO₄)F and CaF₂ from sodium fluoride treatment and Sn₃(PO₄)F₃ and CaF₂, formed after stannous fluoride treatment (Kochavi et al, 1975; Nordquist et al, 1975). These products covered the surface and appeared to fill partly the interprismatic spaces, reducing the "wettability" of the surface and interfering with resin penetration and tag formation, both conditions conducive to lower bond strengths and greater susceptibility to bond disruption by water.
In quite marked contrast, studies by Chow and Brown (1974) and Takahashi (1977) involving the application of fluoride solutions to etched enamel, found that there was no interference with sealant penetration and Low et al (1975) showed that the treatment of etched enamel with eight per cent stannous fluoride solution prior to resin application significantly increased the tensile bond strength. Furthermore, observations by Chow and Brown (1974), using the scanning electron microscope, showed that fluoride treatments produced no discernible precipitates on the surface of the enamel which might interfere with sealant penetration and bonding, although significant enamel-fluoride uptakes had been obtained.

3.6 Prismless Enamel

Ripa, Gwinnett and Buonocore (1966) demonstrated the existence of an outermost layer of enamel composed almost entirely of crystallites whose c-axes were perpendicularly oriented to the outer enamel surface. This layer of enamel does not present the usual prism-structured appearance found in prismatic enamel where abrupt changes in crystallite orientation formed boundaries between prisms (Poole and Brookes, 1961).
In ground sections, the outermost layer of enamel of 70 per cent of permanent and all deciduous teeth showed sites devoid of the usual prism markings (Romanuik and Shroff, 1965; Innes and Shroff, 1966; Gwinnett, 1967). In permanent teeth, two types of prismless enamel have been observed. One type appears as a continuous band, as in deciduous teeth, and the other type appears as an "onion layer". The "onion layer" type described as "scale-like", owes its appearance to the presence of the striae of Retzius which emerge on the enamel surface. Since the striae in deciduous teeth do not generally emerge on the surface but run more or less parallel to it, the "onion layer" type is not usually observed and continuous bands of enamel are seen (Ripa et al, 1966; Gwinnett, 1967). Prismless enamel is the product of amelogenesis and is not acquired post-eruptively; its formation is the result of reduced functional activity during the terminal stages of amelogenesis.

The layer of prismless enamel averages 30 micrometres in thickness and was found by Ripa et al (1966) to occupy the gingival one-third of permanent teeth in 57 per cent of cases, the mid-coronal region in 36 per cent and the entire surface in only 7 per cent of cases. Gwinnett (1973, b) described prismless enamel as having a "unidirectional orientation of
crystallites with a relatively dense arrangement", on which enamel etchants produced a "relatively uniform dissolution with some tissue loss and the creation of limited random porosity". The presence of prismless enamel, might therefore adversely affect the etching of enamel and the quality of the acid-etched surface (Fuks et al, 1977). Without the creation of adequate microspaces in the prismless layer there would be a relative absence of tag formation upon adaptation of resins and a resultant loss in mechanical retention (Sheykholeslam and Buonocore, 1972).

The high percentage of permanent teeth found by Ripa et al (1966) to exhibit prismless enamel in the gingival one-third suggests the possibility of a clinical problem particularly at the gingival margin of some Class III and V cavity preparations. It is not clinically possible to detect and gauge the thickness of the prismless layer and attempts to remove it by prolonged etching times as described by Buonocore (1975) are contra-indicated (Gwinnett, 1973, b).

3.7 Formation of Reaction Products

Chow and Brown (1973) showed that monocalcium phosphate monohydrate formed on the enamel surface during acid-etching and appeared as a white crystalline material. Similarly, Duff
and El-Motayam (1975), using phosphoric acid concentrations of greater than 30 per cent, found that the reaction product was predominantly monocalcium phosphate monohydrate which deposited on the enamel surface and acted as a barrier to minimise further demineralisation. Theoretically, the presence of this compound could, therefore, decrease the depth of etch and limit resin penetration. However, the monocalcium phosphate monohydrate is highly soluble in water and would be completely washed away during rinsing of the etched surface, leaving a clean surface available for mechanical bonding of the resin (Chow and Brown, 1973).

The use of phosphoric acid in a concentration less than 30 per cent produced a different reaction product, dicalcium phosphate dihydrate. Accumulation of this compound could also interfere with the etching effect and, as it is not water soluble, might adversely affect the success of resin penetration (Chow and Brown, 1973 and 1974). Ionic aggregates, present on the etched surface, might also be responsible for inhibited "wettability" and void formation which would effectively reduce bond strengths (Mulholland and de Shazer, 1968).

Considerable attention has been given to the resin tags which penetrate etched enamel. They have been studied in detail
and have been assumed to be of prime importance in bonding. However, studies by Adipranoto et al (1975) and Soetopo et al (1978) have found that etching with 2 per cent phosphoric acid resulted in no detectable resin penetration into the enamel surface, but produced a similar bond strength to that obtained with 40 per cent phosphoric acid, a concentration which is generally considered to allow optimal resin tag formation. These results suggested that the length of resin tags penetrating into enamel bore no correlation with bond strengths. As previously discussed, the use of phosphoric acid concentrations below 30 per cent is associated with the formation of the relatively insoluble compound, dicalcium phosphate dihydrate. An explanation for the similarity in bond strengths might be related to the strength of attachment of the reaction products to the enamel surface. If the reaction products are strongly attached and, in turn, the resin is capable of bonding strongly to them, then a strong resin bond could result with no reliance on tag formation (Sheykholeslam et al, 1972). Adipranoto et al (1975) suggested from their study that "adhesion" arose from the formation of chemical bonds rather than mechanical interlocking. There is, however, at present, no firm evidence to support either of these hypotheses.
The length of wash time following etching has been shown to be of importance by both Adipranoto et al (1975) and Soetopo et al (1978) who observed that washing the etched enamel for 60 seconds substantially increased the bond strength compared with the conventional 15 second wash. This was found with enamel treated with 16 per cent and 30 per cent phosphoric acid concentrations but not with the 2 per cent concentration. This emphasised the need to remove the more soluble, monocalcium phosphate monohydrate to allow optimal mechanical bonding.
CHAPTER 4

SOME CLINICAL ASPECTS OF ACID-ETCHING

4.1 Introduction

Apart from the effects of acid solutions directly on the enamel of the tooth surface or the prepared cavity walls, other clinical aspects of acid-etching include:

(i) Remineralisation of acid-etched enamel (4.2).

(ii) The effect of acid-etching on dentine (4.3).

(iii) Pulpal considerations (4.4).

4.2 Remineralisation of Acid-Etched Enamel

The ability of acid-etched enamel to remineralise has become clinically important with the development of pit and fissure sealants, the use of acid-etching to mechanically retain composite resins to the enamel of the tooth surface and the cavity wall, the use of acid-etching to secure orthodontic brackets and the use of citric acid cavity cleansers. An important factor in the use of these techniques, especially if they are repeated periodically, is the immediate and longer-term damage caused to the enamel surface. If the etched enamel surfaces that are not covered by the dental material are able to return to a "normal" condition within a reasonable period of time, through redeposition of calcium phosphates from saliva, then the acid-etch technique
may not be harmful to any etched enamel left exposed (Albert and Grenoble, 1971). However, much controversy still surrounds the findings of enamel remineralisation studies.

It has been claimed (Albert and Grenoble, 1971; Arana, 1974) that *in vivo* remineralisation of acid-etched enamel takes place rapidly, particularly in younger patients. They found that within 48 hours the enamel had returned to a "normal" appearance and that by 96 hours "remineralisation" was complete. Muhlemann et al (1964) showed that the characteristic prism-end pattern of acid-etched enamel disappeared when exposed to the oral environment for 2 - 48 days. Silverstone (1977, b) found that the solubility rate of acid-etched enamel returned to that of adjacent sound enamel after 48 hours' exposure to oral fluids. This result was interpreted as being due to the uptake of both mineral and organic components from saliva so that a greater acid resistance was conferred on the previously etched enamel, thus returning the value for acid dissolution to approximately normal. These optimistic findings are, however, in conflict with other observations.

Wei and Koulourides (1972) and Pickel et al (1975) found that acid-etched enamel did not show complete remineralisation and, under *in vivo* conditions, did not regain the original surface hardness. Meurman and Asikainen (1976) found that acid-etched
enamel exposed to the oral environment gained up to 84 per cent of its original surface hardness, whereas enamel artificially rehardened by the use of saturated solutions of tricalcium phosphate without fluorides, recovered only 74 per cent. The acid-etching of enamel is responsible for the loss of considerable mineral content, especially calcium and phosphorous. Wei (1971) found that, after remineralisation, these minerals had been recovered to levels approaching those of sound enamel. The original optical appearance of enamel, however, never completely returned after remineralisation and some slight loss of birefringence persisted (McDougall and Adkins, 1966).

Hoffman et al (1969) suggested that fluorides played a role in the remineralisation of etched enamel surfaces by the initial formation of a fluoride complex that involved the less mineralised enamel matrix components. Earlier, Koulourides et al (1961) and Pigman et al (1964) had shown that the rate of rehardening of etched enamel was more rapid in solutions that contained small amounts of fluoride, approximately one part per million. Gron and Amdur (1975) found that the application of sodium fluoride to acid-etched enamel was effective in promoting remineralisation.
Observations using the scanning electron microscope (Meurman and Asikainen, 1976; Meurman and Mikko, 1976) indicated that enamel "repair" following acid-etching was achieved by the filling of etched microspaces with salivary deposits which became calcified into globular substances. Rehardening did not result in an enamel surface histologically consistent with solid, non-porous, intact enamel, since definite defects and micropores could be detected.

In summary, despite somewhat conflicting evidence, it is probable that the clinical appearance of enamel that has been exposed to saliva after etching will return to "normal" within 48 - 96 hours and that the solubility rate of this remineralised enamel will be similar to that of adjacent unetched enamel after approximately 48 hours. On the other hand, the surface hardness, mineral content and microscopic appearance never completely return to their pre-etch condition. The application of certain fluoride solutions is effective in promoting remineralisation.

