An Investigation of the Properties of a New Resin Inlay System.

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Declaration.

This thesis is the original work of the author.

[redacted]

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Summary.

This study was a laboratory based investigation of a new tooth coloured inlay system for posterior teeth; the composite resin inlay. These inlays, although made from conventional composite resin materials are, generally, fabricated outside the mouth and subjected either to hydropneumatic curing in a heat\pressure oven or a secondary cure, following light curing, in a heat\light oven. It has been suggested that this method of fabrication and subsequent curing results in a filling with many advantages over conventional composite resin fillings. Through the study of three proprietary composite resin inlay systems (two light\heat cured and one heat\pressure cured) an attempt was made to assess these proposed advantages and, furthermore, to investigate aspects of the composite resin inlay technique which may be of some clinical relevance.

The differences in materials and curing regimes were examined. Two materials were light curing hybrid composite resins which were post cured using a heat\light oven. The third material was a microfilled composite which did not light cure; it was cured in a heat\pressure oven. Pressure, temperature, light intensity and curing duration within the "post cure" ovens differed between systems. It was considered that these differences may be important in production of the post curing effect.

Investigation of basic physical properties showed that post curing of light cure composite resins could improve certain physical properties in certain materials. These improvements in properties appeared to be material specific for a given post curing regime and could also be influenced by post curing temperature and duration of post cure. Hydropneumatic curing of a microfilled composite resin resulted in superior physical properties to literature values for similar materials cured by light. This was probably due to the reduced porosity of the set material. Although it cannot be excluded, no evidence was

observed for annealing as a process important in improving material properties as no change in physical properties was observed during a storage period of two months.

The physical properties of the dual curing composite resin luting cements, for use in these inlay systems were assessed. Properties for the two hybrid materials were significantly greater than those of the microfilled material and were similar to the light\cured composite with which they were used. However, the properties were significantly less than those of the post cured inlays, suggesting a potential weakness in the system. If allowed to self cure, as would be the case under a thick inlay values of flexural strength and hardness dropped by over 20% in both hybrid materials and by 50% in the microfilled material, suggesting that these materials are not truly dual curing at all. Curing time was assessed for the composite cements both with direct light exposure and when illuminated through 1.5mm of inlay. The introduction of inlay material resulted in a 3-4x increase in setting time. These findings suggest that very long exposure times would be necessary to cure the inlay cement to its full potential.

The proposed mechanism of improvement in physical properties was the increased degree of cure following post curing. Investigation of the materials using Fourier Transfer Infra Red (FTIR) spectroscopy showed this to be the case, with increases of 7% and 12% in methacrylate conversion being observed when light cured and post cured materials were compared. The degree of cure of the heat\pressure cured materials was found to be similar to the light\heat post cured materials with around 65% conversion. None of the materials tested exhibited cure of over 70%.

Fracture toughness testing of the composite resin inlay materials was carried out. The hybrid materials had a higher fracture toughness than the microfil, however no advantage was found in K_{IC} following post curing. This was suggestive of the fact that in addition to the increase in methacrylate conversion occurring during post curing, which increased the flexural modulus, other potentially weakening processes were occurring within the material balancing this effect. No effect was observed in K_{IC} following storage for periods of up to two months.

The increased methacrylate conversion of post cured materials has been cited as a reason for poor bond between inlay and luting resin. This potential for debonding was investigated and found not to be the case in heat\light post cured materials, but to be true of hydropneumatically cured composite. As the degree of cure was similar in all materials an alternative reason for debonding of this group was required. Through the use of scanning electron microscopy and confocal light microscopy differences in the fitting surface morphology and potential for micromechanical bonding were found between the groups. This suggested that the lack of porosity, caused by the use of pressure during curing of the microfilled materials, resulted in lack of micromechanical bonding sites on the fitting surface in this system leaving it liable to debond.

The fit of the three inlay systems was assessed on human teeth by measurement of fit of composite resin lute thickness. The fit of both hybrid, heat\light post cured materials was similar with lute thickness of 100μm or less, whereas that of the heat\pressure cured microfilled system was much poorer with gaps of over 200μm recorded. This was probably due to the single increment build up and sandblasting of the fitting surface of the latter system. This, poorer fitting accuracy, may place extra strain on the inlay\tooth bond and certainly exposes more of the physically inferior composite resin luting cement to the oral environment.

Leakage of the inlay systems was assessed *in vitro*. Surprisingly no differences were detected between inlay types; the increased lute thickness of the microfilled system having no effect. However, the use of a dentine bonding agent proved to be very important, with newer materials which altered or removed the smear layer proving the most effective.

Obviously, clinical trials are required to assess the value of this new technique. However, there were several areas where this study could help guide the dentist in his choice of inlay type and improve his understanding of the important handling characteristics of composite resin inlays necessary to obtain the best results.

Chapter 1. Introduction and Review of the Literature.

1.1 INTRODUCTION

Amalgam is the standard against which any filling material must be compared. Its long history and relative ease of placement make it convenient and efficient to use. Furthermore, relative to its competitors it is inexpensive. However, patients' aesthetic requirements have increased in recent years[1] resulting in more requests for tooth coloured posterior restorations. Coupled with the aesthetic demands, concern in the media regarding the potential toxicity of mercury, has prompted many patients to request non metallic fillings. "Oral galvanism", as this concern is termed by Molin[2], has been particularly prevalent in Scandinavia, while recent television coverage in the USA has heightened concern in that country[3]. Although the validity of this concern has been refuted[2], there is, nonetheless, a need for a strong, aesthetic restorative material not only for anterior but also for posterior teeth, if we are to meet the needs of our ever more demanding patients.

This study investigates the potential of a new aesthetic restorative technique as a filling for posterior tooth cavities.

1.2 AESTHETIC FILLING MATERIALS.

Silicate cements were introduced as an anterior restorative material around 1903 and for many years remained the only available white filling. Despite the fact that there were considerable improvements in these materials, their properties still left much to be desired. They were brittle and, consequently could not be placed in stress bearing areas (such as class IV restorations). Moreover, they eroded in oral fluids resulting in roughened surfaces which picked up stains, and, due to their lack of adhesion to tooth substance, leakage and staining often occurred at margins. They did, however, leach fluoride and this property was probably responsible for the relatively low incidence of secondary caries related to these filling materials[4].

Acrylic resin materials were introduced in the early 1950's. As an insoluble and relatively flexible material they were perceived as a major improvement over the silicate materials. Unfortunately, they suffered from poor colour stability and significant polymerisation shrinkage. This shrinkage, their low stiffness and high coefficient of thermal expansion, coupled with the lack of bond to tooth structure lead to marginal percolation and a high incidence of secondary caries[5]. The potential of inorganic fillers to decrease polymerisation contraction and improve the physical properties of the materials was recognised early on[6]. However, the problems involved in bonding filler to resin were not adequately addressed until the 1960's when the use of silane coatings allowed the polymer to adhere to the inorganic phase. These filled acrylic resins, although they showed improved clinical performance[7], did not become popular as the new Bis-GMA, composite materials, on which all today's materials are based, were introduced at the same time.

1.3 COMPOSITE RESINS.

1.3.1 Definition and Changes in Material.

A composite has been defined as "a combination of two chemically different materials with a distinct interface separating the components and having properties which could not be achieved by any of the components acting alone"[8]. In 1962 Bowen patented the original Bis-GMA based composite resin material[9]. This was synthesised from the reaction product of Bisphenol and Glycidyl methacrylate and had a silanated filler of quartz. A detailed description of the developments leading to the production of this material is given by Bowen[8].

This work formed the basis for production of the first commercial dental composite resins such as Adaptic (Johnson and Johnson Dental Products Inc) which first appeared in America in 1969.

Since the 1960's these materials have been constantly developed and updated in order to improve their physical properties and clinical characteristics. The improvements can be conveniently divided into 4 sections:

1) Resin technology

2) Filler technology

3) Resin\filler interface.

4) Curing procedure.

1.3.2 Resin Technology.

The resin in a composite material acts as a binder holding the filler particles together. In general the resins used in dental composite resins are composed of modified methacrylate or diacrylate oligimers.

Most of the composite resins used today still contain Bowen's original aromatic monomer Bis-GMA[10]. Bis-GMA is the acronym for 2,2-bis[*p*-(2-hydroxy-3 methacryloxypropoxy)phenylene]-propane the structural formula of which is given in Figure 1.1 .

Due to its large size and bulky central chain this molecule combines relatively low polymerisation contraction and high stiffness, when set, with the good reactivity afforded by the methacrylate groups. A very similar monomer, urethane dimethacrylate, is used in some composite resins. This material does not contain the aromatic rings of the central portion of the Bis-GMA molecule but has two large aliphatic moieties containing urethane groups. This

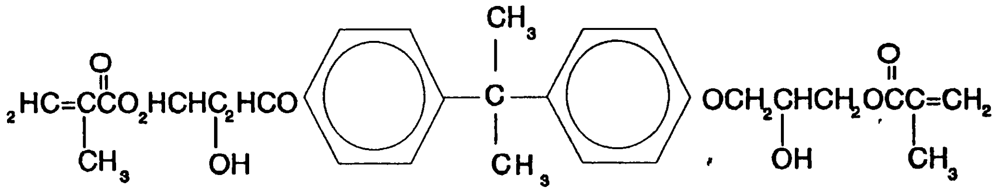
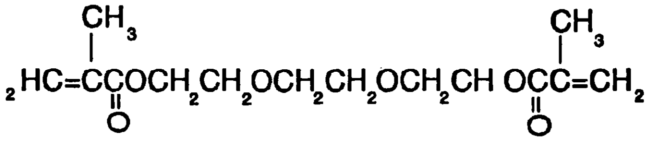


Figure 1.1: Structural formula of Bis-GMA



$$_{2}HC=\overset{CH_3}{C}\underset{O}{C}OCH_2CH_2OCH_2CH_2OCH_2CH\ O\underset{O}{C}\overset{CH_3}{C}=CH_2$$

Figure 1.2: Structural formula of TEGMA

aside there is little difference between the two molecules each having a bulky cental region, for strength and low shrinkage, and a methacrylate group at either end for polymerisation. It has, however, been suggested that urethane containing materials have greater flexibility which may improve their toughness[11].

Due to their bulky nature these resins are very viscous; consequently any restorative material made with them alone would be extremely difficult to handle. It is therefore necessary to dilute them with lower molecular weight monomers. The monomer most commonly used, for this purpose, is also the same as originally described by Bowen[9] i.e Triethylene glycol dimethacrylate (TEGMA) (Figure 1.2), although ethylene glycol dimethacrylate is used in some materials. However, in addition to reducing the viscosity of the material, due to its lower molecular weight, TEGMA also increases the polymerisation contraction[12]. It is therefore, necessary to balance handling characteristics against the detrimental effects of increased shrinkage on setting.

Other simpler methacrylates are often found in composite resin materials such as Bis-MA and Bis-EMA, however they are not greatly different from the original Bis-GMA and do not confer any significantly different properties upon the material. A breakdown of the resin formulation of many of the proprietary composite resin materials has been given by Ruyter[13].

The setting reaction of composite resins is a simple free radical addition polymerisation reaction where carbon-carbon bonds are formed between adjacent methacrylate groups. This has not changed since the conception of composite resin materials.

As can be seen from the above description, resin technology has not changed very much over the past 25 years, with most proprietary materials still relying on Bowen's original resin[9]. The addition of various chemical groups to increase the degree of polymerisation of resins has been suggested,[14-16] as this has been shown to improve physical properties of the set material[17]. However, this process still uses conventional urethane dimethacrylate based materials. Improvements in properties of the set material have also been obtained by use of heat treatment[18-20] although, again conventional type materials were used. Cook suggested the use of fluorinated hydrocarbons, to reduce leakage and marginal staining, and the use of unsaturated spiro-orthocarbons, which would expand on setting thus eliminating the problems associated with polymerisation contraction[21]. It has, however, been reported that these materials are unlikely to be available for at least another five years[22]. At present no great advances in resin technology appear imminent and research appears to be based around optimising the properties of the existing materials.

1.3.3 Filler Technology.

The major component of modern composite resin materials is the reinforcing filler. The type, concentration, particle size and distribution of this filler are major factors controlling the properties. Unlike resin technology there have been significant advances in filler formulation over the last three decades.

The original, "traditional"[23] composite resin materials contained filler particles in the range of 1 - 100μm[24]. These particles were purely inorganic in nature and generally splinter shaped, due to the manufacturing process by which they were produced, as this required crushing or grinding. Although Bowen's original composite actually contained a borosilicate glass filler[8], quartz was the predominant filler material used throughout the 1970's[25]. Quartz was popular, during this period, as it was chemically inert and had an appropriate refractive index. However it was radiolucent, had a relatively high coefficient of thermal expansion and was difficult to polish[26]. With these drawbacks it was not surprising that a transition to other fillers occurred. Barium and strontium glasses have proved to be the most popular replacement for quartz as they are radiopaque and, as they are softer, are more easily polished. Nonetheless, the large filler particles still resulted in difficulty with the finishing of these materials; trimmed materials soon becoming stained. In addition, hydrolysis of interfacial bonds and the fast wear of the organic matrix resulted in protrusion and plucking out of filler particles. This manifested itself, clinically, as poor wear in both contact and contact free areas[23]. In fact this type of composite is no longer promoted, most having been replaced by materials with an improved filler concept[27].

Until 1977 all composite resins were filled exclusively with relatively large inorganic molecules[21]. At this time a new type of composite resin material, the "heterogenous microfilled composite resin"[23], was introduced containing extremely small 0.04μm pyrolic silica particles. The size of these particles resulted in a material which could be polished to a high lustre and would keep this polish during function. It is not possible to produce usable composite resin materials by direct incorporation of this type of filler into resin[23]. Either the filler fraction would be too low for adequate physical properties or the viscosity of the material would be too high to produce a workable material. This problem is overcome by incorporating a maximum amount of filler into dilute monomer and polymerising the mixture. This relatively highly filled material is then crushed or agglomerated to produce filler particles in the 5-30μm range[27] and combined with resin to which more silica has been added to produce the final, workable composite resin[25]. Nonetheless, few of the microfilled materials available today attain a 50% filler loading[27]. Although these materials have good polishability and excellent aesthetics, due to the lower filler fraction, they have poorer physical properties than conventional materials[28] especially flexural modulus[27]. Good adhesion between the prepolymerised polymer blocks and the matrix resin has been questioned with pull out of polymer blocks being observed *in vivo*[29] and chip fractures occurring[30]. There is therefore some doubt regarding the efficacy of these materials in stress bearing areas. Furthermore as the filler in these materials is generally silica they do not have the radiopacity desired[31], although a few materials have specific radiopaque fillers (e.g Ytterbium fluoride in Heliomolar, Ivoclar-Vivadent, Schaan, Liechtenstein). Nonetheless, these materials are used extensively in anterior teeth and one recent study suggests they may even be acceptable for posterior use[32].

The third major class of composite resins, the "Hybrid"[23] materials were developed in an attempt to combine the good physical properties and strength of the traditional materials with the good aesthetics and surface finish of the microfils. A hybrid material basically consists of a traditional material in which the organic phase is reinforced with microfiller. This decreases the differences between filler and matrix resulting in less mismatch in wear characteristics between the two. Consequently, matrix is not removed preferentially and the potential for exposure and exfoliation of filler particle is reduced. These materials also contain softer macrofillers than conventional materials, generally Barium and Strontium glasses, which have the added advantage of being radiopaque. The softer nature of the filler, combined with the microfiller incorporated in the matrix result in a more uniformly polishable material. Nevertheless, the surface which can be produced is not as smooth as purely microfilled materials, nor is it permanent, as the wear mechanism common to all materials containing macrofillers still occurs[23]. The initial hybrid materials contained a large range of particle sizes and had a relatively large average particle size (>10μm). The current trend, however, is to use the smallest, most uniform particle sizes[22]. In the most modern materials the "macrofillers" can now be less than 1μm in diameter[27]. The use of softer filler particles and improved milling techniques have facilitated the production of these smaller filler particles which would not have been possible 10 years ago. This smaller particle size is preferable as it offers a compromise between maximum physical properties and optimum aesthetics. Moreover, it has been suggested that composite resins with filler particles of 1μm, or less, are not as prone to wear *in vivo*[31]. This may be due to the fact that, unlike larger filler particles, they do not extend above the resin surface, they are therefore less prone to masticatory stresses and consequently do not transfer masticatory energy to the matrix to the same degree. Less energy transfer to the matrix results in less stress between matrix and filler and ultimately less wear. At the present time small particle hybrid composite resins appear to be the most suitable materials for use in stress bearing restorations[27].

One slight oddity amongst composite resins is the fibre-reinforced material Restolux SP-4 (Lee Pharmaceuticals, South El Monte, CA, USA). Unlike all other proprietary materials it contains glass ceramic fibres embedded in a conventional resin matrix. The glass fibres are up to 300μm long and, due to this large filler size, the material is relatively rough and difficult to polish. It has been suggested that it is has the potential to wear more than many other materials[27] and at present this particular filler technology has nothing to particularly recommend it.

As can be seen, there have been major changes in composite resin filler technology over the last 30 years. The trend has been from relatively low to very high filler loadings, from large to small particle size and from hard to relatively softer fillers.

These changes have probably been the most important in the continuing improvement of composite resin properties, and performance in the mouth.

1.3.4 Resin\Filler Interface.

Stress transfer from the high strength filler to the relatively ductile resin matrix is necessary if a high strength material is to be obtained. For this transfer to be possible it is necessary that a good bond exists between the two phases of the material[33].

There are two possible methods of bonding resin to filler particles; micromechanically or chemically. Micromechanical bonding can be achieved by etching the filler surface with strong acids (in a manner analogous to etching of porcelain or tooth enamel) as described by Bowen and Reed[34,35]. Alternatively, by sintering particles together, a rough and hopefully porous surface can be produced[36]. The resin flows into these porosities and, during polymerisation, a mechanical bond is formed between resin and filler. Several composites have been produced which utilise this method of bonding e.g. Scinterfil (Teledyne Getz., Elk Grove Village IL60007 USA). Nonetheless, the most commonly used method of producing a bond is chemical.

When bonding an organic material to an inorganic substrate a bifunctional molecule with a polar and a non polar head is required. This allows the molecule to bond ionically to the inorganic material while also bonding in a covalent manner to the organic material. Silanes are the most common group of materials used for this purpose and, of this group γ-methacryloxypropyltrimethoxysilane is the most frequently used[25]. As can be seen from the structural formula (Figure 1.3), it contains a methacrylate group which can bond with the resin during polymerisation and an ionic methoxy group. The ionic bonding is thought to occur by hydrolysis of the methoxy group by bound surface water on the reinforcing filler or, alternatively, with silanol or aluminol groups of the filler itself[33].

Although the silane forms a good bond between the filler and the resin initially and may help protect the filler from water contamination and hydrolysis[37], in the longer term, moisture can weaken the filler resin bond and facilitate its failure. Hence, although the use of silanes has considerably improved the potential for bonding between resin and filler, it may be that this bond is not permanently stable within the oral environment.

With microfilled composite resins the bonding mechanism between filler and matrix is somewhat different, as both materials are essentially organic in nature, the filler consisting of pre-polymerised particles (as described earlier). The difficulty here is that of the potential for a chemical reaction between the resin of the matrix and that of the filler. Due to the highly cured nature of the filler, there are very few remaining methacrylate groups where bonding can occur. Furthermore, due to the large size of the matrix resin molecules micromechanical entanglement of matrix into filler is difficult. This potentially poor bond strength is suggested as a reason for the poor physical properties of such microfilled materials[38]. Nonetheless, Söderholm suggests that there are advantages to this type of bonding between matrix and filler[25]. The bond density can be improved by decreasing the cross-link density of the organic filler and, the bond strength of resin to resin and resin to matrix should be the same. This second point should help reduce internal stresses within the material around the filler particles.

1.3.5 Curing Procedure.

The conversion of oligimers and monomers to a polymer matrix is initiated by chemical or photochemical means to form free radicals[33].

In the original composite resin materials the formation of these free radicals was accomplished by chemical means. Two pastes were mixed together, one containing a tertiary organic amine (N,N-bis(2-hydroxyethyl)-p-toluidine and the other containing an oxidising agent (benzoyl peroxide). Reaction of these two substances results in production of polymerisation initiating benzoyl radicals[39]. Although this activating system could lead to good conversion of the methacrylate groups within the material, approaching 80%[13] in certain materials, there were, nonetheless, various problems with these chemical cure materials. Due to the mixing required to activate the material there is a potential for incorporation of voids into the material. This porosity within the material, which has been suggested to be around 3% for a typical two paste material[24], results in an increase in surface roughness and will result in poorer physical properties which are likely to influence clinical performance[40]. Furthermore, excess of the amine within the set material resulted in a tendency for the material to yellow with time. Finally, the working time of these materials, although in the region of 2 - 4 minutes (depending on the specific one), made the provision of restorations in large or inaccessible cavities difficult. These failings were addressed by the provision of light activated composite resins.

The initial light cured materials were activated by ultra violet light at a wavelength of around 360nm. In these materials a photoinitiator, benzoin methyl ether, undergoes photofragmentation when subjected to U.V. light. This results in free radical formation which, in turn, initiates resin polymerisation[33]. Polymerisation initiation by this method was a major advance with regard to handling of these materials. No mixing was required, reducing the possibility of incorporating voids in the material, and viscosity did not increase significantly prior to light exposure giving very long, although not quite unlimited, working time. However, there were doubts expressed regarding the depth of cure of these materials[41]. In addition, concern, within the dental profession, regarding the potentially harmful effects of near U.V light, as listed by Birdsell *et al.*[42] and Mills *et al.*[43], resulted in the relatively rapid introduction of a visible light curing system for composite resins at the end of the 1970's.

Visible light curing materials normally employ photosensitised free radical initiators, often camphorquinone (an α-1.2 diketone) and an amine reducing agent such as dimethylaminoethyl methacrylate or dimethy p-toluidine[44]. This photosensitiser\reducing agent absorbs energy within the visible light spectrum with peak absorption occurring at around 470nm, in the blue light region (See Chapter 2, Figure 2.5). When a photon of energy is absorbed by camphorquinone the carbonyl group is promoted to an excited singlet state (Q_s). This singlet may then undergo intersystem crossing to the triplet state (Q_t) this acting as an electron acceptor and hence forming an *exiplex* with the amine by transfer of charge from the nitrogen to the carbonyl group. This in turn leads to the formation of two free radical ions which can initiate polymerisation of the resin (Figure 1.4)[45]. Of these two ions it is thought that the amine radical is the most efficient in promoting polymerisation.

These visible light cured materials possess all the advantages that the U.V. cured materials have over the chemical cured composites but also have a greater depth of cure and no U.V. hazard to dentist or patient. Initially some concern was expressed regarding a possible hazard from the intense blue light produced by light curing units[46,47] as the wavelengths implicated in producing retinal damage are in the band 400-500nm and correspond to those which initiate the polymerisation reaction. However, testing would suggest that indirect exposure of the type that would be experienced by dentists or dental hygienists would be required to occur for between 40 and 100 minutes per day for any appreciable hazard to occur. Hence, only a very few dental personnel require to use protective eyewear when these light sources are used and the potential hazard is relatively very small[48].

Problems do still remain with these materials. Although the depth of cure is better in visible than in U.V. curing systems[49]; due to higher light intensity of visible curing units[50] and the greater transparency of composite resin to visible light[51], light cured materials do not produce the infinite depth of cure claimed by manufacturers when these composites were first introduced. With chemically cured materials the degree of polymerisation was roughly uniform throughout the material, hence hardness on the surface was the same as elsewhere. This is not the case in light cured materials where a short exposure to light can result in a fully cured, hard surface with uncured material only a millimetre below this[49,51]. It is therefore more difficult for the dentist to be sure of adequate cure throughout a restoration. This can be a particular problem in interproximal box areas where access for light curing is difficult[53]. The relative merits of light versus chemical curing systems are summarised in Table 1.1.

Method of Cure	**CHEMICAL**	**LIGHT**
Advantages	No light source required Cost of material Uniform cure throughout material Polymerisation contraction uniform throughout material	Long working time Command set Colour stability Good colour matching Low porosity Instant finishing
Disadvantages	Colour stability Colour matching Porosity Short working time Long setting time Must wait for finishing	Need light Premature setting with operating light Cost of material Shrinkage not uniform - towards light source Curing greatest nearest light Potential for uncured material where light does not reach

Table 1.1: Advantages and disadvantages of light and chemically cured materials.

Nonetheless the vast majority of composite resin materials produced today are of the visible light curing type with no U.V cured and very few chemically cured materials surviving.

All these changes have resulted in a strong, colour stable, radiopaque material with improved handling characteristics, good aesthetics and acceptable wear. It is not surprising, therefore, that these materials have achieved universal acceptance amongst the dental profession for the restoration of cavities in anterior teeth. Nonetheless, despite all these improvements, there are still problems associated with these materials particularly with regard to their potential as a posterior restorative.

1.4 CONTINUED PROBLEMS WITH COMPOSITE RESINS.

1.4.1 Polymerisation Shrinkage.

Davidson states that regarding material properties of composite resins "almost only one property counts and that is polymerisation shrinkage"[12]. While this may be an overstatement of the facts, this contraction is nonetheless responsible for many of the failings of this group of materials.

All composite resin materials contract on setting. Increased filler fraction decreases this shrinkage; however a shrinkage of between 1% and 6%[54-56] is still to be expected with proprietary materials. When a filling contracts on setting the tooth\filling interface will be placed under considerable stress (15-20MPa[57]). This will result in excessive stresses being placed on the tooth cusps due to the wall-to-wall contraction which may, in turn, produce cuspal flexure, enamel damage and even cusp fracture[58-60]. Alternatively, the bond between the tooth and resin will fail resulting in a marginal gap. This is particularly the case when cavities have a margin in dentine as, although with the use of the acid etch technique[61] a strong bond to enamel can be expected (over 20MPa[62]), the bond to dentine is much more fragile. These spaces result in marginal microleakage which can cause sensitivity of the tooth to thermal stimuli or may allow bacterial ingress[63], secondary caries and even pulpal pathology[64]. Such marginal microleakage is discussed more fully in Chapter 10.

Polymerisation shrinkage is a more serious problem in posterior teeth as the cavities are generally larger. Increased bulk of material results in greater polymerisation contraction with more stress being placed on the bond between tooth and restoration. Clinical techniques based on layering and incremental curing have been developed to help reduce this problem. These techniques have been shown to decrease the stresses placed on teeth during curing and also decrease microleakage[59].

1.4.2 Wear Resistance.

Wear of anterior composite resin restorations is generally not perceived as a problem as the forces of mastication are relatively much less than in the posterior region[65]. Initial success of traditional composites, anteriorly, led to their use in posterior cavities where, it was supposed, their good mechanical properties would cope with the extra occlusal forces. This was not, however, the case, as wear was a major cause of restoration failure in these early materials[1].

The significant advancements in composite formulation, described earlier, have considerably decreased the wear potential to the point that it has been stated that wear is no longer the most significant shortcoming of the material[66]. Modern composite resins may, indeed, be as much as ten times more wear resistant as earlier materials[67] resulting in materials which have wear characteristics approaching that of amalgam, in conservative cavities[22]. Nonetheless, wear rate can be affected considerably by a number of factors; notably the wear in the contact area can be 3½ times that in the contact free area[68,69]. Moreover, the position of the tooth in the arch and the cavity size are significant in the final wear the material undergoes[67].

With these factors in mind the ADA guidelines for acceptance of composites for use in class I and II cavities now require that at least 50% of the test surface must have an occlusal contact. As conclusive data are not, as yet, available from these cavities[22], the real potential of the modern materials to resist wear is still open to speculation.

1.4.3 Handling Characteristics.

Simplicity and ease of use are essential factors in the success of any filling material. Undoubtedly it is the "user friendly" nature which has been, in a large part, responsible for the success of amalgam. Composite resin materials have proved to be considerably less forgiving in the clinical environment.

The use of the acid etch technique, as mentioned, can produce a good marginal seal with composite resin materials[61]. However, moisture control is of the utmost importance with contamination of the tooth surface with blood or saliva resulting in bond failure. This is in contrast to amalgam which can survive slight contamination of the cavity with no profound effect on the quality of the end product[1].

Adequate tooth morphology is relatively difficult to achieve with composite resin in posterior regions. Occlusally several techniques have been suggested. The use of a mould and syringe technique was described by Jacobsen[70]. However, this was only suitable for use with autopolymerising resins[71]. A system of transparent occlusal matrices was marketed by one manufacturer (Kulzer Gmbh, Friedrichsdorf, Germany). However, they only fit a minority of teeth. In general, occlusal contouring and finishing is carried out following curing by the use of burs and finishing points and discs. Inevitably, the trimming of a white filling in a white tooth results in some tooth destruction or, alternatively, some flash will be left. Even if tooth damage is avoided this is a time consuming and difficult procedure when compared with amalgam carving and contouring[1]. At present there is no ideal method available[72].

The formation of contact points is also difficult as matrix bands and normal wedging do not produce an adequate contour. Unlike amalgam, there is little opportunity to produce contact by condensation of the filling material. Clinical techniques, such as prewedging of teeth and use of ultrathin matrix bands, have been developed[73]. However, these can prove difficult and time consuming.

The final problem regarding tooth morphology is adequate contouring in the interproximal region. Even careful matrix and wedge placement does not always result in a perfect margin free from gingival excess. Roulet[1] stated that even thin layers of composite extending over cervical margins are capable of inducing gingivitis as small composite overhangs are ideal niches for bacteria. If an overhang does occur it is very difficult to remove, involving the use of rotary instruments with the possibility of subsequent tooth damage. This is in contrast to amalgam where carving of amalgam restoratives can remove overhangs without any tooth damage.

The other difficulty with handling of composite resin materials is related to the curing procedure used with most modern materials. Although, as stated earlier, light cured materials are potentially easier to work with, they are not without problems. In anterior teeth, where cavities are relatively small, light curing is adequate to produce cure throughout the whole filling. However, in larger cavities adequate cure to the base of the cavity cannot be assured. This can be a particular problem in interproximal regions in class II cavities where the cavity depth is often greater than 3mm. Inadequate cure at the base of a restoration will inevitably lead to washout of material, leakage and the potential for secondary caries. To overcome this problem, layering techniques have been developed to ensure adequate cure to the base of large cavities. In these no layer of more than 1.5mm is polymerised at any one time[73]. Rees and Jacobsen [53], in fact, suggest an initial increment of 0.5mm in the base of class II cavities. In addition to the potentially inadequate depth of cure, the cure pattern of light polymerising materials can be a problem. Light cured materials cure from the area nearest the light source down. This can result in an upward movement of the material, due to the polymerisation shrinkage, as described by Lutz *et al.*[74]. This is in contrast to chemically cured materials where cure occurs uniformly throughout the materials with no specific force vectors occurring. This shrinkage towards the light source is of particular importance in class II cavities where curing from the occlusal surface can pull the restoration away from the gingival margin. The layering technique has, again, been suggested to help this problem, as have curing techniques beginning interproximally and the use of light reflecting wedges[75]. The efficacy of these light reflecting wedges is, however, open to some question[76].

These manipulative factors all contribute to a relatively complicated and time consuming filling technique, in order to produce an adequate restoration.

1.4.4 Degree of Cure.

Light curing of composite resins does not result in optimum conversion of the methacrylate groups within the material. At best 74% conversion can be expected[77], depending on type of resin and filler formulation[77,78]. This is a source of potential weakness in the material, as it has been shown that the physical properties are related to the cure[17]. Thus, with the present materials, it is not possible to optimise the physical properties with light curing, intraorally, alone. This, theoretically, leads to a material which will not perform to its full potential clinically.

Considering these difficulties and, with any major improvements in material technology apparently some way off, a method of optimising the potential of the existing materials was required. One suggested procedure, with considerable potential, was the composite resin inlay technique.

1.5 RATIONAL FOR THE USE OF COMPOSITE RESIN INLAYS.

1.5.1 Definition.

A composite resin inlay has been defined as "a restoration which is cemented into a dental cavity as a solid mass that has been fabricated from composite resin with a form established either by an indirect or a direct procedure."[79]

The potential advantages of this type of system over any direct composite placement technique are as follows.

1.5.2 Polymerisation Shrinkage.

Virtually all the polymerisation shrinkage of the restoration will take place prior to cementation. The only shrinkage which will take place intraorally is that of the composite resin luting cement. This will place considerably less stress on the tooth\resin interface than that which occurs during polymerisation of a direct filling. Less stress at this interface decreases the potential for cuspal flexure and fracture or alternatively marginal gap formation and its potential sequelae.

1.5.3 Handling Characteristics.

When using an indirect technique for inlay fabrication the occlusal morphology, contact points and marginal fit can all be checked and optimised in exactly the same manner as any other indirect inlay system. This allows for good tooth contact both occlusally and interproximally without the need for prolonged and difficult trimming intra-orally. The use of direct inlay techniques does not have the same benefits with regard to production of correct morphology as the indirect techniques although some precementation trimming occlusally and marginally is possible. Cure of all parts of the restoration is assured as the inlay can be removed from the model\tooth and light applied to inaccessible areas. Adequate cure of all margins, during cementation, is also guaranteed, even if an area such as an interproximal box is inaccessible to light, as inlays are cemented with dual curing composite resin cements. Thus uncured resin is prevented.

1.5.4 Degree of Cure.

The maximum degree of cure by conventional light curing of all parts of the inlay is ensured with any of the proprietary systems, as the inlay can be cured from any angle. This, as has already been stated, is an advantage in itself over direct composites. However, there is potential for increasing the degree of cure of the material. The inlay can be subjected to a secondary curing procedure by heat\light, heat\pressure or heat alone, as suggested by Wendt amongst others[19,20]. This increased cure improves the physical properties resulting in a stronger material. This stronger material should, theoretically, perform better in the clinical situation.

With these potential advantages over direct composite resin restorations many clinicians have been attracted to these systems[67]. In fact, Christianson has suggested that they are the most popular of the tooth coloured inlay systems available at present[80].

A considerable research effort has been directed towards these systems, over the past decade, resulting in the production of a significant amount of literature.

1.6 LITERATURE REVIEW OF COMPOSITE RESIN INLAYS.

1.6.1 Fabrication Methods.

Several of the initial papers concerning composite resin inlays were technique papers describing the fabrication and fitting of various of the inlay types.

The indirect microfilled system Ivoclar Inlay\Onlay, Concept in the USA, (Ivoclar-Vivadent, Schaan, Liechtenstein) was first reported in 1983[81]. A subsequent paper[82] described this technique in more detail. Both of these papers dealt with tooth preparation, inlay fabrication and subsequent cementation and finishing procedures. The latter paper however also gave detailed guidelines on preparation of the tooth. This is something which is lacking in all but the most cursory manner from all the other technique papers sited.

Direct hybrid systems have been described in three papers[83-86]. In these the fabrication of an inlay to fit a single cavity was described. All three studies used the same submicron hybrid composite system (Brilliant, Coltene AG, Altstatten, Switzerland) which is light cured initially intraorally , removed from the mouth, and subjected to a secondary cure using heat and light at 120°C for 7 minutes prior to cementation. This system can also be used in an indirect manner. However, to date, no papers have been published in mainstream English language journals describing this or any other hybrid systems use as an indirect restoration.

The use of an indirect chairside technique was described by Donly[87]. The inlay system used in this study was the Visio-Gem system marketed by Espe (Premier Sales Corporation, Norristown, PA, USA) which utilised a microfilled composite resin. Initial curing of the inlays, was carried out by light in the same manner as the hybrid systems, but subsequent post curing was carried out using light in a vacuum. The interesting fact regarding this system and the closely related EOS system, marketed in Europe by Ivoclar

(Ivoclar-Vivadent, Schaan, Liechtenstein), was the use of a soft die, fabricated in heavy bodied impression material. This die was produced from the impression taken of the inlay preparation intraorally. This soft die technique allows the dentist to fabricate the inlay extraorally without the need for a second appointment to fit the inlay. However, the fit of inlays produced on soft models was questioned by Cassin and Pearson[88], who found that the fit of this type of inlay "was in no way comparable with a conventional inlay". Martin[89] also suggested that distortion of the inlay may occur, due to twisting of the flexible model resulting in poor fit, and Reid *et al.*[90] found that considerable trimming of the fitting surface of this type of inlay was required to make it fit the cavity. Donly[87] went on to suggest that an appointment of 1 hr is necessary to complete a single inlay using this technique. The theme, of fabrication time, is investigated further by Plasmas *et al.*[91] using a similar system (ICS from 3M) which involves fabrication of a chairside model; although in this system the model is made of epoxy resin and hot melt stone. They suggest a time of around 2 hours for a single inlay, although this decreases when more than one inlay is made at once. The relative merits of chairside and laboratory produced methods of fabricating inlays were summarised by Rees and Jacobsen[92]. One further paper which dealt with the fabrication of inlays is that of Wilson and Norman[93]. In this various packing techniques, for inlay production, are discussed. Bulk packing under pressure was found to produce inlays with fewer voids than either a layering technique or bulk packing with no pressure. Furthermore, it was suggested that this lack of porosity following a pressure curing technique may result in improvements in the physical properties of the finished inlay.

1.6.2 Physical Properties Following Post Curing.

The other area on which several of the earliest papers, relevant to the inlay technique, concentrated was the possible improvements in physical properties of the material following secondary curing procedures. This interest has continued up to the present day. Probably the first paper which suggested the use of heat as a secondary curing procedure for composite resins was that by Bausch *et al.*[18]. Using an infra red heating source four proprietary composite resins were heated to temperatures of, up to, 60°C, and the Knoop hardness and diametral tensile strength recorded. Considerable improvements in these properties were obtained and this led to the suggestion that composites should be heated during clinical use to optimise their physical properties. (Exactly how one can heat a composite to 60°C intraorally was not described!). Wendt[19,20] produced similar work, where he tested physical properties of three proprietary light cured composite materials, both before and after subjecting them to heat treatment. In these experiments the heat was supplied by an oven rather than an infra red source and heating to temperatures between 60°C and 200°C was evaluated. The properties of compressive strength, diametral tensile strength, hardness and colour stability were evaluated (marginal dimensional stability and wear were also considered but are dealt with later). Wendt found no improvement in compressive strength, but, like Bausch *et al.*[18], he found significant improvements in hardness and diametral tensile strength. The different materials studied reacted to the temperature of the secondary curing procedure in different manners. For instance diametral tensile strength was only increased in one material at 60°C whereas all three were increased at 125°C. This led Wendt[19] to suggest that the optimum temperature for post curing was around 125°C. Cook and Johannson's[94] work confirmed these findings as, in their study, flexural strength and diametral tensile strength increased with increasing post cure temperatures up to the maximum of 100°C tested. Asmussen and Peutzfeldt[95] also investigated the effect of varying the post curing temperature on the physical properties of four composite resins. However, in this study the resin formulation was varied while the filler type and proportion remained constant. They agreed that post curing could influence physical properties, in this case diametral tensile strength, flexural strength, and modulus of elasticity. However, they went on to suggest that a temperature of around 150°C was optimum for post curing although the improvements in physical properties which could be expected were only modest; in the region of 9%. Moreover the resin formulation employed in the composite was an important factor in the post cure effect which could be obtained; UEDMA and HEMA containing composites performing better than Bis-GMA and TEGMA containing materials.

Covey *et al.*[96] found an average increase of 16.6% in diametral tensile strength when seven composite resins were post cured at 120°C for 7 minutes, however improvement was not noted with every group and differences were noted due to particle type and particle size, ie. the changes were material specific.

Gregory *et al.*[97] compared the physical properties of five composite resin materials two direct and two for use in the inlay technique. Tests were carried out for flexural strength, water solubility, water sorption, radiopacity and depth of cure according to ISO\DIS 4049,1988, for resin based filling materials. They concluded that the laboratory use of pressure and heat or light and vacuum significantly increased flexural strength and Knoop hardness while also decreasing solubility. All materials were acceptable with regard to depth of cure, two were unacceptable radiographically, but this was related to material composition rather than any post curing effect. Flexural repair strength and spectroscopy, also investigated in this paper, are discussed elsewhere in this introduction.

Peutzfeldt and Asmussen[98] investigated the physical properties of three proprietary composite inlay systems both with and without post curing. With one hybrid system no improvements were found following post curing. However, with another hybrid system which was post cured in a similar manner, with heat and light, significant improvements in flexural properties were observed. Interestingly, when the light curing component of the curing process was removed from the post curing of the latter material, the observed improvement in properties did not occur. This prompted the authors to suggest that the effect of additional curing may stem solely from prolonged exposure to light. Although the flexural properties of the microfilled heat\pressure cured materials were inferior to the other materials it had a significantly higher diametral tensile strength. This, it was suggested, was due to the pressure applied during the curing regime.

McCabe and Kägi[99] investigated the hardness, compressive strength and fatigue limit (wear factor, also measured, will be dealt with later) of two closely related hybrid composite resins, with and without post cure at 120°C for 7 minutes. Although some improvement in hardness occurred for one material no other differences were noted and it was considered unlikely that post curing would have any clinical effect.

Most recently Ferracane and Codon[100] have investigated the effect of duration of post cure (10 minutes versus 3 hours); post curing both immediately following light cure and up to 7 days following initial light cure and the effect of changing filler composition on material properties. Results showed that post curing for ten minutes was as efficient as for three hours, and that delaying post curing had little effect on the ultimate physical properties obtained. Moreover, this study was the first to investigate the effect of post curing on fracture toughness (K_{IC}) with specific reference to composite inlays (although it is over a decade since the influence of temperature on K_{IC} was first suggested[101]). Fracture toughness was found to be increased following post curing irrespective of the regime adopted. This confirmed the work of Cook and Johannson[94]. Unfortunately, these results are in conflict with further studies by Ferracane and Marker[102] who suggest a decrease in K_{IC} is to be expected and Uctasli *et al*[103] who suggest that no change in fracture toughness is to be expected following post curing.

1.6.3 Methacrylate Conversion in Post Cured Composites.

Ferracane[100] has also investigated the effect of post curing on the degree of methacrylate conversion of the composites tested, using Fourier Transform Infrared Spectroscopy, and found increased conversion, irrespective of secondary curing regime with all materials. He also noted that with the proprietary brands, even following post curing, 30% or more of the methacrylate groups remained unreacted.

Although it has been shown previously that heat treatment following light curing can increase conversion[78,94] and improve physical properties, this was the first paper to deal with the inlay technique *per se.* In fact although the degree of conversion of composite resin inlays, is often referred to throughout the literature, it has only actually been investigated infrequently. Gregory *et al.*[97] confirmed the presence of unreacted methacrylate groups on all the composites he tested. However, no attempt was made to quantify the percentage conversion. Watts[45] has also shown an increase in degree of conversion following post curing of the resin used in one proprietary material. However, although the curing of the resin alone may be indicative of what happens in the composite resin material, as a whole, this is by no means certain.

One paper which helped to clarify the effect which secondary curing has on the internal structure of composite resins is that of Dioysopoulos and Watts [104], which investigated the dynamic mechanical properties of the material both before and after post curing. This confirmed considerable changes within the material, with less dynamic mobility of the resin phase of the material. These changes, it was suggested, were related to increased cure of the materials and possibly relaxation of polymer segments into lower energy wells. However, irrespective of the exact mechanism, it is suggested that the post cured material is chemically more stable and less likely to exhibit dimensional instability due to internal flow.

1.6.4 Microleakage.

Much emphasis has been placed on the assessment of microleakage of composite resin inlays, as this was perceived as one of the major advantages of the inlay technique.

Initial studies concentrated on the difference in leakage between directly placed composites and composite inlays of various types. Robinson *et al.*[105] found significantly less leakage when an indirect inlay technique was used in MOD cavities. This study utilised a lightly filled composite material with high polymerisation contraction and coefficient of thermal expansion in an attempt to accentuate the differences between the two types of restoration. Nonetheless, this composite could not be considered representative of the materials used in proprietary inlay systems. Hasegawa *et al.*[106] produced a similar result when inlays of both hybrid and microfilled materials were compared with direct restorations of the same material. He also found no difference between the leakage of the two inlay types despite their different formulations. Douglas *et al.*[107] compared microleakage in class V cavities with direct placement and inlays fabricated from a microfilled material. He also concluded that the indirect placement of composite restorations offered considerable improvements in microleakage performance. Furthermore, he found that the use of different dentine bonding agents could have a significant effect on the leakage score observed. Wendt[108] concluded that heat treated inlays showed significantly less leakage than direct composites whichever bonding regime was adopted. However, the use of unfilled resin as luting agent cannot be regarded as representative of normal clinical procedure. Sheth *et al.*[109] evaluated 2 hybrid and 1 microfilled inlay system and found significant improvements, with the use of the inlay technique, over conventional placement of a hybrid material. Moreover significant differences were found between inlay systems with one system, a hybrid, performing worse than the others. However the system that performed worst was "bonded" to dentine using unfilled resin alone, rather than the phosphorylated Bis-GMA dentine bonding agent used with the other systems. This fact may well have been responsible for the relatively high leakage scores seen in dentine with this system. The bonding protocol in the study by Yanikoglu and Scherer[110] resulted in the direct composite being bonded with a dentine bonding agent whereas the inlay was, again, bonded with unfilled resin alone. This is the only study to date which detected no advantage, with regard to microleakage, of the use of an inlay technique. However due to the lack of uniformity of bonding procedure these results must be treated with some caution. In fact a subsequent study by Llena Puy *et al.*[111] found virtually no microleakage associated with enamel or dentine margins when a dentine bonding agent was used with this same inlay system. Cassin and Pearson[88] confirmed earlier observations that leakage is decreased, but not eliminated, when an inlay technique is used. This study investigated a chairside indirect method which uses a microfilled composite resin.

A few later studies have gone into rather more detail comparing various types of inlay and different bonding procedures. Shortall *et al.*[112], in one of the more exhaustive papers on the subject, investigated leakage in direct composites, indirect composite inlays and porcelain inlays in class II cavities. Furthermore, bonding with glass ionomer or a dentine bonding agent were evaluated. They concluded that both porcelain and composite resin inlays provided a superior marginal seal to direct composite resins, although there was no difference between the two inlay types, and, that the use of a dentine bonding agent produced a more reliable bond than a glass ionomer liner. In a subsequent study Shortall[113] attempted to differentiate between the efficacy of two dentine bonding agents and a control, unfilled resin only group. However, no differences were detected except when cleaning of the tooth was not carried out prior to bonding with one system. Interestingly, there was no difference between the inlays bonded with dentine bonding agents and the control in this study. It was suggested that this may have been due to the small size of the inlay cavities (as class V cavities were used) and the good initial fit of the inlays. Karagalclioglu *et al.*[114] compared the leakage of porcelain and a microfilled composite inlay placed in class II cavities on various types of glass ionomer liner. This study concluded that porcelain inlays provide a better seal than composite inlays. However, if one considers the leakage which occurs at the cervical dentine margin, where most leakage has been shown to occur, in all adhesive type fillings there is no difference between the systems. Differences were confined to different glass ionomer types. Occlusally, where the inlays are bonded directly to the tooth enamel with luting composite only there are significant differences between the systems. This is at odds with Shortall and Bayliss[112] and a recent paper by Reid *et al.*[90], who found no difference in leakage between porcelain and two types of composite resin inlays. The composite inlay systems used were very different between Karagalclioglu *et al.*[114] and Shortall and Bayliss'[112] studies, one a hybrid heat\light cure and one a microfilled heat\pressure cure. It is suggested that, due to the higher coefficient of thermal expansion of the microfilled material, increase stressing of the tooth\resin inlay bond may occur during thermocycling. This could be responsible for the increased leakage recorded with this composite system. With regard to difference in glass ionomer lining cements it was concluded that a light\chemical curing material provides a better seal than a light cure only or a chemical cure only material. As this type of glass ionomer has been shown to have a significantly higher bond strength to dentine than the other types[115] this is, perhaps, not a surprising conclusion. Furthermore, it was suggested that etching may have a detrimental effect on the leakage pattern observed, a finding of previous papers[116].

From these papers it would appear that there is general agreement that the inlay technique results in decreased leakage around restorations. Moreover the bonding procedure employed and the used of bonding agent or glass ionomer cement may have a significant effect on the leakage pattern irrespective of the inlay type being studied.

Three closely related papers measured the marginal gap formation rather than microleakage as a measure of marginal integrity. Reeves *et al.*[117] found no difference in marginal gap formation between a direct hybrid composite resin and an indirect chairside system using the same composite resin material. However, as all margins were placed in enamel, this is not really surprising as direct composites placed by the incremental technique, used in this study, are known to exhibit little leakage in enamel and it would hardly be expected that the inlay technique would perform worse. Peutzfeldt and Asmussen[118] compared the gap formation around three proprietary inlay systems cemented in human teeth with and without the use of a dentine bonding agent. They found no gap formation when margins were in enamel. However, gaps occurred when a margin was on dentine. Use of the dentine bonding agent, nonetheless, prevented gap formation in all three systems tested a finding confirmed by Llena Puy *et al.*[111], who investigated marginal adaptation with SEM as part of their microleakage experiment.

1.6.5 Accuracy of Fit.

The accuracy of fit of composite resin inlays has been investigated by several authors. Ariyaratinum *et al.*[119] recorded the variation in resin cement lute thickness around the inlay's fitting surface as a measure of fit and found a lute of between 10μm and 90μm with a heat\light cured indirect hybrid system. It was also suggested that the fit tended to be thinnest and most uniform in the region of the cavo surface margins. Rees and Jacobsen[92] found a much greater lute thickness with a heat\pressure cured microfilled system (over 200μm in some areas), than in a hybrid, light cured system. Moreover, they failed to find any correlation between measuring position and lute thickness. Wendt[19] compared the marginal discrepancy of MOD inlays on a steel die both before and after post curing at temperatures of between 60°C and 200°C. He found no correlation between post curing and accuracy of marginal fit; any changes appearing to be materials related. Peutzfeldt and Asmussen[118] compared the accuracy of fit of MOD inlays produced from three proprietary composite resin inlay systems; two hybrid heat\light cured and one microfilled heat\pressure cured, in a similar manner. In this study axial discrepancy (ie. marginal opening in the approximal area) was assessed using inlays, fabricated both directly, on a milled brass die and indirectly on a dental stone model. They found indirect inlays fabricated from the microfilled material fitted significantly less well than those of either hybrid type. There was no difference between the fit of the indirect hybrid systems. However, direct hybrid inlays tended to fit more accurately than those made by the indirect method in both systems. Moreover, with one direct hybrid and the microfilled systems the inlays fitted less well at 24 hours than at ten minutes. It was suggested that this was due to the fact that curing contraction continues for more than ten minutes following the initiation of polymerisation, although what contraction may still occur following a post curing regime was not discussed. Finally, it was noted that the marginal discrepancy between tooth and inlay may increase when a luting agent is present. This appeared to be the case as cement lutes of "from less than 150μm to 400μm" were cited in this paper although no indication was given with which inlay systems these extremes occurred. Reid *et al.*[90] also considered marginal discrepancy, with regard to cavity convergence angle, and found one chairside indirect composite inlay to be a significantly poorer fit even when cavity convergence was increased to 15°. A recent paper by Qualtrough *et al.*[120] compared two methods of assessing the fit of composite resin inlays; an impression wash technique and a sectioning technique. No conclusion was reached as to which method was best, it being suggested that they provided complimentary information. However, the small number of inlays made for each group, three (apart from control groups), precludes any meaningful investigations between inlay types tested. Furthermore, the use of impression material as the luting agent in the sectioning technique cannot be considered similar to the composite resin luting agents which are used clinically and finally, the concept that aluminium and perspex can be used to simulate enamel and dentine, respectively, is, at least, controversial.

1.6.6 Polymerisation Shrinkage of the Composite Resin Lute.

The effect of composite resin cement lute thickness on the tooth\inlay interface and possible cuspal flexure was investigated by Rees and Jacobsen[121] with the use of finite element analysis. They confirmed earlier work that inlay technique places less stress on the tooth resulting in less cuspal flexure[59]. More interestingly, however, they suggested that the thicker the composite resin lute the greater the stress on the tooth\restoration bond, during polymerisation, and the greater its likelihood of failure. This increase in polymerisation stress in thicker composite resin cement layers was confirmed by the work of Kunzelman *et al.*[122].

1.6.7 Bond Strength to Inlay Surface.

It has been suggested that the increased degree of cure of composite resin inlays results in insufficient remaining methacrylate bonds on the fitting surface to allow a good bond, to the composite resin luting cement, to occur during cementation[20,89,92]. (This suggestion even prompted one manufacturer to produce a bond enhancing agent to increase the potential for bonding of inlay to cement.) However, to date, this assertion seems more theoretical than supported by fact. Swift *et al.*[123] found little difficulty bonding to a sandblasted surface of a hybrid heat\light cured inlay, with bond strengths similar to those of composite to etched enamel being reported. In fact, surface treatments designed to enhance bond strengths in this study (eg HF acid treatment) had a detrimental effect. Ariyaratnam *et al.*[124] also found good bond strength to another heat\light cured hybrid system. If the analogous system of bonding to fractured composites is considered, these bond strengths are not unexpected, as it has been shown that the repair bond strength of composite to itself is similar to that of composite to etched tooth enamel[125]. However, Ruyter[126] suggests that there is a difference between inlay systems depending on whether the inlay is heat\light or heat\pressure cured; heat\pressure cured systems having a considerably poorer bond strength to their luting resin due to their increased degree of methacrylate conversion. This is partly borne out by the work of Tan and McComb[127], who showed lower bond strength and increased adhesive failure between a laboratory processed composite resin veneer, of similar formulation to a heat\pressure cured composite inlay material, and a hybrid system. The situation is confirmed by Gregory *et al.*[97], who investigated the repair flexural strength of four composites following post curing. In this study flexural repair strength of a heat\pressure cured inlay material was much less than those cured by heat and light. Moreover, the plane of fracture was adhesive alone in this group as opposed to adhesive\cohesive in the other groups tested. It was suggested that this difference was due to the increased degree of conversion of methacrylate groups, as any possible difference in surface texture was minimised by standard finishing procedures, prior to bonding. Finally, this study attempted to relate flexural strength and repair strength, suggesting that flexural strength may be used as a measurement of degree of cure and that materials with a high flexural strength will therefore have a poor repair strength. While this may be true for a specific material, it is obviously incorrect when comparing different materials. Materials formulations differ significantly and these will have an enormous effect on flexural strengths of different composite resins irrespective of their degree of cure.

A related study by Peutzfeldt and Asmussen[128] considered the retention of inlays fabricated in either one of two hybrid heat\light or one microfilled heat\pressure cured in standardised class I cavities in human teeth. They concluded that the use of a dentine bonding agent had more effect on the retention of a composite inlay than the choice of inlay type. No assessment of plane of fracture was made in this study, although as the dentine bonding agent improved the bond strength it would appear that the tooth/inlay interface was the weak bond, at least initially.

1.6.8 *In Vitro* Wear Testing.

The wear resistance of composite resin inlay materials has received remarkably little attention, considering it was perceived as one of the major advantages of the post cure technique in Wendt's original paper[19]. Wendt used a Tabler Abraser with a "moderately abrasive carborundum wheel" as the element against which the composite resins were cycled to produce wear. Heating to 125°C produced the maximum improvement in wear resistance with all materials tested. In a subsequent study, using the same technique[129] he found that heating for longer than seven and a half minutes did not result in any further improvement in wear resistance. This 125°C, seven and a half minute cycle which Wendt describes as optimum for improving wear resistance, has been adopted as the ideal post curing regime by at least one manufacturer (Coltene). De Gee *et al.*[130] investigated the effect of post curing on the abrasion resistance of 11 composite resins of varying compositions. An apparatus producing 3 body wear was used in this study with a millet seed slurry being used as the "foodstuff". Wear, in this study, was measured as the material loss/200K cycles measured at intervals over a 70 day storage period. Initially, post cured materials exhibited improved wear. However, over time this advantage diminished until by 70 days there was, in general, no difference in wear rate between conventional light cured and heat\light post cured materials. De Gee *et al*[130]suggest that this is evidence that the heat induced improvements in properties were not due to an increase in degree of methacrylate conversion but, rather, were due to an annealing process in which the polymerisation stresses were released during the heating. This process, it was hypothesised, occurred at mouth temperature over a longer period of time and, consequently, the light cure only materials annealed with time and showed an improvement in wear resistance.

Two body *in vitro* wear of four composite resin inlay materials against enamel, gold and porcelain was described by Bugoyne *et al.*[131]. Using a previously described[132] pin and plate apparatus they considered the wear of both the materials acting as pin (tooth) and plate (filling). Significant differences were found between the composite materials when wear against all three substrates was recorded, with one light cured hybrid composite consistently performing worst in all cases. Interestingly, the hybrid materials all created wear tracks in the opposing materials. However, the microfilled material did not and, generally, suffered least wear itself. It was suggested that this finding was due to the smaller filler particle size and, therefore, reduced friction of the microfil relative to the others.

Most recently McCabe and Kägi[99] compared the wear of two very similar sub-micron hybrid materials for use in the inlay technique using a modified vibrational technique first employed by McCabe and Smith[133]. Weight loss of samples was recorded as a measure of wear produced. Some slight improvement in wear factor was noted in one of the materials. However, McCabe and Kägi[99] suggested that this improvement was unlikely to influence clinical performance.

1.6.9 Colour Stability.

Colour stability of composite resins during post curing has been investigated by two authors. Ruggenberg *et al.*[134] suggested that microfilled resins were more prone to colour changes than hybrid materials subjected to the same post cure regime; this probably being due to the higher resin content of these materials. The tendency was for the materials to become more green and blue following post curing. Nonetheless, from their results, curing within the normal temperature range, up to 125°C, resulted in minimal changes in colour for any of the materials tested. The only cautionary note to be taken from this study was that careful control of oven temperature was necessary, as, if post curing was carried out using an oven not specifically designed for that purpose, "overcooking" at temperatures of around 300°C would cause colour changes. Wendt[19] found no change at all, in colour, when heating one composite to 200°C, and only minimal changes in two others. He suggested that these changes were barely perceptible and would not be clinically relevant.

1.6.10 Cavity Design.

Unlike cavities for direct composites, composite resin inlay cavities have received little attention to date; most recommendations on cavity design coming either directly from the manufacturers[82], or as an adaptation of gold inlay design removing sharp line angles and bevels[89,92,135]. As yet only two papers have dealt specifically with this subject. Wassell *et al.*[136] considered the problems associated with withdrawing composite inlays from cavities with small convergence angles, around 6°, such as would be used with gold inlays. Forces required to withdraw inlays from cavities with varying convergence angles were recorded, leading to the suggestion that a larger angle of around 18° is appropriate, and should be used to facilitate insertion and withdrawal of inlays from the cavity. This finding is supported by Reid *et al.*[90] who considered the effect of cavity wall divergence on ease of seating inlays. They suggested a divergence angle of at least 10° is required to allow seating of either a porcelain or microfilled composite resin inlay system. They also noted that one chairside composite inlay system would not seat even with a 15° divergence angle.

1.6.11 Cuspal Strength Following Restoration.

The potential of composite resin inlays to reinforce teeth which have been weakened by cavity preparation was discussed by Lopes *et al*[137]. Large MOD cavities were cut in human premolars significantly reducing the cuspal rigidity. Cementation of a composite resin inlay restored the tooth to 97% of its original stiffness. This, the authors suggest may mean that large intracoronal restorations would be feasible with this type of restoration. However, they caution that factors such as occlusal wear, material properties and chairside time must also be taken into consideration before a large restoration of this type is undertaken. Furthermore, Wendt[108] suggested that this increase in stiffness, as measured by fracture resistance, was no better than in directly placed composites, although the cementation of inlays with unfilled resin may have some bearing on the results obtained.

1.6.12 Clinical Trials.

There have been disappointingly few clinical reports on composite inlays. Two have been published in English using the SR Isosit inlay\onlay technique (a microfilled, heat\pressure cured system). The longest running trial is that of Bishop[138], a former employee of Ivoclar, who reported the results of 92 inlays placed for periods of between 7 months and 4 years, with only one restoration failing to meet the United States Public Health Service's[139] criteria for a functioning restorative system. Bessing and Lunqvist's[140] preliminary report records 45 inlays over a 1 year period. The guidelines for clinical quality and professional performance of the California Dental Association were used for assessment of the inlays in this study. Of the 30 inlays available for review only one, which had fractured, was unsatisfactory. Some discolouration had occurred at margins which they attributed to inferior conversion of the inlay cement and its subsequent staining rather than leakage. Nonetheless, they stated the short term nature of their study and the limitations of observations over such a limited period.

No clinical reports of proprietary hybrid inlay systems are available in the literature, to date, although Burke *et al.* [135] suggest that trials are going on in several centres throughout the world. However, Wendt and Leinfelder[141,142] have published one and three year results of a hybrid composite resin which has been used as an inlay both with and without secondary heat curing. The composite used, Occlusin (ICI Dental), was, in fact, the subject of an international study as a direct restorative for use in posterior teeth, where it demonstrated satisfactory performance over a five year period[143], and has never been suggested as being particularly suitable for the inlay technique. Nonetheless, this is an important study as it has the potential to investigate differences caused by the heat curing process alone rather than any other complicating factors, such as material type or placement technique. Evaluation, in this study, was carried out using the USPHS system and the Moffa-Luggassey indirect scale as described by Leinfelder *et al.*[144]. In addition, post-operative sensitivity was assessed. At times of up to 3 years there was no difference between light and post cured inlays with regard to colour, interfacial staining, secondary caries and wear. However, marginal integrity and surface texture were superior in the post cured group. Furthermore, there was more initial post-operative sensitivity with light cured inlays than in the post cured group, although, by three months, there was no difference between groups.

It is suggested that the better surface texture of the heat treated material may be due to the increased hardness of the surface following post curing. The fact that this is not translated to improved wear resistance is, it is hypothesised, due to adverse effects on the silane\filler interface caused by the post cure heating. This causes failure of the filler resin bond resulting in an increased wear pattern, reminiscent of traditional composites, negating any improvements which may have occurred due to harder matrix. No explanation is offered for the improvement in marginal integrity, however it seems likely that the higher flexural strength of the post cured material would make the inlay more resistant to the type of marginal fracture described by Rees and Jacobsen[145].

1.6.13 Composite Resin Luting Cements.

The importance of composite resin luting cements for use with tooth coloured inlay materials has not been overlooked. The composition, properties and bonding potential of such cements have been described by Rees and Jacobsen[145] who go on to consider the problems which may be associated with intraoral degradation and loss of lute, by the use of finite element analysis. Attenuation of light and subsequent decreased cure of composite resin cement through porcelain restorations is a well accepted fact[146], and the same has been shown to be true with composite inlays. Breeding *et al.*[147] showed that light cure only materials could not be cured, predictably, through 2mm thickness of composite resin inlay and that a combined light\chemical curing material was preferable. Hasegawa *et al.*[148], when examining three dual cured cements, found that light curing was necessary for them to reach their ultimate hardness and that this did not occur under inlays, although all cements cured to a degree. This hardness, which he related to degree of cure, was material specific with different proportions of light and chemical curing components occurring in different composite cements. A later study, using Fourier Transform Infrared Spectroscopy, by Rueggeberg *et al.*[149], confirmed these findings; with none of the materials tested recording as high a methacrylate conversion when cured by chemical means alone.

The effect of curing mode, chemical, light, or both, was investigated by Zuelliger-Singer *et al.*[150]. MOD inlays were bonded to human dentine with a dual cured resin cement. This cement was either allowed to chemically cure, was light cured or was chemically cured and then light cured. No differences were noted between the groups, suggesting that no advantage is gained, with regard to decreased wall-to-wall contraction, by the slower cure and greater potential for flow of the chemically cured material when compared with the light cured.

Despite the significant amount of work which has already been done with regard to composite resin inlays, there remains scope for a considerable degree of further study.

1.7 AREAS OF FURTHER STUDY

There are now many proprietary composite resin inlay kits on the market, encompassing all types of composite resins and various different secondary curing regimes. It is essential that some comparative testing of these various inlay types is carried out in order to discover any defects in systems and, hopefully, allow improvements. Comparative testing will allow the good aspects of various techniques to be adapted for use with other materials, and, perhaps, result in a better inlay technique. In the short term, at least, this type of study will allow the profession to make a better judgement of which technique they should be using.

The potential of post curing of composite resins using various regimes has been investigated by numerous groups, as already mentioned[18-20,94-100]. In summary it would appear that post curing improves some properties of some materials at certain temperatures. All of the studies cited have concentrated on the physical properties of materials a short time following post curing, no attempt being made to age the materials. If, as DeGee *et al.*[130] suggest, improvements in properties are due to an annealing effect, which will occur at mouth temperature, over time, as well as during the post curing process, it may be expected that light cure and post cured materials should exhibit similar physical properties following storage at physiological temperatures after several months. Alternatively, if the generally held view that post curing increases methacrylate conversion and, hence, increased crosslinking increases physical properties[104] is correct, then differences detected at one day would be expected to continue throughout a testing period. Finally, if Wendt and Leinfelder's[141] hypothesis regarding the possible damage to the filler resin bond following post curing is correct, it may be expected that storage in water will result in increased degradation of post cured materials, leading to decreases in physical properties of these materials.

Therefore testing of physical properties, while a valid criteria by which to assess differences between light only and post cured composites, should be continued for periods of at least two months (De Gee's wear test period) and, preferably, for considerably longer to assess possible longer term effects of post curing.

The results of studies into the property of fracture toughness have been conflicting and clarification of the effect of post curing is required, especially with respect to specific inlay composites and the effect of storage on post cured materials. As this property has been suggested as one laboratory experiment which can be of a direct clinical value in prediction of wear resistance[151] it should be singled out for special attention.

The degree of cure of post cured composite resins, although often discussed has not been specifically investigated with proprietary composite resin inlay brands or curing techniques. This is an obvious gap in the literature at present, and, as it would settle many of the arguments regarding the actual effects of the post curing technique, is an essential study.

It is now well established that composite resin inlays leak less than direct composite resin materials[105-109,112]. However, the potential effect of dentine bonding agents when used with the inlay technique has been given little attention. If, as is expected, the polymerisation contraction force around a composite inlay is much less than around a direct composite, dentine bonding agents should prove more effective with this, indirect technique than with direct placement. Furthermore, each manufacturer of a composite inlay system, naturally, suggests the use of their own dentine bonding agent. With so many different products available there is, therefore, the possibility of compatibility problems between different brands. Finally, different types of composite resin inlay fit with different degrees of accuracy. If, as suggested[121,122], lute thickness has a bearing on stress at the tooth restoration interface, then the thicker lute, of certain systems, may have a detrimental effect on the marginal seal.

The potential for inefficient bonding between inlay and composite resin cement is of considerable interest, as this is perceived as a weakness in the inlay technique. This question would be partly answered by adequate assessment of degree of cure of composites following post cure. However, measurement of bond strength and assessment of plane of fracture are essential if this controversy is to be settled. Furthermore, it seems too simplistic to relate bond strength to degree of conversion alone. Inevitably micromechanical bonding will take place to some extent, and an investigation of this is mandatory for a fuller understanding of the bonding process.

Wear, although one of the main factors for the adoption of the post curing technique, has received remarkably little attention. Two body testing should be carried out as contact areas have been shown to be where most material destruction takes place[68,69]. Only one such study has been carried out[131] and it did not compare the same material pre and post cure to assess whether or not differences in material were due to formulation or curing procedure. Although there are difficulties relating *in vitro* to *in vivo* wear, further laboratory wear studies would perhaps be prudent due to the time required for clinical data to be available. Furthermore, it has been suggested that the luting agents used with tooth coloured inlays are subject to more rapid wear than the inlays themselves and, that this wear is related to the gap between inlay and tooth[152,153]. This area requires further investigation.

Only the hardness of composite resin luting agents has been assessed, and this, only as an indication of the degree of cure. Measurement of the physical properties of these dual curing materials is necessary; as is an indication of the curing process and setting times both with and without light. Furthermore, as it is likely that very little light reaches a proportion of the cement lute some measure of the physical properties of these cements when cured only by their chemically curing component would be interesting.

Ultimately clinical testing is necessary to provide the answers the profession requires. It is obvious that more, varied and well controlled studies are required from multiple centres before the true value of this technique can be known.

As can be seen a considerable amount of further research is required. Through selection of three different proprietary (one submicron hybrid heat\light cure, one conventional hybrid heat\light cure and one microfilled heat\pressure cure) inlay systems, it was hoped to address many of the problems mentioned above.

1.8 AIMS OF STUDY

The aims of this study were as follows:

1. Assessment of the basic physical properties of three composite resins for use in the inlay technique both with, and where appropriate, without a post curing regime. Assessment was to continue for at least two months to assess possible annealing and degradative changes in the materials.
2. Assessment of fracture toughness of the three materials, with and without post-curing, over a 2 month period.
3. Assessment of physical properties of the composite resin cements supplied with each system both with and without light curing.
4. Assessment of the degree of methacrylate conversion of each material with and without post-curing.
5. Measurement of bond strength of inlay to composite resin inlay and investigation of the bonding mechanism with particular reference to the potential for micromechanical bonding.
6. Measurement of accuracy of fit of inlays of different types.
7. Assessment of leakage of inlays in enamel and dentine. Further investigations into the influence of various dentine bonding agents and accuracy of fit on the leakage of the respective systems.

Although some pilot clinical studies have been carried out, unfortunately a full clinical assessment of composite resin inlays was beyond the scope of this study.

Chapter 2: Systems Investigated.

2.1 INTRODUCTION.

Three proprietary systems for production of composite resin inlays were investigated in this study. Each system consisted of a composite resin, a dual curing composite resin cement and an oven or pressure pot in which the "supercuring" was carried out. Two of the materials were light curing hybrid composite resins which were subjected to a secondary cure in a heat\light oven. The third material was a microfil which did not photopolymerise but was cured in a heat\pressure pot.

2.2 COMPOSITE RESINS.

The filler type, filler loading, resin formulations and proposed curing regimes are given in Table 2.1.

MATERIALS	**HYBRID A**	**HYBRID B**	**MICROFIL**
TRADE NAME	BRILLIANT DENTINE	ESTILUX POSTERIOR	ISOSIT INLAY\ONLAY
MANUFACTURER	Coltene A.G., Altstatten, Switzerland.	Kulzer GmbH., Bereich Dental, Wehrheim, Germany.	Vivadent Ets., Schaan, Liechtenstein.
FILLER CONTENT (by weight)	78%	83%	73%-76% (53%-56% inorganic)
FILLER TYPE	90% Barium-aluminium-borosilicate, mean particle size 0.5μm 10% Fumed silica, mean particle size 0.04μm	Bariumaluminium-borosilicate- glass Lithium-aluminium-silicate-glassceramic Siliciumdioxide	Prepolymerised organic resin/highly disperse silylated silica acid, 0.01-0.04μm
RESIN TYPE	Bis-GMA 37.5% Bis-EMA 37.5% TEGMA 25%	Bis-GMA 66% TEGMA 34%	UEDMA 56% 1.4-BDMA 20% 1.10-DDMA 14%
CURING METHOD	Light cure or Heat\light cure	Light cure or Heat\light cure	Heat and pressure only

Table 2.1: Composition of composite resin materials used throughout this study.

The light cured hybrid materials have a very similar resin formulation. Both materials contain a large proportion of Bowen's original Bis-GMA resin (Isopropylidenibis[2-hydroxy-3-(4-phenoxy)]propylmethacrylate. As mentioned in the introductory chapter this is a very viscous monomer. Therefore, in order to produce a composite with reasonable handling characteristics it is necessary to have lower molecular weight monomers present in order to thin the resulting resin and allow it to flow adequately. Hybrid B is a very traditional material, and contains 3,6 Dioxaoctamethylene dimethacrylate, more commonly known as Triethylene glycol dimethacrylate (TEGMA) as it's viscosity regulator; the same as Bowen's original composite[9]. Hybrid A has a more modern resin formulation and in addition to TEGMA it contains Bis-EMA resin Isopropylidenibis[2-(4-phenoxy)]propylmethacrylate. This resin is very similar to Bis-GMA however, it is slightly less viscous and consequently less TEGMA is required to decrease the viscosity in hybrid A. This is an advantage as the short chain TEGMA is responsible for increasing the polymerisation contraction of the resin.

However, as can be seen from Table 2.1 the main difference between the two hybrid materials is the size of the filler particles. Although both materials contain Barium-Aluminium-Borosilicate glass filler in hybrid A the mean particle size is only 0.5μm whereas hybrid B is an older type hybrid with a mixture of large and small particles up to 10μm in size. Both materials are marketed as suitable for the restoration of class I and II cavities as well as for use in the inlay technique.

The microfilled material is of entirely different resin formulation to the two hybrids. It is mainly a Urethane Dimethacrylate resin (UEDMA), 1,6-bis(methacryloyloxy-2-ethoxycarbonylamino-2,4,4,-trimethylhexane. Like Bis-GMA, UEDMA is a highly viscous monomer, requiring lower molecular weight comonomers to regulate the viscosity of the final resin. In this case two short chain dimethacrylate resins are the viscosity regulators, 1,4-

butanediol dimethacrylate (BDMA) and 1,10-decanediol dimethacrylate (DDMA). The filler is also quite different in this material being composed of fumed silica of <0.04μm diameter and relatively large prepolymerised particles (>20μm) containing the same sized filler particles.

This material cannot be used as a direct restorative due to its lack of photoinitiator; it is however closely allied to other Ivoclar composites such as Heliomolar.

This difference in formulation results in a difference in the method of fabrication of the inlay types. The two hybrid materials are built up in layers, light curing 1mm increments until the inlay is complete. At this stage it is subjected to a secondary cure in its respective heat\light oven. Due to the lack of photoinitiator this method of fabrication is impossible for microfilled inlays. Consequently they are bulk packed in a single increment and cured entirely in the heat\pressure oven.

The filler type, loading and resin formulation of the composite resin cements supplied are given in Table 2.2.

MATERIAL	**HYBRID CEMENT A**	**HYBRID CEMENT B**	**MICROFIL CEMENT**
TRADE NAME	DUO CEMENT	ADHESIVE CEMENT	DUAL CEMENT
MANUFACTURER	Coltene A.G., Altstatten, Switzerland.	Kulzer GmbH., Bereich Dental, Wehrheim, Germany.	Vivadent Ets., Schaan, Liechtenstein.
FILLER CONTENT (by weight)	67.4%	75.4%	60.1%
FILLER TYPE	90% Barium-aluminium-borosilicate, mean particle size 0.5μm 10% Fumed silica, mean particle size 0.04μm	Bariumaluminium-borosilicate- glass Lithium-aluminium-silicate-glassceramic Siliciumdioxide	Prepolymerised organic resin/highly disperse silylated silica acid, 0.01-0.04μm, Ytterbium trifluoride
MAXIMUM FILLER SIZE	5-10μm	38-59μm	58-87μm
RESIN TYPE (Mixed cement)	Bis-GMA 37.5% Bis-EMA 37.5% TEGMA 25%	Bis-GMA 55% TEGMA 45%	UEDMA 80% DDMA 20%

Table 2.2: Composition of composite resin luting cements used throughout this study.

The formulation of these cements closely mimics that of their respective composites, although the filler loading is less. They are all dual curing, that is they will cure either with light or alternatively chemically in the absence of irradiation.

The final component of each system is an unfilled resin. All three unfilled resins (Duo Bond for hybrid A, Adhesive Bond for hybrid B and Heliobond for the microfil) are Bis-GMA, TEGMA mixtures. All three unfilled resins can be cured with light. However, the resin for use with hybrid A is dual curing and will also cure chemically in the absence of light.

2.3 CURING OVENS.

2.3.1 Light Cure Only.

For initial light cure of the hybrid materials a Translux curing light model (Kulzer GmbH., Bereich Dental, Wehrheim, Germany) was used throughout the experiments. The light intensity which has been shown to decrease with use[154] was checked prior to beginning each experiment with a lamp checker. When the light intensity fell below 80% of its starting value the bulb was replaced.

2.3.2 Heat and Light Cure.

It has been shown that varying the temperature at which post curing occurs can have an effect on the final mechanical properties achieved for a specific material[18-20,95]. The effect of light, and light intensity have not been investigated, specifically, although Peutzfeldt and Asmussen[98] have suggested that it may be of more importance than the heating which occurs. The effect of duration of secondary curing cycle has also been investigated by Wendt[129] who suggested an optimum curing time of 7.5 minutes.

It would appear, therefore, that the temperature, light intensity and duration of cycle may be of importance in determining the final properties of the material being subjected to the regime. Consequently, an introductory study was undertaken to measure these parameters for the curing ovens used in later experiments.

2.3.3 Heat and Pressure Cure.

The curing apparatus for use with the microfilled material is completely different from the other curing units. It is basically a pressure cooker which heats the material under water to a high temperature under a constant pressure.

2.4 AIM.

To determine, where appropriate, the light intensity, temperature, pressure and duration of cycle of the curing ovens recommended by the manufacturers for curing each material.

2.5 MATERIALS AND METHOD.

One heat\light oven is supplied for use with hybrid A (D.I.500). This oven consists of a curing chamber with a single halogen lamp which supplies both the light and heat to the composite. The oven also has a fan which circulates the air across the chamber. Two different ovens are supplied for use with hybrid B. The Unilux A.C. is an oven designed to be used with any of Kulzer's light or U.V. curing materials. It has a large curing chamber with six U.V.A. and six visible blue light tubes arranged circumferentially around the chamber. These tubes supply both light and heat to the oven. A fan directed from above circulates the air within the chamber to create a constant temperature throughout. The Translux Lightbox is a small mirrored attachment which connects to any conventional Translux light curing unit. The heat and light are transmitted from the halogen bulb of the curing unit down a perspex rod into the chamber. There is no method of circulating the air in this system.

The pressure pot, for use with the microfilled material, is heated to a set temperature by an electric element at the base of the apparatus. Pressure is supplied from an external compressed air supply.

Measurement of temperature within the curing ovens was carried out using a thermocouple (R.S. Components) attached to a multimeter (Fluke 8000A John Fluke MFG Co Inc., P.O. Box 43210, Mountlake Terrace, Washington 98043, USA). The thermocouple was covered in a 1.5mm cube of composite material appropriate to the oven being tested, to mimic the temperature change which would occur within an inlay. Temperature was recorded at 10 second intervals up to 2 minutes and then at 30 second intervals until the end of the cycle. Measurements were made at the base, middle (2.5cm from base) and the top of each curing chamber to assess any variation in temperature with specimen placement. A series of five measurements were carried out in each position within each box. Mean and standard deviation values were calculated for each position in each oven at each time.