There appears to be wisdom in the suggestion by Meurman and Asikainen (1976) and Meurman and Mikko (1976) that unnecessary acid-etching of intact enamel by acid solutions used in the acid-etch technique should be avoided.
4.3 The Effect of Acid Solutions on Dentine

Dentine is different in structure from enamel, possessing a high organic content, 18 per cent by weight, and 12 per cent by weight of water. Much of the water content is freely available to form a wet dentine surface, which is one of the chief barriers to the development of an adhesive dental restoration (Barnes, 1977, c).

During cavity preparation, a layer of debris consisting of organic and inorganic material, with particles varying in size from less than one micrometre to more than 15 micrometres, was found to coat the prepared cavity walls and floor (Brannstrom and Johnson, 1974). This debris, from cavity instrumentation, formed "plugs" by blocking the openings into the dentinal tubules sufficiently to prevent bacterial growth down the tubules (Vojinovic et al, 1973). Following cavity preparation, both water spraying and gentle scrubbing with water-soaked cotton pellets failed to remove the "plugs". Application of acid-etchants, however, resulted in the removal of the layer of debris including the dentine "plugs". Openings into the tubules increased from the normal 1 - 2 micrometres to 3 - 5 micrometres and in profile, the tubules were widened and funnel-shaped to a depth of 10 - 20 micrometres below the surface (Vojinovic et al, 1973; Brannstrom et al, 1974).
This acid-treatment, therefore, was found to increase the liquid area of the treated dentine surface from 10 per cent to approximately 25 per cent and, thereby, markedly impair "adhesion"; by removing the "plugs" of debris which afforded a certain degree of protection to the pulp, acid-treatment had a generally undesirable effect (Brannstrom et al, 1974; Evans et al, 1976).

Clinically, in cases of tooth-brush abrasion, where areas of dentine and enamel have been removed, a restoration is often required. Problems then arise because of the difficulty in placing a restoration and finishing the margins on dentine of the root surface. These problems are caused by the relatively high content of water available on the cut dentine surface and the lack of success in etching dentine to produce a surface suitable for the mechanical bonding of resins. In these situations, a glass ionomer cement may be the aesthetic material of choice.

4.4 Pulpal Considerations

Increasing popularity with the technique of acid-etching tooth enamel to improve the retention and marginal seal of restorative resins has resulted in an increasing application, either intentionally or unintentionally, of acid-etchants to vital dentine. Investigation has shown that the use of
concentrated acid solutions on vital dentine may seriously aggravate the pulp and increase its response to composite resins (Retief et al, 1974; Stanley et al, 1975; Eriksen, 1976; Cotton and Siegel, 1978). Composite resins, although free of methacrylic acid and of a neutral pH, have been found to be responsible for pulp damage. The intensity of the pulpal response can be increased by acid pre-treatment procedures—a result, probably, of increased dentine permeability (Stanley et al, 1975). Acidic solutions remove the debris left by cavity preparation, dissolve the peritubular dentine and expose and enlarge the tubule openings. These actions effectively increase the permeability of dentine to fluids and to the growth and penetration of bacteria (Brannstrom and Nyborg, 1972; Vojinovic et al, 1973).

Goto and Jordan (1972) and Heys et al (1977) studied the pulpal response to composite resin materials in dogs and monkeys, respectively. Both found that, over a period of eight weeks, the pulps initially showed a severe inflammatory response which diminished with time to a mild inflammation, indicating that the pulpal response to composite resin may be reversible. It has been suggested that the main cause of pulpal damage from composite resin materials is not irritation from the material itself, but irritation from bacteria and their toxins that
penetrate between the cavity walls and the restorative material, usually as a result of poor adaptation between the restoration and the tooth (Brannstrom and Nyborg, 1972; Vojinovic et ál, 1973). In contrast, a study by Cotton and Siegel (1978) found no evidence of bacterial penetration, from which they concluded that the adverse pulpal reactions were the result of the acid-conditioner (in their case, citric acid), rather than bacterial penetration.

Over many years the irritating effects of two phosphoric acid-based materials, zinc phosphate cement and silicate cement, have been reported. As early as 1944, Harvey and others considered a major cause of pulp damage to be the very high acidity of zinc phosphate and silicate cements at the time of their insertion into the cavity. Brannstrom and Nyborg (1960) studied pulps with moderate to strong cellular infiltration, which appeared to be due to the irritant action of zinc phosphate cements, and believed that these changes might result in irreversible damage. Swartz et al (1968) found that components of silicate cements were capable of penetrating a considerable thickness of dentine. Ihara et al (1973) studied, in vivo, the detrimental effects of silicate cements on the pulp and found clinical symptoms ranging from mild to prolonged discomfort and pathological sections which suggested pulpal injury ranging from either no damage or slight damage to severe destructive changes in the pulp.
In contrast, Lee et al (1973) applied 50 per cent phosphoric and citric acid solutions, in vitro, to one millimetre thick dentine discs and found that a five minute exposure resulted in surface etching only, with little sub-surface effect, and that colourimetric detection and scanning electron microscopy showed that the etchants did not penetrate the thickness of the dentine discs. Goto and Jordan (1973) found that pulp damage from the application of 50 per cent phosphoric acid to vital dentine was slight and concluded that this procedure did not produce harmful effects. Previously, Johnson et al (1970) had compared the effect of phosphoric acid with that of distilled water on the dentine of deep cavities and found that both had similar mildly irritating properties.

In spite of these several rather surprising and, perhaps, dubious findings, the results of many studies have indicated that the use of acid-etchants and composite resins do aggravate the pulp to some degree, and have indicated the need for a protective layer of a calcium hydroxide-containing base material to cover the exposed dentine (Buonocore and Davila, 1973). Jedrychowski and Reisbick (1974) and Eriksen (1976) found that the use of a base reduced the pulpal irritation attributable to acid-etching.
The protective capacity of the calcium hydroxide base may be due partly to its forming a simple barrier, partly to the ability of the calcium hydroxide content to neutralise the acid before it reaches dentine (Brannstrom and Nyborg, 1972; Eriksen, 1976), or it may be due partly to its adherence to the dentine. Bases containing calcium hydroxide adhere more securely to dentine than to the composite resin; should the resin shrink from the cavity walls and bacteria penetrate under the restoration, the dentine would remain protected (Brannstrom and Nyborg, 1972; Grajower et al, 1976). Mjor and Furseth (1968) found that normal dentine covered with a calcium hydroxide base became further mineralised by an intratubular deposition of minerals, thereby further protecting the pulp.

In most respects the calcium hydroxide base is compatible with the pulp and the composite resin. The only reported problems are a possible small decrease in hardness of the composite resin and a report by Grajower et al (1974) of a greyish discolouration of the composite resins when placed against a calcium hydroxide base. Other bases, however, are not as acceptable. Zinc oxide and eugenol materials are not recommended for use with composite resins because free eugenol may inhibit polymerisation of the composite resin and reduce its transverse bending strength and hardness (Griffith and Cannon, 1973; Grajower et al, 1974).
Zinc phosphate cements are compatible with composite resins but their initial low acidity may irritate the pulp and exacerbate the effect of acid-etching the cavity walls.

In summary, there are several potential causes of pulpal damage from acid-etched composite resin restorations; these include the composite material itself, the possible penetration of bacteria and its toxins at the margins and the acid-etchant. There is considerable difference of opinion concerning the damage caused by the acid, but it is generally considered wise to protect the pulp by the application of a quick-setting calcium hydroxide base.
CHAPTER 5

COMPOSITE RESIN RESTORATIVE MATERIALS

Composite resin restorative materials were a development of resins based on poly(methyl methacrylate) (PMMA) because certain inherent characteristics of the PMMA resins limited their use and effectiveness as a restorative material. These characteristics included a high polymerisation contraction, a low degree of hardness and strength and a high coefficient of thermal expansion.

By definition, a "composite material" is the combination of at least two chemically different materials with a distinct interface separating the components (Phillips, 1973). Such a combination of materials provides properties that could not be obtained with any one of the components acting alone. In Dentistry, the term composite resin has been given to a material in which an inorganic filler has been added to a resin matrix in such a way that the properties of the matrix have been improved (Phillips, 1973).

Compared with the PMMA resin restorative materials, composite resins possess both an improved resin matrix and a high percentage of inorganic filler. These differences markedly alter the physical properties.
Research into an improved resin matrix system suitable for use in a dental composite resin finally led to a compromise between an epoxy resin and a methacrylate resin. Developed by Bowen (1965), the resin is based on the dimethacrylate monomer, BIS-GMA, which is essentially the reaction product of bisphenol-A and glycidyl methacrylate. This resin was shown to be suitable for use as a binder for reinforcing fillers because it has a relatively low polymerisation shrinkage and hardens rapidly under oral conditions (Bowen, 1963). For use as the resin matrix of commercial composite resins, the original Bowen's resin was modified to reduce the viscosity of the material at room temperature by the addition of methyl methacrylate or other monomers (Bowen, 1965).

Requirements of the filler component of composite resins are high hardness, chemical inertness, low thermal expansion and a refractive index and opacity close to that of tooth structure. A number of different types of inorganic fillers have been used in commercial composite resins and include fused silicon, crystalline quartz, lithium aluminium silicate and borosilicate glass. The fillers have been used in numerous forms, as spheres, whiskers or ground powders, the last being best retained in the matrix. Beech and Brown (1972) indicated that the particle shape should provide the maximum surface area for
bonding, the size distribution should give maximum packing efficiency and the overall size of the particles should be as small as possible, consistent with the need to wet the particles with monomer. The particle size of the filler varies from 1 - 40 micrometres in maximum dimension, the majority of the particles being in the range 15 - 20 micrometres.