Measurement of visible light intensity within the box was carried out using an SED033 #2764 photodiode with a W#5713 diffuser and a TBLU filter (International Light Inc., Dexter Industrial Green, Newburyport Mass. USA.). This resulted in a system sensitive to light of wavelength between 420nm and 480nm. The absorption wavelength of camphorquinone, the photoinitiator in these materials is approximately 470nm. Consequently the light measured by this detection system is that which is important in producing the curing reaction.

The light intensity was only measured in the middle of the ovens as the size of the detector prevented placement elsewhere.

As previously mentioned the Unilux A.C. also produces light in the ultra violet spectrum. This is not necessary for the curing of the composite resin in this study, but is important in curing of other Kulzer materials. The U.V. radiation was measured using an IL 730A Actinic Radiometer (International Light Inc., Dexter Industrial Green, Newburyport Mass. USA.). Only the constant value of this light intensity was recorded.

Finally, the pressure within the Ivomat was measured using the units own pressure monitor.

2.6 RESULTS.

2.6.1 Heat\light Systems

The functions of the curing ovens are summarised in Table 2.3. Light and heat intensities are also shown in Figures 2.1-2.4

The D.I. 500 oven heated to a temperature of 110°C - 120°C over a period of two minutes. It then remained at this temperature for a further four minutes before cooling gradually, with the heat\light source switched off, until the cycle ended at eight and a half minutes. The higher up the box the sample was, the more quickly its temperature rose. Moreover, at the top of the curing chamber the final temperature was higher at 140°C. The light intensity within this oven was 1.34W/cm^2 initially, and dropped to 1.15W/cm^2 over the six and a half minutes during which the oven was illuminated.

The Unilux A.C. heated to a temperature of around 55°C over a period of 8 minutes and subsequently remained at around this temperature until 20-22 minutes had elapsed. There was no temperature variation in different areas within the curing chamber. The light intensity rose to around 1.9W/cm^2 over three minutes and remained constant for the rest of the cycle. The U.V.A. radiation was constant at around 6.65W/cm^2 throughout the cycle.

The Translux Lightbox heated to around 47°C at the base, 53°C in the middle and 82°C at the top of the curing chamber over the six minute curing cycle. The temperature was still rising, slowly, at the end of the cycle. The light intensity was 3.95W/cm^2 initially, dropping to 3.32W/cm^2 after six minutes.

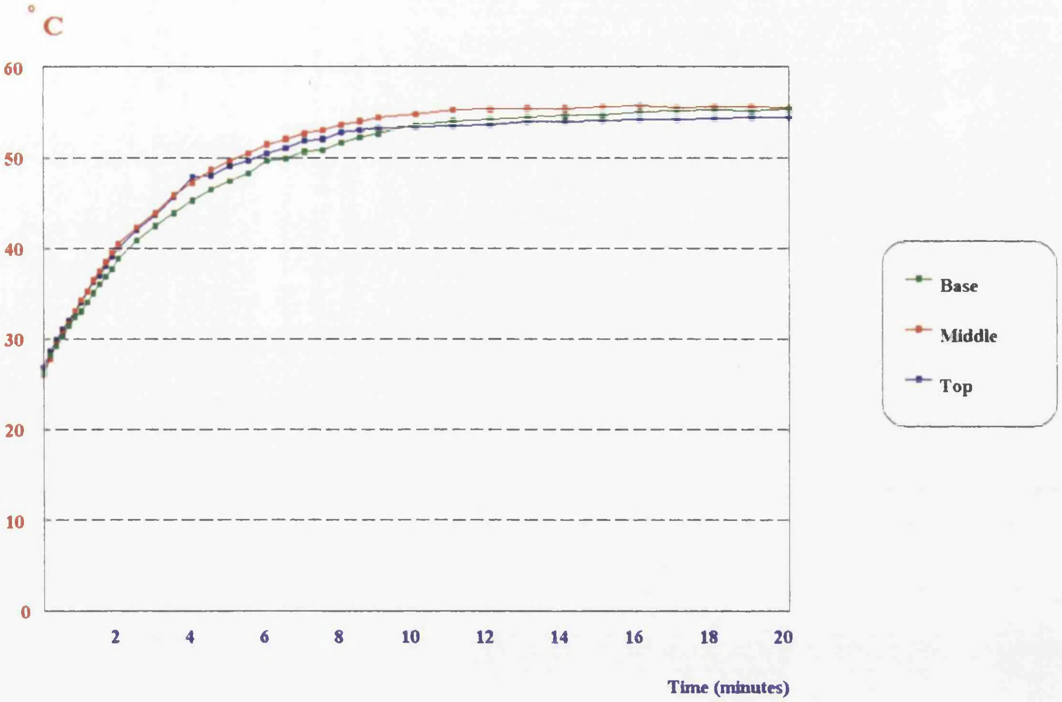


Figure 2.3: Temperature variation within Unilux A.C. oven during curing cycle.

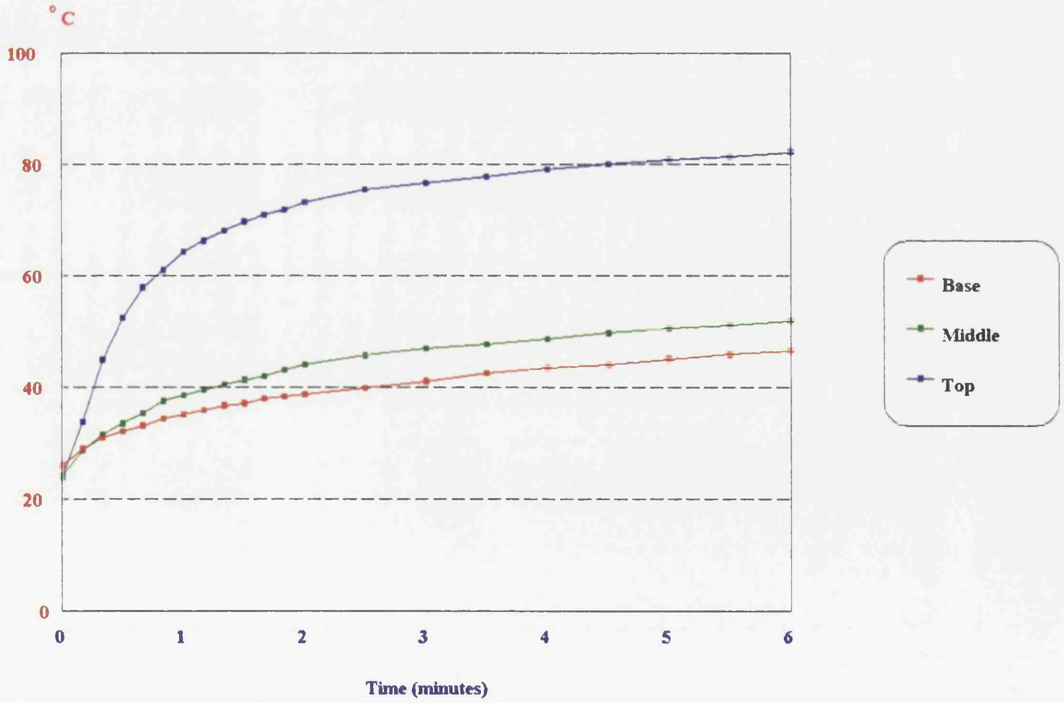


Figure 2.4: Temperature variation within Translux lightbox during curing cycle.

OVEN	**D.I. 500**	**Unilux A.C.**	**Translux Lightbox**	**Ivomat**
COMPOSITE	**Hybrid A** BRILLIANT DENTINE	**Hybrid B** ESTILUX POSTERIOR	**Hybrid B** ESTILUX POSTERIOR	**Microfil** ISOSIT INLAY/ONLAY
MANUFACTURER	Coltene A.G., Altstatten, Switzerland.	Kulzer GmbH., Bereich Dental, Wehrheim, Germany.	Kulzer GmbH., Bereich Dental, Wehrheim, Germany.	Vivadent Ets., Schaan, Liechtenstein.
MAXIMUM TEMPERATURE (In centre of box)	120°C	55°C	53°C	120°C
MAX\MIN LIGHT INTENSITY (mW/m^2)	1.34 - 1.15	1.9	3.95 -3.32	N/A
PRESSURE	Atmospheric	Atmospheric	Atmospheric	6 bar
CYCLE DURATION	8.5 minutes	20-22 minutes	6 minutes	10 minutes at 120°C

Table 2.3: Temperature, Light Intensity, Duration and Pressure in the curing chambers of the ovens for use in the composite resin inlay technique.

2.6.2 Heat\pressure System.

The heat and pressure variation within the Ivomat is shown in Table 2.3 and Figure 2.5.

The temperature rose gradually to 120°C over 5-10 minutes, depending on the initial water temperature. It then remained at this temperature for 10 minutes at which point the water was evacuated and the temperature dropped rapidly. The pressure increased to 6 bar immediately on switching the unit on. It remained at this level until the heating cycle was complete.

2.7 DISCUSSION.

2.7.1 Heat\light Cured Systems.

The temperature achieved in the D.I.500 oven was considerably greater than that which occurs in either of the other ovens. In fact this oven produced a similar regime to that described by Wendt[129] as desirable when post curing composites. Nonetheless the 120°C, suggested by Wendt, only occurred for around five minutes of the eight and a half minute cycle; rather than for the seven and a half minutes he described and the seven minutes claimed by the manufacturer.

The two Kulzer ovens produced temperatures of around 50°C when samples ~~are~~ were * placed in the centre of the curing chamber as suggested by the manufacturer. Post curing effects have been shown to occur at temperatures of this order[18], however, higher temperatures have produced more marked improvements in physical properties[18,19,95]. The significance of these differences, in temperature, with respect to properties of the post cured materials will be discussed in the following chapter.

The curing cycle of the Unilux A.C. was much longer than those of the other two ovens (approximately 3x the D.I.500 and 4x the Lightbox). This increased duration of curing cycle may be important in producing the post cure at a lower temperature. However, the light intensity in the lightbox was 1.5X as high as in the Unilux and this may have a balancing effect allowing for a shorter curing time. It must be mentioned that the light intensity of the lightbox was measured directly from the light source. This may give a higher reading than will actually occur within the curing chamber, on surfaces not directly facing the light source. Although the internal surface of the box is mirrored 100% reflection of light cannot be expected. It is therefore likely that the light intensity falling on areas following reflection will be somewhat decreased. This is in contrast to the Unilux where the arrangement of the bulbs results in all light falling directly from the light source onto the sample being cured.

The ultra violet light produced in the Unilux A.C. is for curing of other Kulzer products, not composite resin materials. However, camphorquinone not only absorbs visible blue light but absorbs light energy over a relatively wide part of the visible and UV spectrum (Figure 2.6). Hence, although the main absorption and subsequent activation will be caused by the visible light the UV radiation will also play a part in activating the photoinitiator and, possibly, increasing the degree of cure[13].

Temperature within the Unilux A.C. and the D.I. 500 ovens is controlled by a thermostat and fan. This leads to a more controlled and constant temperature throughout the curing chamber. The D.I. 500 fan is situated at the side of the chamber about half way up the wall and it is here that its maximum effect is felt. Further up, towards the top of the box, the cooling effect is less and the temperature gets higher. This higher temperature is also due to the closer proximity of the heat source. There is no variation in temperature with altered position within the Unilux A.C. This is because the fan is directed from above and the light\heat sources are distributed around the walls rather than in a single position.

There is considerable variation in the temperature within the Translux Lightbox, depending on how high up within the chamber the thermocouple is placed. In general the higher up within the box the thermocouple is placed the hotter it gets. There are two reasons for this. Firstly the heat\light source is at the top of the box and causes direct heating and, secondly, as no fan is present convection causes the hot air to rise also increasing temperature at the top.

The relative importance of heat, light and duration of cycle are examined more fully in the next chapter.

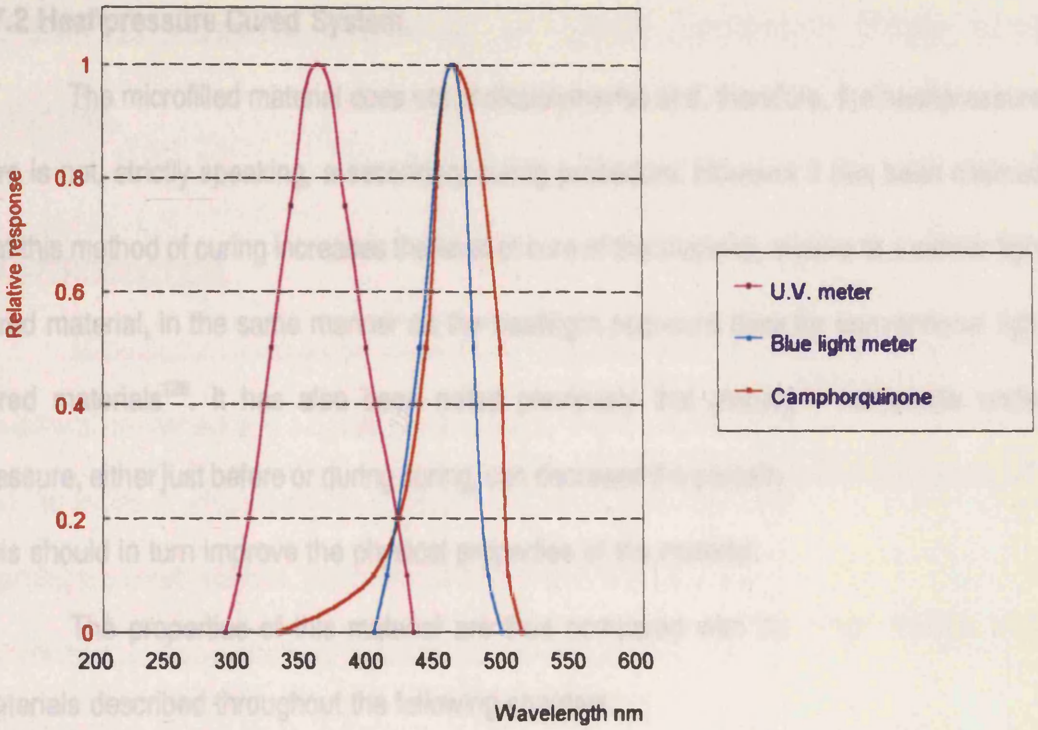


Figure 2.6: Relative absorbance of camphorquinone, blue light and ultraviolet light filters.

2.7.2 Heat\pressure Cured System.

The microfilled material does not photopolymerise and, therefore, the heat\pressure cure is not, strictly speaking, a secondary curing procedure. However it has been claimed that this method of curing increases the level of cure of this material, relative to a similar light cured material, in the same manner as the heat\light post-cure does for conventional light cured materials[126]. It has also been noted previously that placing a composite under pressure, either just before or during curing, can decrease the porosity within its substance[33]. This should in turn improve the physical properties of the material.

The properties of this material are thus compared with the other potential inlay materials described throughout the following chapters.

Chapter 3: Physical Properties of Three Composite Resin Inlay Materials.

3.1 INTRODUCTION.

Many factors must be considered when deciding which properties are important to the clinical performance of a dental filling material. The environment in which it will be placed and the forces to which it will be subjected are crucial in assessing which tests and testing regimes are most relevant. Innumerable laboratory tests have been developed to simulate the conditions *in vivo* and help elucidate the clinical performance without the need for long and expensive clinical trials. (Examples of these types of test are seen in Chapters 8 and 10 where bonding and leakage are investigated). Nonetheless, simple mechanical properties, such as compressive strength, remain a valid criteria by which to evaluate dental filling materials, as such properties are fundamental to an understanding of their clinical performance[33]. In addition simple testing methods remove many of the variables present in more complicated "lifelike" regimes and allow more exact comparison between very similar materials, as is the case in this study.

As with any filling material, composite resins have to be strong enough to withstand biting forces without fracture, while, at the same time, being rigid enough to maintain their shape under load. The properties important in producing such a material are best quantified by determination of compressive, diametral tensile and flexural strengths. From flexural strength the flexural modulus of the material can also be determined. This gives a good indication of the rigidity of the material and hence its ability to accept load without permanent deformation. The hardness of a material is also of great importance as it gives a measure of its ability to resist scratching; a material with a high hardness number is likely to retain a polish better than one with a low number. Hardness has also been used to give an indication of the wear resistance of a material[155] where the wear process is thought to include scratching, as in abrasive wear. There is , however, some doubt whether this correlation is valid[156].

The mechanical properties of dental composite resins are dictated by several factors.

1) Filler volume.

2) Filler type and size.

3) Resin formulation of the matrix.

4) Bond between resin and filler.

5) Degree of cure of the material.

Of these the filler content is the most important factor[22]. However, although materials with very high filler contents can be produced, there is a point at which the handling characteristics of the material preclude further filler additions. The materials become too viscous to use easily and exhibit poor surface wetting. Moreover very highly filled composites, by necessity, contain a mixture of large and small filler particles. These large particles, although contributing significantly to the strength of the material, can lead to a surface which is very difficult to polish. The current trend is, therefore, to use the smallest, most uniform particles for hybrid materials. Often particles are less than 1 μm, resulting in a "sub micron" hybrid material (hybrid A in this study). Particles of this size allow good filler loadings (approaching 80% by weight) while still maintaining a degree of polishability not possible in earlier hybrid materials.

The maximum level of filler loading, while maintaining a workable material, has probably been reached. The bond between filler and resin has been optimised by the use of silane coupling agents, such that a strong and permanent bond can be obtained. Resin chemistry, although slowly modified over the years, is still largely based around Bis-GMA as described by Bowen over thirty years ago[9]. New improved resins containing different structures are being developed however their introduction seems some way off[14-16]. Interest has, therefore, increased in methods of improving the physical properties, and, hence, hopefully, the clinical performance of existing materials.

It has been known for many years that the physical properties of methacrylate based resins, such as composite resin filling materials, are related to the degree of cure of the material[17,157]. It is also known that conventional curing of composite resins, by either light or chemical means, does not result in maximal methacrylate conversion[158,159]. Furthermore, because of polymerisation contraction, which occurs during curing, composites develop internal stresses which may weaken the material[160].

In an attempt to improve the physical properties, Bausche *et al*[18] suggested heating composites to 60°C. At this temperature they suggested that extra crosslinking of methacrylate groups occurred and, hence, physical properties were improved. Wendt produced similar work in which improvements were noted in some physical properties in certain materials subjected to secondary curing[19,20]. It has also been suggested that heat treatment following initial light curing may allow relaxation of the internal stresses in a manner similar to annealing of metals[128]. This concept was discussed further suggesting that this annealing process could take place at mouth temperature, such that, after a period of time in the mouth, the conventionally cured material would perform in the same manner as that subjected to post curing.

3.2 AIM.

The potential of secondary or post curing as a method of improving the physical properties of composite resins is examined in this section. To this end the mechanical properties of compressive strength, diametral tensile strength, flexural strength, flexural modulus and Knoop hardness were measured in three composite resin materials recommended for this technique. The aim was to establish if the secondary curing procedure did result in an improvement in the physical properties of the materials tested. Furthermore, it was hoped to ascertain if a similar improvement in physical properties could be expected in a non post-cured material placed in the mouth. Consequently, samples were stored in water at mouth temperature and tested at periods of up to two months.

3.3 MATERIALS AND METHOD.

3.3.1 Materials Used.

Each manufacturer produced their own system with a heat/light or heat/pressure oven for curing. These were used accordingly in the initial part of the investigation. One manufacturer, however, produces two similar "post-cure" boxes, both utilising heat and light and their relative efficacy was assessed at this time. The heat and light intensities of the curing ovens used in this study are described in the previous chapter. The composite resin materials used are described in the previous chapter and batch numbers are listed in Table 3.1.

MATERIAL	TRADENAME	BATCH NO.	MANUFACTURER
HYBRID A	BRILLIANT	210590-02	COLTENE AG ALTSTATTEN SWITZERLAND
HYBRID B	ESTILUX POSTERIOR CVS	30/6/93 38	KULZER GmbH BEREICH DENTAL, WERHEIM, GERMANY
MICROFIL	IVOCLAR INLAY/ONLAY	240087	VIVADENT DENTAL GmbH, ELLWAGEN, LEICHTENSTEIN

Table 3.1: Composite resin inlay materials.

3.3.2 Specimen Fabrication.

Specimens for compressive and diametral tensile strength were fabricated in glass rods of 3.5 mm internal diameter and 5 mm in length. The light cured materials were cured for 60 seconds from both ends of the glass rod and for a further 60 seconds from both front and back of the rod. Due to shrinkage of the materials, they could then, generally, be pushed out of the rod. The samples for testing of flexural properties were produced according to the ISO Standard 4049 part 7[161]. Samples for hardness testing were produced 1cm in diameter and 1 mm thick by compressing the composite between two glass slides separated by a steel mould with a 1 cm internal diameter. The light cured materials were cured on one surface only, designated the top, for 1 minute. The secondary curing of hybrid A was carried out in the Coltene D.I. oven (Coltene AG, Altstatten, Switzerland.) recommended for this material. For hybrid B secondary curing was carried out either in the Unilux A.C. (Kulzer GmbH, Werheim, Germany) heat/light oven for the recommended time (setting 45 which corresponds to 20 minutes) or the light box connected to a translux dental curing light (Kulzer GmbH) for six minutes. In all cases the microfilled material was cured in the Ivomat heat pressure oven (Vivadent Dental GmbH. Jagst, Leichtenstien) as it does not light cure.

3.3.3 Groups Tested.

Light cured: both hybrid materials at one day, one week, one month and two months.

Heat\light post-cured: both hybrid materials at one day and two months.

Heat\pressure cured: microfilled material at one day and two months.

Ten samples were produced for each of the groups tested to fracture. Only two samples were produced for hardness testing of each group, as preliminary testing had shown that the standard deviation was low.

All samples were stored in the dark, in water at 37°C prior to testing.

3.3.4 Testing Procedure.

Compressive and diametral tensile strengths were measured at crosshead speed of 0.5 mm/minute using a universal testing machine (Nene M3000: Nene Instruments, Wellingborough, Northampton, UK). The flexural properties specimens were tested using the apparatus described in ISO No.4049 (subsection 7.8.1.7.)[161] mounted on the Nene at crosshead speed of 0.5 mm\minute.

Knoop hardness was measured using a Leitz miniload 2 (Ernst Leitz Wetzlar GmbH, D-6330 Wetzlar, Germany) at a load of 1.961N for 20 seconds. Ten measurements were made for each sample at each time.

Mean values and standard deviation were calculated for all physical tests. The individual values were compared by one-way analysis of variance, and, where differences were detected, these were identified by Student's *t* test.

3.4 RESULTS.

The results of the investigations are shown in Table 3.2, one day results for normal post curing are shown in Figures 3.1 to 3.5.

3.4.1 Compressive Strength. (Figure 3.1)

There was no significant increase ($p<0.05$) in compressive strength for either of the hybrid materials when they were subjected to a secondary cure (e.g 200±16MPa for light cured vs 197±37MPa for post cured hybrid B). At all times measured the microfilled material had a superior compressive strength to the other materials tested ($p<0.01$). For example post cured hybrid A was 250±36MPa whereas, the microfil was 427±57MPa following storage for one day.

3.4.2 Diametral Tensile Strength. (Figure 3.2)

Diametral tensile strength (DTS) was significantly ($p<0.05$) improved in hybrid A following post curing, with both the manufacturers recommended post curing regime at 120°C in the D.I.500 oven. No significant improvement was noted in hybrid B when it was post cured in any either of the recommended heat\light ovens. At all times the diametral tensile strength of the microfilled material was significantly greater than that of the hybrid materials($p<0.05$)(e.g 62±9MPa for the microfil vs 51±7MPa for post cured hybrid A at one day).

3.4.3 Flexural Properties. (Figures 3.3 and 3.4)

Flexural strength and flexural modulus were significantly increased by secondary curing in hybrid A. Post curing of hybrid B in the Unilux A.C resulted in a significant increase ($p<0.05$) in properties with flexural strength increasing from 107±16MPa to 166±18MPa. However, post curing in the Translux lightbox did not result in any significant change in the flexural properties of the materials.

The flexural properties of the microfilled material were inferior to those of the other secondarily cured materials at both times measured ($p<0.05$). They were similar, however, to those of light cured hybrid B.

3.4.4 Hardness. (Figure 3.5)

The Knoop hardness of hybrid A was significantly increased following post curing ($p<0.05$). The hardness of hybrid B was significantly increased following curing in either the Unilux A.C.($p<0.01$) or the Translux lightbox ($p<0.05$), however, there was a significant difference in hardness depending on which curing regime was adopted. Post curing in the Unilux A.C. resulted in a significantly harder material ($p<0.01$) than curing in the lightbox; with a mean Knoop hardness of 85 and 47 respectively, at one day post curing.

The hardness of the microfilled material was significantly greater than that of all other samples ($p<0.05$) apart from hybrid B cured in the Unilux A.C. where it was significantly softer ($p<0.01$).

3.4.5 Effect of Storage.

There was no significant change in any of the measured values of the materials following 2 months aging in water at 37°C.

	CURING REGIME	TIME	COMP. STR. **MPa**	FLEX. STR. **MPa**	FLEX. MOD. **MPa**	DTS. **MPa**	HARD-NESS
HYBRID A	LIGHT CURED	1 Day	228 (26)	133 (18)	8444 (1547)	41 (9)	37.3 (1)
		1 Week	224 (35)	123 (14)	8462 (441)	34 (11)	37.7 (2.4)
		1 Month	229 (33)	122 (15)	8380 (1221)	39 (8)	38.1 (1.1)
		2 Months	228 (58)	127 (17)	8302 (1130)	43 (9)	38.8 (2.3)
	POST CURED	1 Day	250 (36)	168 (24)	10926 (1486)	51 (7)	43.5 (1.4)
		2 Months	254 (35)	165 (19)	11672 (1923)	48 (9)	45.9 (2)
HYBRID B	LIGHT CURED	1 Day	200 (16)	106 (16)	7732 (1162)	46 (8)	37 (2)
		1 Week	211 (34)	112 (14)	8124 (1868)	46 (6)	39 (1.8)
		1 Month	202 (35)	116 (28)	8235 (2092)	47 (8)	38 (1.9)
		2 Months	200 (35)	115 (10)	8200 (1788)	43 (6)	37 (2.2)
	POST CURED UNILUX	1 Day	196 (37)	166 (18)	16510 (1404)	46 (6)	85 (9.2)
		2 Months	217 (44)	150 (18)	15059 (1744)	48 (4)	89 (7.1)
	POST CURED LIGHT-BOX	1 Day	216 (42)	121 (21)	8491 (873)	44 (4)	47 (2.9)
		2 Months	223 (42)	124 (13)	9246 (1560)	45 (12)	50 (3.6)
MICROFIL	POST CURED	1 Day	427 (57)	117 (31)	7279 (1191)	62 (9)	55 (3.3)
		2 Months	451 (94)	112 (30)	8419 (1748)	58 (17)	54 (3.7)

Table 3.2: Composite resin inlay materials; physical properties. Figures represent mean scores for 10 samples ( ) = Standard deviation

3.5 DISCUSSION.

By secondary curing of a composite resin material the expectation is that the conversion of methacrylate groups will increase[104] and that relaxation of internal stresses will occur[128]. Thus, the physical properties of the material should improve.

The fact that there is no improvement in compressive strength of either of the hybrid materials with secondary curing is, therefore, surprising. It is, nonetheless, in agreement with previous studies by Wendt[19], Peutzfeldt and Asmussen[98], and McCabe and Kägi[99]. McCabe and Kägi[99] offer a possible explanation for these findings. With secondary curing there is a possibility that the process will not only produce beneficial effects in the material but may also be detrimental to it; if taken to excess secondary curing could result in a stiffer but at the same time more brittle material. There must, therefore, be a balancing of possible good and bad effects of the post curing process. Consequently manufacturers will adjust their post curing regimes such that they obtain maximum benefit with minimum loss of properties. Thus, it may be that, in order to produce the increases seen in the other physical properties, it was not possible to produce any appreciable increase in compressive strength.

Compressive strengths of microfilled composite resins are, in general, lower than those of hybrid materials [6]. Therefore, the high strengths found for the microfilled material, are surprising. Nonetheless, they are in agreement with Peutzfeldt and Asmussen[98]. It seems likely that, the pressure under which this composite is polymerised may decrease the amount of porosity within the material and hence accounts for the high compressive strength observed in these samples. Porosity is known to decrease the strength of composite resins, the lack of porosity being one of the reasons for the improvement in properties of single paste over two paste composites[162]. Craig[33] also mentions that the application of pressure can markedly reduce porosity within a composite. Wilson and Norman[93] noted that there was a decrease in the number of voids in inlays produced when pressure was incorporated in the curing process, relative to conventional light curing procedures. They concluded that this was due to the pressure effect as it occurred with several different composite materials and suggested that an improvements in physical properties may be expected when a pressure curing regime was adopted. The lack of porosity of the material is also, indirectly, confirmed by the difficulty experienced in bonding to its surface. As a result of its less porous nature when compared with the two hybrid materials it does not allow a good micromechanical bond to form between inlay and composite resin cement during its cementation. Further details of this and SEM of the inlay surface are shown in Chapters 7 and 8. It would appear likely that the non-porous nature of the material may also account for the high DTS observed with the microfilled material.

The DTS of hybrid A was increased by secondary curing, in agreement with Covey *et al*[96]. However, hybrid B showed no improvement in this property when post cured in either of the suggested post curing ovens. A possible explanation for this lies in the design of the 'ovens' used with the two systems. The Coltene oven, used with hybrid A, heats the material to a temperature of approximately 120°C over a period of 7 minutes. The Unilux AC. and Translux lightbox only heat to about 60°C although the Unilux does have a longer cycle time of around 20 minutes, see Chapter 2 for further details. The work of Asmussen and Peutzfeldt[95], showed a linear increase in DTS with increasing temperature of post curing. Hence, the higher the secondary curing temperature the greater the increase in DTS. This is also supported by Wendt[19] who showed an increase in DTS in all three of his test materials at 125°C, but only in one at 60°C. It may be that heat treatment above the temperature occurring in the hybrid B curing systems would improve the DTS of this material. This premise is investigated further in Section 7 of this chapter.

The hardness of both hybrid materials was increased by secondary curing. This is, again, in agreement with Wendt[20]. However, in hybrid B, the secondary curing technique used had a significant effect on the results obtained, with curing in the Unilux AC. resulting in a significantly greater increase in hardness. Flexural properties of hybrid A were increased by secondary curing as would be expected from the work of Dionysopoulos and Watts[104], which showed an increase in stiffness of the same material following post curing. The flexural properties of hybrid B were also increased by secondary curing in the Unilux AC. but not in the Translux lightbox. Flexural strength and hardness are related for a given material[163]. During flexural testing the surface layer is under most stress when bending occurs. Fracture initiates at the surface, in these samples, hence any increase in strength at the surface, will lead to an improvement in flexural properties. It is not surprising therefore, that the flexural strength for the materials was increased in the same manner as hardness. Hybrid A, and hybrid B cured in the Unilux increased significantly while hybrid B cured in the lightbox also increased but to a lesser extent. The results of the flexural and hardness testing, taken together, suggest that the differences in curing regimes between the two Kulzer heat\light box systems is the important factor influencing the material properties.

The curing cycle in both boxes heat the composite to approximately the same temperature, 55°C in the centre of the box . The visible light intensity in the light box is 1.5 times as great as in the Unilux, yet the cycle is only one quarter of the time (Chapter 2 section 6.1). It seems likely, therefore, that the increased duration of cure i.e. maintenance of a raised temperature for a longer time, is the most important factor in increasing the hardness in this material, and that the light intensity within the box is of secondary importance. This is in conflict with the work of Peutzfeldt and Asmussen[98] who found no improvement in Hybrid B when it was cured by heat only. It does, however, agree with the work of Reinhard and Smolka[164] who indicated that the extra oral curing was a purely thermal effect. A further investigation into this point follows in Section 7 of this chapter.

The hardness and flexural properties of the microfilled material are again greater than might be expected for such a composite[28]. This again is probably due to the pressure during the curing process. The values obtained for flexural modulus are rather higher than those of Peutzfeldt and Asmussen[98], however they admitted that their method of testing tended to underestimate this property.

There is no improvement in physical properties of the light cured materials over a two month period when stored in water at 37°C. This would seem to refute the idea that improvements in physical properties are to be expected due to slow stress relaxation at mouth temperature discussed by De Gee *et al.*,[16]. This may only be the case with the materials tested in this study. It does, nonetheless, disagree with De Gee's *et al.*'s result for Coltene Brilliant (Hybrid A in this study).

3.6 CONCLUSIONS.

The results of this study support the findings of Bausche *et al.*[18], Wendt[19,20], Asmussen and Peutzfeldt[95], Covey *et al.*[96], Gregory *et al.*[97], and Peutzfeldt and Asmussen[98] that some physical properties of composite resin materials can be improved by secondary curing with heat and light. It seems likely that the duration of cycle used to cure the hybrid B material is important in the final mechanical properties obtained. Moreover, it may be that the heat rather than the light is the important factor in the secondary curing process. Finally it appears that the temperature at which curing is carried out may be important with regard to which physical properties are improved. For example, diametral tensile strength may only improve at higher temperatures than that required to improve hardness or flexural properties.

The following section investigates these hypotheses.

3.7 FURTHER TESTING, AIMS.

The aims of this part of the study were to investigate if both the heat and light produced by the different curing systems were necessary for post curing or whether heat alone was sufficient to produce the improvement seen in physical properties. Furthermore, changing the duration of heating within one of the systems (hybrid B) was investigated in the hope that this may explain the differences between the two curing ovens used with that system. Finally, by changing the curing temperature, it was hoped to discover if diametral tensile strength is dependant on the temperature of post cure or is a material dependant property.

3.8 MATERIALS AND METHOD.

Fifty specimens for flexural properties and twenty for diametral tensile strength testing were prepared in the same manner as in Section 3.3.2, to the light cure only stage. Subsequently the following groups of ten samples each were processed.

Hybrid A: (1) Flexural - Post cured in Unilux A.C.

(2) Flexural - Post cured in D.I.500 oven wrapped in tin foil to prevent further light curing.

(3) DTS - Post cured in Unilux A.C.

Hybrid B: (1) Flexural - Post cure in D.I.500 oven.

(2) Flexural - Post cure in Unilux A.C. Wrapped in tin foil to prevent further light curing.

(3) Flexural - Post cure in Unilux A.C. for 6 minutes (equivalent to post cure time in the Translux lightbox)

(4) DTS - Post cured in D.I.500 oven.

All samples were stored in the dark, in water at 37°C for 24 hours prior to testing. Testing was carried out using the Nene testing machine in an identical manner to section 3.3.4.

Statistical analysis was carried out in the same manner using ANOVA and Student's *t* test.

3.9 RESULTS.

The results from this section are shown in Tables 3.3 and 3.4 and in Figures 3.6-3.8.

3.9.1 Flexural Strength.

Curing of either material in their own post cure box, when covered in tin foil, led to the same increase in flexural strength seen when light was also present. Curing in the other manufacturers oven also resulted in the same increase in flexural strength (hybrid A 168±24MPa when post cured in the D.I.500 oven vs 160±121MPa when post cured in the Unilux A.C.). However, curing of hybrid B for a shortened cure cycle did not increase the flexural strength $p<0.05$, rather the flexural strength was comparable with that following post curing in the lightbox (166±18MPa when post cured in the Unilux A.C for the full cycle vs 118±15MPa for short cycle in the Unilux vs 121±21MPa for lightbox).

3.9.2 Diametral Tensile Strength.

Diametral tensile strength was not increased by post curing hybrid B at a higher temperature in the D.I.500 oven, conversely post curing hybrid A at a lower temperature in the Unilux oven did not result in the increase in DTS seen when its own post curing regime was followed ($p<0.05$).

	HYBRID A				HYBRID B					
POST CURING REGIME	Light Cure only	D.I.500 oven	D.I.500 oven & Tin foil	Unilux AC	Light Cure only	D.I. 500	Unilux	Unilux & Tin foil	Unilu x 6 mins	light box
FLEXURAL STRENGTH **(MPA)**	133 (18)	168 (24)	160 (21)	164 (25)	107 (16)	154 (21)	166 (18)	150 (22)	118 (15)	121 (21)

Table 3.3: Flexural strength following different post curing regimes. Mean values for ten samples. ( ) = standard deviation.

	HYBRID A			HYBRID B		
POST CURING REGIME	Light Cure only	D.I.500 oven	Unilux AC	Light Cure only	D.I.500 oven	Unilux AC
DIAMETRAL TENSILE STRENGTH **(MPA)**	41.0 (9.0)	50.8 (7.7)	42.6 (7.5)	45.8 (8.0)	46.6 (6.5)	45.3 (6.6)

Table 3.4: Diametral tensile strength with Different Curing Regimes. Mean values for ten samples. ( ) = standard deviation.

3.10 DISCUSSION.

From these results it would seem that the major factor important in producing a secondary curing effect on the materials studied in this section is heat. Removal of the light curing component of the post curing regime does not appear to have a deleterious effect on the observed improvement in the properties of the materials following post curing. This is in agreement with most of the early work done on this subject[18-20] as, initially, secondary curing was done in heat only ovens with similar improvements seen in physical properties. Only Peutzfelt and Asmussen[98] have suggested that the light component of the secondary cure is the most important factor although they, themselves concede that their results are at odds with previous studies. Moreover the size and shape of specimens used in their study did not conform to the ISO standard[161] and had been shown to give lower values than normal. As yet no other papers have been produced which have attempted to separate the light and heat components of the post curing process and their relative importances, to a degree, remains unresolved at this time. Further work is required to discover the importance of secondary light curing and, if necessary, the optimum light intensity and time interval for this process. However, from this study, it would appear that the light plays very little part in the secondary curing process for the two materials tested. The amount of time the material is exposed to the post curing regime also appears to be important as the samples cured for a reduced amount of time in the Unilux did not attain the same flexural strength as those cured for the recommended time. Wendt[129] has suggested an optimum time for curing of around 7.5 minutes but this was using temperatures of around 120°C. The Unilux box only reaches about 60°C during curing so it seems reasonable to assume that a longer curing cycle will be required. Consequently, when the curing cycle is shortened, insufficient time has elapsed for significant secondary curing to have occurred hence the flexural strength was not improved. It is interesting to note that using a curing regime similar to that recommended by Wendt[129] (i.e the D.I.500 oven - see Chapter 2) results in the same increase in flexural strength as the slower Unilux secondary cure. Thus, it would appear that post curing of composites can be accomplished in more than one way with a resulting improvement in flexural properties. This is borne out by the fact that the flexural strength of Hybrid A, for use with the D.I.500 oven is also improved, to the same degree, by secondary curing in the Unilux at a lower temperature for a longer time (60°C for 20 minutes as opposed to 120°C for 7 minutes).

Asmussen and Peutzfeldt[95] have suggested that the DTS of a material following post cure is linearly related to the temperature at which it is cured. Wendt[19] also noted a greater increase in DTS at higher post cure temperatures. It was therefore expected that an improvement in this property would be noted for Hybrid B when post cured at 120°C as opposed to 60°C. However this was not the case. The position is further complicated by the fact that curing Hybrid A at the lower temperature resulted in no improvement in DTS relative to light cure only samples (Fig 9). It would, therefore, appear that there is a temperature dependant relationship to DTS for Hybrid A but not for Hybrid B. The only explanation available for this phenomenon is that the properties following heat treatment are, like those before treatment, material specific. Not all composites will react in the same manner to heat treatment of any type. It may be that DTS of this material cannot be improved by secondary curing. Alternatively, if DTS can be improved in Hybrid B, it is at a temperature and time regime different from those tested in this study and extensive further study would be required to discover the optimum regime.