The filler component has an important role in influencing the properties of the composite resin. In order to inhibit deformation of the matrix a high filler content is necessary and although the filler concentration varies with different products, it usually constitutes 70 - 80 per cent. The higher the ratio between the dimensionally stable filler and the relatively dimensionally unstable resin, the lower will be the coefficient of thermal expansion of the composite resin.

One of the important properties of any restorative material is its cavity sealing ability and research is currently being conducted in an attempt to minimize the lack of adaptation of composite resins to tooth structure. Several factors are associated with the marginal adaptation of a composite resin restoration to tooth structure. Factors tending to improve marginal adaptation include
conventional retentive undercuts in cavities, the use of lingual locks and pins in cavity preparations, acid-etching procedures and water absorption by the resin particularly during the first 48 hours after placement. Factors tending to reduce marginal adaptation are polymerisation contraction of the composite resin and dimensional changes associated with the coefficient of thermal expansion. However, not all techniques aiming to improve marginal adaptation have proved to be successful; restorations with lingual locks often failed because the modulus of elasticity of the composite resin was inadequate. Composite restorations retained only by pins frequently failed because, although they provided retention of the composite to the tooth, they did little to improve marginal adaptation which usually deteriorated as a result of polymerisation shrinkage of the composite and dimensional changes associated with the difference in coefficient of thermal expansion and contraction between the composite resin and tooth. Failures occurred clinically despite claims by Chan et al (1977) of excellent adaptation of the composite around the pin and claims by Dilts et al (1973) of only a slight decrease in compressive and tensile strengths of the set composite incorporating pins.
Recently, the clinical relevance of the difference in coefficient of thermal expansion between tooth and composite resin has been questioned (Asmussen and Jorgensen, 1978; Glyn Jones et al, 1978). Evidence has shown that "provided there is a relatively small wall-to-wall polymerisation contraction and an adequate expansion due to water absorption, temperature changes within realistic limits will not influence the marginal integrity" (Asmussen and Jorgensen, 1978).

Other factors which may influence marginal adaptation include the removal of weak or already fractured enamel at the cavity margin during cavity preparation and the different finishing techniques which include the placement of bevels on all cavity margins and the use of surface glazes over the contoured composite resin.

More recent attempts to improve marginal adaptation and overcome the retentive problems of composite resins have led to the introduction of the technique of acid-etching enamel cavity walls, which has been successful in minimizing some of the problems of retention and marginal adaptation. This has, in turn, led to the widespread use of acid-etching as a relatively routine procedure in cavity preparation either in conjunction with conventional retentive undercuts or as the sole means of retention.
Unfilled resins of low viscosity have been suggested by some authors to be a necessary addition when restoring acid-etched enamel with composite resins. Buonocore et al (1968), and McLundie and Messer (1975) suggested that improved surface penetration would be possible with a resin of reduced viscosity. However, other authors have considered the unfilled resin to be unnecessary (Jorgensen, 1975, a; Pahlavan et al, 1976).
CHAPTER 6

LABORATORY STUDIES RELATED TO MICROLEAKAGE

6.1 Introduction

Close contact between composite resin and tooth enamel is an important factor influencing retention, marginal seal and the life expectancy of composite resin restorations. The failure of many existing resin restorations to seal cavities adequately and minimize microleakage may be responsible for pulp pathology, post-operative sensitivity, discolouration, recurrent caries and the hastened breakdown and dissolution of materials.

Recently, the technique of acid-etching enamel for the purpose of increasing the mechanical retention of various types of composite resins to enamel and thus reducing microleakage, has been extensively studied. Optical and scanning electron microscopic examination of phosphoric acid-etched surfaces reveals a fairly uniformly roughened, pitted surface, caused by preferential dissolution of prism cores. Etching the tooth converts a previously hydrophobic, low energy surface to a more wettable, hydrophilic, high energy surface (Newman and Facq, 1971) possessing an increased surface area with numerous retentive sites for tag formation.
6.2 Adaptation of Resins to Tooth Structure

A study by Mitchem and Turner (1974), noted two basic factors determining the retention of resins after acid-etching of the enamel; first, the "wettability" of the surface or the ability of the resin both to adapt closely to the etched surface and to penetrate into available micropores and, secondly, the inherent strength of the adapted resin tags.

Many studies have indicated that there is a marked increase in bond strengths of resins applied to acid-etched enamel, compared with resins applied to unconditioned surfaces (Laswell et al, 1971; Lee et al, 1971; Brauer and Termini, 1972).

Studies of the resin enamel interface have shown the presence of numerous projections emanating from the under-surface of the resin and interdigitating with the adjacent enamel irregularities (Buonocore et al, 1968; Gwinnett and Matsui, 1967; Gwinnett, 1972 and 1973, b; Sheykholeslam et al, 1972; Silverstone, 1974; Myers et al, 1974).

Buonocore (1955) was probably the first to study these projections or tags and concluded that they were an extension of the resin into enamel micropores created by acid-etching. Later, Gwinnett and Matsui (1967) and Buonocore et al (1968) noted the presence of filamentous tag like "extensions" of resin 15 – 20 micrometres in length at the resin-enamel interface. Since 1955 there have been a wide range of values reported for
tag lengths. Gwinnett and Buonocore (1965) noted an average tag length of 10 micrometres using transmitted light microscopy and polarised light for observation, and Sharp and Grenoble (1971) found, using light microscopy, tag lengths of 15 - 30 micrometres, after etching with 50 per cent phosphoric acid for two minutes. Gwinnett and Ripa (1973), in an \textit{in vivo} study, reported average tag lengths of 25 micrometres using light microscopy. Silverstone (1974 and 1975) observed, by light microscopy and ultra violet light fluorescence microscopy, average tag lengths of 50 micrometres produced after acid-etching with 30 per cent phosphoric acid for one minute, and Asmussen (1977, c) also observed tag lengths of 50 micrometres, with the light microscope, following acid-etching with 35 per cent phosphoric acid for one minute.

In contrast, tag lengths measured by scanning electron microscopy differed markedly from those seen using light microscopy. The scanning electron microscope provides a means of studying the resin-enamel interface by direct observation of the adaptation of restorative materials to the margins and walls of cavity preparations (Boyd and Knight, 1969). Studies by Jorgensen(1975, b) and Jorgensen and Shimokobe (1975) found tag lengths of 8-9 micrometres, and Pahlavan et al (1976) observed tag lengths ranging from 5-10 micrometres in length after etching for one minute with 50 per cent phosphoric acid.
A number of researchers have reported a difference in tag length measurements observed by light and scanning electron microscopy and considered that the technique of demineralisation of enamel, subsequent handling, coating and examination of the specimen for scanning electron microscopy might have led to tags fracturing (Gwinnett and Ripa, 1973; Silverstone, 1975; Pahlavan et al., 1976). Silverstone (1975) indicated that many published photomicrographs demonstrated tags that had been inadvertently damaged so that only the base of the tag remained. He suggested that demineralisation of enamel, during specimen preparation, should be halted before all enamel is removed, so that the bases of the tags are exposed but their extremities are still supported and embedded in enamel. Tags prepared in this way measured 50-60 micrometres in length before disappearing into the enamel.

Previous discussion indicated that enamel exposed to 30-40 per cent phosphoric acid for short periods provided a suitable surface for bonding. Once this surface was prepared, deep penetration of the resin into the etched enamel was necessary to ensure effectiveness of the procedure. Considerable controversy exists, however, concerning the need for an intermediary resin of low viscosity to penetrate the acid-etched surface prior to the placement of the restorative resins.
Longer tag lengths and better surface penetration by materials with a lower viscosity have been suggested (Buonocore et al, 1968; McLundie and Messer, 1975). Dogon (1975 and 1976) found that the frequency of tags as well as their lengths increased as the viscosity of the resins decreased. It was observed that the penetration of resins was markedly reduced if its viscosity rose above 350 centipoise.

However, many authors found no such correlation when comparing the tag lengths of low viscosity, unfilled resins with those of the more viscous composite resins (Jorgensen, 1975, a; Jorgensen and Shimokobe, 1975; Pahlavan et al, 1976; Barnes, 1977, b; Asmussen, 1977, c; Breakspere and Wilton, 1977; Low et al, 1978). They also observed that composite resins replicated the detail of the etched enamel as accurately as the non-composite resins and concluded that adaptation of the resin to the microrelief was not dependent on the viscosity of the composite material as a whole but on the viscosity of its resinous components.

Some researchers reported no changes in tag lengths with changes in viscosity, but did find a significant improvement in bond strengths for a composite system incorporating a low viscosity resin (Lee et al, 1971; Meurman and Nevaste, 1975;
Short et al, 1976). In contrast, Low and von Fraunhofer (1976), Rider (1977) and Rider et al (1977, a and b) found no significant increase in the bond strengths of composite resin to etched enamel when using a low viscosity resin.

Lee et al (1971) suggested that the superior retention and adaptation provided by acid-etching could also result in a reduction in microleakage. The use of a low viscosity resin in addition to a composite resin has considerably improved marginal adaptation and inhibited microleakage (Hembree and Andrews, 1976, c; Forsten, 1977; Qvist and Qvist, 1977; Luscher et al, 1978; Brannstrom and Nordenvall, 1978).

6.3 Microleakage Occurring at the Interface between Tooth and Resin

The lack of adaptation between the restorative material and tooth structure almost inevitably leads to the seepage of harmful agents along the interface between the restoration and the tooth. Microleakage has been defined as the clinically undetectable passage of bacteria, fluids and molecules or ions between a cavity wall and the restorative material applied to it (Kidd, 1976, b). Microleakage has been implicated in a variety of clinical conditions including recurrent caries, tooth discoloration, hypersensitivity, pulp pathology and hastened
breakdown of filling materials (Kidd, 1976, a and b; Torney et al, 1977).