3.11 CONCLUSIONS.

The following conclusions can be drawn from this study.

(1) Secondary curing of the hybrid composites, tested in this study, results in significant improvements in some of the physical properties tested.

(2) The improvement in physical properties appears to be related to the temperature of the secondary curing process. The light component appears to be of secondary importance.

(3) The improvement in physical properties appears to be related to the duration of the secondary curing cycle. Shortening of the curing cycle is therefore not recommended.

(4) The improvement in properties appears to be dependant on the material being tested.

(5) Curing of the microfilled material by heat and pressure results in improvements in physical properties when compared with values of similar light cure materials, from the literature.

The relative importance of the heat and the pressure is not known at this time.

Chapter 4: Physical Properties of Composite Resin Cements for use in the Inlay Technique.

4.1 INTRODUCTION.

Composite resin cements have been used extensively throughout restorative dentistry for cementing of both intracoronal and extracoronal restorations and prostheses. More recently they have been used for bonding of porcelain and composite resin laminates and inlays to teeth. Initially the composite resin cements used for this purpose were chemically cured. However, in the same way as the original chemically cured direct composites were inconvenient to use, these materials proved unacceptable. Light cured materials were adopted for these applications, but the curing of such materials has proved to be susceptible to attenuation of light when passing through porcelain and composite restorations[146-148,165]. This led to the potential problem of uncured resin remaining beneath restorations, with its subsequent sequelae. To overcome the problem of inadequate cure, dual-curing composite resin materials were introduced. These combined the photosensitive, camphorquininone with the conventional chemical amine\peroxide system. Generally, a relatively slowly acting amine\peroxide system was employed to allow an adequate working time while still assuring curing[66]. Theoretically, at least, these materials provided the clinician with an adequate working time, command setting in areas exposed to light; allowing stabilisation of the restoration, while any area which did not receive sufficient light, was guaranteed cure by the "backup" chemical system. With these potential advantages, dual curing materials were rapidly adopted and have now become the material of choice for cementation of translucent resin bonded restorations.

The ideal properties of a luting agent are well known and have not altered much since described by Skinner in 1946[167]. However, when considering luting agents for use with composite resin inlays, the physical properties and ability to resist degradation in the oral environment are, perhaps, of greater importance than in those for use with cast metal inlays. Composite resin inlays do not incorporate bevelled margins[82,89,135]. Consequently, unlike metal inlays where these bevels protect the cement lute, in composite resin inlays, the lute is directly exposed to the oral environment. Moreover, the cement lute has been shown (Chapter 9) to be at least double the accepted, normal value of 25μm-50μm[92] for cast metal restorations, therefore an even larger area of lute is exposed. Furthermore, the greater cavity divergence, acceptable with resin bonded inlays, places more stresses on the cement lute than would be the case in a more parallel type cavity, where retention is achieved, mainly, by near parallelism[168]. It has, therefore, been suggested that the ideal lute, for resin bonded inlays, should possess similar properties to the inlay it cements in order to support it during function and resist degradation[145].

When considering the properties of a dual curing composite resin luting cement specifically, however, there are certain extra parameters which may be of some importance. The setting time of the materials both with and without light curing is extremely important. The degree of light attenuation caused by the inlay material, and its subsequent effect on setting time, is also crucial as it enables the dentist to modify his cementing technique to take this factor into account. Furthermore, as it has been reported that the degree of methacrylate conversion following chemical curing of a dual cured cement is, generally, less than when cured with light[149], and that light attenuation, by composite resin inlay materials, may be a problem[147], the properties of the luting cement when cured by chemical means alone will be important as they may, in fact, be the properties of a large proportion of the cementing material.

Conversely, the film thickness of luting agents for polymeric restorations may not be as critical as for cast metal inlays as it has been shown that the lute is generally considerably thicker, as mentioned earlier.

4.2 AIMS.

The physical properties of three composite resin luting cements for use with the composite resin inlay technique were examined in this section. The properties chosen for examination were the same as in the previous chapter for the inlay materials themselves, to allow direct comparison of the inlay and its respective cement. All these properties (compressive strength, diametral tensile strength, flexural strength, flexural modulus and hardness) were recorded for light cured materials. However, further testing of hardness and flexural properties was carried out on samples which had been allowed to chemically cure in the absence of light; as these properties were the most sensitive to change in the previous chapter and have been shown to be a good indicator of degree of cure[17,163]. From this it was hoped to discover the relative importance of the chemical and light curing components and assess any differences between the respective materials. Additional properties important to composite resin inlay cements were also considered. Film thickness was assessed as, theoretically, this may have some effect on ease of seating of restorations. Furthermore, the setting time of the inlay cements was recorded. This property was measured both with direct light exposure and with no light exposure, the latter time relying entirely on the chemically curing component of the material. Finally, the effect of attenuation of light by inlay material was assessed in an attempt to provide some guide on what increase in curing time is required when curing through an inlay.

4.3 MATERIALS AND METHOD.

4.3.1 Materials Used.

A description of the composition of the three proprietary composite resin inlay cements used in this study is presented in Chapter 2. Batch numbers are listed in Table 4.1.

MATERIAL	TRADENAME	BATCH NO.	MANUFACTURER
HYBRID CEMENT A	DUO CEMENT	9112467	COLTENE AG ALTSTATTEN SWITZERLAND
HYBRID CEMENT B	ADHESIVE CEMENT	93.03 038	KULZER GmbH BEREICH DENTAL, WERHEIM, GERMANY
MICROFIL CEMENT	DUAL CEMENT	160115 (catalyst) 160116 (base)	VIVADENT DENTAL GmbH, ELLWAGEN, LEICHTENSTEIN

Table 4.1: Composite Resin luting cements used.

4.3.2 Physical Properties and Film Thickness.

Specimens for compressive and diametral tensile strength were fabricated in almost exactly the same manner as light curing samples in Chapter 3. Equal amounts of base and catalyst of each respective material were mixed together for 30 seconds. The mixed cement was then packed into glass rods of 3.5 mm internal diameter and 5 mm in length and subsequently light cured for 60 seconds from both ends of the glass rod and for a further 60 seconds from both front and back of the rod. Due to shrinkage of the materials, they could then, generally, be pushed out of the rod. The samples for testing of flexural properties were produced according to the ISO Standard 4049 part 7[161]. However, in addition to the light cured samples, specimens for each material were produced utilising only the chemical curing component of the material. These samples were allowed to cure in the mould, in darkness for 15 minutes before being removed. Unlike in the previous chapter, where separate hardness specimens were produced, hardness testing was carried out using the flexural strength specimens following their fracture. Ten samples were produced for each material, for each testing procedure. All samples were stored in the dark, in water at 37°C for one day prior to testing.

Film thickness, of the three materials, was assessed according to British standard no. 6039[169]. No specific standard exists for composite resin luting cements. However, as the testing regime is the same for all the other types of luting cement available, it seemed logical to adopt this method.

Mean values and standard deviations were calculated for all physical tests and film thickness. The individual values for each test were compared by one-way analysis of variance, and, where differences were detected, these were identified by Students *t*-test.

4.3.3 Setting Time.

The setting time of the composite resin luting cements was measured by the acoustic transmission method first described by Young *et al.* in 1977[177]. Equal amounts of base and catalyst pastes were mixed for each material and a droplet, approximately 1mm in diameter, was placed between two steel pins housed in two steel cylinders (Figure 4.1). A 1kHz continuous sound signal was generated at one end of the steel cylinders. The increase in sound level transmitted by the resin as it set was measured using a vibrational transducer and associated amplifiers (Bruel and Kjaer precision sound level meter type 2203). The setting time was calculated from the definition of Main *et al.*[171]: the time taken from the start of light exposure to the time at which the extrapolated initial slope of the curve of transmitted sound level against time intercepts the back-extrapolated plateau region of the curve (Figure 4.2). This definition was slightly modified when the chemically cure only materials were being tested where setting time was assessed from the time when light exposure would have occurred in a light cured sample. Thus, the measurement of setting time began immediately following the 30 second mixing time and the ten seconds required for placement of the resin on the metal pins, as this was when light curing began with appropriate samples.

(a) Light and Chemically Cured Samples

Composite resin discs of 10 mm diameter and 1.5 mm thickness were produced for each of the inlay materials. Only one shade of composite was used for each inlay material, a similar universal shade being selected for each material as large differences in shade have been shown to affect curing times in testing of similar materials[147]. The same translux curing light was used throughout this study. Measurements were made with and without these discs between the light source and the resin. In all cases the light source was positioned 3mm from the centre of the resin droplet. Five measurements were made for each resin cement both with and without the inlay material in place.

(b) Chemical Cure Only Samples

These samples were mixed and placed on the metal pins in the same manner as the light and chemically cured samples. Thereafter curing took place in the dark. Five measurements were taken for each composite resin luting cement in this series.

Presence of differences in setting time, under each condition, were assessed by one-way analysis of variance. A Student's *t*-test was then used to test for differences between specific pairs.

4.4 RESULTS.

4.4.1 Physical Properties and Film Thickness.

The results of the investigations into physical properties are shown in Table 4.2 and in Figures 4.3-4.7.

	CURING PROCEDURE	COMP. STR. MPa	DTS MPa	FLEX. STR. MPa	FLEX. MOD. MPa	HARD-NESS
HYBRID A	LIGHT CURE	207 (53)	44 (5.1)	109 (21)	7512 (1325)	39 (1.4)
	CHEMICAL CURE			83 (14)	5732 (720)	29 (2.3)
HYBRID B	LIGHT CURE	195 (29)	44 (5.1)	104 (17)	7421 (1657)	31 (3.6)
	CHEMICAL CURE			80 (12)	5745 (720)	24 (2.6)
MICROFIL	LIGHT CURE	158 (44)	37 (4.4)	77 (14)	3109 (506)	24 (1.2)
	CHEMICAL CURE			49 (6)	1564 (263)	12 (2.4)

Table 4.2: Physical properties of composite resin cements. Figures represent mean scores for ten samples. ( ) = Standard deviation.

For the light cured materials, there were no statistical differences in compressive strength (207±53MPa for hybrid A vs 195±29MPa for hybrid B), diametral tensile strengths (43±5MPa for hybrid A vs 44±5MPa for hybrid B) or in flexural properties between the two hybrid materials ($p<0.05$). All properties, measured to fracture, were poorer in the microfilled material than in either of the hybrid cements, at the $p<0.01$ level (eg. Flexural strength was 77±14 for the microfil but 108±21MPa and 104±17MPa for hybrids A and B respectively).

The hardness values for all three materials were significantly different from each other ($p<0.05$); hybrid cement A being hardest (39±1.4 Knoop hardness number) and the microfilled material least hard (24±1.2 Knoop hardness number), with hybrid B having an intermediate value (31±3.6 Knoop hardness number).

The physical properties of the chemically cured only materials showed significant reductions in both hardness and flexural properties for all three materials ($p<0.01$). Hybrid A showed a 26% reduction in properties, hybrid B showed a reduction of 23% while the microfilled material showed the greatest drop in properties of almost 50%.

The results of the film thickness testing are given in Table 4.3 and in Figure 4.8.

	HYBRID A	HYBRID B	MICROFIL
FILM THICKNESS (μm)	106 (4)	137 (7)	45 (8)

Table 4.3 Film thickness of composite resin cements. Figures represent mean scores. ( ) = Standard deviation.

There were significant differences in film thickness of the three resin cements; with the microfilled material having a much lower film thickness than the two hybrid materials ($P<0.001$ vs hybrid B, $p<0.01$ vs hybrid A). Although the two hybrid materials had more similar values, there was still a significant difference between these two ($p<0.01$).

4.4.2 Cement Setting Times.

The results of these investigations are shown in Table 4.4 and Figure 4.9.

	CURING PROCEDURE	Time
HYBRID A	Chemical cure	5 mins 10 ± 34 seconds
	Light cure	46 ± 4.5 seconds
	Light cure through inlay	123 ± 17 seconds
HYBRID B	Chemical cure	4 mins 35 ± 37 seconds
	Light cure	40.8 ± 4.2 seconds
	Light cure through inlay	116 ± 12 seconds
MICROFIL	Chemical cure	10 minutes 6 ± 54 seconds
	Light cure	68 ± 7.2 seconds
	Light cure through inlay	228 ± 32 seconds

Table 4.4: Setting times of composite resin cements ± standard deviation following curing by chemical means, light and light through 1.5mm of composite inlay material.

There were significant differences between the setting times of all three cements both with and without light curing. Hybrid A and hybrid B have similar setting times following light exposure of 46±4.5 seconds and 40±4.2 seconds respectively. However the setting time of the microfilled material is significantly greater ($p<0.001$) at 68±7.2 seconds.

The extremely, long chemically induced, setting time of the microfilled material (over 10 minutes) was more than twice that of the other materials tested (hybrid A 5 minutes 10 seconds, hybrid B 4 minutes thirty five seconds). The attenuation of light caused by the presence of inlay material between light source and inlay had a significant effect on the setting times of all three materials tested ($p<0.001$). The setting time of the two hybrid materials was increased to around 2 minutes in each case, whereas the microfilled material increased by more than three times to almost four minutes.

All setting times were recorded at ambient room temperature (around 23°C) the setting times could be expected to be less at body temperature, although, it seems likely that the differences between groups would remain.

4.5 DISCUSSION.

4.5.1 Physical Properties

The physical properties of the light and chemically cured hybrid materials were not greatly different from the inlay material, for which they were manufactured, when it has only been light cured. This is not very surprising as they are essentially the same materials with only slight modifications in filler content (only 5.5% difference with hybrid B). However if the cementing agents are compared with the post cured inlay materials significant differences occurred. Wherever the post curing process produced an improvement in mechanical properties of the light cured inlay material a significant difference was observed between inlay cement and post cured inlay material. Hence, hardness and flexural properties of both hybrid cements were significantly lower ($p<0.05$ for hardness and flexural properties of hybrid cement A vs Hybrid A post cured inlay material and, $p<0.01$ for hardness and flexural properties of hybrid B cement vs hybrid B post cured inlay material). The fact that these properties are less in the inlay cement than in the inlay itself could prove a potential area of weakness around the inlay as has been suggested by Rees and Jacobsen[145]. However, as the properties of the cements are similar to those of the light cured only inlay materials, both of which are marketed as posterior composite resins for direct placement, it seems likely that these particular composite resin luting cements should perform adequately. This is only the case when the composite resin cements are adequately light cured. Where the chemical curing process is allowed to act alone the properties of the set material are markedly inferior to even the light cure only inlay materials. Under these circumstances the cement lute must be considered an area of potential weakness in the composite resin inlay restoration.

The microfilled luting cement exhibited significantly poorer physical properties in all parameters tested. This result, which is supported by Breeding *et al.*[147] and Hasegawa *et al.*[148], is not unexpected as, in general, the physical properties of microfilled materials are poorer than hybrid materials[27,172]. However, very large differences occurred between the physical properties of the microfilled composite resin inlay cement and its respective composite resin inlay material. In none of the measured parameters, did the luting cement achieve even half the values of the heat\pressure cured inlay material. This is probably due, in part, to the lower filler content of the luting cement (only 60% for the cement as opposed to 76% for the inlay material) however, the role of the heat\pressure curing process in improving the physical properties of the inlay material (as described in the previous chapter) is undoubtedly at least as important. It therefore seems likely that this is not a particularly weak microfilled luting agent, rather it is the inlay material which is unexpectedly strong. Nonetheless, the mismatch in physical properties between the inlay and its luting agent may be a problem with this inlay type. This is of particular importance as the microfilled inlays have been shown to exhibit the largest marginal gaps, and, consequently, have most luting agent exposed to the oral environment[90,145] (Chapter 9). The physical properties of this material are decreased by a further 50% by lack of light during the curing process, producing the weakest material of any of those tested and, consequently an even greater mismatch between properties of lute and inlay would result.

Production of ultimate physical properties of all three dual cured composite resin cements relied on exposure to light. In all the materials tested there was a significant decrease in hardness and flexural properties when cured by chemical means alone. This drop in properties is probably due to a decrease in the methacrylate conversion of the material when cured in this manner; as both of these properties have been shown to be related to degree of cure in composite resins[17,163]. This situation is confirmed by Rueggenberg *et al.*[149], who used Infrared Spectrometry to assess degree of cure of composite resin luting cements with both light and chemical cure or chemical cure alone. In only one of the four materials tested did chemical cure result in the same degree of methacrylate conversion as when light was employed. Furthermore, there were variations between materials in the differences between light and chemical cured and chemical cured only samples. This is in agreement with Hasegawa *et al.*[148], and was, presumably, due to differences in the relative amounts of light and chemical activators within the materials. The materials with most chemical activator came closest to their observed light and chemical cured properties when cured by chemical means alone. Materials which approach their light cured values give the operator a larger margin of error when curing and are, surely, to be preferred. However, as no material in this study achieved its full curing potential without the application of light, the concept that any of these materials is a truly dual curing material must be called into question.

4.5.2 Film Thickness.

Film thickness gives an indication of a cements ability to flow during seating of a restoration. This property is influenced by several factors, notably the viscosity of the material.

The difference in film thickness between the two hybrid materials can be explained by the different formulations of the two materials. Hybrid A is a less heavily filled material than hybrid B; only 67% as opposed to 75%. Furthermore the filler particles are of much larger size in hybrid cement B, up to ten times the size. Finally, hybrid A cement contains a smaller proportion of the highly viscous monomer Bis-GMA than Hybrid B. These factors lead hybrid B to be a relatively more grainy and viscous cement, which flows less well under load, and, consequently, exhibits a higher film thickness.

The difference between hybrid A and the microfilled composite resin cement is slightly more difficult to understand. The microfilled material only contains 60% filler as opposed to hybrid A's 67%. This filler is composed of highly dispersed silyated silica acid and ytterbium fluoride with a particle size of 0.01μm to 0.04μm. Finally, the microfilled material has a totally different resin formulation. These factors combine to produce a cement which, clinically, appears less viscous than hybrid A cement and in turn produces a low film thickness. However, although the inorganic filler size is extremely small, this material contains prepolymerised filler particles of up to 87μm which Inokoshi *et al.*[173] have suggested may interfere with seating of restorations. It is difficult, therefore, to account for the fact that a film thickness of 45μm was obtained for this material.

4.5.3 Cement Setting Times.

Working time of these materials is dependant on the chemical curing time. All three materials recorded what could be considered an adequate working time of over four and a half minutes. This time would be shortened by working at mouth temperature and also by ambient light, especially the dental operating light, as shown by Jacobsen[174]. However, in none of the cements tested would inadequate working time appear to be a problem. Rather, the opposite would appear to be true, particularly if the microfilled material is considered.

The light curing times for the composite resin cements tested are comparable with previous results for a two composite luting agent[146] and several light curing composite resins[175] tested in the same manner. However these times were recorded with direct exposure of the composite to light which would only occur at an inlay margin. The increase in setting times caused by inlay or porcelain facing materials is also well known[146,165] and is borne out by this study. Increases in setting time from 3-4x were recorded following curing through 1.5mm of composite resin inlay material. Obviously, in order to obtain the best from any of these materials, clinically, the ultimate degree of cure and, therefore, optimum physical properties are desired. While dual curing resins do offer an advantage over light cure only materials, in that some degree of cure is assured in all areas, whether exposed to light or not. From this study, at best only 75% of the ultimate properties are likely to be obtained by self curing. Therefore, the idea that light curing can seal the edges quickly while the chemical cure will deal with any residual uncured material, in inaccessible areas or beneath the inlay is incorrect. Consequently, it is desirable to light cure as much of the resin cement as possible. This will inevitably result in extended light curing times as light exposure must be increased by at least three times that suggested by the manufacturers if adequate curing of the luting agent is to be achieved.

Chapter 5. The Fracture Toughness of Three Composite Resin Inlay Materials.

5.1 INTRODUCTION.

The results of Chapter 3 have shown that post curing of light cured composite resin materials with heat and light results in improvements in certain physical properties of two hybrid composite resins; while curing of a microfilled material with heat and pressure resulted in higher values for all physical properties than reported in the literature for light cured microfilled materials. Despite the importance of these properties in assessment of the materials and the potential of the post curing processes, it has been suggested that they are not particularly suitable for explaining clinical observations[176].

Resistance to fracture is important in prevention of marginal breakdown and chipping of restorations[94,177]. Furthermore, any wear mechanism which involves crack propagation must include fracture resistance as a major controlling factor. Consequently, a fracture mechanics approach to testing of dental materials has been proposed[178], with critical stress intensify factor or fracture toughness (K_{1C}) being the criterion assessed as a measure of a materials fracture resistance.

5.2 THEORY OF FRACTURE TOUGHNESS.

Dental restorative materials are all brittle and, therefore, suffer from an inability to absorb appreciable quantities of elastic strain energy prior to fracture. This failing manifests itself as low tensile strength, flaw sensitivity and tendency to catastrophic failure[179].

The basic study of the fracture mechanics of brittle solids was carried out by Griffiths[180]. His concept was that where a solid was fractured, energy was required to form the newly cleaved surfaces and that this energy was supplied by relaxation of stored energy around the growing crack. This can be quantified either as the critical strain release rate (G_{IC}) or as the stress intensification at the onset of unstable crack propagation (K_{IC}). These are equivalent concepts, being related simply through the elastic modulus. The critical stress intensification factor has been the preferred method of fracture toughness measurement, in the dental literature, as it is directly related to the fracture stress of materials (a more widely used and understood parameter e.g. compressive stress or tensile stress) in an expression of the type:

$$K_{IC} = \sigma_F(\pi a)^{0.5}$$

where σ_F= fracture stress and a= the size of the flaw initiating failure. This relationship is only valid provided the extraneous defect is larger than the critical size of flaws inherent to the material. This inherent flaw size (a_0) of a material is the equivalent of Griffiths crack which initiates failure in a highly polished surface and, if inserted in the above equation, will produce the fracture strength of a polished material. i.e. If extraneous flaws are less than a_0 in size then the fracture stress is constant and dictated by the value of a_0.

If, as assumed in the original derivation of the equation, the material is an elastically isotropic and homogeneous solid in which the only deformation is elastic (obeying linear laws), this method of fracture analysis would be restricted to single phase glasses. The obvious inhomogeneity of dental filling materials would preclude its use . However, linear elastic fracture mechanics has been applied to all classes of materials. Common properties and microstructural features producing deviations from the assumed elastic response, have not restricted use in commercial systems.

In metals and some polymers the high stress produced at the crack tip produces plastic flow that precedes the front. This is, nonetheless, as much a part of the "work to fracture" as that produced at the new surface. Providing the extent of this plastic zone is considerably less than the crack length or specimen width the measurement of K_{IC} is not affected. Similarly, filler particle may also disturb the crack front. However, this does not invalidate the results provided the specimen is large enough to reduce the size of the inhomogeneity relative to crack length and specimen width.

Fracture mechanics has been used successfully to quantify the resistance to failure of a wide variety of constructional materials for many years. However, it is only in the last 20 years that limited use has been made of this approach in dental research.

5.3 FRACTURE TOUGHNESS IN DENTISTRY.

5.3.1 Introduction.

Initially, interest in fracture toughness centred around its applications in testing of denture base materials[181-184]. However, increasing interest has been shown in dental cements, amalgam, porcelain and, most notably, in composite resins. The fracture toughness of teeth has even been calculated with that of dentine[185] being considerably higher than that of enamel[186], due to the blunting effect of the dentinal tubules on the crack front. Representative values of fracture toughness for teeth and dental materials are presented in Table 5.1 at the end of this section.

5.3.2 Dental Cements.

Lining cements have been investigated by Lloyd and Anderson[187]; whose study considered the fracture toughness of calcium hydroxide cements, and Hill and Labok[188]; who discussed the influence molecular weight of polyacrylic acid may have on the properties of polycarboxylate cements. Dental cements for use as filling materials have also been studied. Lloyd and Mitchell[189] investigated the fracture toughness of several proprietary silicate and glass ionomer cements and found glass ionomer to have a generally higher K_{IC}, particularly in the case of Chemfil (AD International Ltd., Weybridge, England), which achieved a value of 0.451 $MNm^{-1.5}$ due, it was suggested, to the high powder water ratio possible with this material. A fracture toughness value of around $0.5MNm^{-1.5}$ for glass ionomer cements was confirmed by Goldman[176] and a subsequent study by Lloyd and Adamson[190]. This latter paper investigated the toughness of a metal reinforced glass ionomer material however, this modification in formulation did not result in any improvement in K_{IC}.

5.3.3 Dental Amalgam.

Fracture toughness of dental amalgam was first described by Roberts *et al.*[191]. More recently, Lloyd and Adamson[192] considered the fracture toughness of sixteen proprietary amalgams. K_{IC} values of between 0.955 and $1.602MNm^{-1.5}$ were obtained with decreased fracture toughness values noted for materials with higher copper content. A subsequent paper[190] studied the effect of storage on fracture properties and showed an increase in K_{1C} over two days followed by a gradual decline over the following three months, although never falling to its initial value. Inherent flaw size (a_0), however, was greatly decreased over this period; from 121μm to 20μm. It was suggested that this decrease in flaw size was due to the diffusion within the amalgam allowing initial flaws to partially "heal" over time. Cruickshanks-Boyd and Lock[193] investigated the fracture toughness of high and low copper amalgams. Similar results to those of Lloyd and Adamson[192] were obtained with high copper materials exhibiting lower fracture toughness. Furthermore, this paper considered three methods of fracture toughness testing, concluding that single edge notch beam specimens (described later in this chapter) gave the most reliable values. Hassan *et al.*[194] investigated the fracture toughness of six amalgams using a microindentation method. The values for K_{IC} recorded in this study were lower than those of Lloyd and Adamson[190,192] varying from 0.65MNm$^{-1.5}$-0.75MNm$^{-1.5}$. Interestingly, however fracture toughness was shown to decrease as the margin of the amalgam was approached with a drop of 30% being recorded. This factor was variable between amalgams tested and, it was suggested, may be related to the differences in marginal breakdown seen *in vivo* with different materials. However it is questionable whether the conditions required for indentation testing were met when indentation was made near the specimen edge. It would appear likely that the fact that less constraint of the amalgam occurred at the edge of a specimen in turn allowed longer crack growth. Consequently a lower K_{IC} value was recorded. It may be, therefore, that the observed effect was due to experimental design rather than any true difference.

5.3.4 Porcelain.

Fracture toughness of porcelain and porcelain bonded to metal has been investigated by several authors, by both indentation techniques and using three point bending. The fracture toughness of porcelain has been shown to vary between 0.9MNm$^{-1.5}$ and 1.56MNm$^{-1.5}$ with aluminous porcelains having significantly higher values than feldspathic porcelains[179]. This importance of alumina content was confirmed in a recent paper by Kvam[195] who found a new high alumina content core material to have an even higher value of K_{IC} than conventional aluminous porcelains, with values of 2.1MNm$^{-1.5}$ and 1.6MNm$^{-1.5}$ respectively. However, this toughness can be significantly increased by bonding the porcelain to a metal substrate[196]. A fracture mechanics approach has subsequently been used to evaluate this bond strength[197,198]

5.3.5 Composite Resins.

Fracture mechanics has been applied to the testing of composite resins more frequently than any other dental material.

The fracture toughness of composite resins has been shown to increase with increased filler fraction[178,179,199-202]. Various mechanisms have been proposed for this observed increase. Filler particles can cause the crack to deviate from a straight line, resulting in a crack of greater surface area. Hence more energy can be dissipated. The deviation of the crack from the straight line can be seen as increased surface roughness of the fractured surface. This appearance is characteristic of highly filled composites[189,200].

Another model for determining the fracture path and, subsequently, the fracture energy, has been proposed by Lloyd and Mitchell[189]. This theory is based upon the mismatch between moduli of matrix and filler particle. In this, the application of a tensile stress to the matrix creates a stress concentration at the interface between particle and matrix. The crack front advances through these regions of high stress, however the exact path is dictated by the degree of adhesion between filler and matrix. Where adhesion is perfect, the crack travels around the particle; where it is lacking, the crack occurs at the interface between matrix and particle resulting in separation. This model suggests differences in appearance of fractured composite surfaces as being related to the matrix\particle adhesion; a fact confirmed by this and other studies[189,202].

A further mechanism involving, pinning and bowing of the crack front has been suggested[94,189,199,200,202]. From observed crack front/particle interactions and the assumption that the crack front possesses a line energy, Lange[203] proposed that the momentary pinning of the crack front by particles would result in bowing of the crack front between particles. This would increase the crack length and, thus, the fracture energy. There are, however, various factors which are of relevance, should Lange's model be applied to dental composite resins.

1. Matrix phase dispersion toughening does not occur until the particle interspace is less than the inherent flaw size of the material.

2. Fracture energy then increases as particle interspace decreases either by increase in filler loading or particle size decreases.

3. As the particle interspace decreases even further, the stress fields from adjacent bowing crack fronts overlap and the energy required for crack propagation decreases.

4. The matrix strain imposed by the crack must be accommodated elastically by the particles; if not the particle may fracture before the bowing can take place.

As can be seen, this mechanism will only be effective in filled resins where the particles are, relatively widely spaced (i.e. low filler loadings or small particles). The higher filler loadings of many dental composites places them beyond the range over which Lange's model holds. In fact, several workers[204-206] have shown that the fracture energy reaches a maximum at around 30% by volume. Above this the fracture energy falls.

On the basis of filler content only microfine filled composites may be controlled by this mechanism. However, Lange and Radford[205] have shown that small particles, such as occur in microfilled composites, are less effective at crack pinning and, consequently, produce a lower increment in the fracture energy. This fineness of the dispersed phase has been suggested[189] as the reason why such composites have a low fracture energy. Nonetheless, another explanation of the high K_{IC} of composites with high filler loadings is required.

Various authors have noted that the fracture toughness is the square root of the product of the fracture energy and the elastic modulus. Elastic modulus increases with increased filler loading due to interaction of stress fields around the particles[206]. Therefore, even though the fracture energy may be decreasing, due to increasing filler load, the fracture toughness will continue to increase.

The ability of the resin matrix to absorb energy by plastic deformation and, consequently, increase fracture toughness has been recognised as an important factor[4,15,23-26]. Davis and Waters[200] relate K_{IC} to the yield stress of the material; a low yield stress resulting in a high fracture toughness. The yield stress represents a diffuse yielding around the crack tip blunting it. Thus, a plastic zone forms at the crack tip which can absorb energy increasing fracture toughness. They went on to identify several factors which may make it easier for the matrix to undergo plastic deformation (the presence of water, slow crosshead speed during testing and, controversially, lack of adhesion between matrix and filler).

The aging of composites in air, water, saline, sucrose solution and ethanol has been studied by various authors. Lloyd[101,207] and Lloyd and Ianetta[178] reported a significant rise in toughness, over one week, of a composite stored wet rather than in air. However by 30 weeks this difference had become negligible. These changes have been attributed to the function of the leaching and absorption of plasticisers by the composite. Initially plasticiser, in the form of water is gained by the composite, resulting in increased energy absorption and hence, increased fracture toughness. However, with extended storage monomer, which acts as plasticiser within the set material, is lost. This decreases the energy absorbing capacity of the material and the fracture toughness falls.

In addition to its plasticising action, water sorption by composites has the potential to effect the resin filler bond. The silane coupling agent may be effected by the corrosive nature of water, resulting in debonding of filler particles and a lower fracture toughness[177,190,208]. The resulting fractography of composites affected in such a manner reveals clean particles and pits on the fractured surface. However, the resistance of a composite to water damage varies significantly between materials with no effect noted in some[102,177], whereas some materials show substantially greater deterioration[190].

Storage of composites in solutions of saline and sucrose produced no effect on the fracture toughness of composites. This led Lloyd[207] to conclude that the transient changes in environment which occur in the mouth were unlikely to affect failure of restorations through fracture.

Storage of composite resins in ethanol, however, has been reported to both increase and decrease the fracture toughness of composite resins. Pilliar *et al.*[177] suggested that ethanol produced an environmental crazing around the crack tip which reduced stress intensification and consequently increased fracture toughness. Ferracane and Marker[102], in contrast, suggested that the resin matrix is softened and degraded by prolonged exposure to ethanol; a finding supported by the decrease in K_{IC} of unfilled resin when subjected to the same regime. Moreover, it was suggested that the lower K_{IC} may also be due to the drop in elastic modulus observed in composites stored in alcohol. The difference between these two studies remains unresolved. However, perhaps the different testing geometry of the specimens and the different composite resin materials used may offer some explanation.

In addition to moisture and chemicals, composite resin restorations are subjected to a range of temperatures in the mouth. Lloyd noted little variation in fracture toughness of composites tested at temperatures between 20°C and 40°C. Furthermore, temperature cycling of up to 10^4 cycles between 3°C and 60°C has been shown to have no effect on K_{IC} of 6 proprietary materials[209]. These results would suggest that variation of temperature *in vivo* will have little effect on the fracture toughness of composite resins.

The development of fracture toughness has been described by Lloyd and Ianetta[4]. An initial increase in toughness has been interpreted as a post curing phenomenon resulting from a strengthening of the resin. This peak, which occurs after a few days, is however, followed by a slight decline in the long term, as plasticising monomer is either used up in a continued process or is lost to the atmosphere.

Finally, fracture toughness has been used to investigate composite repairs and layering techniques. Dhuru and Lloyd[210] and Lloyd and Dhuru[211] reported a significant decrease in fracture toughness of the repaired material and, recently, the fracture toughness of layered composite has been investigated[212]. Layering perpendicular to the crack front had no effect with either composite tested. However, K_{IC} was reduced when layering was parallel to the crack in a hybrid material.

5.3.6 The Effect on Fracture Toughness of Post Curing in Composite Resins.

Post curing, as a method of increasing fracture toughness, was first described by Cook and Johanssen[94]. In this study the degree of conversion was increased by post curing of a light cured composite at different temperatures. The higher the post cure temperature, the greater the degree of conversion and, consequently, the higher the fracture toughness. They proposed that this increase was due to higher elastic modulus which resulted from the increased conversion. This, it was suggested, more than compensated for the loss of plastic deformation of the matrix at the crack tip. However heating in a dry oven for 24hrs is a very different regime from those now advocated, and results obtained may not be comparable.

With the interest in post curing of composite resins to optimise their physical properties, as part of the composite inlay technique, the potential effect of this process on fracture toughness has recently been described in four papers. Unfortunately, results of these studies are conflicting (despite three of them having an author in common). Ferracane and Marker[102] suggested that post curing of 4 experimental composites decreased the fracture toughness initially but the value would increase with storage in water over a 14 month period. Ferracane and Codon[100], however, suggested that an increase in K_{IC} can be expected following heat treatment and, in a subsequent abstract, Ferracane *et al.*[213] suggest that a fall in fracture toughness can be expected over 180 days. The most recent paper on this subject[103] further complicates the issue by suggesting that post curing has no effect on fracture toughness and that no change in properties can be expected over a three month period. A possible explanation for differences observed between these studies must be the use of different post curing regimes and different composite resin materials. Nonetheless, there is an obvious need for clarification of this situation; particularly with regard to proprietary composite resin inlay systems.

MATERIAL	FRACTURE TOUGHNESS ($MNm^{-1.5}$)
Enamel	0.7 - 1.3
Dentine	3.08
Denture Base Acrylic	≈1.5
Calcium Hydroxide Cement	0.02 - 0.16
Zinc Polycarboxylate Cement	0.21 - 0.80
Silicate Cement	0.12 - 0.3
Glass Ionomer Cement	0.25 - 0.5
Glass Cermet Cement	0.42
Amalgam (< 6% copper)	1.29 - 1.58
Amalgam (> 6% copper)	0.95 - 1.29
Microfilled Composite Resin	0.7 - 1.0
Hybrid Composite Resin	1.5 - 2.0
Porcelain (feldspathic)	0.9 - 1.1
Porcelain (aluminous)	1.48 - 1.56
Metal Bonded Porcelain	1.91

Table 5.1: Representative values of fracture toughness of dental tissues and dental materials.

5.4 AIMS.

This study investigated the effect of secondary curing on the fracture properties of three composite resins for use in the inlay technique. These properties were measured both with and without secondary curing at 1 week, for two hybrid materials and one hydropneumatic curing microfilled material. From this it was hoped to establish whether the secondary curing process had an effect on the initial fracture toughness and inherent flaw size of the materials. Additional testing was carried out following ageing of the composites in water at 37°C for 2 months to assess whether any initial effect was maintained.

5.5 MATERIALS AND METHOD.

The three composite resin materials used in this study and are described in detail in Chapter 2. Batch numbers are listed in Table 5.2.

MATERIAL	TRADENAME	BATCH NO.	MANUFACTURER
HYBRID A	BRILLIANT	9203943	COLTENE AG ALTSTATTEN SWITZERLAND
HYBRID B	ESTILUX POSTERIOR CVS	93.12.31 28	KULZER GmbH BEREICH DENTAL, WERHEIM, GERMANY
MICROFIL	IVOCLAR INLAY/ONLAY	240087	VIVADENT DENTAL GmbH, ELLWAGEN, LEICHTENSTEIN

Table 5.2: Composite Resin Materials used.

Fracture toughness was determined using Single-edge-notch (SEN) specimens, with standard geometry conforming to the British standard BS.5447[214] (Figure 5.1). To minimise the volume of composite required for each specimen but ensure that plane strain conditions prevailed at the crack tip, a thickness B=3.00mm was used. From this all other dimensions were set. The critical value for width (B) has previously been calculated to be 0.1mm for heavily filled composites as used in this study[189]. Hence the values used in this study are an order of magnitude greater than the minimum value actually required.

The specimens were produced in a split PTFE-lined brass mould; constructed such that no force was required to remove the set specimen. The central 'crack' was produced by the insertion of a straight scalpel blade into a close fitting slot which extended half way down the height (W) of the specimen.(ie. to give a/W = 0.5W).

Forty samples of each of the hybrid materials were produced by light curing the materials in the mould with 60 second exposures from a Translux light (Kulzer) in a series of overlapping circles, to both upper and lower surfaces and to both side surfaces in turn. Twenty samples of each hybrid material were then subjected to a secondary curing procedure, using their respective curing 'oven'. Twenty samples of the microfilled material were produced: curing in the heat/pressure oven recommended for this material. Following curing, all samples were stored in water at 37°C until required for testing.

Pre-cracking of the samples as advocated by Ferracane *et al.*[202] was not included in the experimental protocol, as Lloyd[215] has suggested that although, in theory, it should be included to sharpen the moulded crack, it is "not necessary and in practice may introduce errors". In fact, in recent publications Ferracane and his coworkers have abandoned pre-cracking[100,102,213].

The fracture test was carried out on an Instron TTCM machine (Instron Ltd., High

Wycombe England), in three-point bending, at a crosshead speed of 0.1mm/sec. Following testing the dimensions B and W were measured with a micrometer and (W-a) with a travelling microscope. From these the fracture toughness K_{IC} was calculated using the formula below

$$K_{IC} = \frac{3PLY_1}{BW^{1.5}}$$

where P is the fracture load and L is the half loading span. The value of the factor Y_1, which depends upon the value of a/W is most easily obtained from standard tables.(e.g. Knott[216])

Values of fracture stress in flexion σ were taken from the results of Chapter 3 and the inherent flaw size a_0 was calculated from this data with the equation

$$a_0 = \frac{1}{\pi}\left(\frac{K_{IC}}{1.12\sigma}\right)^2$$

Mean values and standard deviations were calculated for all physical tests. The individual values were compared by ANOVA and where differences were detected, these were identified by the Students *t* test.

Selected fractured surfaces were examined by SEM to determine any differences in the nature of fracture between materials, curing regimes and time of storage post curing.

5.6 RESULTS.

The fracture toughness and inherent flaw size of the materials at one week and 2 months are shown in Table 5.3, and Figures 5.2 and 5.3.

For the one week samples there was no significant improvement in fracture toughness following secondary curing. Both hybrid materials, however, had significantly higher ($P < 0.001$) fracture toughness than the microfilled material. Storage of the materials for 2 months did not result in any statistically significant change in the fracture toughness of any of the materials tested.

At one week, the inherent flaw size of both hybrid materials was significantly decreased by postcuring ($p<0.05$). This difference was not altered by storage. The inherent flaw size of the microfilled material was constant during the testing period. However, at both measuring times it was significantly less than any of the light cured and post cured hybrid materials.

The SEM images of the fractured surfaces failed to show any differences in crack propagation between light and post cured materials nor were any differences observed between 1 week and 2 month samples. However, differences were noted between materials, with hybrid B showing evidence of fracture at the resin/filler interface resulting in debonding of filler particles (Figure 5.4,5.5). Contrastingly, in hybrid A crack propagation would appear to be entirely through the matrix, as no exposed filler particles were seen (Figure 5.6,5.7). In the microfilled material, a very flat fractured surface was observed, with no evidence of interfacial debonding of the prepolymerised resin fillers. Rather, the crack would appear to have propagated through any prepolymerised particles which may have been in its path (Figure 5.8,5.9).

MATERIAL	CURING REGIME	TIME	FRACTURE TOUGHNESS (K_{IC}) $MNm^{-1.5}$	INHERENT FLAW SIZE (a_o) microns
HYBRID A	LIGHT CURED	1 WEEK	1.78 (0.60)	53 (17.9)
	POST CURED		1.83 (0.38)	30 (6.2)
	LIGHT CURED	2 MONTHS	1.72 (0.13)	47 (3.5)
	POST CURED		1.84 (0.45)	31 (7.5)
HYBRID B	LIGHT CURED	1 WEEK	1.50 (0.17)	45 (4.1)
	POST CURED		1.70 (0.35)	27 (5.5)
	LIGHT CURED	2 MONTHS	1.32 (0.23)	33 (5.7)
	POST CURED		1.47 (0.30)	25 (5.0)
MICROFIL	HEAT AND PRESSURE CURED	1 WEEK	0.74 (0.10)	10 (1.4)
		2 MONTHS	0.67 (0.14)	9 (1.9)

Table 5.3: Fracture toughness (K_{IC}) and inherent flaw size (a_o) of materials. Standard deviations in ( ).

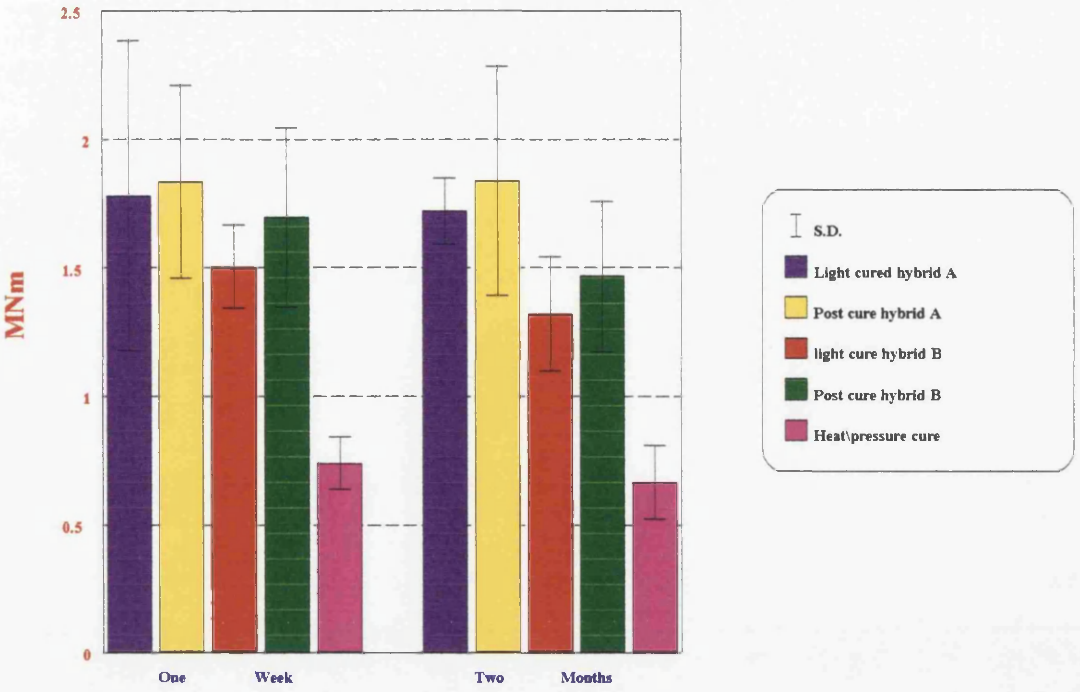


Figure 5.2: Fracture toughness (K_{IC}) of three composite resin inlay materials, with and without post curing, at one week and two months.

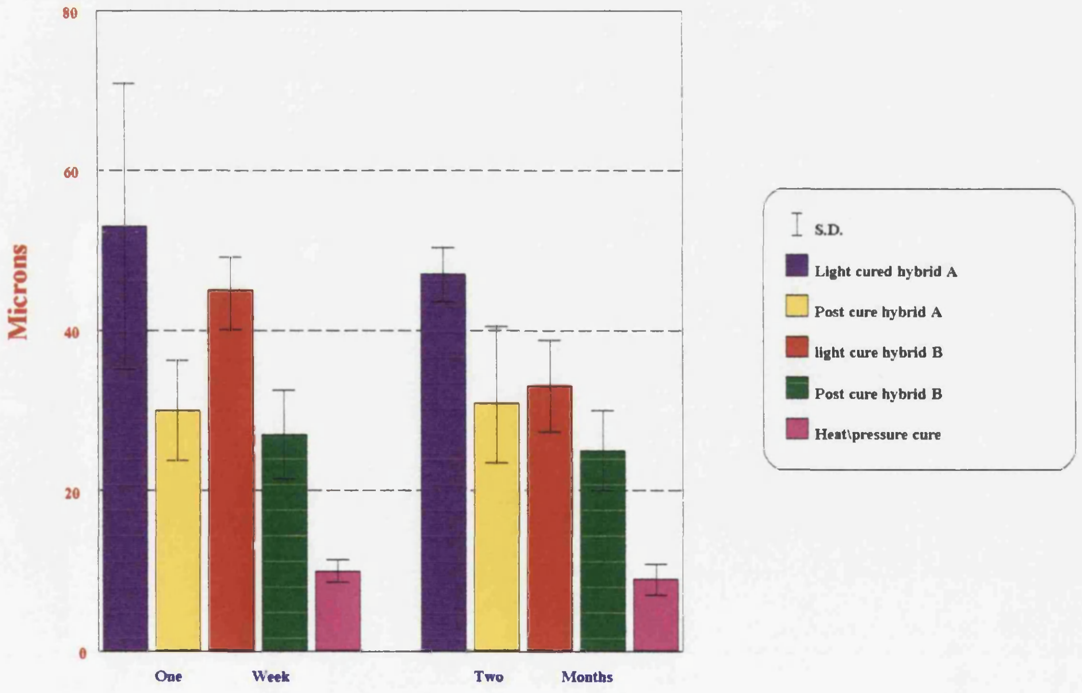


Figure 5.3: Inherent flaw size (a_0) of three composite resin inlay materials, with and without post curing, at one week and two months.

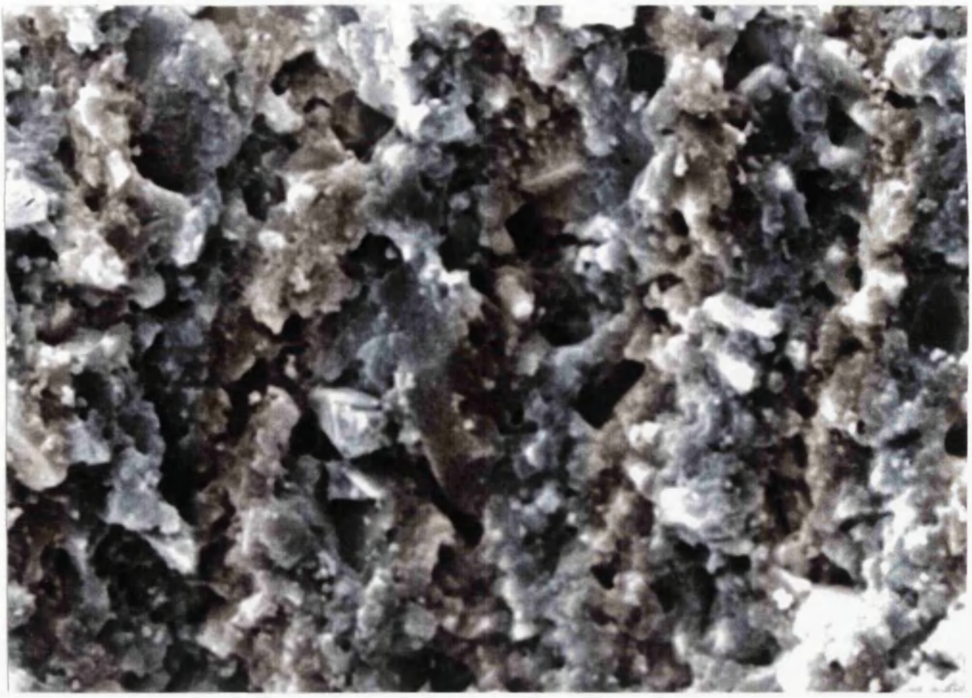


Figure 5.4 SEM image of fractured surface of hybrid B composite resin inlay material. Very rough surface with some exposure of filler particles obvious. Original magnification x500. Field width 200μm

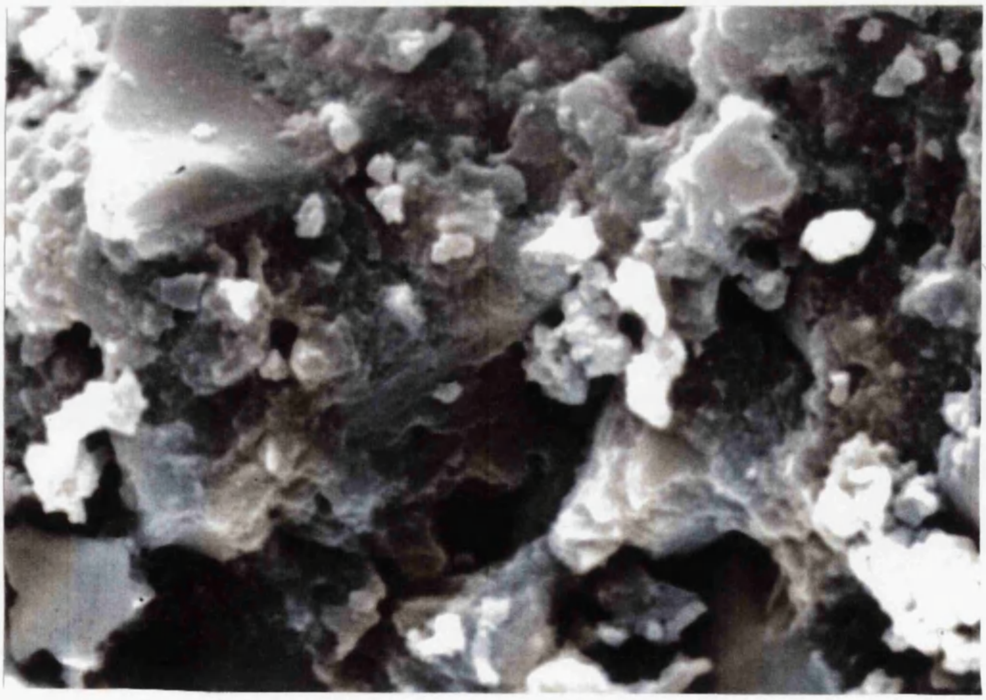


Figure 5.5 SEM image of fractured surface of hybrid B composite resin inlay material. A large filler particle can be seen exposed in the top left hand corner of the photograph. Original magnification x3500. Field width 30μm

Figure 5.6 SEM image of fractured surface of hybrid A composite resin inlay material. A considerably smoother surface than in hybrid B is evident. No obviously exposed filler present. Original magnification x500. Field width 200μm

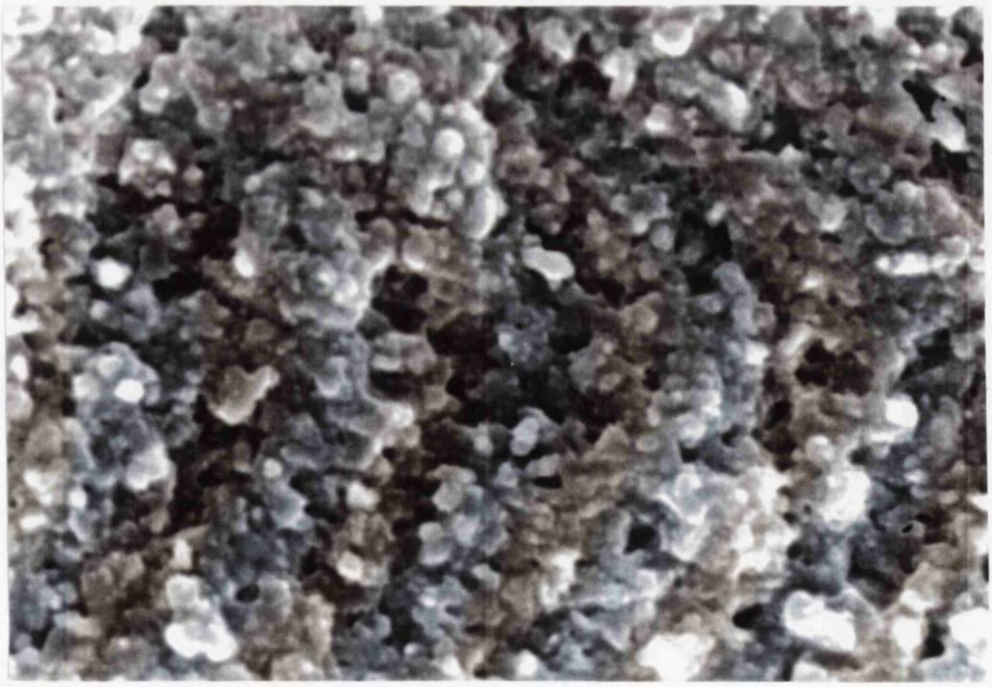


Figure 5.7 SEM image of fractured surface of hybrid A composite resin inlay material. Small filler particles present. However these are completely covered in resin, suggesting a plane of fracture through the matrix. Original magnification x3500. Field width 30μm

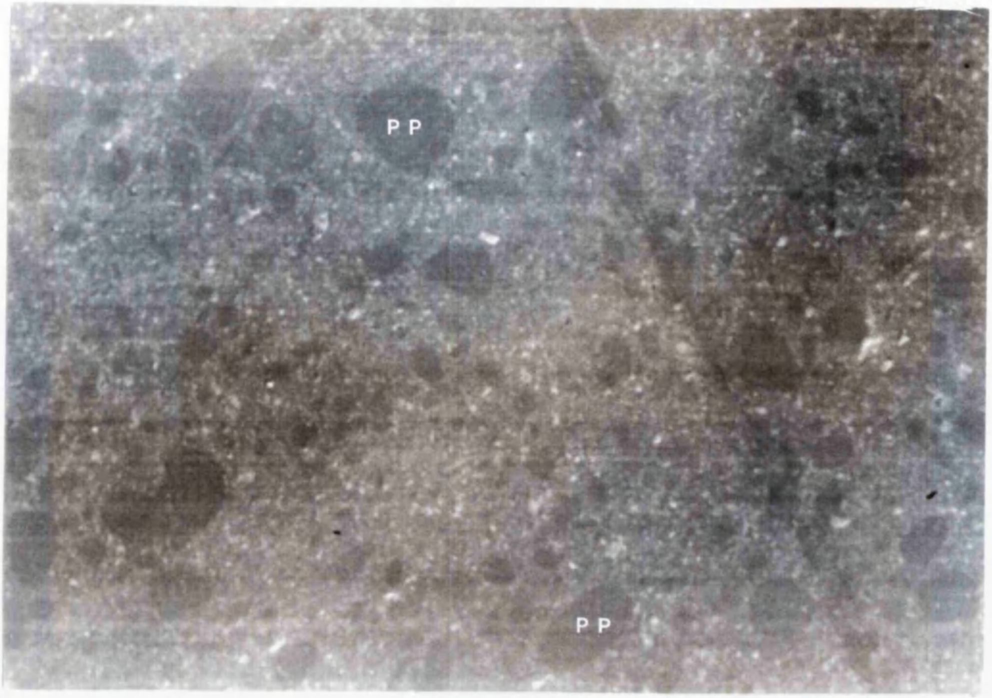


Figure 5.8: SEM image of fractured surface of microfilled composite resin inlay material. Very flat surface evident. Large prepolymerised particles (pp) are present. Original magnification x100. Field width 1000μm.

Figure 5.9: SEM image of fractured surface of microfilled composite resin inlay material. Very smooth surface with plane of fracture through the prepolymerised particles. Original magnification x500. Field width 200μm.

5.7 DISCUSSION.

5.7.1 Fracture Toughness of Light Cured Materials Following Post Curing.

The fracture toughness values obtained for the light cured composites used in this study are in agreement with previously published work examining similar, heavily filled, hybrid composite resin materials[94,100,176,190,201,202,209,217] (Table 5.1). However, no improvement in fracture toughness was recorded following the post curing of either of these materials in their respective heat\light ovens.

The proposed mechanism of action of post curing is that heating to around the glass transition temperature increases molecular mobility and encourages crosslinking of unreacted methacrylate groups. This increased degree of cure, following post curing, is confirmed in the following chapter with hybrid A undergoing a further 7% and hybrid B undergoing a further 12% conversion of methacrylate.

An increased degree of cure has been shown to result in an increase in flexural modulus of dimethacrylate resins[218] and composites[163] due to the denser polymer matrix. As fracture toughness is related to the square root of the product of fracture energy and elastic modulus, it might be expected that a higher modulus should result in a higher fracture toughness, as occurs with increasing filler fraction. However, as no increase in K_{IC} was noted in the composites studied (despite the increase in modulus observed in Chapter 3), some mechanism must be acting to decrease the fracture energy and, therefore, balance the increase in modulus.

It has been suggested that increased methacrylate conversion of composite resins, in addition to increasing the stiffness of the material, is also likely to result in a decrease in the potential for plastic deformation of the matrix, and, hence, its ability to dissipate energy at the crack tip[94]. This hypothesis is supported by the work of Lloyd and Ianetta[178] and Lloyd and Adamson[190]. In these studies the fracture toughness of composites was seen to fall following storage in water. This, it was suggested, was due to leaching out of plasticiser in the form of monomer. If, in the case of post curing, this monomer is used up during the additional polymerisation, then its potential plasticising action is lost, reducing the materials ability to absorb fracture stress. A further mechanism for reduction in ability to absorb fracture stress has been proposed by Ferracane *et al.*[202]. It is suggested that post curing at high temperatures produces significant tensile stresses within the composite, particularly at the resin/filler interface, where the differences in thermal expansion coefficient between matrix and filler are most important. This increased stress within the material would result in a decrease in the fracture stress, as the energy required to propagate the crack through such a stressed region would be lower than that through an unstressed region. This theory is supported by Truong *et al.*[219], who suggested that the fracture resistance of highly cured composites, as used in this study, may be reduced by high temperature curing due to internal stress formation. This theory is obviously diametrically opposed to De Gee *et al.*'s annealing hypothesis[130].

The fact that post curing does not result in any alteration in fracture toughness is supported by the work of Cook and Johansson[94]. Although it was noted, in their study, that post curing at elevated temperatures increased fracture toughness, this was only related to composites maintained at low temperatures following initial light curing. If the results are re-examined and comparison made between physiological temperatures of around 37°C and their highest post cure temperature of 100°C, very little change in fracture toughness is observed. Moreover, a recent study[103], which recorded the fracture toughness of the materials used in this study when subjected to similar post curing regimes exhibits very good correlation of post cure K_{IC} values and concluded that post-curing had a minimal effect.

The results of Ferracane and Codon[100], Ferracane and Marker[102], and Ferracane *et al.*[213] are rather more difficult to explain. However, it seems likely that the different materials and post curing regimes used may have resulted in the variations between these studies and between them and the present study.

5.7.2 The Fracture Toughness of a Heat\pressure Cured Composite.

The heat pressure cured material, used in this study was a microfilled composite resin. As microfilled materials have been shown to have lower values of K_{IC}[176,189,202,209,217], the finding that the fracture toughness of this material was around half of the hybrid materials was not unexpected. The small particle size of the filler, 0.04μm in this material, is less effective at crack pinning[101] and, consequently, less energy is required to propagate a crack and fracture toughness is low. The hydropneumatic curing regime used with this material does not produce a higher degree of cure than conventional light curing of related materials (Chapter 6). Nonetheless, it may have been expected that fracture toughness would increase due to the decrease in porosity noted when curing occurs under pressure[93]. However, Yoshimoto[220] has suggested that fracture toughness is unaffected by porosity, even when levels of up to 2.5% (volume) are deliberately introduced into a material.

The results for this material are at odds with those of Uctasli *et al.*[103]. There is no obvious reason for this discrepancy as both studies were carried out using the same materials and curing apparatus. Differences between individual curing ovens and batches of material are possible. However, these are unlikely to account for the two fold difference between the results of this and Uctasli's study.

5.7.3 The Effect of Storage on Fracture Toughness.

No change was noted in K_{IC} values over the two month storage period. This is in agreement with the results of the previous chapter, where no variation was noted, in the more conventional fracture stress parameters, in any of the tested materials, either with or without post curing during this period. Little variation in fracture toughness of conventionally cured composite materials, stored in water, for the times measured in this investigation, is in accordance with previous studies. Lloyd[101] and Lloyd and Ianetta[178] noted an increase in K_{IC} with storage in water for periods of 1 week; the initial testing period in this report. However, in a later study Lloyd and Adamson[190] compared K_{IC} values of five composites over a 3 month period. No statistical analysis was reported in this paper. Nonetheless, it would appear that for periods between 1 week and 3 months no difference was recorded in fracture toughness for four of the five materials tested. This finding was supported by the work of Pilliar *et al.*[177], who found no significant differences in four composites aged for up to 28 days in water, and Ferracane and Marker[102], who found no significant difference in four composites stored for 14 months in water at 37°C.

Aging of post cured composites, in water, has been reported by Ferracane and Marker[102], who suggested that by 14 months a significant increase in K_{IC} could be expected. However, at two months no differences have been observed. This is in agreement with the present study. However, it is the opposite result to that recorded by Ferracane *et al.*[213] in a subsequent study. In this abstract a decrease, in fracture toughness, over thirty days, followed by a constant value for the remaining period (180 days) was reported. The different materials and curing regimes used must be the important factors in explaining this apparent contradiction, as all other factors were constant. Consequently, some caution must be used when applying these findings to the present study, where different materials and curing regimes were used. The report of Uctasli *et al.*[103] is, therefore, the most comparable to the present report. In this the same materials, and similar post curing regimes were used, results are therefore directly comparable. No change was noted in K_{IC} of any of the materials tested over a three month period of storage in water at 37°C, in agreement with the present study

5.7.4 Inherent Flaw Size.

The values for inherent flaw size of the light cured hybrid materials are within the range previously recorded by Lloyd and Adamson[190] for similar materials. The inherent flaw size of both hybrid materials is reduced by post curing with heat and light. This is consistent with the work of Cook and Johansson[94]. It may be that the increased aliphatic C=C conversion following post curing in these materials (Chapter 6) is the important factor in producing this increase, as it has been suggested that it is the crosslinking density of the matrix which is the important in factor in determining the inherent flaw size in methacrylates[51].

Previous studies have shown the inherent flaw size of microfilled materials to be lower than for materials with larger filler particles[176,217]. Therefore the lower inherent flaw size of the microfilled materials was not unexpected.

Only light cured, hybrid B showed any change in inherent flaw size following storage. There is no obvious explanation for this observation. It may be that some degree of post curing occurs over the two month period. However, this seems unlikely as no other properties are altered during this time. FTIR analysis of this particular composite, following storage, would be required to assess any possible changes in methacrylate conversion accurately. This was beyond the scope of the present study.

5.7.5 Fractography.

SEM analysis of the fractured surfaces of materials provides information on the mode of crack propagation through the material.

In hybrid A crack propagation would appear to be entirely through the matrix. This is indicative of good bonding between resin matrix and filler particles. In hybrid B, however, there was evidence of some breakdown of the resin\filler bond during fracture as "clean" filler particles were visible. No obvious changes were observed following post curing or storage of the hybrid materials.

The fractography of the microfilled material was more interesting. A very flat surface was produced with this material. There was no evidence of the prepolymerised filler being exposed or plucked out, as has been described previously in light cured microfilled materials[212]. This is indicative of either improved matrix/filler adhesion or, alternatively production of a matrix with improved fracture properties; very similar to those of the prepolymerised filler particles. Either mechanism is possible. Heating of the composite during curing will increase molecular mobility, and possibly, allow more diffusion of matrix polymer into the filler improving adhesion. Furthermore, heating will encourage crosslinking, possibly resulting in a matrix with a more similar degree of cure to the prepolymerised filler particles and, consequently, similar fracture properties.

5.8 Conclusions.

Fracture toughness of two light cured composite resin inlay materials was not improved by post curing; the properties remaining constant for the period tested. The inherent flaw size of both materials was reduced by post curing. However, whether or not this has any clinical implication is open to speculation as both materials already exhibited low inherent flaw sizes[176] following light curing. The fracture toughness of the microfilled heat\pressure cured inlay material was significantly lower than that of the hybrid materials, which would suggest that it may be more prone to chipping and fracture. Furthermore, the K_{IC} value recorded for the microfil was comparable with previously published data on light cured microfilled materials.

This study suggests that post curing either with heat and light or a heat\pressure technique has little part to play in improving the fractural properties of posterior composites.

Chapter 6: The Influence of Post curing on the Degree of Conversion of Three Composite Resin Inlay Materials.

6.1 INTRODUCTION.

6.1.1 Degree of Cure in Dental Composites.

It is a generally agreed concept that resin-based restorative materials do not reach an optimum cure during polymerisation under normal clinical conditions[13]. Moreover, it has been suggested that composite resins undergo a progressive cure effect with time resulting in improved physical properties[221]. Furthermore, several authors have shown[18-20,95-99] that heating of composite resins, following their initial conventional cure, results in an improvement in physical properties, (see Chapter 3 for further details). The proposed mechanism of action of the post curing process is that heating allows movement of molecules within the polymer facilitating a progressive crosslinking of unreacted methacrylate units[104]. The potential for post curing to increase methacrylate conversion is confirmed by the work of Cook and Johannson[94]. However, it has been suggested that an annealing process may take place with relaxation of internal stresses which form during polymerisation[13]. However, evidence for this process is rather more scant and, while it may be of some importance, it seems likely that the major influence on the properties of post cured composite resins is the increased methacrylate conversion[100].

The increased conversion, following post curing may improve physical properties, but, it has been suggested, may also prejudice the potential bond between inlay and composite resin cement[20,89,92], as this bond is reliant on chemical bonding of methacrylate groups in inlay and cement.

The degree of conversion, following post-curing, of composite resin inlay materials is, therefore, of considerable importance to the potential clinical performance of such inlays. Recently Ferracane *et al.*[100,102] has reported increases of around 10% in methacrylate conversion following several different types of post curing regimes. These two studies were, however, carried out mainly using specially made experimental composites and may not correspond to values which would occur for commercially available composites. In fact, of the commercial composites used in the latter study, one microfilled material showed an increase in degree of cure of almost 20%. However, none of the materials studied were designed to be used with a post curing technique, and to date no study has been reported (in English) to assess the post curing potential of specific composite resin inlay materials using their various curing regimes.

Investigations of multifunctional methacrylate polymerisation have been conducted for more than thirty years. In the early stages of polymerisation titration of polymer solution was used for determination of residual monomer[222]. Calorimetric and thermometric analysis have been used for many years as have chromatographic techniques, to study the residual monomer after polymerisation[223-225]. The advent of Fourier Transform Infrared Spectroscopy has brought about renewed interest in infrared spectroscopy as a characterisation technique and has proved very popular in dental research as a method of determining the degree of conversion of various dental restorative materials[10,17,77,78,94,100,102,158-160,226,227].

6.1.2 Theory of Infra Red Spectroscopy.

When infrared light is passed through an organic compound some of the frequencies of light are absorbed while others pass through without change. The frequencies of light absorbed are related to the chemical groups which occur within the compound being tested. Different chemical groups absorb light energy at different wavelengths, this light energy corresponding to that required to cause vibration within that chemical group. The vibration can occur in the form of stretching or bending of the group, each requiring energy at a particular wavelength and each being characteristic of a particular chemical group.

If the infra red absorbance of a particular molecule is plotted it is possible to identify the chemical groups which make up the molecule as each will absorb light at its characteristic wavelength resulting in an absorption band in the infrared spectrum. For example C=C bonds characteristically absorb light at a wavenumber of 1638cm^{-1}, consequently any spectrum of a molecule containing C=C will have an absorption band in this area. Wavenumber of peak absorbance for other chemical groups is readily available from tables.

There are two basic types of infrared spectrometer, characterised by the manner in which the infrared frequencies are handled. The first type has a long history, and in it the infrared light is separated into its individual frequencies by dispersion, using a diffraction grating. In the second type, however, the infrared frequencies are allowed to interact to produce an interference pattern, and this pattern is subsequently analyzed mathematically, using Fourier Transforms, to determine the individual frequencies and their intensities. Details of mode of action of the two types of spectrometer is covered adequately by Kemp[228].

Fourier Transform Infrared Spectroscopy (FTIR) is the type which has been used extensively in dental research. This method has several advantages over dispersive infrared spectroscopy. It is much quicker, (scans can be recorded in very short times, even a few milliseconds), spectra can be obtained from picogram quantities of a sample, spectral summation and subtraction are possible with these machines, and, finally through coupling of a suitable optical microscope to the spectrometer, sample size of only a few micrometers can be examined. These advantages are not, however obtained without significantly greater cost.

It has been suggested that multiple internal reflective FTIR is the most appropriate technique for studying dental composite resins[227] as the filler particles interfere with transmission. However, several authors have carried out successful studies using transmission techniques[77,78,94,100,102,158,226] the results of which are very similar to those using the more complicated reflectance method. This would suggest that the difficulties of producing good transmission spectra of composite resin materials are more perceived than actual.

6.2 AIM.

Through the use of FTIR Spectroscopy this study investigated the potential of post curing with heat and light to increase the degree of methacrylate conversion in two composite resin materials for use in the inlay technique. Secondly, the degree of cure of a third inlay material cured using heat and pressure was examined as it has been suggested that this method of curing may result in a superior degree of conversion[126].

6.3 MATERIALS AND METHOD.

6.3.1 Introduction

The three dental composite resin materials used in this investigation are listed in Table 6.1 and described in more detail in Chapter 2. A FTIR spectrometer was used to determine the degree of conversion of these systems both with and without post curing.

MATERIAL	HYBRID A	HYBRID B	MICROFIL
TRADE NAME	BRILLIANT DENTINE	ESTILUX POSTERIOR	ISOSIT INLAY/ONLAY
RESIN TYPE	Bis-GMA/TEGMA	Bis-GMA/TEGMA	UEDMA
CURING METHOD	Light or Heat/Light	Light or Heat/Light	Heat/Pressure
BATCH NO.	101290-51	93-12-31 28	240087

Table 6.1: Composite resins used.

6.3.2 Composite Sample Preparation.

Samples of the unpolymerised pastes were produced by placing approximately 1x2mm of the resin paste on a KBr disc and smearing it to a very thin layer of between 25μm and 75μm. Spectra of the two light curing materials were run immediately following sample preparation to prevent daylight causing unwanted cure of these materials. Although the microfilled material does not light cure and, consequently, there is no danger of premature setting of the material; spectra of these samples were also run immediately following preparation.

Samples of the polymerised and post cured materials were produced by placing the composite resin (approximately 2mm in diameter) between two pieces of mylar strip. This "sandwich" was subsequently placed between two sheets of perspex and a load of 5kg was applied to press the perspex together. After 2 minutes the load and perspex were removed and polymer samples of approximately 10mm in diameter and 25μm to 75μm in thickness were produced. Ten samples of each of the hybrid materials were light cured for 60 seconds. Subsequently five samples of each material were subjected to the post curing regime appropriate for that material; hybrid A in the D.I.500 oven and hybrid B in the Unilux A.C.

Five samples of the microfilled material were cured in the Ivomat heat/pressure oven, as this material does not light cure. Following curing all samples were removed from between the mylar strips and stored dry at 37°C for 24 hours prior to testing. The 24 hour storage prior to testing was important as it allowed the light-cure-only samples to reach their maximum cure by light alone. Had testing been carried out prior to this a significantly lower degree of conversion could have been expected in these samples[159].

6.3.3 Spectroscopy.

Transmission spectra of the three composite resin materials were recorded using a Bomem P.A.3 FTIR spectrometer (Bomem Inc., 625, rue Marais, Vanier, Quebec, Canada). The spectrometer was used with a potassium bromide beam splitter and Mercury\Cadmium|telluride detector operating at 77°K. Furthermore, the spectrometer used in this study was connected to a I.R. Plan light microscope (Spectra Tech Inc., Stanford, Connecticut, USA) which allowed accurate location of the samples, to be scanned, within the IR beam, ie. away from flaws and voids which may have occurred in the composite samples during fabrication. The spectra were recorded in transmission and converted to absorption spectra by the microprocessor. One hundred and twenty eight scans were made of each sample and an average of these scans was produced. This number of scans resulted in a very high signal to noise ratio producing very "clean" easy to interpret spectra with a spectral resolution of 2 wavenumbers. Spectra were stored on disc prior to further interpretation.

6.3.4 Analysis of Spectra

The analysis of degree of conversion of Bis-GMA containing composite materials is based upon the calculation of the ratio of the aliphatic C=C absorption at 1638cm^{-1} to that of the aromatic C=C absorption at 1609cm^{-1}. The aromatic group does not take part in the polymerisation reaction and can thus act as an internal standard for subsequent calculations of degree of polymerisation. Calculation and drawing of baselines from which to measure the C=C peaks has generally been carried out using a standard baseline technique as described by Heigl *et al.*[229]. However, not all researchers draw baselines in the same position, resulting in possible inaccuracies and differences between laboratories. This potential problem was addressed by Rueggeberg *et al.*[230] who suggested that a baseline drawn between the trough before 1638cm^{-1} and that after 1609cm^{-1} demonstrated the best correlation between C=C concentration and C=C IR absorption. Although Rueggeberg's study was conducted using attenuated total reflectance FTIR, it is suggested that results for transmission FTIR are likely to be similar, and, consequently, this method for baseline placement was adopted during the present study.

Using the spectrometer's microprocessor it was possible to recall spectra and superimpose them upon each other. By superimposing the spectra of uncured, cured and post cured materials it was possible to ensure that the baseline and recorded peak values were accurate for each sample.

A record of the aromatic and aliphatic absorption values for the two hybrid materials was made and calculation of the degree of cure of these two composite resins was carried out using the equation described by Ferracane and Greener[226].

$$\frac{[\text{absorption(aliphatic C=C)}]/\text{absorption(aromatic C=C)}]\text{polymer}}{[\text{absorption(aliphatic C=C)}]/\text{absorption(aromatic C=C)}]\text{monomer}} \times 100$$

The degree of conversion is calculated by subtracting the residual percentage of aliphatic C=C from 100%.

The microfilled heat/pressure cured material had a completely different chemical composition from the two hybrid materials. It was mainly composed of a urethane dimethacrylate resin the chemical name of which is 1,6 bis(methacryloyloxy-2-ethoxycarbonylamino-2,4,4,-trimethylhexane. As can be seen, this material contains no aromatic groups and consequently, the normal internal standard cannot be used. Cook and Johannson[94] suggested using the carbonyl adsorption at 1720cm^{-1} as an internal reference when similar problems were encountered during testing of Occlusin (another urethane dimethacrylate-containing composite). However in the composite tested this group produced a very strong absorption band which generally went off scale. An alternative is to use the N-H group which produced a strong absorption band in the 3380cm^{-1} region. As this group is locked within the backbone of the monomer molecule it will not take any part in the polymerisation reaction and can be used as an alternative internal standard. Subsequent calculation of degree of cure was carried out using the same formula, with N-H absorption pre and post polymerisation being substituted for the aromatic C=C.

Student's t-tests were used to compare the degree of cure before and after post curing and between materials (see Discussion).

6.4 RESULTS.

Spectra of the three materials tested are shown in Figures 6.1-6.3. These Figures illustrate the decrease in aliphatic C=C following light and post curing.

The degree of cure of the three inlay materials with and without post curing is shown in Table 6.2 and Figure 6.4. There was no significant difference between the two hybrid materials following light curing $p<0.001$. Following post curing, both hybrid materials exhibit significant increases in degree of cure $p<0.001$. Hybrid A increases by 7% while hybrid B increases by 12%. The microfilled material, which does not light cure, attained a conversion of 63% following heat\pressure curing.

None of the materials attain 70% conversion, even following post curing, although hybrid B exhibited significantly greater cure than the other two materials ($p<0.05$).

	MATERIAL	**HYBRID A**	**HYBRID B**	**MICROFIL**
CURING PROCEDURE	Light Cure only	55±4%	55±3%	N/A
	Light\Heat Post cure	62±3%	67±2%	N/A
	Heat\Pressure cure only	N/A	N/A	63±3%

Table 6.2: Mean degree of conversion of methacrylate ± standard deviation (%) following curing by light, light and heat and light and pressure.