A number of causes for microleakage have been suggested; most important is the lack adaptation of the restorative material to tooth structure which could result from any one of a number of clinical events, including loss of resin from the restorative material by dissolution, fracture of either the restorative material or the tooth (Roydhouse, 1968), polymerisation contraction of the restorative material, breakage of the marginal seal during thermal cycling or porosity of the material (Al-Hamadani and Crabb, 1975). Other contributing factors which may initiate microleakage include the performance of the operator (inadequate cavity preparation or poor manipulation of the material), the behaviour of the fluid penetrant and the pressure differences between the external surface and the internal face of dentine (Roydhouse and Weiss, 1967).

The detection of microleakage has involved many different techniques, these include:
(i) Dye tracers (6.31);

(ii) Radioisotope tracers (6.32);

(iii) Air pressure (6.33);

(iv) Bacteria (6.34);

(v) Neutron activation analysis (6.35);

(vi) Artificial caries/Gelatin gel (6.36).

6.31 Dye Tracers

The use of organic dyes as tracers is one of the oldest but most common methods of microleakage detection. Early experiments were carried out in glass tubes roughened to simulate dental tissue surfaces (Grossman, 1939; Massler and Ostrovsky, 1954), but more recent studies have used extracted human and bovine teeth (Tani and Buonocore, 1969; Baharloo and Moore, 1974; Kopel et al, 1975; Eriksen and Buonocore, 1976, a and b; Speiser and Kahn, 1977; Luscher et al, 1977 and 1978). The method involves placing a restoration in an extracted tooth and immersing the specimen in dye solution. After an interval of time, the tooth is removed, washed, sectioned and examined to establish the extent of dye penetration.
6.32 Radioactive isotope tracers

Extensive use has also been made of radioactive salts - usually $^{45}$Ca (Hawkins et al., 1976; Hembree and Andrews, 1976, a, b and c; Eliasson and Hill, 1977), but $^{14}$C (Powell, 1975) and $^{131}$I (Galan et al., 1976) have occasionally been used as the tracer. The technique is similar to that for the dyes except that autoradiographs are taken of the cut surfaces of the specimen to detect the degree of penetration by the isotope tracer.

Both dye and isotope experimental results have been quantified by assigning numerals to defined depths of penetration (Going et al., 1960, b). This somewhat subjective method of quantification is one disadvantage of these techniques (Going, 1972). In addition, Roydhhouse (1968) has questioned whether the potential for microleakage demonstrated by experiments carried out in vitro was a reality in vivo, where such factors as pulpal hydrostatic pressure must be taken into account. However, Swartz and Phillips (1961) and McCurdy et al (1974) found a close correlation between results obtained from both in vitro and in vivo studies and supported the suggestion of expanding in vitro studies to obtain further information regarding both the materials and the effects of various
manipulative techniques on the marginal integrity of dental restorations.

6.33 Air pressure

This method of examining microleakage delivers air to the base of the restoration by way of the pulp chamber. It has been developed so that there now exists a reproducible method which has the particular advantage of studying microleakage over a period of time, since examination of the specimen does not necessitate its destruction. The main limiting factors of this technique are its restriction to in vitro use and that it does not simulate conditions present in the tooth or the mouth (Going, 1972; Kidd, 1976, a).

6.34 Bacteria

The use of bacteria in the study of microleakage would seem to be a more clinically oriented test, because it can be related both to the caries process and to its recurrence around restorations. Unfortunately the bacterial tests have the disadvantage that the results are qualitative rather than quantitative (Going, 1972; Kidd, 1976, a).
6.35 Neutron activation analysis

This technique was used by Going et al (1968) to study microleakage both \textit{in vitro} and \textit{in vivo}. It has the advantage that results can be quantified but also has limitations; the teeth must be extracted for irradiation and analysis, the path and depth of tracer penetrations are not well defined unless serial sections are made, and experimental costs are high.

6.36 Artificial caries

Artificial secondary caries-like lesions have been produced \textit{in vitro} using either bacterial cultures or a chemical system, the acidified gelatin gel technique. The use of this method in the study of microleakage has the advantage that microleakage may be linked directly with the development of the artificial lesion (Kidd, 1976).

6.4 Some Factors Influencing the Microleakage around Composite Resin Restorations

A number of authors have demonstrated that etching the enamel walls of intracoronal cavities prior to the placement of resins enhances adaptation and retention and either greatly reduces microleakage (Rafei and Moore, 1975; Dogon, 1975; Oritz et al, 1976; Hembree and Andrews, 1976, b and c; Eliasson and Hill, 1977), or eliminates it (Retief, 1973, b).
These results have led many investigators to recommend routine etching of all cavity walls prior to the placement of resin restorations (Lee et al, 1971; Retief, 1973, b; Adipranoto et al, 1975; Jorgensen and Shimokobe, 1975; Kopel et al, 1975; Eames et al, 1976). However, work by Baharloo and Moore (1974) failed to establish that a durable, adhesive bond was formed between the etched enamel and the composite resin. Furthermore, Luscher et al (1977) noted that in conventional cavities, neither was dye penetration decreased nor marginal adaptation improved when enamel margins were etched prior to placement of the restoration. The results of a study by Speiser and Kahn (1977) indicated that acid pre-treatment of the enamel of butt joint cavities significantly increased microleakage.

Several factors have been found to influence significantly the microleakage pattern around composite resin restorations:

(i) The effect of different cavity designs (6.41).

(ii) The effect of different finishing times (6.42).

(iii) The effect of thermal cycling and ageing (6.43).
6.41 The Effect of Different Cavity Designs

The introduction of the technique of etching cavity walls prior to the placement of composite resins has resulted in controversy over the design of cavity preparation and in particular, the finishing of the cavo-surface angle; some have recommended the placement of a bevel on all cavity margins, others have associated the need for a bevel with the use of acid-etching or an unfilled resin layer.

Buonocore and Sheykholeslam (1973) suggested that the acid-etch technique may be used to greater advantage with certain modifications to conventional cavity design, such as bevelling the cavo-surface angle. In the absence of acid-etching, however, Gilmore and Lund (1973) suggested that the cavity form for all tooth coloured materials should be box shaped with a cavo-surface angle refined to a right angle; a bevel should be avoided in order to prevent an overlay of material on the tooth which would be susceptible to fracture and staining. Microleakage studies carried out by Kopel et al (1975) and Kempler et al (1976) using organic dyes as tracers, indicated that the least amount of microleakage was found in those teeth restored with composite resin to a 90 degree butt
joint against etched enamel cavity walls which were first coated with a low viscosity resin. On the other hand, work by Buonocore et al (1973), using an organic dye as the tracer, and Eriksen and Buonocore (1976, a and b), using both an organic dye and radio-isotope to detect microleakage, showed that restorations, placed in etched cavities prepared with a butt joint and first coated with an unfilled resin, failed to significantly reduce microleakage.

Two alternative cavity designs have been suggested in order to eliminate or reduce microleakage; the first was the application of the etchant not only to the walls but also to the tooth surface immediately adjacent to the cavity with a butt joint cavo-surface angle and the subsequent application of an unfilled resin before and/or after the insertion and finishing of the composite resin. Buonocore et al (1973), Buonocore (1975), Hembree and Andrews (1976, c), Eriksen and Buonocore (1976, a and b), found, using either basic fuchsin dye or isotopes ($^{45}\text{Ca}$ or $^{35}\text{S}$), that this method of "featheredging" either the composite resin and/or the low viscosity resin onto the etched enamel surrounding the cavity eliminated leakage. The second suggested modification was the placement of a bevel at the cavo-surface angle; both the bevelled area and the cavity wall were etched, followed by the application of an unfilled
resin before and/or after the insertion and finishing of the composite resin. Although this technique was suggested by many, including Denehy and Torney (1975), Hawkins et al (1976), Oilo and Jorgensen (1977) and Torney et al (1977), as a means of reducing microleakage, only Hawkins et al (1976) investigated the microleakage; they used $^{45}$Ca and commented that "a bevel preparation appeared to reduce the amount of leakage and seemed especially effective with etching".

The aims of these two alternatives were several: to seal the surface enamel surrounding the cavities where hair-line cracks might have been produced during cavity preparation, and to enhance bonding of the composite resin by providing a surface composed of enamel prisms more nearly in transverse than in longitudinal section (Gwinnett and Matsui, 1967; Buonocore et al, 1968; Buonocore, 1975; Hals and Laegreid, 1976). Clinically, problems with these two techniques include the difficulty of limiting the extent of the etch in inaccessible areas, for example, at the gingival margin of a Class III cavity, the difficulty of placing a bevel in inaccessible areas, the problems of contouring and removing resin that has extended beyond the etched area and the problems of maintaining the integrity of a thin film of unfilled or filled resin when it is subjected to wear, temperature fluctuations and physical and chemical deterioration (Kopel et al, 1975; Kempler et al, 1976).
6.42 The Effect of the Finishing Time

Composite resins, like other restorative resins, contract during polymerisation; volumetric contraction of composite resins is 2.0 per cent, and that of unfilled resins is 7.0 per cent (Phillips, 1973). Under normal conditions this results in the formation of a microspace between the material and the cavity wall, making possible the ingress of bacteria into the cavity (Asmussen, 1975; Barnes, 1977). Immersion in water causes these materials to expand as a consequence of water absorption — composite resins 0.3 mg/cm² and unfilled resins 0.6 mg/cm² over 24 hours (Phillips, 1973). This hygroscopic expansion, which commences four to six hours after insertion, reduces the width of the spaces created by polymerisation contraction but does not completely close all interspaces (Jacobsen, 1975).