6.5 DISCUSSION.

Various authors[159,227] have produced calibration curves relating concentration of aromatic C=C bonds in TEGDMA/Bis-GMA and TEGDMA/Bisphenol-A solutions to infrared absorption. They have stated that these are essentially similar and that conversion values obtained from them will yield similar values. From this it is extrapolated that these calibrations are relevant to composites which have similar resin formulations. It is further assumed that there is a linear relationship between C=C concentration and IR absorption. However, the work of Rueggeberg *et al.*[230] calls these assumptions into question. It is suggested that TEGMA/Bisphenol-A produce quite different calibration curves from TEGMA/Bis-GMA and that the former is unsuitable for calibration purposes. Furthermore, the non-linearity of the calibration curve is emphasised when high aliphatic to aromatic ratios are obtained, although

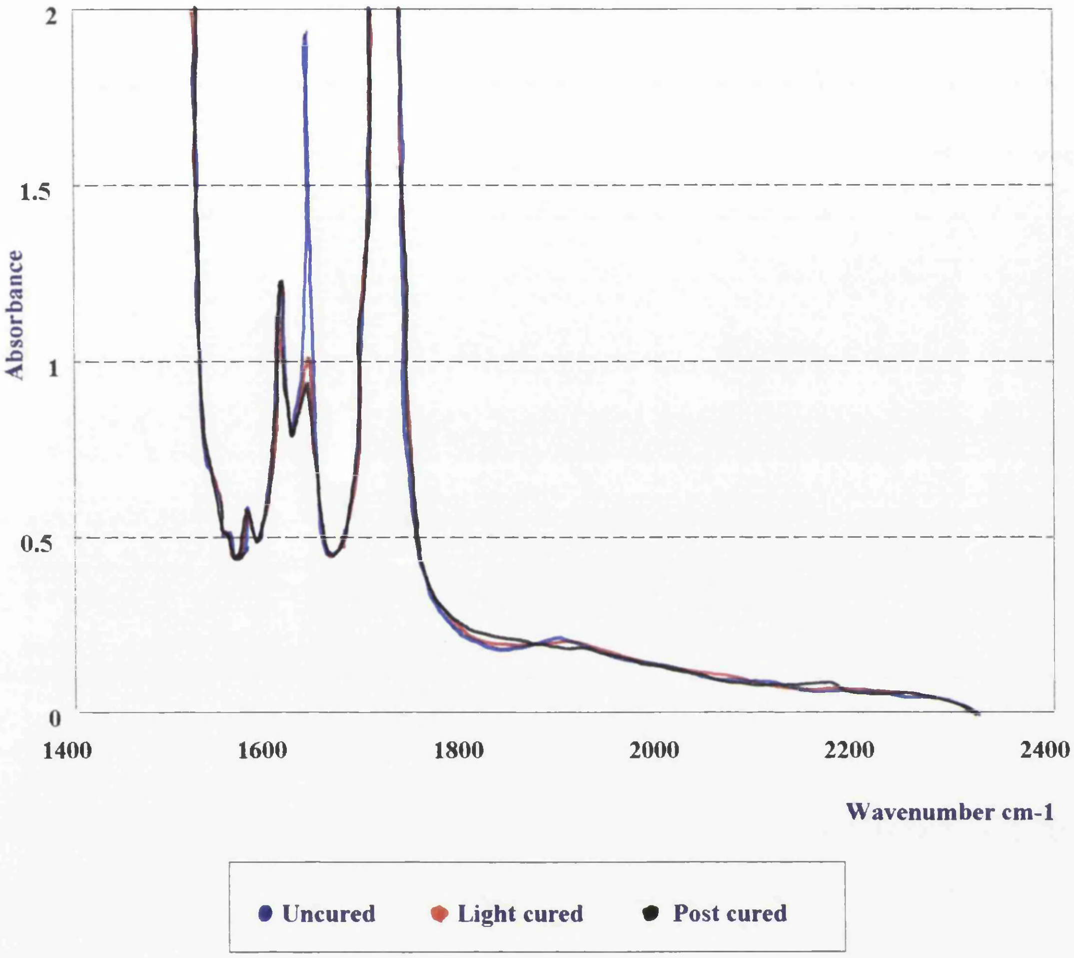


Figure 6.1: FTIR absorption spectra of hybrid A. Spectra of uncured, light cured and post cured materials are superimposed about the aromatic C=C peak at 1608cm^{-1} to show the diminution of the aliphatic C=C group, at 1638cm^{-1}, during curing.

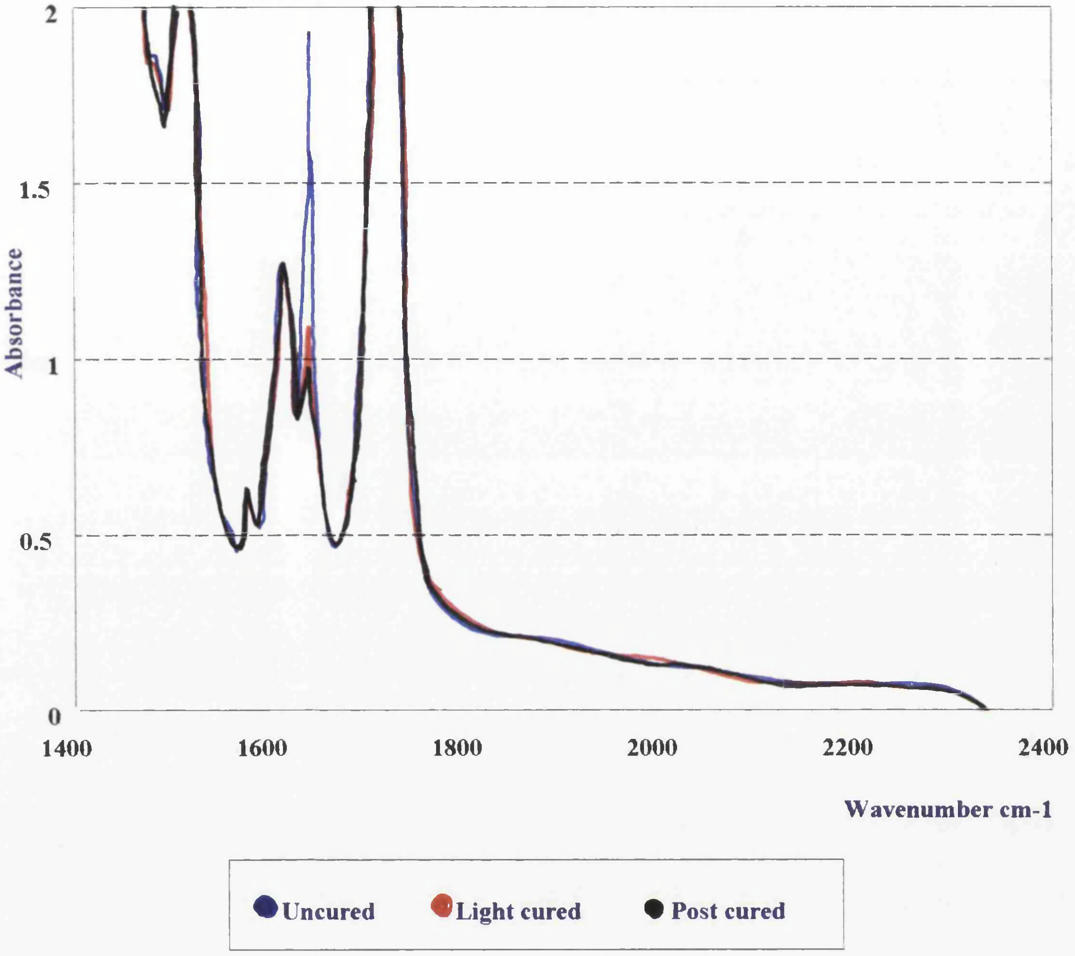


Figure 6.2: FTIR absorption spectra of hybrid B. Spectra of uncured, light cured and post cured materials are superimposed about the aromatic C=C peak at 1608cm^{-1} to show the diminution of the aliphatic C=C group, at 1638cm^{-1}, during curing.

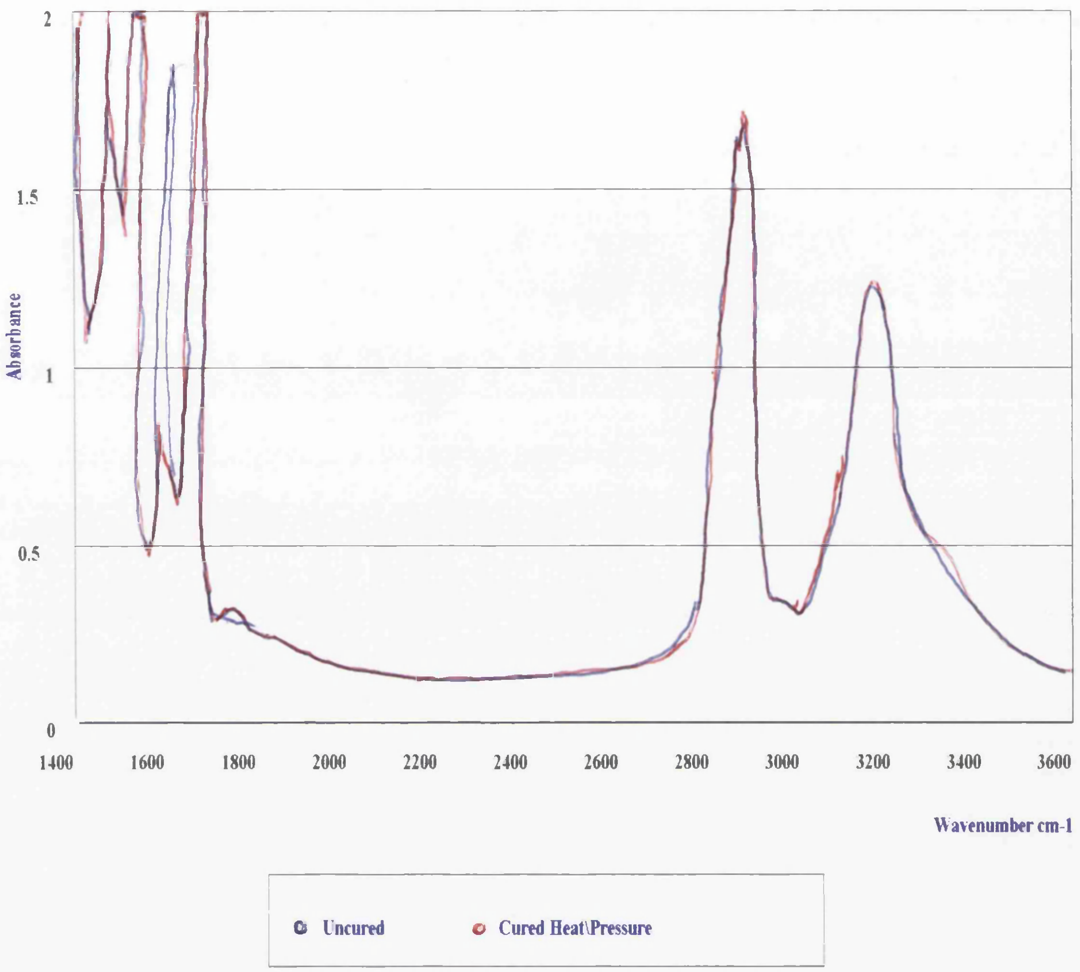


Figure 6.3:FTIR absorption spectra of the microfilled material. Spectra of uncured and heat\pressure cure materials are superimposed about the N-H peak at 3380cm-1 to show the diminution of the aliphatic C=C group, at 1638cm-1, during curing.

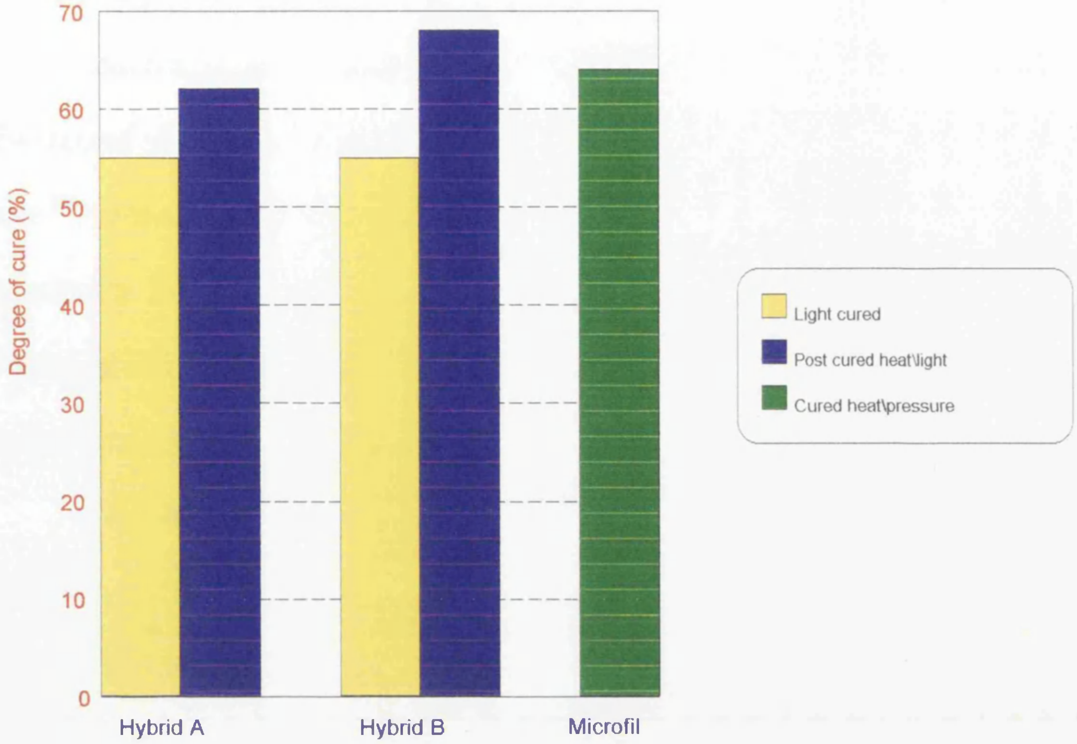


Figure 6.4: Mean methacrylate conversion (%) of three composite resin inlay materials following light curing, post curing with heat and light and curing using a heat\pressure technique.

Vankerckhoven *et al.* suggest that this need not be a problem[159].

Ideally a calibration curve should be produced for each composite investigated, using the uncured resin present in each material, hydrogenated, as described by Vankerckhoven *et al.*[159], up to an aliphatic to aromatic ratio of around 6:1[230]. This curve should then be used to calculate the mole ratio's of C=C bonds and conversion calculated thereafter. This procedure was, however, beyond the scope of the present experiment and the previous calibration curves were accepted and linearity assumed for all three materials.

Accepting these limitations, the results obtained for the two light cured materials (55%) are within the range of those obtained in earlier studies. In fact Chung and Greener[77] reported a conversion of 52.8% and Ruyter and Oysead[10] reported a conversion of 55% for Estilux Posterior (hybrid B in this study), suggesting extremely good correlation with previous studies.

Light polymerisation does not result in uniform cure throughout the material. Rather, cross-linked microgels are formed, these microgels having a higher conversion than the surrounding matrix, which contains unreacted monomer. Dynamic Mechanical Thermal Analysis is indicative of this situation with two thermal transitions being present in light cure only materials heating[104]. Heating of a light cured composite results in increased mobility of monomer and probably greater flexibility of polymer chains in the network. This, in turn, allows further reaction of monomer and polymer resulting in the observed single peak when Dynamic Mechanical Thermal Analysis is carried out on post cured material. This single peak is indicative of homogenisation of cure between the matrix and the microgels. It has been suggested that an increase in conversion of between 5% and 15%[231] is responsible for this change. Results in this range have been recorded by Ferracane and Codon[100] and by Cook and Johannson[94].

However, in neither of these studies were inlay composites used, nor were commercial heat\light ovens. It was therefore disappointing to find that even when a specialised inlay system was used increases in conversion of only 7% for hybrid A and 12% for hybrid B were obtained. However, as it seems likely from the results of Chapter 3, that the light component plays little part in the post curing process. It is therefore not unexpected that Ferracane's post curing regime of 120°C for 10 minutes, which closely approximates that of the D.I.500 oven, used with hybrid A, produces a similar result.

The use of very thin composite resin specimens could be responsible for a higher degree of cure, in the light cure only specimens, than might occur in thicker composite resin samples, more akin to inlays. Therefore, there is a possibility that an inlay may benefit more from secondary curing than the samples used. Nonetheless the ultimate degree of conversion following secondary curing would be no greater than recorded for a thin section.

A conversion of only 63% for the heat pressure cured material is surprising as Ruyter[126] suggests that a conversion of over 90% may be obtained with such a curing regime. However, no details of the experimental method are included in this paper. It may be that the calibration curve for this material is markedly different from a Bis-GMA based material leading to the assumptions made earlier being invalid. However, previous results from light cured urethane dimethacrylate containing composites do not differ markedly from Bis-GMA based materials[77]. It seems likely that the post curing process in urethane based materials is similar to that in Bis-GMA based materials, i.e. heating to around the glass transition temperature allows increased molecular movements within the polymer and subsequent increased crosslinking. The temperature reached in the Ivomat is the same as that in the D.I.500 oven and duration of cycle is similar (Chapter 2). It could, therefore, be hypothesised that the potential for increased molecular movement and hence crosslinking is comparable in both regimes. The pressure present during the microfilled materials curing cycle may be responsible for elimination of voids and porosities within the material[93], resulting in better physical properties (Chapter 3). However, its potential to improve methacrylate conversion is open to speculation. Moreover, a conversion of over 90% as suggested by Ruyter[126] is chemically very unlikely. Due to the spatial arrangement of molecules within the polymer it is difficult to imagine that over 90% of the available methacrylate bonds could be close enough to each other to react irrespective of the conditions under which polymerisation took place. A maximum potential conversion of around 80% for any dimethacrylate polymer is altogether more likely, and, to date, no other reports of methacrylate conversion above 80% have been published.

The fact that in all three materials investigated over 30% of the methacrylate remains unreacted suggests that a significant number of potential methacrylate bonding sites will remain on the surface of all three inlay types. Consequently, although it has been suggested that there is little potential for bonding of composite to inlay surface there would seem to be at least some potential for chemical bonding to take place in all three inlay types. This hypothesis is investigated further in Chapters 7 and 8.

The findings of this study support the concept that post curing with heat and light significantly increase the degree of methacrylate conversion of composite resins for use in the inlay technique. This increase in conversion may be responsible, at least in part, for the increase in physical properties recorded in Chapter 3. Curing of a composite with heat and pressure did not result in a increased degree of conversion relative to the heat\light method.

Chapter 7: The Plane of Fracture and Shear Bond Strength of Composite Resin Inlays.

7.1 INTRODUCTION.

Composite resin inlays are cemented into tooth cavities using a composite resin luting cement of very similar composition to the inlay material itself (See Chapter 2). As this is a composite to composite bond the potential exists for chemical bonding between methacrylate radicals on both the inlay surface and in the composite cement. It has been suggested by several authors[20,88,89,92] that, due to the increased degree of cure of composite resin inlays, following the post curing procedure (see Chapter 6), there are too few C=C bonds remaining to allow good chemical union, and that a poor bond is to be expected. This suggestion even prompted one manufacturer to produce a bond enhancing agent to increase the potential for bonding of the inlay to the cement (Special Bond, Vivadent Dental GmbH.). It has also led at least one author to investigate methods of treating inlay surfaces to improve bond strength[123]. However, treatment of a hybrid inlay surface with hydrofluoric acid, to etch the glass filler and produce a retentive surface, was significantly less effective than merely roughening the surface with a sandblaster.

The problem of adhesion, however, may not be common to all inlay types. Ruyter[126], for instance, reported that most problems of this nature occur only with the heat\pressure cured inlay type, while Ariyartanam *et al.*[124] have previously reported a good bond strength in both shear and flexural testing, to a heat\light cured system. The possibility of differences between systems is also supported by the work of Tam and McComb[127] and Gregory *et al.*[97], both of whom observed lower bond strengths to heat\pressure cured composites compared with heat\light post cured materials.

7.2 AIMS.

This study measured the shear bond strength of three commercially available composite resin inlay systems when bonded to enamel using their respective composite resin cement. Furthermore, the effect of unfilled resin and, where appropriate, bond enhancers was studied. Finally, the plane of fracture of these restorations was recorded in an attempt to discover if the inlay\cement bond was the weak link.

7.3 MATERIALS AND METHODS.

7.3.1 Enamel Preparation.

Bovine incisors were used in this study as they are readily available and have a large surface area for bonding[231]. Each crown was embedded in autopolymerizing polymethylmethacrylate resin in an aluminium square such that between 3 and 5 mm of the labial surface protruded. This surface was ground flat using P300 and P600 grade silicon carbide paper. These samples were stored in water at 5°C until required.

7.3.2 Inlay Fabrication.

The composite resins, composite resin cements and unfilled resin bonding agents are described in detail in Chapter 2, batch numbers are listed in Table 7.1.

Composite inlays, with a surface area of 25 mm^2 and 1 mm thick, were prepared using a specially made perspex mould which was placed on a base of dental stone (Jade Stone). Both the mould and the stone were coated with the appropriate separating medium. The composite was packed into the mould and a glass slide placed onto the surface of the composite. The light curing materials, hybrids A and B were cured for 60 seconds with a Translux (Kulzer) curing light. After removal from the mould, the flash was removed from the samples. They were then subjected to the appropriate post curing process; hybrid A samples in the D.I.500 oven and hybrid B samples in the Unilux A.C. The Ivoclar samples were cured in the mould entirely in the Ivomat heat\pressure oven, as they do not light cure.

On removal from the oven, the fitting surface of the Ivoclar samples was sandblasted with 50μm Al_2O_3. No surface treatments were carried out on the other samples prior to bonding. At this stage all inlay samples were stored, dry, for 1 week prior to bonding.

MATERIAL	TRADENAME	BATCH NO.	MANUFACTURER
HYBRID A	BRILLIANT	210590-02	COLTENE AG ALTSTATTEN SWITZERLAND
HYBRID CEMENT A	DUO CEMENT	080890-26	
UNFILLED RESIN A	DUO BOND	9112437	
HYBRID B	ESTILUX POSTERIOR CVS	30/6/93 38	KULZER GmbH BEREICH DENTAL, WERHEIM, GERMANY
HYBRID CEMENT B	ADHESIVE CEMENT	92.02 026	
UNFILLED RESIN B	ADHESIVE BOND	93.06 706	
MICROFIL	IVOCLAR INLAY/ONLAY	240087	VIVADENT DENTAL GmbH, ELLWAGEN, LEICHTENSTEIN
MICROFILLED CEMENT	DUAL CEMENT	360324 (BASE) 360326(CAT)	
UNFILLED RESIN C	HELIOBOND	240789	
BOND ENHANCER	SPECIAL BOND	240327	

Table 7.1: Composite resin inlay materials.

7.3.3 Bonding Procedure.

The enamel was etched for 60s with phosphoric acid gel, washed for 30s and then dried with oil free air for 10s.

Immediately prior to bonding, all samples were cleaned with phosphoric acid gel for

30 seconds, to remove finger grease, washed with water for 30 seconds and then dried.

Ten inlays were bonded for each of the following groups.

1) Hybrid A inlays bonded using Dual cure hybrid cement A.

2) Hybrid A inlays bonded using Dual cure hybrid cement A and unfilled resin A.

3) Hybrid B inlays bonded using Dual cure hybrid cement B.

4) Hybrid B inlays bonded using Dual cure hybrid cement B and unfilled resin B.

5) Microfilled inlays bonded using Dual cure microfilled cement.

6) Microfilled inlays bonded using Dual cure microfilled cement and unfilled resin C.

7) Microfilled inlays bonded using Dual cure microfilled cement, unfilled resin C and bond enhancer (Special Bond, Ivoclar/Vivadent FL9494, Schaan, Liechtenstein) dried.

8) Microfilled inlays bonded using Dual cured microfilled cement, unfilled resin C and bond enhancer wet.

All materials were used as recommended by the manufacturers. In all cases the bonding agent/resin was applied to both the enamel and inlay surfaces and cured for 60 seconds using a Translux light. The bond enhancer (Specialbond II) was applied to the Ivoclar inlay fitting surface only. Some confusion arose during the marketing of the bond enhancer. Initial supplies suggested drying the surface prior to applying the unfilled resin. However, later instructions were to leave the surface wet with bond enhancer while applying the unfilled resin. Both methods were, consequently, tried in this study. The bonding resin was applied and cured for 60 seconds.

Although all the cements and some of the unfilled resins are dual curing, it has been shown by Hasegawa *et al.*[148] that dual curing materials obtain better hardness properties when they are light cured (this improvement in properties was confirmed in Chapter 4).

Consequently, every sample was cured for 2 minutes with a Translux light. Two minutes was chosen as the curing time, as there is decreased light penetration through these materials[147] resulting in a need for increased exposure time (Chapter 4).

During bonding a constant force of 10N was applied to each sample. Following bonding the excess resin was removed from around the inlay samples and they were stored in water at 37°C for one week. Prior to testing the samples were thermocycled between 5°, 37° and 55°C for 500 cycles with a dwell time of ten seconds.

7.3.4 Testing Procedure.

Testing was carried out using a specially constructed jig mounted in a Nene testing machine (Figure 7.1). A crosshead speed of 0.5mm\min was used. Shear bond strengths were recorded and debonded specimens examined under a dissecting microscope at x40 magnification to assess the gross plane of fracture. Following this a representative number of samples were sputter coated and viewed using a JEOL T300 scanning electron microscope (Jeol Ltd. Japan) in order to assess, more accurately, the exact fracture plane of the bonded system.

The data obtained for shear bond strengths was analyzed using a two way ANOVA to determine if there was any difference between the groups. Comparison of groups was then carried out using a Students *t* test.

7.4 RESULTS.

7.4.1 Shear Bond Strength.

Table 7.2 and Figure 7.2 show the shear bond strengths of the samples tested. These results showed that the hybrid A, composite cement/unfilled resin, specimens had the highest shear bond strength of 27.1 ± 11.4 MPa (±1 S.D.). This was significantly greater than any of the other samples tested ($p < 0.05$ for hybrid cement A alone and $p < 0.01$ for all other samples tested). There were no significant differences for any of the other samples tested. The use of unfilled resin had no effect on bond strength of the other systems tested nor did the application of the bond enhancer, either wet or dry, to the microfilled system.

COMPOSITE RESIN	COMPOSITE CEMENT	UNFILLED RESIN	BOND ENHANCER	MEAN (S.D.) SHEAR BOND STRENGTH (MPa)
HYBRID A	Y	N	N	17.0 (6.5)
	Y	Y	N	27.1 (12.0)
HYBRID B	Y	N	N	8.3 (3.2)
	Y	Y	N	11.5 (8.4)
MICROFIL	Y	N	N	11.6 (5.7)
	Y	Y	N	14.0 (4.8)
	Y	Y	DRY	9.3 (5.2)
	Y	Y	WET	11.6 (3.5)

Table 7.2: Shear bond strengths of composite resin inlays bonded to bovine enamel.

7.4.2 Plane of Fracture.

With both the hybrid, heat\light curing inlay systems the plane of fracture was between enamel and composite, with the composite resin remaining firmly attached to the inlay surface (Figures 7.3-7.6). A little resin can be seen, still attached, to the enamel when the hybrid B inlays were fractured. However the plane of fracture is mainly adhesive at this interface.

In the microfilled heat\pressure cure inlay system however, the cement always remained attached to the enamel surface, with the inlay surface appearing cement free (Figures 7.7 and 7.8). No difference was observed in the debonded inlay surface whether the inlay had been treated with bond enhancer or not.

In none of the systems did the use of an unfilled resin alter the appearance of the debonded surface.

7.5 DISCUSSION

For the two hybrid heat\light cured systems the bond between the inlay and enamel did not break at the inlay\cement interface as anticipated. From this result it would appear that the composite inlay to composite cement bond in these systems is not as low as was thought.

The bond between a luting cement and a composite inlay is essentially the same as that to a fractured composite resin. It follows that a comparable bond strength would be expected.

The bond strength of a composite repair to a fractured composite has been shown to be only in the region of 25% to 75% of that of the unbroken material[232-234]. This is nonetheless in the region of 15-30 MPa, which is comparable with the bond strength of composite to etched enamel[125]. If this is true, the bond strengths obtained with the hybrid A

Figure 7.3: SEM image of enamel surface following debonding of hybrid A composite resin inlay. Enamel surface with etched pattern is visible. Original magnification = x1000. Field width 100μm.

Figure 7.4: SEM image of inlay surface following debonding of hybrid A composite resin inlay. Luting composite is firmly attached to inlay surface and an impression of the etched enamel surface is visible, where debonding has occurred. Original magnification = x500. Field width 200μm.

Figure 7.5: SEM image of enamel surface following debonding of hybrid B composite resin inlay. Enamel surface with etched pattern is visible. A little resin (arrowed) can be seen attached to the enamel. However the plane of fracture is mainly adhesive. Original magnification = x500. Field width 200μm.

Figure 7.6: SEM image of inlay surface following debonding of hybrid B composite resin inlay. Luting composite is firmly attached to inlay surface and an impression of the etched enamel surface is visible, where debonding has occured. Original magnification = x500. Field width 200μm.

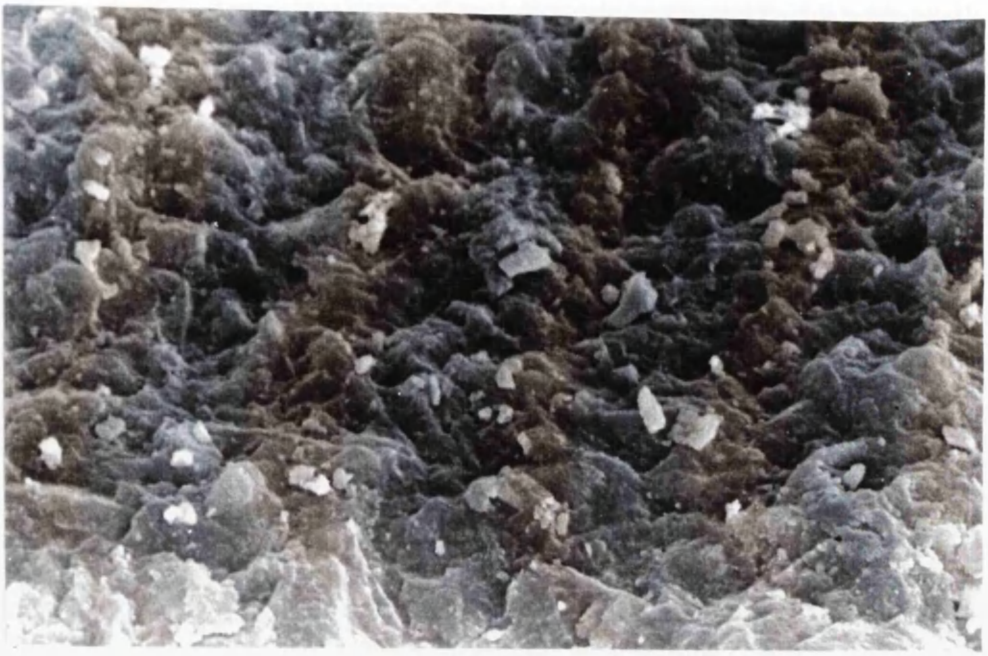


Figure 7.7 SEM image of enamel surface following debonding of microfilled composite resin inlay. Luting composite has remained firmly attached. No enamel morphology is discernable. Original magnification = x500. Field width 200μm

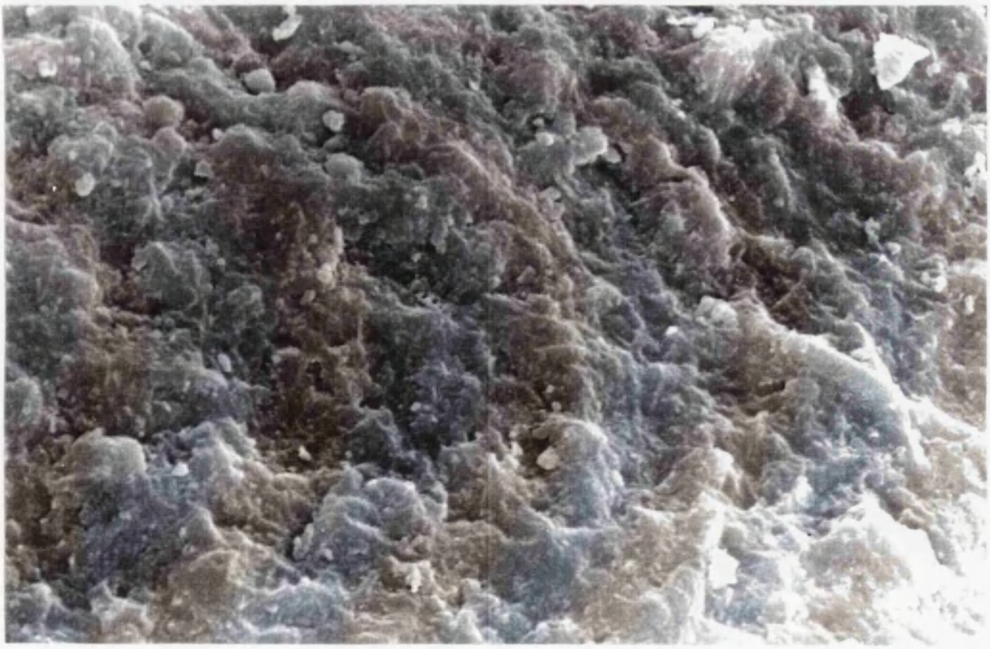


Figure 7.8 SEM image of inlay surface following debonding. This surface appears exactly the same as Fig 7.7. It is not possible to completely distinguish the inlay from cement, however it appears that adhesive failure has occurred at this interface. Original magnification = x500. Field width 200μm

system, which are comparable to the highest bond strengths obtained by Swift *et al.*[123], are similar to those which might be expected. In this case, the bond strength of the resin cement to the inlay surface exceeded that to the enamel surface and the bond fractured at the resin enamel interface. Application of unfilled resin increased the bond strength to the etched enamel, resulting in a higher shear strength. However, this was still less than that of the inlay to resin cement and hence, despite the increase in bond strength, no alteration in fracture plane occurred.

With the hybrid B system, the bond strengths were significantly lower than for hybrid A, although the plane of fracture remained the same. The decrease in bond strength between these two inlay systems could well be explained by the granular and relatively viscous nature of the luting cement used with hybrid B when compared to hybrid A which, although of similar filler content, flowed much more easily. The higher viscosity of composite cement B is borne out by its significantly higher film thickness, 137μm for hybrid B against 104μm for hybrid A (see chapter 4 for details). This may have prevented adequate wetting of the enamel surface and led to the relatively low bond strength between the two. The fact that the luting cement remained attached to the inlay at this shear strength was not unexpected as Crumpler *et al.*[234] in studies, using Estilux posterior, (hybrid B) have shown that it can achieve up to 88% of its strength following repair, with a shear strength of 27 MPa being recorded.

The fact that both these systems show the fracture plane to occur at the enamel composite bond suggests that there is no problem in bonding to these inlays, despite the decrease in C=C bonds available (see Chapter 6). It would also seem probable from these results that there may be other factors important in allowing bonding to occur between the two composites. Surface roughness, porosity and wettability of the inlay surface, with the cement leading to micromechanical interlocking, could be as important as purely chemical bonding.

The microfilled inlays fractured in the manner which would be expected if there were insufficient bonding sites remaining due to supercuring. It has been suggested that this is the case in the Ivoclar material, as it is cured in a different manner which results in an increased degree of methacrylate conversion relative to the other systems[126]. However the results of Chapter 6 do not bear out this suggestion with similar conversion being noted in all three inlay types. The sandblasting, as recommended by the manufacturer, will lead to a relatively rough surface which should encourage bonding. However, from the plane of fracture it would seem that, despite this treatment, the surface is less retentive than either of the hybrid materials.

The bond enhancer, developed to increase the bond strength of inlay to cement, did not improve the bond strength in this study. The intended mechanism of action of this material has not been disclosed, so no further comment on its efficacy is possible at present.

The variation in bond strengths between hybrid heat\light cured and microfilled heat\pressure cured systems is in general agreement with previous authors[97,124,126,128]. There are several probable reasons for the differences in plane of fracture observed between the systems:

1) The pressure cured system uses a microfilled composite resin. As such it has a higher coefficient of thermal expansion than the two hybrid materials. This means that it will be subjected to more stress during thermocycling and, hence, be more prone to bond breakdown[6]. Peutzfeldt and Asmussen have shown a greater decrease in retention of Ivoclar inlays following thermocyclng, than of two hybrid composite systems[128].

2) The tensile strength of the microfilled composite resin cement is less than that of either of the hybrid materials used (See Chapter 4). As a positive correlation has been shown

between tensile strength and bond strength of a composite[235] this may have some bearing on the result.

3) The possibility of micromechanical adhesion should not be overlooked. Wilson *et al.*[93] has noted a decrease in porosity of composites cured under pressure. This lack of porosity is also, indirectly, supported by the high values obtained for physical properties of the heat\pressure cured material, tested in this study, compared to similar light cure only microfilled materials. While a less porous material may be strong, the pressure curing may also lead to a lack of appropriate areas on the fitting surface where resin cement can flow into the inlay surface and provide micromechanical interlocking.

The potential importance of micromechanical adhesion in promoting bonding between inlay and composite resin cement is investigated in the next chapter.

Chapter 8: An Investigation of the Composite Inlay, Composite Resin Cement Interface.

8.1 INTRODUCTION.

The results of the previous chapter would suggest that factors other than chemical bonding, between residual methacrylate moieties, may be important to the adhesion between a composite resin inlay and its luting cement. This chapter investigates the possibility of micromechanical interlocking between composite resin inlays and luting resin: it can be inferred from the preceding chapter that bonding of this nature may play an important role in the adhesion achieved. Furthermore, by investigating inlays fabricated using different techniques, the role of the curing procedure in affecting micromechanical bonding may be elucidated. Scanning electron microscopy and confocal light microscopy, combined with fluorescent markers, were used to characterise the interface between inlay and composite resin cement.

8.2 MICROSCOPIC TECHNIQUES.

Various microscopic techniques have been used for investigation of hard tissues and restoration interfaces.

Conventional light microscopy can be conveniently divided into transmitted and reflected light microscopy. Transmitted light microscopy can be used to examine hard tissue and restoration interfaces. However, this requires the samples to be processed into very thin sections. When dealing with potentially fragile interfaces, sectioning techniques, which often involve the use of saws and subsequent polishing, inevitably lead to damage and disruption to both tooth and biomaterial. The problems of sectioning can be partially avoided by the use of a single cut. However, this generally results in a bulky sample, too thick to transmit light, which can consequently only be viewed using reflected light. In order to obtain images of reasonable quality, samples for reflected light microscopy must be polished to give adequate light reflectance. This polishing process firstly, requires the sample to be embedded in a suitable medium, usually a resin polymer and secondly may itself cause damage to the areas of interest. Even if it is possible to process a sample for reflected light microscopy without disruption of the area of interest it can be difficult to obtain a satisfactory image. At best information can be gained from the surface layer, from which most of the light is reflected. Sparse information can be obtained from deeper layers[236] as much of the light is reflected back into the image plane by both the image surface and lenses within the microscope, resulting in degradation of the image contrast[237]. Epifluorescence reflected light microscopy can give information about deeper layers, however the depth resolution is poor[238].