Polishing composite resin restorations immediately after the initial set may result in a zone of fractured enamel 20–40 micrometres wide, beyond the margin of the restoration (Asmussen and Jorgensen, 1975). Regardless of whether the enamel at the margin is supported by sound dentine, or, as frequently occurs clinically, the enamel is unsupported, due to the incomplete removal of either unsupported or already fractured enamel, the contraction of the resin during
polymerisation ensures that, in the absence of acid-etching, the enamel at the margin is not supported by direct contact with resin during immediate finishing procedures. Should the enamel have been acid-etched, evidence suggests that the resin does remain in contact with the enamel, but, due to the natural tendency of the resin to contract when polymerising, the resin-enamel interface becomes stressed. In each case, therefore, whether the enamel was acid-etched or not, there is the likelihood of fracture and either subsequent loss of enamel or leakage around the fractured prisms when the abrading forces used in finishing are applied immediately. For this reason, it has been suggested (Asmussen and Jorgensen, 1975; Mitchem and Granum, 1976; Gjerdet, 1976) that, once gross removal of excess has been carried out, further finishing is delayed for at least 24 hours. It was reasoned that the absorption of fluid into the resin during this time and the related expansion would improve adaptation of the resin to the enamel at the margin, reduce stress at the resin-etched enamel interface and, in each case, tend to reduce the potential for enamel to fracture and cause subsequent microleakage.
The technique of acid-etching enamel cavity walls was introduced in an attempt to overcome the problems of adaptation of composite resins to cavity walls and the presence of microspaces resulting from polymerisation contraction. However, it has been suggested that acid-etching may itself be responsible for microleakage; Jorgensen et al (1975) showed that etching resulted in such effective bonding of the resin to the cavity walls of Class V cavities that subsequent strains, attributable to polymerisation contraction, thermal dimensional changes and elastic hysteresis caused the enamel surrounding the cavity to fracture. Sela et al (1975) observed that a composite resin restoration placed in a cavity with etched walls, was retained to the walls of one side of the cavity but on the opposite side there was a wider than normal gap between the composite resin and the etched cavity wall. Sela's findings have not been supported; it is conceivable, however, that this situation could arise when part of the cavity margin is on enamel, to which there is good retention, and part is on dentine, to which acid-etched retention is relatively poor.
6.43 The Effect of Thermal Cycling and Ageing

One of the essential physical properties of a dental restorative material is that it should retain its size and form after it has been inserted and contoured to the tooth.

In 1929, Fraser suggested the possible importance of variations in temperature on the marginal adaptation of restorative materials. In 1952, Nelson et al carried out some classic experiments studying the opening and closing of margins of acrylic resin restorations which were subjected to temperature changes. They concluded that "temperature changes of teeth and restorations in the mouth caused a fluid exchange between the teeth and restorations" and that "this marginal percolation was caused in part by a difference in the coefficients of thermal expansion of the fluid occupying the crevice between the tooth and restoration";

the coefficient of thermal expansion for tooth structure is $11.4 \times 10^{-6}/^\circ C$ and for unfilled resin is $81 \times 10^{-5}/^\circ C$ (Phillips, 1973). Asmussen (1974, a) has since demonstrated that marginal percolation may be at least partly caused by a difference in the coefficient of thermal expansion of tooth structure and composite resin, $30 \times 10^{-6}/^\circ C$ (Phillips, 1973). However, the relatively high coefficient of thermal expansion of
unfilled resins does not necessarily imply a high degree of marginal percolation during thermal changes (Asmussen and Jorgensen, 1978).

Asmussen (1977, a) investigated the polymerisation contraction of composite resins placed in acid-etched cavities and the effects of water absorption and temperature cycling on the formation of marginal gaps around these restorations. He found that acid-etching the cavity walls resulted in a bond between the composite resin and the enamel which counteracted the polymerisation contraction as long as the contraction forces remained smaller than the bonding forces. A similar restoration placed in an unetched cavity, however, would have resulted in the formation of gaps between the restoration and cavity margins, due to the lack of bonding forces. Asmussen reasoned that storage of the "etched" restorations in water resulted in hygroscopic expansion which reduced the stresses formed after the polymerisation contraction and also reduced the likelihood of marginal gap formation during subsequent thermal changes; it appeared that restorations that had expanded against cavity walls as a result of water absorption, could be cooled through a certain temperature range without the formation of marginal gaps. The temperature range that could be tolerated, however, was dependent upon the degree of polymerisation contraction, the hygroscopic expansion and the coefficient of thermal expansion (Asmussen, 1974, a and b). In summary, these
results indicated that if an acid-etching technique was used, polishing was postponed for at least 24 hours and limits of 15ºC and 50ºC were taken as the temperature range that composite resins experienced under oral conditions, marginal gaps due to polymerisation contraction and temperature changes might not form (Asmussen, 1977, a).

A study by Tani and Buonocore (1969) using basic fuchsin as the tracer, found that all materials tended to show essentially maximum leakage following temperature cycling.

More recently, however, Kidd et al (1978), using an artificial caries technique, found that specimens that had been temperature cycled showed either no change or a reduction in leakage compared with the controls which had not undergone temperature cycling. These results differed markedly with previous results from both dye and isotope tracer studies. This difference might be explained by the storage of the restorations in water for one week before temperature cycling in the experiments conducted by Kidd et al (1978). Glyn Jones et al (1978), using an artificial caries technique, examined a composite resin, an unfilled restorative resin and a resin containing an organic filler and found that there was no deterioration in the cavity sealing ability of the
resins lacking an inorganic filler after temperature cycling compared with the composite resin. Thus the relevance of the coefficient of thermal expansion to the marginal sealing ability of resin materials should, perhaps, be questioned. They concluded "that to condemn a material on the basis of its coefficient of thermal expansion may be an over-simplification of a more complex problem".

It is of some importance that very few studies have been conducted on the microleakage of specimens that have been "aged" for greater than one week before testing. Microleakage studies conducted by Hembree and Andrews (1976, a, b and c) indicated that marginal leakage after one year's ageing was significantly reduced or eliminated in those restorations where the enamel had been etched and coated with an unfilled resin compared with restorations placed in unetched cavities. Although a direct relationship has not been established between bond strength, ageing and microleakage, several workers studied bond strengths between resin and etched enamel; Lee et al (1971) found no degradation of the resin-enamel bond after six months storage in water; Joos et al (1976) showed that the bond between acid-etched enamel and composite resin became stronger with age or stressing.
6.5 Failure at the Resin-Etched Enamel Interface

Many factors are probably associated with failure at the resin-etched enamel interface.

Insufficient etching, causing failure at the interface, may result simply from inadequate exposure of the enamel to acid. Alternatively, the presence of either prismless enamel or enamel rich in fluoride content is responsible for altered etch patterns and, by causing a reduction in the number and length of resultant resin tags, reduces overall adaptation and retention of the composite resin (Sharp et al, 1971; Sheykholeslam et al, 1972). Although excessive etching can result in the formation of longer tags, if the tags become too long, rather than providing additional retention they may fracture when subjected to stress. Alternatively, exaggerated etching may weaken the tooth structure between the long tags causing the enamel itself to fracture under stress (Retief, 1976).

As previously discussed, finishing of the restoration immediately after placement may be a factor associated with failure at the resin-enamel interface. Although a sufficient etch may have been achieved initially and the composite adequately adapted to the etched surface, the bond may be
disrupted during finishing. Complete finishing, therefore, should be postponed for at least 24 hours to allow the composite resin to expand as a result of water absorption and reduce stress at the resin-etched enamel interface.

Although classical principles of cavity preparation, first formulated by Black, dictate the removal of all unsupported and fractured enamel (Blackwell, 1955), recently there has been some acceptance of the clinical fact that in certain cases, enamel not supported by sound dentine should be retained in preference to a greater display of composite resin (Denehy and Torney, 1976). In spite of this, enamel already fractured during cavity preparation must be removed. In addition, it should not be forgotten that the use of the acid-etch technique in combination with an unfilled resin may be sufficient to fracture enamel that appeared relatively strong, although unsupported. It is important to recognise the role of adequate cavity preparation on failure at the margins of composite restorations. The use of an acid-etch technique may therefore place greater emphasis on the need for judicious finishing of the enamel. A modified cavity design associated with the placement of a bevel, although removing weakened enamel, may predispose to greater failure, related to manipulative problems and the inherent properties of the resin (6.41).
Thermal cycling of restorations has also been implicated in failure at the resin-enamel interface probably as a result of eventual permanent deformation of the composite resulting in an increase in the size of the spaces formed between the composite restoration and the enamel.

Tensile loading tests carried out by Retief (1974, a and 1975, c) to determine the resin-enamel bond strength, found that the site of failure could occur within the resin, partly within the resin at the resin-enamel interface, within the enamel or at a combination of these sites. In a series of tests involving experimental bonds, only one specimen broke within the enamel, 24 partly in the resin and partly at the interface and 80 failed at the interface. In addition, Jorgensen et al (1975) and Asmussen (1977, a) observed that polymerisation and thermal contraction stresses of restorative resins bonded to the etched enamel of cavity walls could fracture the enamel at the cavo-surface angle.
6.6 Summary

From the discussion, it does appear that the acid-etching of cavity walls improves retention and overall adaptation of the composite resin to the enamel, together with a possible increase in bond strength. A majority of authors have concluded that microleakage is reduced, but not completely eliminated, by the use of an acid-etching procedure. Results vary, however, depending on the use of different cavity designs and the application of an unfilled resin. There is a general consensus that, for the restoration successfully to reduce microleakage, final finishing procedures should be postponed for at least 24 hours.
ORIGINAL INVESTIGATION
SCOPE OF THE INVESTIGATION

Inadequate sealing around composite resin restorations is responsible for microleakage which, in turn, may cause discolouration, recurrence of caries, sensitivity to thermal changes and other irritants and, eventually, pulp pathology. The technique of acid-etching enamel was introduced in an attempt to overcome some of these problems by producing a restoration with improved adaptation and seal at the margins. The review of the literature, however, indicated some controversy concerning the use of an etchant, the etching time, the type of etching solution, the design of the cavity and the use of a low viscosity resin; in addition, relatively little work has been carried out either on the effects of ageing the restorations for a period of twelve months or on the effects of particle size of the composite resin on the microleakage.

This investigation was in two parts. Part A examined microleakage in teeth using tracer dyes and a selection of composite resin materials. Part B examined the etched enamel surface available for bonding and the adequacy of adaptation of the composite resin to this surface, using a scanning electron microscope.
CHAPTER 7

METHOD - A

AN INVESTIGATION OF THE MICROLEAKAGE AROUND COMPOSITE RESIN RESTORATIONS

7.1 Materials used in the investigation

Teeth

Two hundred, sound, upper and lower premolar teeth extracted for orthodontic reasons were used. Immediately after extraction, the teeth were washed in water and then placed in a one per cent sodium hypochlorite solution for five minutes to dissolve any plaque and attached periodontal ligament. Each tooth was examined and any tooth showing visible evidence of cracking or bruising as a result of the use of extraction forceps was discarded. The proximal surfaces of the teeth were examined for the presence of calculus deposits which were removed using a suitable scaler. The teeth were then washed in water and stored in physiologic saline at 37°C until required.

Enamel etching solution

Concise Etching Liquid*, a 37 per cent ortho-phosphoric acid, was used to etch the enamel cavity walls prior to the application of the composite or unfilled resin.

* 3M Center, St. Paul, Minnesota, U.S.A.
Composite resin

Concise composite resin* was supplied in two 16 gram containers labelled Concise Paste A and Paste B. The low viscosity, unfilled resin was supplied in two 2.5 gram bottles, labelled Concise Enamel Bond System Resin A and Resin B (Fig. 7.1). When required, approximately equal quantities of Pastes A and B or an equal number of drops of Resins A and B were dispensed and mixed.

Estic composite resin** was supplied in the form of a 27 gram jar of Estic Paste and a 3 gram tube of Estic Catalyst (Fig. 7.2). A mixing slab was supplied on which the required amounts of Paste and Catalyst could be measured.

Dye

The tracer dye, used to detect microleakage, was 0.25 per cent toluidine blue dye which has a particle size of approximately 1.33 x 0.35 nm. (Craig, 1969).

7.2 Method of investigation

Forty teeth were assigned at random to each of five treatment groups; each group was subsequently subjected to a different testing regimen (Fig. 9.1).

* 3M Center, St. Paul, Minnesota, U.S.A.

** Kulzer - Ivoclar, West Germany.
Fig. 7.1 - Photograph of Concise/Enamel Bond composite resin material.
Fig. 7.2 - Photograph of Estic composite resin material.
Preparation of the cavity

Cavities of a Class V form were prepared on the mesial and distal surfaces of each tooth using a water-cooled tungsten carbide bur* at high speed; hand instruments were not used (Boyde, 1976; Lester, 1978). The mesial and distal surfaces were chosen instead of the buccal and lingual, because they were less likely to have been cracked during the forceps extraction. The external dimensions of each cavity were approximately 1.5 mm, occluso-gingivally, and 5.0 mm, bucco-lingually, and internally, the axial wall was placed just into dentine, an average depth of 1.5 mm. Ideally, the gingival margin of the cavity was placed at least 2 mm from the cemento-enamel junction, but in some cases, because of the curvature of this junction, it was a little closer. All cavity margins were checked to ensure continuity without irregularities and a cavo-surface angle of approximately 90°. Retention was placed, using a small round steel bur** (No. ½) at low speed, along the occluso-axial and gingivo-axial line angles. A reference groove was made on each tooth with the water-cooled tungsten carbide bur approximately 5 mm apical to the cemento-enamel junction to identify the surface.

* ELA, Germany.

When indicated in the experimental regimen, acid-etchant was applied for one or two minutes to the prepared enamel surface of the cavity walls. A small cotton pellet soaked in the etchant was gently rubbed against the cavity walls and although no specific attempt was made to etch the enamel of the tooth surface adjacent to the cavity, some etching was occasionally seen to have taken place. After the prescribed etching, the cavities were sprayed first with water, then with air and water for a total of fifteen seconds and finally dried with an uncontaminated air stream for five seconds (Dogon and Silverstone, 1975). All cavity walls were examined for the characteristic dull, matt appearance of adequately etched enamel (Buonocore, 1970).

Clinically, a layer of cement base is required to cover the exposed dentine of the axial wall; the base not only affords protection for the pulp against the restorative material but also restricts the pulp-ward passage of ions associated with marginal microleakage (Phillips, 1973). In this study, however, no base was applied because the subsequent interpretation of results was confined to an examination of the ability of the etching procedures and materials to confine microleakage to the region of the enamel cavity wall. The ability of a base to limit the pulpal spread of ions within dentine was not under investigation.
Preparation, placement and finishing of the composite resin

The 40 teeth allotted to each of the five treatment groups were divided into two (Fig. 9.2). Twenty teeth were restored with Concise composite resin in an unetched cavity on one proximal surface and with Concise composite resin in an etched cavity on the other surface; cavities in the remaining 20 teeth in each group were etched and restored, on one proximal surface with the low viscosity resin, Enamel Bond, followed by Concise composite resin and on the other proximal surface with Estic composite resin. In this way, within each group of 40 teeth, each restorative technique was used in 20 proximal cavities. The etching time varied according to the particular treatment group (Fig. 9.1).

Both the Concise and Estic were dispensed and mixed according to the manufacturers' directions. The viscosity of the mix made it necessary to force the material into the cavity to ensure the closest possible adaptation with the cavity walls. A small, tightly rolled pellet of cotton wool held in a pair of tweezers was used to force the first portion of the mixed resin into the retentive areas and line angles (Craig, 1969). The cavity was then slightly overfilled and a polyester matrix* band wrapped firmly around the tooth and secured. This was

left in place until the material had completely set, approximately five minutes later.

When Enamel Bond was required, the resin was dispensed, mixed according to the manufacturer's directions and applied to the etched cavity walls in a thin layer with a tightly rolled cotton pellet held in tweezers. The composite resin, Concise, was then immediately mixed and placed as already described, before the Enamel Bond had polymerised.

All restorations were finished approximately 15 minutes after the material had set. A standardised finishing routine was used; after the removal of any thin "flash" with a probe, the contour was adjusted and the margins of the restoration smoothed using an aluminium oxide white stone* at low speed by a fine grade composite finishing disc** containing a zirconium silicate abrasive. In this way, all the restorations were finished to the cavo-surface angle of the cavity and the marginal integrity was checked with a probe.

Experimental regimen

The restored teeth were subjected to different procedures prior to immersion in the dye; a summary is given in Fig. 9.1.

* Meisinger, Germany.

** 3M Center, St. Paul, Minnesota, U.S.A.
Two groups, Groups I and II, were stored in physiologic saline at 37°C for 24 hours before immersion in dye. Those cavities that required etching in Group I, were etched for one minute and those in Group II were etched for two minutes.

Two groups III and IV, investigated the effects of thermal changes on the integrity of the marginal seal. Group III cavities that required etching, were etched for one minute and those in Group IV were etched for two minutes. After finishing, teeth from both groups were stored in physiologic saline at 37°C for approximately half an hour before being thermally cycled. Thermal cycling was carried out by alternately immersing a number of gauze bags containing the teeth in water baths maintained at temperatures of 4°C and 60°C. The immersion time in each bath was 30 seconds and the teeth were subjected to a total of 100 cycles.

The remaining group, Group V, investigated the effects of "ageing" on the microleakage. Cavities in this group that required etching were etched for one minute. Teeth were stored in physiologic saline at 37°C for twelve months before immersion in dye.

**Immersion in the dye**

The teeth were removed from the physiologic saline storage baths or from the thermal cycling baths and were allowed to
dry for one hour. The entire tooth surface, except for the restoration and an area approximately one millimetre wide beyond the margin was covered with a layer of nail varnish in order to confine exposure of the dye. When the nail varnish had dried, approximately 30 minutes later, the specimens were immersed in stoppered bottles containing 0.25 per cent toluidine blue dye and stored for 24 hours at 37°C; the specimens were then removed from the solution, washed in running water for one minute to remove any excess dye, dried and prepared for sectioning.

Tooth sectioning

The roots of the teeth were removed and discarded and the crowns were embedded in blocks of dental plaster, leaving the occlusal surfaces exposed. Parallel cuts were made mesiodistally, in a longitudinal plane, through the restorations and the pulp chamber with a diamond-edged blade at 1,425 r.p.m. under a continuous water spray (Fig. 7.3). Two or three sections approximately one millimetre thick were obtained from each tooth, the number of sections depending on the size of the restorations and the shape of the tooth. In this way, in each group, from the original 80 proximal surfaces restored using the four techniques, a varying number
Fig. 7.3 - Photograph showing varnished tooth crowns embedded in dental plaster for sectioning.
of sectioned surfaces were obtained. The sections were allowed to dry and then stored dry in small stoppered vials, in a dark place, for subsequent examination.

Evaluation of the microleakage

The sections obtained from each experimental and restorative technique (for example, 24 hour-storage before immersion in the dye, Concise, with no acid-etching of the cavity) were numbered.

The restorations in each sectioned surface were examined using an optical microscope* at a magnification of x9 and the degree of penetration evaluated using a standardised system, similar to that used by Tani and Buonocore (1969). The system is diagramatically represented in Fig. 7.4. A zero (0) value was assigned where there was no evidence of dye penetration. When the section showed penetration of dye confined to an enamel wall, the grade of microleakage was assigned a value of one (1). When the dye had spread beyond that part of the cavity wall in enamel and onto a dentine cavity wall but remained short of the axial wall, the microleakage was assigned a value of two (2). When the dye had spread further to involve the axial wall of the cavity and associated dentinal tubules, the grade of microleakage was assigned a value of three (3). Finally, in

Fig. 7.4. - Photograph illustrating the grading system used in the evaluation of dye penetration.
Fig. 7.5. - Photographs showing examples of the different grades of microleakage: a, Grade 0; b, Grade 1; c, Grade 2; d, Grade 3; e, Grade 4.
cases showing maximum microleakage, when the dye had spread to involve the enamel and dentine of the cavity wall and along the dentinal tubules to the pulp, the microleakage was assigned a value of four (4). Figures 7.5, a to e, show examples of each grade of microleakage.