Electron microscopy, both scanning and transmission, have also been used to give information on interfaces between teeth and restorations. Transmission electron microscopy suffers from the same drawbacks as transmitted light microscopy, except to a greater extent. Very thin, electron lucent films are required with all the concomitant problems of their fabrication. Furthermore, the dehydration and fixation required in this technique can distort already extremely fragile samples. Scanning electron microscopy (SEM) is an altogether simpler procedure; although not without its drawbacks. In this technique the sample is placed in an electron beam and the image is produced by collection of secondary electrons emitted by the surface and volume of the sample. Scanning electron microscopy is capable of producing images of high resolution and large depth of field at very high magnifications. However, there are disadvantages. Firstly the surface to be examined must be made electrically conductive. This is generally accomplished by coating with a thin metallic film (normally gold) in a vacuum; a process termed sputter coating. Loss of surface detail is a theoretical consideration during this process, however as the metal film is only of around 30nm in thickness it is generally considered inconsequential. Of more importance is the potential for desiccation of the sample when placed under vacuum. This is the case with certain tooth filling materials such as glass ionomer cements[239] and tooth substance itself particularly the dentine[240]. This dehydration places considerable stresses both within the tooth itself and on the bond between the tooth and any adhesive attached to it. Watson[238] suggests that this dehydration effect can be used to good advantage in observing the fractured interface between tooth and restoration. However, distortion and fracture of specimens under vacuum can make interpretation difficult for all but the most experienced. One possible method of investigating the interface between bonded surfaces is to dissolve away the substrate. In the case of tooth tissue hydrochloric acid and sodium hypochlorite can be used. This removal of the tooth reveals the fitting surface of the restoration giving information on the shape of the bond[238]. The most successful method of avoiding the problems associated with desiccation is not to desiccate the sample at all. This is accomplished by the use of a replica technique. Briefly, this technique involves the casting of an epoxy resin replica from an accurate impression of the specimen and the subsequent microscopic analysis of the replica; Barnes providing a thorough review of this subject[240,241]. The replica technique allows SEM of *in vivo* specimens and serial examinations of samples over time. However very fine surface detail may be lost with this technique. All scanning electron microscopy is by definition the examination of the surface of the specimen although the image may be affected by subsurface features as the electron beam interacts with a volume of the sample; not only the surface. It is, nonetheless generally interpreted as the surface which is seen. Although it is possible through careful preparation of samples to examine bonded surfaces it is not possible to look beneath the surface and examine the interface between tooth and biomaterial *in situ.*

Recently a new SEM has been developed which operates at normal pressures and does not require dehydration of samples. This "Environmental Scanning Electron Microscope" may well be able to overcome many of the problems of conventional machines. However, no studies have yet been reported in the dental literature to assess its potential in this field. Nonetheless, despite these failings, scanning electron microscopy remains an excellent way of examining surfaces of dental tissues and restorative materials and is used extensively throughout dental materials research.

A new microscopic technique has recently become available. The introduction of the **confocal scanning optical microscope** has made reflected light or high resolution epi-illumination microscopy viable for the study of dental hard tissues and biomaterials. Essentially these instruments are capable of producing thin optical section of samples, at high resolution, below the surface of translucent samples. This occurs in a manner analogous to radiographic tomography.

8.3 THE PRINCIPLE AND MODE OF ACTION OF THE CONFOCAL MICROSCOPE.

Confocal microscopy is based around the principle of eliminating all the scattered and reflected light from planes outwith the plane of focus. Confocal operation can be achieved by combining the illumination with the imaging of one spot in one focal plane at one time. In order to produce this effect an aperture is placed in the illuminating beam such that the image of this aperture can be focused in a plane within the specimen. Light returning from the sample along the imaging pathway can pass an equivalent aperture only if it is derived from the plane of focus within the specimen. The solid, opaque region surrounding the aperture will prevent transmission of scattered, reflected or fluorescent light from planes which are out of focus and thus does not degrade the image (Figure 8.1). In order to produce an image of a whole plane of section (an optical tomogram) rather than just a single spot, as described, it is necessary that the light scans the sample. This can be accomplished by moving the sample beneath a stationary beam, scanning the beam by the use of mirrors, by moving the objective lens or by scanning the apertures.

The first description of a confocal light microscope was that of Minsky, whose original patent was filed in 1957[242]. Petran *et al.*'s "Tandem Scanning Reflected Light Microscope" was the next to be cited in 1968; interestingly, both these microscopes were developed for use in neurological studies although the latter was used *in vivo*[243].

Two types of confocal microscopes have been described. Confocal scanning laser microscopes were initially based on the principle of a stationary laser and moving sample. This scanning method produces very high resolution images. However, it is an extremely time consuming process with a frame rate of minutes duration[244]. Recent systems have been developed, using galvanometric mirrors, by which the laser beam can be scanned across the specimen[245,246]. The beam itself is introduced from a "black box" through a conventional light microscope. The reflected or fluorescent signal is returned to the box for descanning and subsequent image processing. No direct image is possible with this type of instrument, in scanning mode. Following image processing the sample is viewed on a TV screen, in a similar manner to SEM. The first generation confocal laser scanning microscopes had slow frame rates and could not be used for real time imaging. This was due to their reliance on mechanical scanning with mirrors, which could not match TV frame speed[238]. This problem can be solved by the use of acousto-optical deflection of the beam, as described by Draaijer and Houpt[247] or by slit scanning systems, described by Brackenhoff and Visscher[248] which although increasing frame rate do result in loss of confocal resolution in one axis. Complex image processing devices and frame stores generally used with confocal microscopes bring with them an advantage. By recalling serial scans, it is possible to construct three dimensional images of objects using modern computing techniques[236].

The second type of confocal microscope available is the Tandem scanning microscope (or TSM). These are also termed real time, or direct view confocal microscopes. This variant has many thousands of apertures, and the confocality is produced by the movement of these apertures. This automatically produces very high frame rates, much faster than video rate, and allows observation of high speed events and searching within a specimen in confocal mode. The original tandem scanning microscope used exactly matched apertures on opposite sides of a spinning disc to separate the incident and reflected light. Recently, a one sided, unilateral tandem scanning microscope has been developed which transmits and receives the light through the same hole in the disc. This approach was described by Xiao *et al.*[249,250] and Boyde *et al.*[251], and significantly simplifies the alignment of the microscope components, although requiring some extra components to remove stray reflected light.

One of the great advantages of confocal microscopy is the reduced need for specimen preparation. Providing the sample is slightly translucent, it is possible to undertake a subsurface examination, as has been done with human enamel/restoration interfaces *in vivo*[252] with no preparation at all. However, interpretation of the image may be rather difficult due to surface irregularities. These irregularities can prevent deep penetration by the microscope simply due to lack of lens clearance. Consequently, it is preferable to produce a flat, polished surface on the sample. This acts as a reproducible reference point for subsequent examination. Any surface disruption, or damage inflicted by the flattening procedure will be apparent during examination of the specimen. However, the advantage of the confocal system is that this surface disruption can be focused through to an undamaged area below. Moreover, specimens for examination using this type of microscopy can be placed in a virtually normal environment, without the need for vacuum or the dehydration required for SEM.

In order to obtain good quality images below the surface of translucent samples it is necessary to use high quality objective lenses designed to work while immersed in liquids. The liquid immersion is important as it reduces the strong reflections from the surface of the specimen and also, importantly improves the light gathering ability (numerical aperture) of the lens, because of the higher refractive index of the liquid relative to air. Typical immersion media are water, glycerine and oil. A lens with a high numerical aperture is necessary as this allows a thin optical section to be obtained. At present 60X oil immersion lenses of numerical aperture 1.4 are the most commonly used in confocal microscopy[238]. The working distance within the section is also controlled by the lens. The most commonly manufactured lenses have a working distance of around 200μm to allow a glass coverslip of 170μm to be used. While, theoretically, a working distance of more than 200μm would be preferable, the transparency of most dental samples is such that penetration beyond this depth is not possible.

Fluorescence imaging has been used extensively in both biological and dental research, for labelling cells or different components in tissues or biomaterials[239,253-258]. This technique works on the principle of a material absorbing light energy at one wavelength and subsequently emitting it at a different longer wavelength. An example of this type of material is Rhodamine which absorbs green light at 546nm while emitting red light at 600nm. The use of optical filters of known wavelength can block all but the fluorescent light, allowing identification of the areas in which the fluorophore is present. Incorporation of fluorescent dyes into components of dental materials can give an insight into their distribution onto and within dental tissues, giving new information on the interfaces formed between these materials and dental hard tissues. Moreover, by combining different dyes with the different components of a restorative material and using filters within the microscope, the relative position of these components within the bonded interface can be visualised[236,238,239,253,255]. Furthermore, examination over time allows tracking of the fluorescent labels which, in turn, indicates dynamic events taking place at the restoration interface[239].

It has been suggested[236,238], that leakage studies could be carried out using fluorescent markers in conjunction with confocal microscopy. This could give much more information on the position and degree of marginal failure than the low resolution, destructive sectioning techniques used at present and described in detail in Chapter 10. Recently, the imbibition of fluorescent marker into carious enamel has been used to allow rapid histological assessment, without recourse to polarised microscopy of thin sections and its attendant difficulties[254].

It is possible to produce images with an extended plane of focus with the confocal microscopes. In the tandem scanning microscopes this is accomplished by focusing through a section of the specimen while the image is being recorded[236]. With the laser scanning microscopes the addition of several focal planes can be accomplished using the frame store facility of the microscope's image processing software[238]. This ability to add focal planes to make the final image allows surface images to be obtained which resemble SEM images.

Confocal microscopes are all able to produce stereo images to the resolution of conventional light microscopy[259,260] through reconstruction of two slanted, extended focus views through the same slice of a specimen.

Image recording of static events with these tandem scanning microscopes is simple. A photomicrograph can be taken using a suitable camera and 35mm film. Recording of dynamic events is more problematic requiring the use of video cameras and perhaps specialised stages. Furthermore the light intensity derived from biological specimens is seldom sufficient for use with colour cameras. This necessitates a black and white image and often requires the use of image intensifiers to increase the camera's sensitivity. These intensifiers may tend to degrade the image, resulting in a more grainy appearance. Until very recently, laser scanning microscopes did not have a direct view facility; images were therefore recorded from TV screens following suitable image analysis.

Comparison of the two types of confocal microscopes is problematic as each has its advantages. Both microscopes allow good images to be obtained with a minimum of damage to the specimen, due to the simple and, relatively, non destructive sample preparation required. However, Watson and Boyde[238] have suggested that for problem solving in dental materials research tandem scanning confocal microscopes are more convenient and efficient, having direct viewing of colour images and confocal imaging at faster than video rate. Furthermore, the advantage of the laser scanning system is more apparent in samples with low levels of fluorescent dye: not generally a problem when considering dental materials.

8.4 AIMS OF STUDY.

Through the use of scanning electron microscopy and tandem scanning confocal light microscopy, the latter utilising fluorescent markers, it was hoped to investigate the interface between composite resin inlays and their luting resin.

8.5 MATERIALS AND METHOD.

8.5.1 Composite Inlay Preparation.

Composite resin inlay samples were prepared in a similar manner to that described in the previous chapter. Twelve samples were produced for each material (six for SEM and six for confocal microscopy). They were produced with a surface area of 25 mm^2, as before, however they were made 3mm thick, for easier handling. Again a specially made perspex mould which was placed on a base of dental stone was used. Both the mould and the stone were coated with the appropriate separating medium. With the light cured materials (hybrid A and B) the composite was packed into the mould in 1mm increments and light cured for 60 seconds for each increment. After removal from the mould, the flash was removed from the samples. They were then subjected to the appropriate post curing process, hybrid A samples in the D.I.500 oven and hybrid B samples in the Unilux A.C. The Ivoclar samples were cured in the mould, in a single increment, entirely in the Ivomat heat\pressure oven as they do not light cure.

On removal from the oven the fitting surface of the Ivoclar samples was sandblasted with 50μm Al_2O_3. No surface treatments were carried out on the other samples. At this stage all inlay samples were stored, dry, for 1 week prior SEM analysis or fluorescent resin application.

8.5.2 SEM Sample Preparation.

Samples for SEM analysis were cleaned with 37% phosphoric acid etching gel, to remove finger grease, washed with water and wiped finally with alcohol to remove any surface moisture; the same procedure as carried out prior to bonding. They were then placed under vacuum and sputter coated with gold.

Following this treatment they were viewed using a JEOL T300 scanning electron microscope (JEOL Ltd., Japan) at original magnifications of up to 3500X.

8.5.3 Confocal Scanning Microscope Sample Preparation.

Fluorescent resin was produced by mixing a trace of fluorescein or Rhodamine B with the unfilled resin recommended for use with each inlay system as described previously by Watson[253]. The resin used with hybrid A is dual curing, hence the fluorescent marker was combined with the base component alone, in this case; to avoid premature curing of the resin. The special bonding agent for use with the microfilled inlay system was also labelled with Rhodamine B. The fluorescein labelled resin is exited by light at 450nm producing yellow/green light at 520nm wavelength, while rhodamine B labelled resin produced red light at 600nm wavelength having been excited by green light at 546nm.

The inlay specimens were cleaned in the same manner as above. Prior to fluorescent resin placement a clear matrix (Automatrix. L.D. Caulk, Millford DE, USA) was placed around the sample to provide a well into which the unfilled resin could be flowed. An initial layer of unfilled resin was painted onto the inlay surface and blown to a thin layer. This was subsequently light-cured for 30 seconds. Following curing a second layer of fluorescent resin, of around 2mm in thickness, was flowed onto the first layer and light-cured; this second layer was used to make the samples easier to handle. Where the bond enhancer was used it was applied prior to resin application, it being labelled with a different fluorophore than the subsequent resin layer to allow its differentiation later. All samples were stored dry until tested.

Immediately prior to testing further inlay samples (three per material), which had been cleaned as previously described, were dipped directly into aqueous solutions of

Rhodamine B for 5 minutes as it was thought that an aqueous dye solution may provide further information on the potential porosity and penetrativity of the inlay surface. This information, while perhaps not being as clinically relevant as that produced with the fluorescent resin would, it was hoped further delineate the inlay surface.

The low sample numbers used throughout this study were an unfortunate consequence of the limited access afforded to the confocal microscope, there only being two in dental use in the U.K. Nonetheless, the results described below were representative of all samples viewed. The appearance of the samples was identical within the groups studied, indicating little variation within the samples. Consequently, while the viewing of further samples would have been preferable, it would probably have resulted in very similar findings.

All samples for viewing with the confocal microscope were sectioned using a slow speed diamond saw under water (Labcut, Agar Scientific, Stanstead, Essex). The sectioned surfaces were examined with a tandem scanning confocal light microscope (Noran 2551W. Beltline Highway, Middleton, W.I. USA) using oil immersion objectives with original magnifications ranging from 20 to 100 in conjunction with a 10X eyepiece. The samples were either illuminated with the entire spectrum from a high pressure mercury arc lamp or with specific wavelengths of light when excitation of the fluorescent labels was desired. Colour images were recorded by an automatic exposure 35mm camera. Images were also recorded using a low light level SIT video camera. The images were digitised and stored using an image processing system (MISIS Image-3D Plus, St Etienne, France). These stored images could then readily be measured and low light level signals enhanced.

8.6 RESULTS.

8.6.1 SEM Images.

Scanning electron micrographs of the fitting surfaces of the three inlay types are shown in Figures 8.2-8.7. All three inlay systems showed generally rough surfaces. However, there were notable differences between the systems. Hybrid A exhibited areas of roughness where the small (< 1μm) filler particles were clearly demarcated. Furthermore, holes between and around the filler were obvious, suggesting porosity within the fitting surface. Hybrid B had a similar appearance. However, in this material, the filler particles, which were much larger (up to 10μm), were visible sticking out from the surface and creating undercut areas. The holes and pores around the filler particles were also visible in this material. Moreover, porosity in the form of air bubbles was present within the matrix of this material. The microfilled heat\pressure cured material, although superficially rough following sandblasting, did not exhibit porosity or undercuts around filler particles. In fact, an altogether more uniform surface texture was observed.

8.6.2 Confocal Microscopy.

Photomicrographs of the tandem scanning confocal microscopic examination of the interface between inlay and fluorescent labelled bonding resin are shown in Figures 8.8-8.10.

There was evidence of uptake of fluorescent resin into the fitting surface of the hybrid A inlay material. Resin penetration varied between 1μm and >10μm depending on where the interface was scanned. In the combined reflectance, fluorescent image (on the right side of Figure 8.8) the filler particles were visible as bright dots and a degree of resin penetration could be seen where the green, of the composite, became lighter. However, with the use of a filter the fluorescent only image was produced (left of Figure 8.8), more clearly demarcating the degree of resin penetration into the inlay surface.

The confocal image of hybrid B confirms its SEM appearance. A very rough surface was present with large filler particles sticking out from the fitting surface (arrowed in Figure 8.9). As was expected, large filler particles could be seen throughout the body of the inlay with the reflected image. Both this and the fluorescent only images confirmed the impregnation of the inlay surface by resin as filler particles were clearly demarcated within the substance of the fluorescent resin. This was suggestive of flow into porosities and undercut areas within the material.

The reflected image of the microfilled material confirmed its structure, with large dark prepolymerised particles (>40μm) (pp in Figure 8.10) being present along with very small particles within the matrix. The uniform, non-porous nature of this material was suggested by the fluorescent image where, although the surface is demarcated, there is no obvious penetration of fluorescent marker into the material. The use of the bond enhancer did not make a significant difference to the observed interface.

Photomicrographs of the confocal scanning microscopic images of the inlays dipped in aqueous solutions of fluorescent labels are shown in Figures 8.11 and 8.12. Impregnation of the hybrid A sample showed dye penetration into the inlay surface to be possible. This is also the case, although to a lesser extent, with hybrid B where the outlining of filler particles can be observed. No obvious penetration into the microfilled material appeared to have occurred, however this technique did outline the surface of the inlay very well and Figure 8.12 taken at higher magnification shows cracks which were formed in the material during the sandblasting procedure.

Figure 8.2: SEM image of fitting surface of hybrid A composite resin inlay. Generalised roughness and porosity of surface is evident. Original magnification = x500.
Field width 200μm.

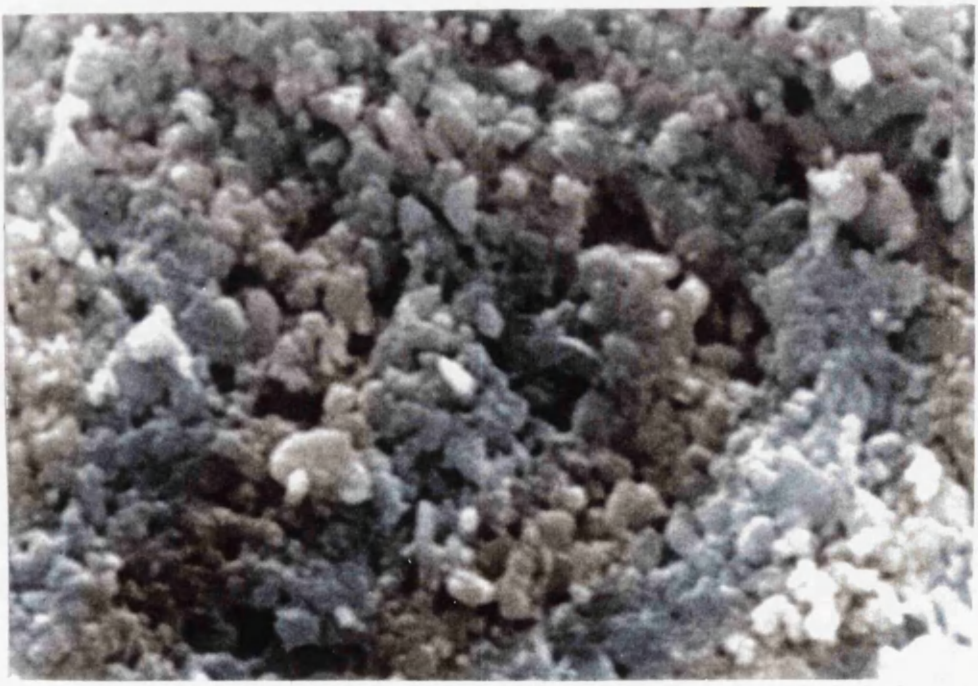


Figure 8.3: SEM image of fitting surface of hybrid A composite resin inlay. Filler particles can be seen. Spaces and undercut areas between particles are evident.
Original magnification = x3500. Field width 30μm.

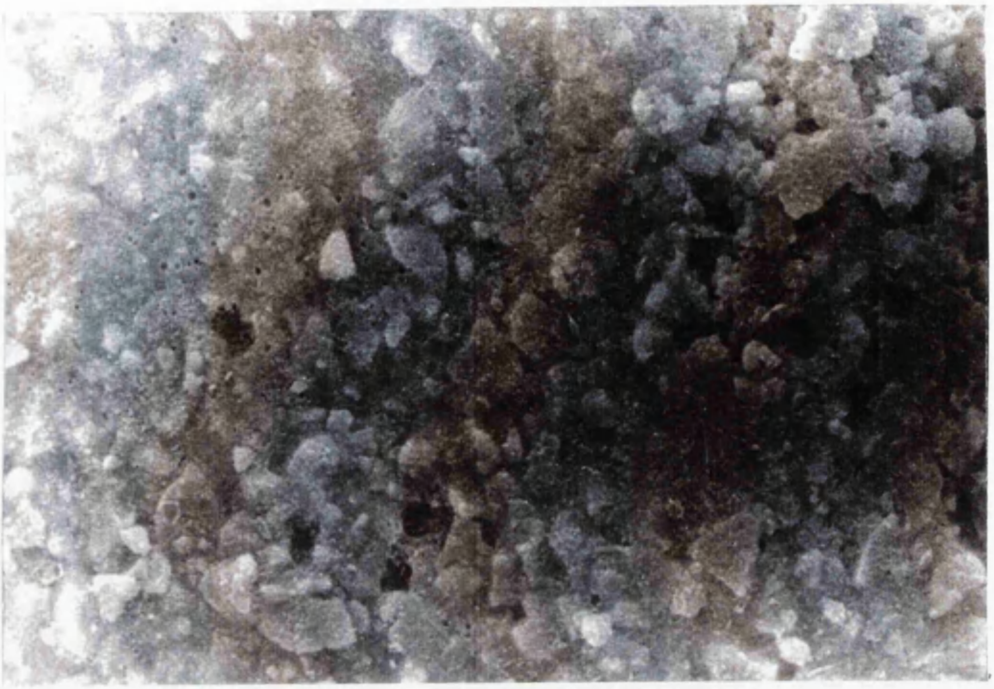


Figure 8.4: SEM image of fitting surface of hybrid B composite resin inlay. Filler particles sticking out from the surface with spaces around them are obvious. Also evident is porosity within the matrix.Original magnification = x500. Field width 200μm

Figure 8.5: SEM image of fitting surface of hybrid B composite resin inlay. A very rough surface with large filler particles (10μm), porous and undercut areas is present.
Original magnification = x3500. Field width 30μm

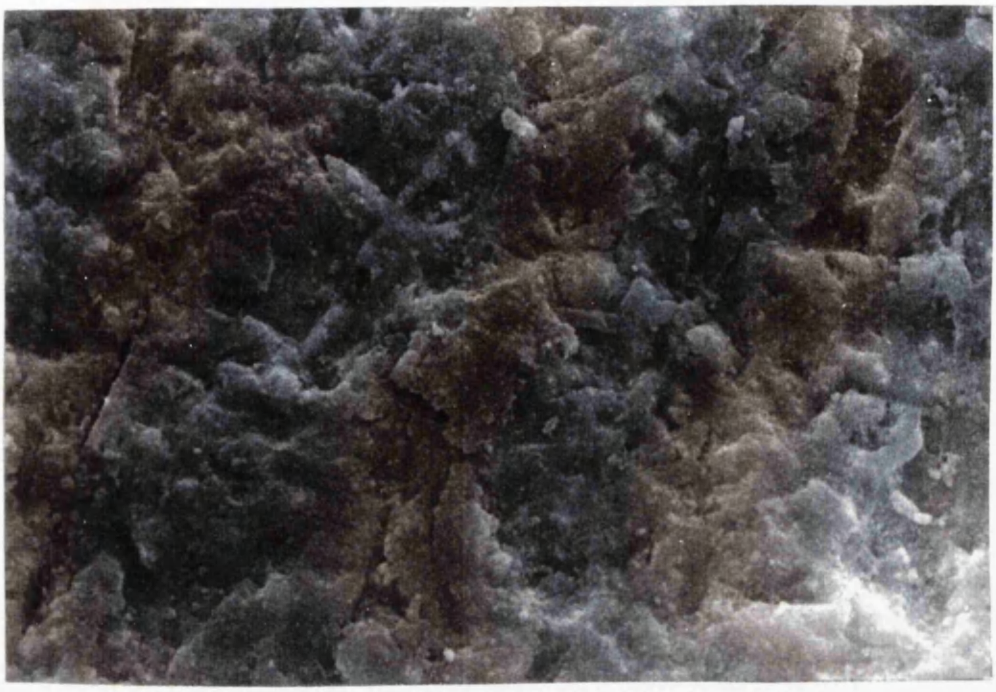


Figure 8.6: SEM image of fitting surface of the microfilled composite resin inlay. Although the surface is relatively rough there is no evidence of porosity, exposed filler particles or undercut areas. Original magnification = x500. Field width 200μm

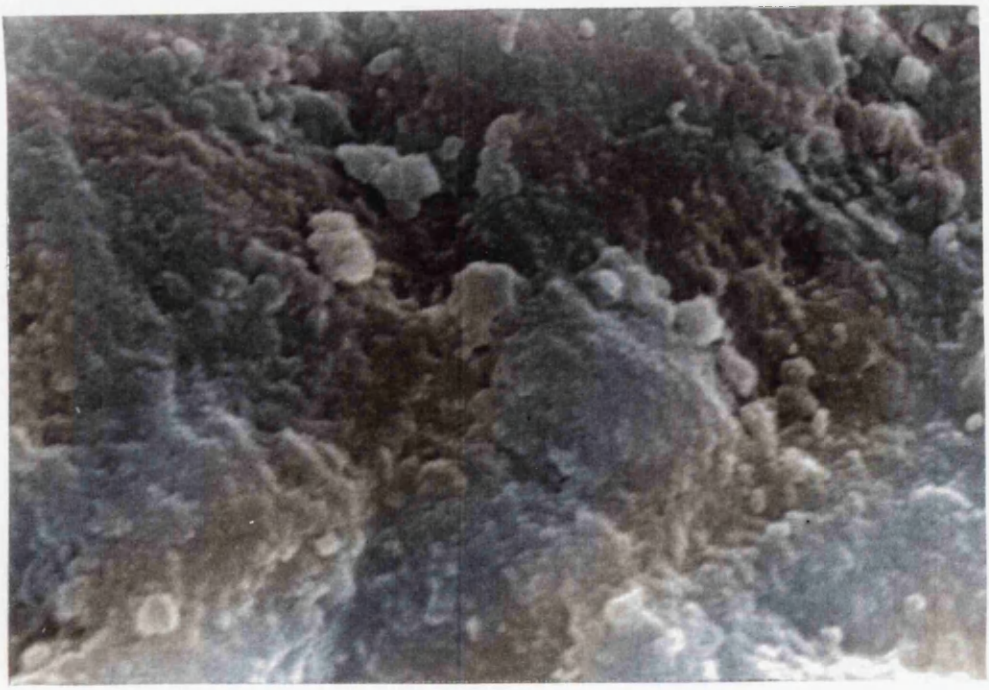


Figure 8.7: SEM image of fitting surface of microfilled composite resin inlay. Relatively flat, smooth surface with no evidence of the porosity and undercuts common to the other materials. Original magnification = x3500. Field width 30μm

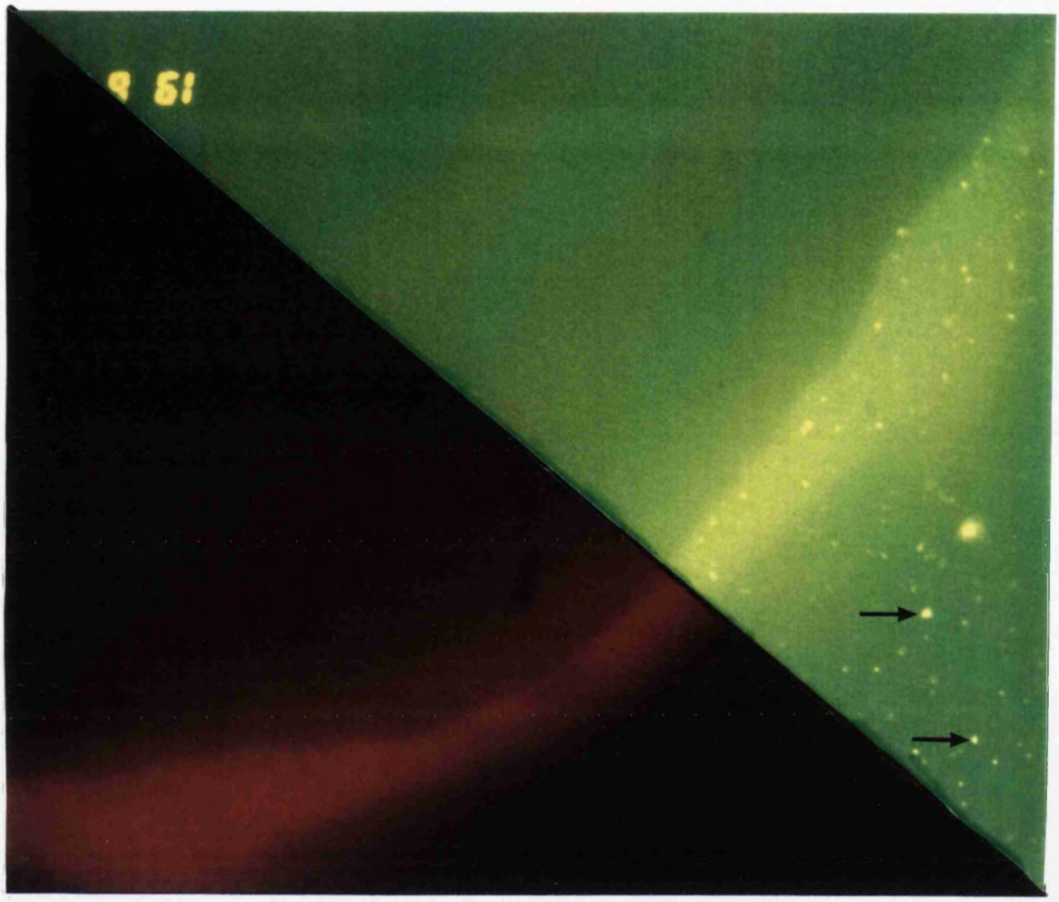


Figure 8.8: Confocal image of the interface between hybrid A inlay and fluorescent resin labelled with Rhodamine B. Filler particles can be seen as specks of light at the bottom right of the picture (arrowed). Resin penetration into the inlay surface can be seen on the left of the image where the barrier filter allows the fluorescence of the unfilled resin to be seen alone. Resin penetration to a depth of around 10μm is evident. 546nm green excitation filter, 600nm red barrier filter on left. x60/1.4NA, oil immersion objective. Field width 150μm.

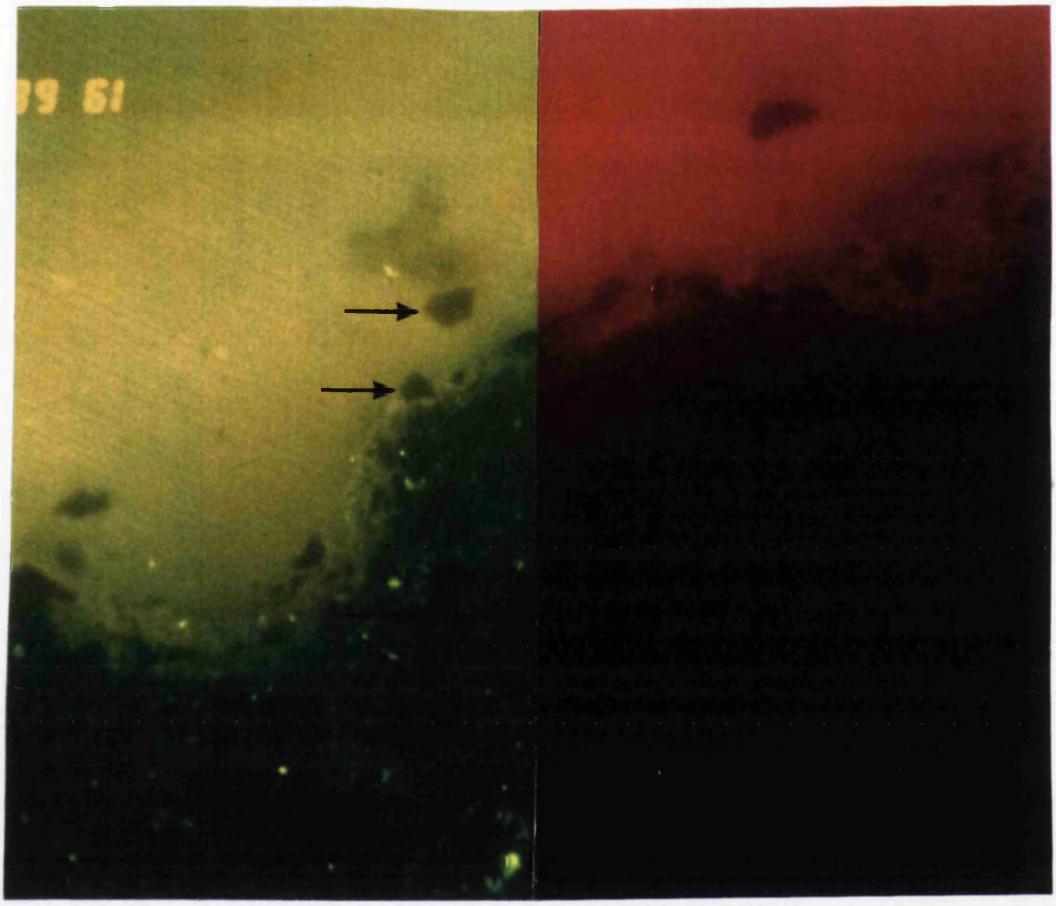


Figure 8.9: Confocal image of the interface between hybrid B inlay and fluorescent resin labelled with Rhodamine B. Large filler particles (arrowed) of up to 10μm can be seen within the material, on the left, and protruding along its whole fitting surface. Resin penetration in and around the filler particles is evident both with and without the barrier filter. 546nm green excitation filter, 600nm red barrier filter on right. x60/1.4NA, oil immersion objective. Field width 150μm.

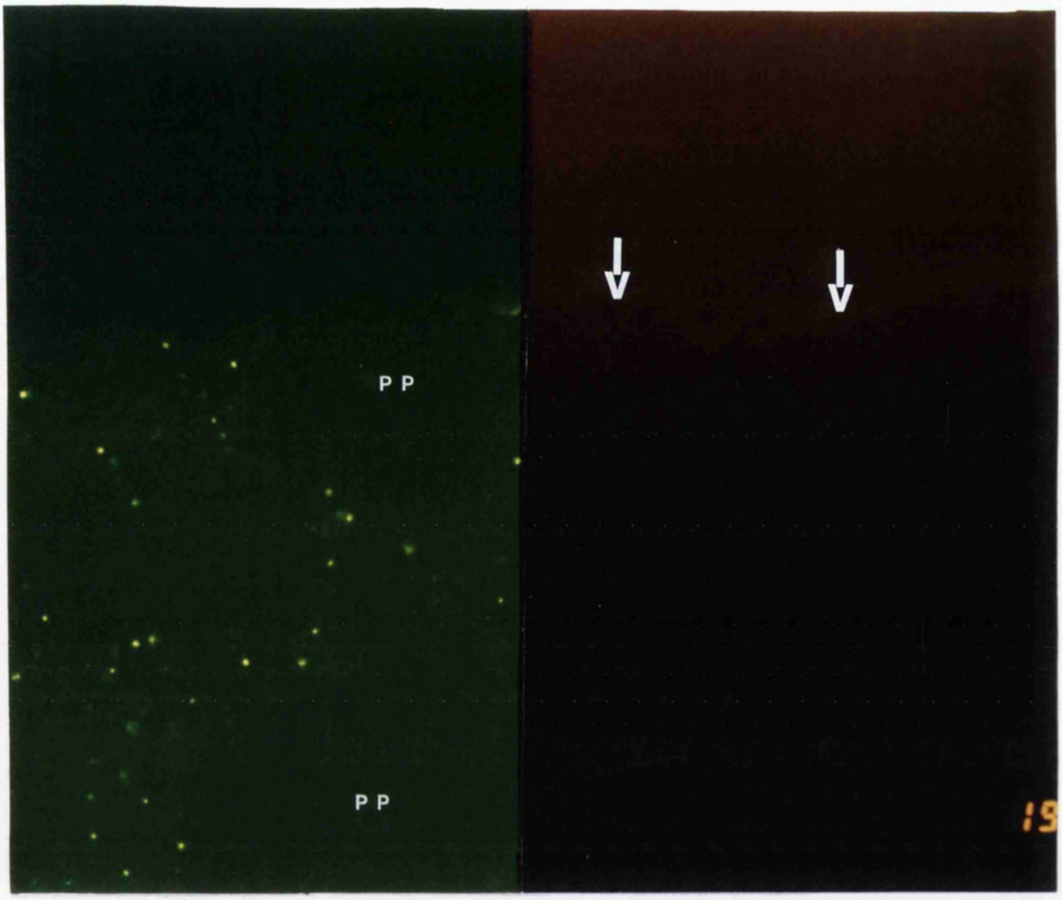


Figure 8.10: Confocal image of the interface between microfilled inlay and fluorescent resin labelled with Rhodamine B. Large prepolymerised particles (pp) of over 40μm can be seen as darker areas within the substance of the composite. The very small silica particles also show up as specks of light on the left of the picture. Although the surface is clearly delineated following placement of the barrier filter no impregnation of the inlay surface is evident (arrowed). 546nm excitation filter, 600nm red barrier filter on right. x60/1.4nm, oil immersion objective. Field width 150μm.

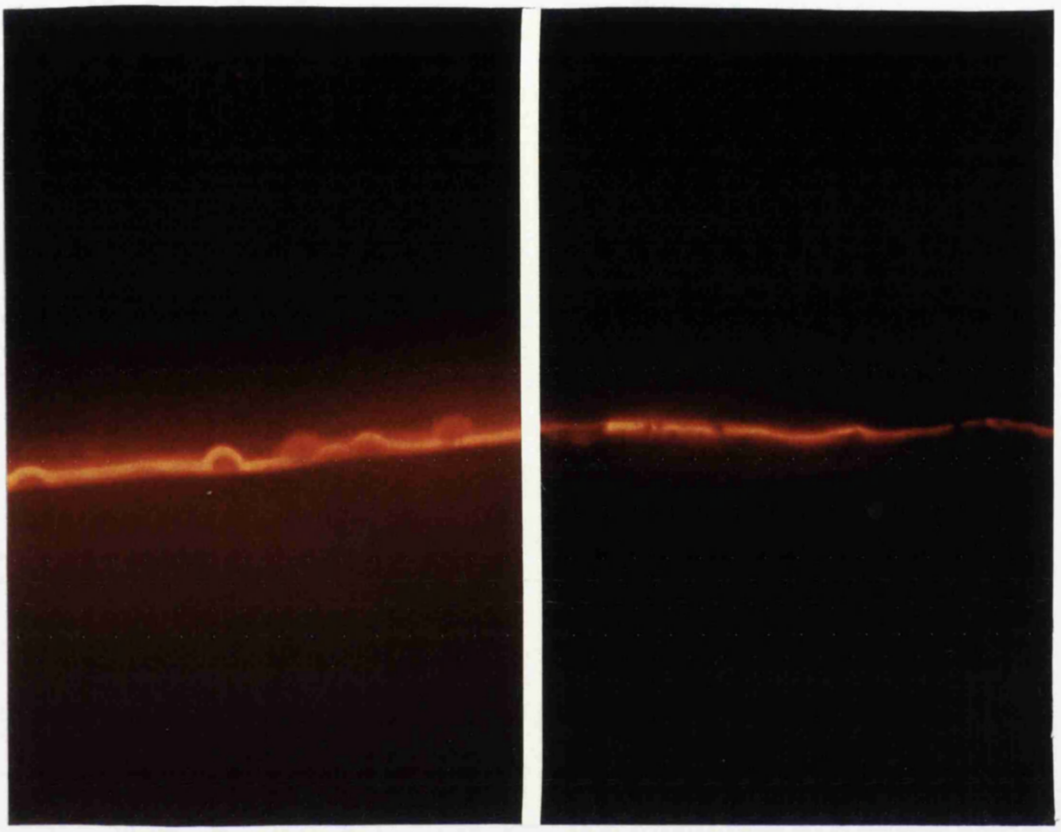


Figure 8.11: Fluorescent confocal image of hybrid A (left) and hybrid B (right) inlays following immersion in aqueous Rhodamine B with subsequent removal of excess dye, inlay at top. Both inlays show a degree of resin impregnation into the surface suggesting porosity. Hybrid A appears more porous as more dye is taken up. However, in the hybrid B image it is possible to see the outline of filler particles by the fluorescent dye. 546nm green excitation filter, 600nm red barrier filter. x20/0.8NA, oil immersion objective. Field width 500µm.