For each restoration that exhibited microleakage, the site of the microleakage was recorded; that is dye tracer was observed along the gingival wall, the occlusal wall or both these walls. In all these restorations, microleakage was found along the gingival wall, none showed microleakage only along the occlusal wall and approximately eight per cent showed microleakage along both walls.

Each surface was labelled and the restorations in the sectioned surfaces were evaluated on two different occasions to examine the reproducibility of the gradings. Table 7.1 shows the number of disagreements between the first and second evaluations of the microleakage occurring around the restoration. A total of 1292 surfaces of sectioned restorations were examined in the five treatment groups; the number of surfaces, in which there was disagreement between the first and second evaluation, was 53 (or 4.1 per cent). Of these disagreements, 29 (or 55 per cent) occurred between Grades 0 and 1; that is, on one evaluation the microleakage was assigned a Grade of 0 and, on the other evaluation, a Grade of 1. The remaining disagreements
TABLE 7.1.

Number of disagreements recorded between the first and second evaluations of the microleakage.

<table>
<thead>
<tr>
<th>Group</th>
<th>Number of surfaces</th>
<th>Number of disagreements</th>
<th>Number of disagreements recorded between grades of microleakage</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Grades</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0-1</td>
</tr>
<tr>
<td>I</td>
<td>194</td>
<td>6</td>
<td>2</td>
</tr>
<tr>
<td>II</td>
<td>270</td>
<td>8</td>
<td>6</td>
</tr>
<tr>
<td>III</td>
<td>276</td>
<td>12</td>
<td>1</td>
</tr>
<tr>
<td>IV</td>
<td>296</td>
<td>14</td>
<td>10</td>
</tr>
<tr>
<td>V</td>
<td>256</td>
<td>13</td>
<td>10</td>
</tr>
<tr>
<td>Total</td>
<td>1292</td>
<td>53</td>
<td>29</td>
</tr>
</tbody>
</table>
occurred between Grades 1 and 2 (8 disagreements), 2 and 3 (11) and between Grades 3 and 4 (5). In no instance was the disagreement between non-consecutive grades. The low (4.1 per cent) level of disagreement between the first and second evaluations was considered to indicate an acceptable degree of consistency in evaluation; results expressed subsequently in this research represent, in each case, the findings of the first evaluation.

Results for each material and etching time within each of the five treatment categories have been grouped in the following manner, similar to the classification used by Speiser and Kahn (1977):

1. Microleakage confined to the enamel cavity wall.
   Those results indicating that, as a result of the etching time and/or because of the material type, microleakage was confined to the enamel cavity wall, Grades 0 and 1, have been grouped together.

2. Microleakage extending along both the enamel and dentine cavity wall.
   The remaining results (Grades 2, 3 and 4) indicating failure, because of the etching time or material, to limit microleakage to the enamel cavity wall, under the
conditions of testing, have been grouped together.

Of the 1292 surfaces evaluated twice, only eight (0.6 per cent) showed a disagreement occurring between Grade 1 and Grade 2 microleakage suggesting that this method of grouping the results was not only clinically relevant but also generally well defined in the experimental technique.

Statistical analysis of the data

Because of the non-parametric nature of the grading system, the determination of an average (mean) microleakage value for each material and etching time within each of the five treatment categories was not appropriate.

The fourfold (2 x 2) form of the Chi-square test was used to test the significance of differences occurring between different materials and etching times in their ability to limit microleakage to the enamel cavity wall. A value for Chi-square ($\chi^2$) was obtained by application of the equation (Maxwell, 1967, p.21):

$$\chi^2 = \frac{N(|ad-bc| - 0.5N)^2}{(a+b)(c+d)(a+c)(b+d)}; \hspace{1cm} \text{d.f.} = 1$$

in which a correction for continuity, the Yates correction, was applied at all times, where there was one degree of freedom (for 2 x 2 form of the test) and where N, a, b, c and d were
derived from the general 2 x 2 contingency table for the two attributes, A and B

<table>
<thead>
<tr>
<th></th>
<th>Not-A</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Not-B</td>
<td>a</td>
<td>b</td>
<td>a+b</td>
</tr>
<tr>
<td>B</td>
<td>c</td>
<td>d</td>
<td>c+d</td>
</tr>
<tr>
<td>Total</td>
<td>a+c</td>
<td>b+d</td>
<td>N</td>
</tr>
</tbody>
</table>

Differences between materials and etching times in their ability to limit microleakage were regarded as significant when the calculated value for $\chi^2$ exceeded the tabulated value at the one per cent level of confidence; that is, $\chi^2 > 6.635$, where d.f. = 1 (Documenta Geigy Scientific Tables) and the null hypothesis, that there was no difference between the observed and expected frequencies that could not be due to chance, was disproved.
CHAPTER 8

METHOD B

AN INVESTIGATION OF ETCHED ENAMEL SURFACES AND THE
ADAPTATION OF COMPOSITE RESINS

8.1 Materials used in the investigation

The enamel etching solution and the composite and unfilled resins used were identical to those in the first part of this study (An Investigation of the Microleakage around Composite Resin Restorations).

Teeth

Twenty sound premolar and incisor teeth, extracted for orthodontic reasons, were cleaned as previously described, examined for damage occurring during extraction and stored in physiologic saline at 37°C until required.

8.2 Method of investigation

Specimen preparation

In the first part of the study, enamel surfaces were used to investigate the etched walls of prepared cavities and the effects of different etching times. The surfaces examined were transverse and longitudinal sections of teeth, bevelled cavo-surface angles and the enamel tooth surface; in each case tooth preparation was carried out using a water-cooled
tungsten carbide bur at high speed. The transverse sections were prepared by sectioning the crown of the tooth approximately 4 mm occlusal to the cemento-enamel junction and the longitudinal sections were prepared by sectioning through the crown mesio-distally at the approximate position of the labial wall of a Class III cavity. The bevelled cavo-surface angle was prepared by placing a 45° bevel on one of the transverse sections. In addition to examining the intact enamel tooth surface, the labial surface of a tooth was investigated after the removal of approximately 0.3 mm of surface enamel before application of the etchant.

From the transverse sectioning procedure two surfaces were obtained for examination from each tooth; each cut surface was divided into four parts, demarcated by lines prepared with a sharp chisel (Fig. 8.1 and 8.3, a); one part was examined unetched and the remaining parts were etched for one, two and four minutes. The longitudinal sectioning of lower incisor teeth produced one usable surface that was divided into two parts by a vertical line (Fig. 8.2 and 8.3, b); one part was examined unetched and the other was etched for one minute; a
Fig. 8.1. - Diagram illustrating the preparation of a transverse section through a premolar tooth.
Fig. 8.2. - Diagram illustrating the preparation of a longitudinal section through a premolar tooth.
Fig. 8.3. - Photographs showing - a, Transverse section through a premolar tooth; b, Longitudinal section through an incisor tooth. Mounted and coated for examination in a scanning electron microscope.
second tooth, from the same mouth, was used to examine the effect of the etching for two and four minutes. Both the labial tooth surface and reduced enamel surface were divided by two horizontal lines (Fig. 8.4) into three parts which were etched for one, two and four minutes. Two teeth were used, one to examine the etched tooth surface and the other to examine the etched surface after the removal of the small amount of enamel.

Each surface that required etching was etched with 37 per cent phosphoric acid using a small cotton pellet and a gentle rubbing action. The etchant was removed by washing with water followed by a water and air spray for a total of fifteen seconds; the surface was dried with an uncontaminated air stream for five seconds.

In the second part of this study, transverse surfaces were prepared, etched and cleaned as previously described. Three composite resin materials - Estic, Concise, and the unfilled resin, Enamel Bond, used in combination with Concise - were applied to the prepared enamel surfaces in order to investigate adaptation to the etched enamel and the nature of the fitting surface. Each material was prepared according to the manufacturer's directions and adapted to the etched enamel surface in a manner similar to that described in Part A for
Fig. 8.4. - Diagram illustrating the preparation of subsurface labial enamel of an incisor tooth.
the placement of material into the prepared cavity. For each specimen, a small stainless steel orthodontic bracket with a pre-welded mini-mesh bonding pad* was positioned on the adapted resin to facilitate later handling of the specimen and to provide improved conductivity in the scanning electron microscope (Fig. 8.5). Preparation of the resin negative of the etched enamel surface was completed by dissolving all enamel in a five per cent nitric acid solution; complete dissolution was achieved in approximately four days after which the remaining softened dentine separated easily from the resin surface.

In the third part of this investigation each of the resin materials was adapted to the surface of a metal slab possessing machined grooves of different widths ranging in size from 25 micrometres to 300 micrometres. In turn, each material was mixed and adapted to the grooves on the surface of the slab in the manner previously described. A plastic ring 8 millimetres in diameter and 1.5 millimetres high, loaded with additional material was used to confine the material as it set under a load of 150 grams (Fig. 8.6).

* Unitek, Melbourne, Australia.
Fig. 8.5. - Photograph showing a composite resin negative mounted on a stainless steel orthodontic bracket.
Fig. 8.6. - Photographs of - a, The metal slab with machined grooves; b, A composite resin negative prepared from the metal slab, mounted and gold coated.
Examination of the specimens

The prepared specimens were cleaned ultrasonically in distilled water for ten seconds and then transferred to trays lined with lint-free tissue paper. After a drying period of 30 minutes (longer drying times caused cracks to develop in the teeth), the specimens were mounted onto metal stubs using silver conducting paint (colloidal silver) and were then coated with gold to a thickness of approximately 20 - 40 nanometres in a sputter coating unit*.

The specimens were examined in a scanning electron microscope**, by the mode of secondary electron emission, using accelerated voltages in the range 5 - 15kV. Specimens were examined initially on the television screen for area location and then on the cathode ray tube screen at a scanning speed of 2.5 seconds per frame for final area location and focussing. Photographs were taken at magnifications of x200, x600 and x2,000, using the

* Dynavac - Dynavac High Vacuum Pty. Ltd., Victoria, Australia.

** JSM-U3 - Japan Electron Optics Laboratory Co. Ltd., Tokyo, Japan.
photographic attachment SMU3-CS1 with Kodak Plus-X pan professional film (5-PXP 120), at f/11 and a scanning speed of 50 seconds per frame. Original magnifications of the photographs were one-half that which was shown on the scanning electron microscope cathode ray tube screen.

* Eastman Kodak Co., Rochester, New York, U.S.A.
CHAPTER 9

RESULTS - A

AN INVESTIGATION OF THE MICROLEAKAGE AROUND COMPOSITE RESIN RESTORATIONS

9.1 Introduction

Using the method described, the microleakage occurring around composite resin restorations, \textit{in vitro}, was investigated by examining:

(i) The microleakage around composite resin restorations placed in unetched cavities (9.3).

(ii) The influence of etching the enamel cavity wall on the microleakage (9.4).

(iii) The influence of the etching time on the microleakage (9.5).

(iv) The influence on the microleakage of the type of composite resin material — the particle size of the inorganic filler and the viscosity of the resin component (9.61).

(v) The influence on the microleakage of ageing and thermal-cycling the restorations (9.62).

Tables 9.1 - 9.5 present data for the microleakage of restorations from the five experimental groups (Figure 9.1), namely,
Figure 9.1. Experimental regimen.

200 teeth

- NaHClO solution for 5 minutes
- Scaling
- Stored in physiologic saline at 37°C

Divided randomly into five groups

<table>
<thead>
<tr>
<th>Group</th>
<th>Group</th>
<th>Group</th>
<th>Group</th>
<th>Group</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>II</td>
<td>III</td>
<td>IV</td>
<td>V</td>
</tr>
</tbody>
</table>

Each tooth in each group restored in one of two ways (Fig. 9.2).

- Etching time, when required:
  - 1 minute
  - 2 minutes
  - 1 minute
  - 2 minutes
  - 1 minute

Storage time in physiologic saline at 37°C after restoration and prior to immersion in dye:

- 24 hours
- 24 hours
- 30 minutes
- 30 minutes
- 12 months

Thermal-cycled:

- No
- No
- Yes
- Yes
- No
I. Teeth in which cavities were etched, when this was indicated, for one minute and stored for 24 hours before immersion in dye (Table 9.1).

II. Teeth in which cavities were etched, when indicated, for two minutes and stored for 24 hours before immersion in dye (Table 9.2).

III. Teeth in which cavities were etched, when indicated, for one minute and thermal-cycled before immersion in dye (Table 9.3).

IV. Teeth in which cavities were etched, when indicated, for two minutes and thermal-cycled before immersion in dye (Table 9.4).

V. Teeth in which cavities were etched, when indicated, for one minute and aged for 12 months before immersion in dye (Table 9.5).

In these tables, the results are presented for each of the four types of restorations placed (Figure 9.2),

A Concise placed in an unetched cavity

B Concise placed in an etched cavity

C Concise with Enamel Bond placed in an etched cavity

D Estic placed in an etched cavity.
Figure 9.2. Restorations* placed in the 40 teeth from each of Groups I - V.

*One restoration in each proximal surface.
For example, Table 9.1, row three, shows that 61 surfaces were examined from the 20 teeth that had been restored with Concise composite resin and Enamel Bond placed in cavities etched for one minute, and immersed in dye after storage for 24 hours. Of these surfaces, 16 showed microleakage graded at 0, eight showed microleakage graded at 1, eight showed microleakage graded at 2, 14 showed microleakage graded at 3 and 15 showed microleakage graded at 4. In these results, 24 surfaces (39.3 per cent) exhibited microleakage confined to the enamel cavity wall (that is, Grades 0 and 1) and 37 surfaces (60.7 per cent) exhibited microleakage extending beyond the enamel and onto the dentine cavity wall.

Table 9.6 summarizes the data presented in Tables 9.1 to 9.5 by indicating — for a particular material, etching time and test procedure — the percentage of surfaces examined which showed microleakage confined to the enamel (Grades 0 and 1). This data is used subsequently to illustrate the results by means of histograms.
TABLE 9.1

Microleakage of Group 1 restorations - cavity etched for one minute, storage for 24 hours.

<table>
<thead>
<tr>
<th>Material</th>
<th>Total number of surfaces evaluated</th>
<th>Number of surfaces in each grade of microleakage</th>
<th>Surfaces with microleakage confined to enamel</th>
<th>Surfaces with microleakage in enamel and dentine</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Grade</td>
<td>Number</td>
<td>Per Cent</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0  1  2  3  4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Concise, unetched</td>
<td>36</td>
<td>0  2  4  21  9</td>
<td>2</td>
<td>5.6</td>
</tr>
<tr>
<td>Concise + etch</td>
<td>36</td>
<td>10  7  11  5  3</td>
<td>17</td>
<td>47.2</td>
</tr>
<tr>
<td>Concise + E.B.* + etch</td>
<td>61</td>
<td>16  8  8  14  15</td>
<td>24</td>
<td>39.3</td>
</tr>
<tr>
<td>Estic + etch</td>
<td>61</td>
<td>11  8  22  6  14</td>
<td>19</td>
<td>31.1</td>
</tr>
</tbody>
</table>

*Enamel Bond.
TABLE 9.2

Microleakage of Group II restorations - cavity etched for two minutes, storage for 24 hours.

<table>
<thead>
<tr>
<th>Material</th>
<th>Total number of surfaces evaluated</th>
<th>Number of surfaces in each grade of microleakage</th>
<th>Surfaces with microleakage confined to enamel</th>
<th>Surfaces with microleakage in enamel and dentine</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0 1 2 3 4</td>
<td>Number Per Cent</td>
<td>Number Per Cent</td>
</tr>
<tr>
<td>Concise, unetched</td>
<td>68</td>
<td>0 4 7 40 17</td>
<td>4 5.9</td>
<td>64 94.1</td>
</tr>
<tr>
<td>Concise + etch</td>
<td>68</td>
<td>41 21 4 2 0</td>
<td>62 91.2</td>
<td>6 8.8</td>
</tr>
<tr>
<td>Concise + E.B.* + etch</td>
<td>67</td>
<td>40 19 8 0 0</td>
<td>59 88.1</td>
<td>8 11.9</td>
</tr>
<tr>
<td>Estic + etch</td>
<td>67</td>
<td>26 33 2 3 3</td>
<td>59 88.1</td>
<td>8 11.9</td>
</tr>
</tbody>
</table>

*Enamel Bond.
<table>
<thead>
<tr>
<th>Material</th>
<th>Total number of surfaces evaluated</th>
<th>Number of surfaces in each grade of microleakage</th>
<th>Surfaces with microleakage confined to enamel</th>
<th>Surfaces with microleakage in enamel and dentine</th>
</tr>
</thead>
<tbody>
<tr>
<td>Concise, unetched</td>
<td>65</td>
<td>0 5 23 21 16</td>
<td>5 7.7</td>
<td>60 92.3</td>
</tr>
<tr>
<td>Concise + etch</td>
<td>65</td>
<td>0 3 48 8 6</td>
<td>3 4.6</td>
<td>62 95.4</td>
</tr>
<tr>
<td>Concise + E.B.* + etch</td>
<td>73</td>
<td>8 5 17 14 29</td>
<td>13 17.8</td>
<td>60 82.2</td>
</tr>
<tr>
<td>Estic + etch</td>
<td>73</td>
<td>2 16 45 0 10</td>
<td>18 24.7</td>
<td>55 75.3</td>
</tr>
</tbody>
</table>

*Enamel Bond.
TABLE 9.4

Microleakage of Group IV restorations - cavity etched for two minutes, thermal-cycled.

<table>
<thead>
<tr>
<th>Material</th>
<th>Total number of surfaces evaluated</th>
<th>Number of surfaces in each grade of microleakage</th>
<th>Surfaces with microleakage confined to enamel</th>
<th>Surfaces with microleakage in enamel and dentine</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Grade</td>
<td>Number</td>
<td>Per Cent</td>
</tr>
<tr>
<td>Concise, unetched</td>
<td>75</td>
<td>0, 1, 2, 3, 4</td>
<td>6</td>
<td>8.0</td>
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<tr>
<td>Concise + etch</td>
<td>75</td>
<td>0, 7, 50, 8, 10</td>
<td>7</td>
<td>9.3</td>
</tr>
<tr>
<td>Concise + E.B.* + etch</td>
<td>73</td>
<td>28, 6, 39, 0, 0</td>
<td>34</td>
<td>46.6</td>
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<tr>
<td>Estic + etch</td>
<td>73</td>
<td>26, 17, 30, 0, 0</td>
<td>43</td>
<td>58.9</td>
</tr>
</tbody>
</table>

*Enamel Bond.
TABLE 9.5

Microleakage of Group V restorations - cavity etched for one minute, aged for 12 months.

<table>
<thead>
<tr>
<th>Material</th>
<th>Total number of surfaces evaluated</th>
<th>Number of surfaces in each grade of microleakage</th>
<th>Surfaces with microleakage confined to enamel</th>
<th>Surfaces with microleakage in enamel and dentine</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td>Grade</td>
<td>Number</td>
<td>Per Cent</td>
</tr>
<tr>
<td></td>
<td></td>
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<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Concise, unetched</td>
<td>68</td>
<td>25</td>
<td>20</td>
<td>13</td>
</tr>
<tr>
<td>Concise + etch</td>
<td>68</td>
<td>24</td>
<td>29</td>
<td>14</td>
</tr>
<tr>
<td>Concise + E.B.* + etch</td>
<td>60</td>
<td>13</td>
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</tr>
<tr>
<td>Estatic + etch</td>
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</table>

*Enamel Bond.