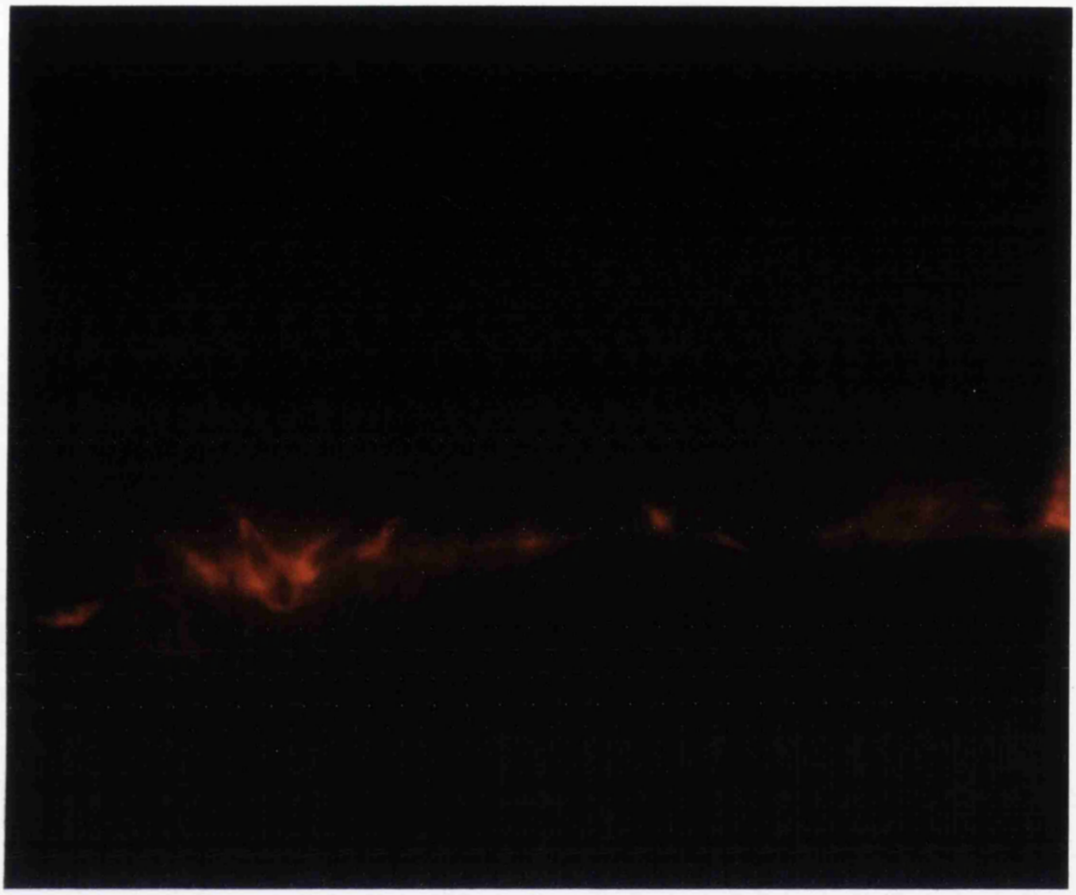


Figure 8.12: Fluorescent confocal image of the microfilled inlay fitting surface following immersion in Rhodamine B, inlay at top. The surface is clearly delineated with cracking of the surface evident. 546nm green excitation filter, 600nm red barrier filter. x100/1.4NA, oil immersion objective. Field width 90μm.

8.7 DISCUSSION.

The confocal images produced for hybrid A (Figure 8.8) and the microfilled material (Figure 8.10) were slightly surprising. Fluorescent resin was present above the inlay in both these samples, however no fluorescence was observed. An image more like that of hybrid B, labelled in the same manner, would have been expected with fluorescence visible wherever fluorescent resin occurred. It seems likely that this appearance was due to quenching of the fluorescence[261]. This phenomena is caused by too much fluorescent dye being present in the resin. When this occurs light energy produced by one dye molecule is taken up by another dye molecule in very close proximity. This results in absorption of the light energy before it can escape and produce fluorescence of the sample. If the concentration of dye decreases, as happened when the resin penetrated the inlay surface in Figure 8.8, fluorescence will occur as quenching of the energy by adjacent flurophore molecules will no longer occur. This phenomena has been observed previously, in a similar type of experiment, with a heavily labelled fluorescent labelled dentine bonding agent[262].

Scanning electron microscopy is a well established technique used extensively throughout dental research. In this study it was used successfully to examine the surface detail of three composite resin inlay materials. These samples were well suited to SEM analysis as they are relatively strong and are not affected adversely by the drying process required during preparation for this technique. However viewing of interfaces between composite resin inlay materials and their respective composite resins luting cements would prove problematic, when using SEM, as both materials are essentially the same. This lack of difference between inlay and inlay cement would result in difficulties in differentiation between the two materials and potential errors in assessing where one ends and the other begins. Consequently a new technique, confocal light microscopy, utilising fluorescent markers, was used in this study. This allowed rapid and easy differentiation of the inlay surface from unfilled resin, giving information on the nature of this interface which could not have been obtained previously.

The small particle nature of hybrid A was clearly visible in the scanning electron micrographs of the inlay fitting surface; with particles of less than 1 μm present (Figure 8.2,8.3). The fitting surface was remarkably rough with filler particles and porosity obvious over most of the surface. There were, however certain areas where the roughness was considerably greater than others and voids were present in the surface. This non-uniform surface texture was confirmed by the confocal microscopic examination. As can be seen in Figure 8.8 the infiltration of fluorescent resin was not constant over the surface of the inlay. Areas were present where resin infiltrated only to a minimal extent, around 1μm. However in a significant number of areas there was deeper resin impregnation to a depth of around 10μm. It seems likely that this deeper impregnation corresponded to the more porous areas observed on the surface during the SEM analysis.

The flow of bonding resin into the inlay surface to a depth of 10μm will inevitably be responsible for a micromechanical bond occurring between inlay and bonding resin. Even the 1μm infiltration may be sufficient to promote bonding between the inlay and its cementing medium. Similar degrees of resin penetration have been reported to produce good bonding in other dental situations[263,264].

Hybrid B is clinically a much rougher material than hybrid A fact confirmed by SEM analysis of the fitting surface. The difference in surface roughness was not unexpected as this material is a much older, more traditional type of hybrid material with filler particles of up to 10μm in size. The size of the filler particles was confirmed by the SEM appearance. Interestingly, combined with the inherent surface roughness caused by the size of the filler particles protruding from the surface, there were also many small holes, or pores, a situation similar to that observed in hybrid A.

Confocal microscopic analysis confirmed the extremely rough nature of the inlay fitting surface and the large particle size of this material. It appeared that some penetration of fluorescent resin occurred, as filler particles were outlined by fluorescence on the inlay surface. Nonetheless, good adaptation in and around the rough surface was obvious and a degree of mechanical interlocking appears inevitable when using this system. Whether micromechanical adhesion is actually gained from porosity in this system, or solely from the extremely rough nature of the surface is open to speculation. However, with such a rough surface the good adhesion recorded in the previous chapter is not surprising.

The lower power scanning electron micrographs of the microfilled samples showed a fairly rough and potentially retentive surface following sandblasting. Notably, however, there was no porosity obvious, nor were there any filler particles protruding from the surface which could provide mechanical retention. The lack of obvious filler was not surprising as this material is filled with 0.04μm particles. Furthermore, any prepolymerised particles, which are also present, would be expected to abrade at a similar rate to the matrix during sandblasting. With no differential removal of matrix versus filler a relatively uniform surface will be produced. This was, in fact, the case with this material where although a roughened surface was produced, it lacked the porosity and undercut areas obvious in the other materials (Figure 8.3,8.5,8.7). This is particularly obvious if the three higher powered scanning electron micrographs are viewed. Both hybrid materials have many rough undercut areas whereas the microfil appears relatively smooth at this magnification.

The lack of surface porosity was confirmed by the confocal microscopic analysis of this inlay type. Although the labelled resin could be seen to flow right up to the inlay surface there was no obvious infiltration present. Nonetheless the inlay surface was rough and some degree of cracking of the surface was obvious, probably as a result of the sandblasting process. This cracking is shown particularly well in Figure 8.12 which was taken using aqueous Rhodamine B. These cracks and surface roughness will provide some degree of micromechanical interlocking and undoubtedly some bonding of this nature will be able to take place. However, there appears to be less potential for this type of bonding to occur with this inlay type.

The use of the bond enhancer did not appear to have any effect on the degree of resin penetration obvious on the inlay surface. Initially it proved difficult to separate the fluorescent unfilled resin from the bond enhancer as, although the fluorescent dyes used produce light of different wavelengths, there remains some crosstalk of emitted signals, which makes differentiation of very thin layers difficult[261]. Further study using labelled bond enhancer alone produced slightly better results, with the use of a low light level video camera and image analysis system. However, even with this technique little difference in surface impregnation was observed when using this material.

It seems likely that the variations in surface roughness and porosity between the inlay groups must be, to some extent, responsible for the differences in plane of fracture observed in the previous chapter. Both heat\light cured materials, which remained attached to their luting resin, during shear testing, exhibited marked surface roughness with porous areas present and many undercuts into which resin could flow and produce micromechanical bonding. The heat\pressure cured material, which, contrastingly, debonded from its resin cement during shear testing, although rough, did not have the same degree of porosity or undercut areas present to help during the bonding process.

The difference in porosity between heat\light and heat\pressure cured materials is supported by the work of Wilson and Norman[93] who found significantly less porosity in composite resins cured under 6 bar pressure (the pressure used in this study). Further, indirect, evidence of the non porous nature of the microfilled material comes from the high values obtained for its physical properties when compared with a conventionally cured microfilled material (Chapter 3), as porosity has been shown to be responsible for reduced physical properties in composite materials[40].

However the relative roles of micromechanical and chemical adhesion cannot be extrapolated fully from this study or Chapter 7. If, as has been suggested, by Ruyter[126], heat\pressure cured materials undergo a greater methacrylate conversion than heat\light cured materials, the remaining C=C on the surface of the microfilled inlay will be fewer than on the hybrid materials studied. This will further compromise the bond between inlay and resin. Which of these two reasons is of most importance could only be assessed from bonding of inlays of known surface methacrylate conversion, from calibrated standards, and standard surface texture in an attempt to separate the two components. Although the results from Chapter 3 cannot be used to give numerical values for the number of remaining C=C bonds on each inlay, nor can they be used to directly relate the three materials, the proportion of remaining C=C bonds in each material was around 30% for all three materials. This is, at least, suggestive of the fact that there is potential for chemical bonding in all three inlay types and that the differences between groups may be caused by the differences in availability of micromechanical bonding sites.

Nonetheless, at present, there appears to be a problem with regard to bonding to heat\pressure cured composite resin inlays. This study would appear to indicate decreased bonding due to lack of surface roughness and porosity resulting from the pressure curing of the material. This leads to fewer undercut areas being present where micromechanical adhesion may occur and a reduced possibility of resin impregnation of the inlay fitting surface, inadequate bonding between inlay and resin cement being the end result.

Chapter 9: Accuracy of Fit of Composite Resin Inlays.

9.1 INTRODUCTION

9.1.1 Importance of Accuracy of Fit.

The fit of indirect restorations directly bonded to tooth structure is at least as important as in those cemented in the conventional manner. Metallic restorations rely on marginal bevels and chamfers for protection of the cement lute. In composite resin inlays, however, bevels are not possible. This is due to the possibility of marginal fracture of thin composite layers during fabrication and subsequent cementation[135]. It is also very difficult for the technician to finish such restorations to a fine bevelled edge due to loss of material during polishing. Consequently, the butt joint margin which is employed in these restorations exposes the cement lute fully to the oral environment and to masticatory forces occlusally. This is a possible weakness in the resin inlay technique. Although the composite resin luting cements are highly filled, they are not as filled as the inlay materials (Chapter 2), nor are they subjected to the secondary curing procedure to improve their physical properties. Consequently they have relatively poorer physical properties than the inlay (see Chapters 3 & 4 for further details). The resin lute will, therefore, inevitably be more susceptible to the degradative processes which occur in the mouth.

The fit, and hence the composite resin cement lute thickness is of great importance to the longevity of the restoration. An interface of more than 100μm is believed to result in occlusal wear of the luting agent with washing out of some of the lute evident by six months[152,153]. Loss of lute results in the inlay itself becoming unsupported. This edge will in turn be prone to flexure and eventual fracture leading to ditching around the margin[145]. This situation has been confirmed clinically[265].

The thickness of the resin cement layer may also be important with regard to sealing of the restoration during cementation. A thin layer of composite, as in an inlay lute, will contract less than a thick layer, as in a direct composite restoration. However, even with the thin layer, problems may occur. Polymerisation contraction still occurs and may place severe stress on the tooth/resin interface with forces in the region of 4-17.6 MPa being suggested[79,266,267].It has also been suggested that an increase in cement lute thickness results in an increase in shrinkage stress during contraction. Using Finite Element Analysis Rees and Jacobson suggested an increase from 7.2MPa with a lute of 100μm to 10.7MPa with a thickness of 300μm[121]. This stress increase has also been supported by the work of Kunzelman *et al.*[122] who found a significant increase in stress when the lute increased from 100-300μm.

9.1.2 Methods of Assessing Fitting Accuracy.

Several methods of *in vivo* evaluation have been suggested for evaluation of accuracy of fit of indirect restorations. The most widely used method is the United States Public Health Service method[139]. However, various other methods using radiographs and impressions have been reported[268,269]. These have been described more fully by Qualtrough and Piddock[270].

In vitro methods of evaluation have also been developed. S.E.M. analysis of restoration margins, generally using a replica technique, have been used for evaluation of porcelain and metal ceramic restorations[271,272]. The three dimensional fit of various types of restorations has also been assessed using an elastomeric wash technique[120,273,274].

With regard to resin inlays three techniques have been suggested previously. Peutzfeldt and Asmussen[118] used a milled brass die and stereomicroscope to assess the marginal fit of proximal boxes of three composite inlay types. They also measured the cervical composite resin cement lute thickness and marginal gap size of inlays cemented into human teeth *in vitro* using the same technique. This technique only gives an indication of fit at the margin. This may be unrepresentative of the fit along the rest of the fitting surface of the restoration. Also, with the inlay placed on a brass die, no account is taken of the influence of the resin luting cement on the final seating of the restoration. Reid *et al.*[90] used a similar technique to assess the fit of 2 composite and one porcelain inlay system. In this study the maximum marginal gap was assessed prior to cementation in a natural tooth. Although this study used natural teeth and, it could be argued, is therefore more clinically relevant it still suffers from the same problems as the previous study.

Secondly Qualtrough *et al.*[120], have compared the use of an elastomeric wash and a sectioning technique. No advantage was discerned for either technique. Rather it was suggested that each technique provided complimentary information. However, in view of the small sample numbers and the inappropriate use of impression material as a composite resin luting cement substitute in the sectioning technique, little information can be drawn from this study.

The final technique which has been described involves production of inlays and their cementation into prepared teeth. The teeth were subsequently sectioned and the thickness of composite resin cement lute was measured[92,119]. This gives an indication of the accuracy of fit. This method has the advantage of being more clinically relevant, as the film thickness and other handling properties of the resin cement are taken into account. It also allows assessment of the fit of the inlay along a whole section of the fitting surface. It does, however, result in destruction of the tooth.

9.2 AIMS

The aim of this experiment was to determine the accuracy of fit of the three composite resin inlay systems being studied. Cement lute thickness was taken to be the most clinically relevant measure of this. It was also hoped to determine if there were any particular areas along the fitting surface of the inlay which fitted more accurately than others.

9.3 MATERIALS AND METHOD.

Mesio-occlusal and disto-occlusal cavities were cut in 30 intact, recently extracted human third molar teeth. The teeth were then divided into three groups of ten, one for microfilled inlays and one of 10 for each of the two hybrid systems and stored in water at 5°C until used. Teeth were selected to minimize the variation in inlay volume caused by tooth morphology.

The standardized cavities (Figure 9.1) were prepared initially using a 1.5mm tungsten carbide twist drill in an industrial milling machine (Bridgeport Universal Milling Machine, Bridgeport Machine Ltd., Leicester, U.K.) under water spray. The cavities were then finished using fine diamond burs in a conventional speed dental handpiece. All internal line angles were rounded and all margins were finished to a butt joint.

Impressions were made in an addition silicone impression material (Provil, Bayer Dental, Leverkusen, Germany) using a one stage putty\wash technique. The impressions were stored for 24 hours to allow for stress relaxation before dies were poured in high strength dental stone (Jade Stone, Whip Mix Corporation, Louisville, KY, USA). Composite inlays were fabricated in either a hybrid or microfilled composite resin. Hybrid A inlays were built, light curing the material in 1mm increments, according to the manufacturers instructions. These inlays were subjected to a secondary cure in the D.I. 500 oven. The hybrid B material was built up in the same manner except for the final cure which was carried out using the Unilux A.C. heat/light oven at the end of their fabrication. The microfilled inlays were built up in a single increment and cured in the Ivomat heat/pressure oven. Following curing the microfilled inlays were sandblasted with 50μm aluminium oxide. All the inlays were stored dry, overnight at room temperature and then bonded to the tooth using the composite resin luting cement recommended by the manufacturer.

The teeth were stored in water for 1 week. Following this they were sectioned through the centre of the cervical floor of each restoration. The thickness of the cement lute was recorded at nine points around the fitting surface of each of the two inlays in each tooth (Figure 9.2) using a travelling microscope.

Statistical analysis of differences in lute thickness was determined using ANOVA. Comparison of pairs was carried out using a Students' t-test.

9.4 RESULTS.

The mean thickness of cement lute for the different materials is shown in Table 9.1 and Figures 9.3 and 9.4.

The mean lute thickness, and hence accuracy of fit, was significantly better in the hybrid A (100.7μm, standard deviation 80.9μm) and hybrid B(88.7μm, standard deviation 40.9μm) than the microfilled material (282.4μm, standard deviation 123.2μm) ($p<0.01$). There was no significant difference between the two hybrid materials tested ($p<0.05$). Although there was considerable variation in the thickness of the lute around the fitting surface of the inlay there were no particular areas in which inlays of any type exhibited better or worse fit.

INLAY TYPE	Position on fitting surface									MEAN VALUE
	1	2	3	4	5	6	7	8	9	
Hybrid A	86	79	123	154	118	121	92	72	61	100
S.D.	97	86	68	83	77	80	70	53	57	81
Hybrid B	78	89	91	73	91	103	110	90	83	89
S.D.	43	43	23	31	37	38	38	48	37	41
Microfil	296	264	324	233	250	305	350	321	269	282
S.D.	133	110	98	69	124	126	126	106	77	123

Table 9.1: Mean and standard deviation values of composite resin luting cement thickness (μm) around inlay fitting surface.

9.5 DISCUSSION.

The results show there to be a great difference in the lute thickness and hence accuracy of fit between the hybrid and the microfilled systems tested in this study. The results for the microfilled material are comparable with those of Rees and Jacobson who found lute thicknesses of over 250 μm beneath the same type of inlays[92] and those of Reid *et al.*[90] who found marginal gaps of between 196μm and 266μm. Peutzfeldt and Asmussen also support the findings that the microfilled material tested in this study produced much less accurately fitting inlays than the hybrid materials used[118]. Although, in that study the values for gap size were lower than those recorded in the present one it was axial discrepancy without cement lute which was recorded and it would seem likely that an increase in axial discrepancy would be expected as a result of presence of cement. In fact in the same paper lute thicknesses of "less than 150" and over "400 microns" were mentioned although it was not recorded with which inlay system these occurred.

There are several probable reasons for the differences in fit of the inlay systems. First, the hybrid systems are built up incrementally rather than the single, bulk curing of the microfilled system. This allows some compensation for the curing contraction which occurs during fabrication. Second, the hybrid systems are built directly onto the model with only one layer of separating medium: the microfil has a layer of separator and an activator layer prior to composite build up. Finally, and probably most importantly, the microfilled material is sandblasted following fabrication to ensure a good bond to its luting cement, although this has been shown to be ineffective (see Chapters 7 and 8). This appears, clinically, to remove a significant amount of material from the fitting surface and must adversely affect the fit of the inlay. Recording of the lute thickness as a measurement of fit of the inlay is also influenced by the resin composite luting cement, particularly its film thickness. This was measured according to British Standard no 6039[169] and, in fact, the film thickness of the cement used with the microfilled system was significantly ($p<0.01$) lower than that of the cement used in the hybrid system (mean value of 45μm for the microfil as opposed to 106μm for hybrid A and 137μm for the hybrid B). (See Chapter 4 for details). It appears unlikely, therefore, that this was an important factor in producing the difference noted between the two inlay types. If anything it would be expected to make the result obtained closer than if the same cement was used for both hybrid and microfilled systems.

It has been suggested that the fit of composite inlays is best in the vicinity of the occlusal margins[119]. This was not the case in the present study where no particular area fitted better than any other. Differences in inlay type and sectioning technique may account for the differences between the two studies. However,in Ariyaratnam *et al.*'s[119] paper there was actually no significant difference between the occlusal margins and various other areas, despite the inference to the contrary.

The increased thickness of the lute in the microfilled system may be expected to result in increased wear around these inlays[152,153]. However the microfilled nature of the composite resin luting cement may help to prevent this as it has been shown that this type of cement can be expected to exhibit considerably less wear than the coarser hybrid types as used with hybrid B[275]. The luting resin used with hybrid A can be expected to have a similar wear profile to the microfilled material as it contains very small (sub-micron) particles. Consequently with its thinner lute thickness and potentially more wear resistant cement this system should theoretically, at least, perform best with regard to lute wear *in vivo.* Nonetheless more wear of cement lute is to be expected relative to inlay wear.

The significantly greater lute thickness with the microfilled material may also be expected to place greater stresses on the tooth/restoration interface[121,122]. This will result in a greater chance of breakdown of the bond between tooth and restoration, particularly when a margin is placed on dentine. If the bond breaks and the restoration pulls away from the margin post operative sensitivity would be expected and the potential for occurrence of secondary caries would be increased.

This problem is investigated more fully in the next chapter.

Chapter 10: Microleakage of Three Composite Resin Inlay Systems.

10.1 INTRODUCTION.

10.1.1 Definition and Importance of Microleakage.

Microleakage has been defined as "the passage of bacteria, fluids, molecules or ions between a cavity wall and the restorative material applied to it"[276].

This phenomenon is particularly relevant to composite resin restorations for the following reasons:

(1) they undergo significant polymerisation shrinkage[277,278]

(2) they have a coefficient of thermal expansion greatly different from tooth structure[279]

(3) they are frequently subjected to occlusal forces[280]

(4) they lack a self sealing ability[281]

as described in more detail in chapter one.

Clinically this leakage can cause tooth sensitivity to hot/sweet/cold, or alternatively bacterial penetration[63] and secondary caries can occur which may lead, ultimately, to pulpal damage[64].

Control of leakage has consequently been a major preoccupation for dental researchers, the goal being a leak free filling.

Relatively few studies have been carried out into leakage *in vivo*[282-285] and results have been conflicting[276]. Furthermore, as with any clinical study, they are expensive and time consuming to do results taking several years before they are available. This time delay is of particular importance in the field of dental materials where rapid turnover of products may result in the product being obsolete before meaningful clinical results can be reported. The investigation of leakage has therefore been largely laboratory based with many different techniques being used to assess the sealing ability of dental filling materials.

10.1.2 Methods of Measuring Microleakage *in vitro.*

(a) Air Pressure:

The use of air pressure to test the marginal integrity of restorations was first described by Harper in 1912[286] when he used Class II amalgam restorations placed in a steel die. Compressed air applied through the die resulted in air bubbles being produced where leakage occurred. It was 40 years, however, before this method was adapted for the study of restorations placed in human teeth. Fiasconaro and Sherman[287] and later Pickard and Gayford[288] used compressed air for measurement of leakage around acrylic and amalgam restorations, respectively. In both cases compressed air was applied through the root canals of the teeth in a closed system, and loss of pressure measured as the degree of leakage. Moreover, examination of the restoration margins, for release of air bubbles underwater, gave a view of where the leakage was occurring as well as how much pressure was escaping. This method had the advantage of being non-destructive and also allowed measurement of leakage over a period of time. However, no attempt was made to simulate *in vivo* conditions during experimentation. Also no account was taken of the drying effect of compressed air upon the tooth or restoration. With certain filling materials this could be expected to have a marked effect on long term leakage. Glass ionomer cements for instance are particularly prone to crack on drying and would result in aberrant results as the air could escape through the filling rather than just around its margin.

(b) Bacteria:

The use of bacteria to measure the marginal leakage of a dental restoration would seem, superficially at least, to be the ideal method. Bacteria are, after all, the important factor in causing caries and tooth destruction around restoration margins.

Initial studies used amalgam packed into glass tubing. Fraser[289] looked for bacterial penetration through or around fillings by assessing clouding of culture medium placed below the restoration.Seltzer[290] and Rose *et al.*[291] placed filled teeth in bacterial broth mixtures later removing the fillings and attempting to culture bacteria from dentine scrapings from below the restorations. Bacterial techniques of assessing leakage are still used today[63,292,293] however, they tend to be qualitative rather than quantitative and result in destruction of the tooth. Marginal gaps required for bacterial penetration are large (around 1μm). Therefore smaller gaps which would allow acids or bacterial toxins to penetrate the restoration margin go unrecorded. Finally these studies are difficult and time consuming to carry out and are considered unreliable, by some[294], due to the many variables experienced.

(c) Neutron Activation Analysis:

This method has been used both *in vivo* and *in vitro* by Going *et al.*[285], and, more recently *in vitro* by Douglas *et al.*[295]. Restored teeth are soaked in non-radioactive manganese solution. They are then placed in the core of a nuclear reactor which converts the manganese to its radioactive form. Subsequent measurement of X-ray emission from the teeth is a measure of isotope uptake and hence microleakage.

This method has several obvious problems.

(1) Although supposedly quantitative it does not take into account the uptake of isotope in areas other than at the tooth/filling interface.

(2) It is very expensive requiring access to a nuclear reactor and services of a nuclear scientist.

(3) The use of radioactivity makes it relatively more hazardous than a simple dye or chemical tracer study.

(4) It does not show which areas along a restoration leak ie. does leakage occur when a margin is on dentine or enamel?

(5) Some filling materials can contain manganese or absorb manganese into their surface eg. silicate.

(d) Electricity:

Electrical conduction through marginal gaps in restorations has also been studied[296-300]. An electrode is placed within a filled tooth and the tooth, in turn, is placed in an electrolyte solution. Following production of a voltage between tooth and bath, the current flowing is a measure of the leakage present. Although this method can be used to test leakage over time *in vitro* it does not show where the leakage occurs. Obviously this method cannot be used with metal containing restorations.

(e) Artificial Caries:

Secondary caries like lesions can be produced *in vitro* by the use of cultured bacteria[301] or artificial caries media such as acidified gelatin gel[302-304]. It has been suggested that this method is clinically relevant since it links microleakage with one of its consequences ie. cavity wall demineralisation. Quantification of the result is also feasible using microdensitometry or polarised light microscopy. Usually, depth of lesion penetration is the measured parameter[276].

Grieve and Glyn Jones[305], however, noted that acid etching alone was sufficient to produce a mild demineralisation along the cavity wall and hence artificial caries may not be suitable for examining margins of acid etched retained materials.

(f) Microscopy (Direct Visualisation):

Although not directly a measure of leakage microscopic analysis of marginal gap formation is a popular way of assessing potential seal of a restoration. Assessment may be made using either light or scanning electron microscopy.

Asmussen and his co-workers have used light microscopy extensively in their assessment of gap formation following use of various dentine bonding agents[306,307].

Scanning electron microscopy has also been used but can introduce errors and artifacts due to the drying necessary during sample preparation[308]. If, however, replicas are used, as described by Barnes[240,241] these problems can be overcome. With this method good visualisation of defects is possible due to the high magnification and good depth of field.

Although Qvist[309] has found good correlation between marginal gap size and bacterial microleakage *in vivo*, microscopy does not show definitively where leakage will occur only where a gap is on the surface. It is for instance quite possible that a filling may debond at the surface but be perfectly sealed 10μm from the surface, allowing only minimal leakage. This would not be detectable by microscopy alone.

(g) Radioactive Isotopes:

Many different radioisotopes have been used for assessment of marginal leakage. ^{45}Ca, ^{131}I, ^{35}S, ^{22}Na, ^{32}P, ^{86}Rb,^{14}C have all been used at one time or another with no particular rational for the use of any specific one. ^{14}C in sugar molecules was used, for instance, in one study[310] as sugar usage leads to caries. However penetration of sugar molecules *per-se* does not cause caries, also microleakage of this particular molecule is no more valid than any other radioactive molecule of a similar size.

It has been suggested by Going *et al.*[282] that the small size of radioisotopes provides a more accurate measure of leakage than other methods. However, later he reported[311] that the accuracy is too great and that leak free fillings are virtually impossible with this method. Another possible failing with this method is that ions may react with filling material or tooth structure resulting in incorrect leakage scores. The procedure is relatively complex and costly, generally requiring autoradiography of sectioned specimens and, as with many of the other techniques described leakage is only measured qualitatively.

Two closely allied techniques which allow some quantification of the leakage occurring have also been described.

(1) Radiochemical diffusion[312] which uses a diffusion cell in conjunction with a radioactive ion.

(2) Reverse Radioactive Absorption[313] in which radioactive leucine is placed beneath a restoration. Subsequent monitoring of the radioactivity of a surrounding solution gives a measure of the amount of leakage occurring.

(h) Chemical Tracers:

Chemical tracers are distinct from dyes in that they do not show leakage on their own but require a second chemical to cause a visible effect.

An early use of chemical tracers was described in 1953 by Kornfield[314]. In this barium sulphate solution reacted with lead glass in a filling material to form a black precipitate where the restoration leaked.

The most commonly used solution today is 50% aqueous silver nitrate in which the teeth are immersed *in vitro*[315-317]. The silver is subsequently reacted with photographic developer so that a black stain is seen.

A similar technique is described by Lienfelder *et al.*[318] in which soluble calcium hydroxide from a lining cement is used to demonstrate leakage at restoration margins with sensitive pH paper being used as indicator.

(i) Dye Penetration Studies:

This is the most commonly used method for microleakage detection. In general the restored tooth is placed in a dye for a predetermined period before washing and sectioning. The dye produces a clear record of leakage in a contrasting colour to both tooth and restoration without need for further chemical application, or autoradiography.

The main problems with this technique are:-

(1) Often only a single section is taken through a tooth. Hence one may pick up the only area where leakage occurs or alternatively the only area where it does not.

(2) Assessment is often carried out using a scoring system. This can be somewhat subjective; even when more than one observer is used.

(3) Leakage depends on the size of the dye molecule or particle.

Dye techniques are therefore generally qualitative, or at best semi quantitative, rather than quantitative a problem shared with many chemical tracer and radioisotope studies. Attempts have been made to use dyes quantitatively by Douglas *et al.*[319] and notably Retief and his co-workers[320,321]. In this method the tooth is dissolved in a known amount of acid solution and the resulting coloured solution is examined spectrophotometrically to determine the dye concentration and hence the amount of leakage which has occurred. This technique has the obvious problem of dye penetration elsewhere than at the tooth/restoration interface, e.g. cracks in and around the cavity or in the restoration itself, giving a falsely high leakage value. Retief, however, claims that this is not a problem(?).

Recently a technique for three dimensional microleakage assessment[322] has been reported. In this method a typical dye penetration study is carried out. Serial sections are then produced and the sections are subjected to image analysis. From this it is possible to quantify the volume of dye penetration and the areas of the cavity where leakage occurs. Cavity area and tooth volume are also recorded. This technique has, as yet, only been reported as a pilot study, but shows potential as a more quantitative method of assessing leakage *in vitro.*

Many different dyes have been used to assess microleakage of composite restorations and a summary of those used commonly is presented by Taylor and Lynch[323]. No one dye appears to be more suitable than any other although variation has been shown between different dies and even between different concentrations of the same dye[324]. There does not appear to be an 'ideal' dye at present. Providing the same dye and same concentration of dye are used throughout an experiment and no attempt is made to compare, it except in the broadest terms, with any other study using a different dye any of those used routinely should be acceptable.

When choosing a dye the following criteria are important:

(1) Must not bind to tooth or restoration.

(2) Must be small enough to penetrate dentinal tubules (1-4μm).

(3) Must be stable. Aniline blue, for instance, is discoloured by alkali such as calcium hydroxide lining cements[308].

10.1.3 Mouth Simulation *in vitro*:

Attempts have been made to simulate some of the conditions which occur in the mouth to make laboratory based testing more representative of the conditions in which restorations have to function. These procedures have mainly involved mechanical abrasion, mechanical cycling and, most often, thermal cycling.

(a) Mechanical abrasion has generally been carried out using toothbrushing machines[325,326]. The latter two of these studies suggested that glazing restorations with unfilled resin could decrease microleakage. However, any improvement inevitably proved short term as the resin was worn away relatively quickly. Furthermore, this technique is rather impractical as margins which leak most[327], i.e. cervical margins, are both inaccessible and often have no suitable enamel for etching.

(b) Mechanical cycling or loading of teeth results in deformation of the cavities and 'mechanical percolation' as described by Jorgenson *et al.*[280]. In non-bonded cavities he found cycling led to transient or permanent gaps. Bonded, acid etched, cavities were much less likely to develop gaps when loaded. These results were supported by Raadal *et al.*[328] and by Stewart *et al.*[329] who found load cycling had no effect on composite bonded to etched enamel. Similar results were obtained by Derbyshire *et al.*[330] when glass ionomer cement and a dentine bonding agent were applied to dentine. Mandras *et al.*[331] found no difference in leakage in Class V cavities in canine teeth, with or without load cycling, when Scotchbond 2 dental adhesive was used. In larger cavities in molar teeth some difference was noted. However as marginal gap dimensions are related to cavity diameter[332] the increased cavity size may have been the cause.

(c) Thermal cycling is by far the most commonly used method of simulating oral conditions and stressing the tooth/filling interface. The acceptance of this procedure as an important part of microleakage testing is due in a great part to the work of Nelsen *et al.*[333], Coy[334] and Paffenburg *et al.*[335]. In the early 1950's they showed that the differential coefficients of thermal expansion between tooth and restoration caused "percolation" and subsequent microleakage of these materials.

The range of temperatures used by different researchers varies considerably but is generally chosen to simulate the upper and lower ranges of temperature found in the mouth (4-8°C minimum and 45-60°C maximum).

The dwell times in the baths at the temperature extremes have also been variable. Times of one minute have been used by some authors[315,336-338] whereas others have used much shorter times of 15 seconds or less[339-341]. The use of shorter dwell times would seem to be of more clinical relevance as, work done *in vivo* to assess normal intra-oral temperature variation suggests that teeth are in contact with temperature extremes for only a very short time[342,343]. The idea of allowing a tooth to equilibrate in temperature with a solution at 5°C or 55°C is unreasonable. No patient would tolerate such temperatures applied to their teeth for more than a few seconds as attested by the use of heat and cold in sensitivity testing of teeth.

The number of temperature cycles also shows a great range, from 1 to 2500[276]. However, short term cycling (100's) has been shown to be equally effective in showing microleakage as longer term (1000's)[344]. It would seem logical, therefore, from a purely pragmatic viewpoint to opt for a less time consuming short term cycling regime.

The final variable which occurs in thermocycling regimes is whether or not to cycle in the dye or isotope being used as tracer. At present there is no consensus however it has been argued that there is no difference between the two methods[345].

As mentioned at present the use of thermal cycling as part of a leakage experiment is accepted by most researchers as it has been shown to increase the degree of leakage observed[346]. Nonetheless, there have been some papers which question its use as a method of stressing the tooth/restoration interface. Wendt *et al.*[347] for instance states "thermocycling microleakage specimens for the purpose of analyzing the effects of coefficients of thermal expansion in composites should be discontinued". While it is true that the thermal diffusion through composite restorations is relatively slow, and consequently dimensional change due to the coefficient of thermal expansion will be less than expected, when clinically relevant (i.e. short dwell time) thermocycling regimes are used[343], it would be unreasonable to stop thermocycling. It is obvious that some degree of differential expansion and contraction of restorations occurs in the mouth, and consequently some attempt should be made to mimic this *in vitro*. Dwell times should be kept short and baths should include a return to a normal physiological temperature between hot and cold. Even accepting these constraints many of the procedures used will still be more severe than conditions occurring in the mouth. At best an *in vitro* experiment can hope to correctly compare the materials within the bounds of that experiment. Hopefully this will allow some conclusions to be drawn which may guide towards a clinical procedure. However until all the factors important in restoration leakage *in vivo* can be quantified, any *in vitro* experiment is, at best, a rough guide to what may occur in the mouth. In the meantime a procedure, such as thermocycling, which can justifiably be expected to produce a more lifelike result should be adopted.

10.1.4 Microleakage of Composite Resin Inlays.

The rational for the use of composite resin inlays is described in the Introductory Chapter.

Previously mentioned studies have shown that the marginal seal of the inlay restorations is indeed superior to direct composite resins[88,105-109,112]. However, leakage although it is reduced with the inlay technique still occurs, particularly when a margin is placed on dentine. The use of some dentine bonding agents in conjunction with these restorations has also been shown to decrease microleakage, however it is still not completely eliminated[112,113].

It would seem obvious that the thin layer of composite resin cement, used in the inlay technique, will contract less than a direct composite resin filling. This in turn should produce lower contraction forces on the bond between resin and tooth. However as mentioned in the previous chapter there may still be problems associated with the contraction of this thin layer. Previous work by Feilzer *et al.*[266], pointed out that the increased wall-to-wall polymerisation contraction in thin resin layers may be substantial, and went on to calculate a shrinkage stress of 17.6 MPa for a class I composite resin inlay. Other workers have also estimated likely forces to be between 4 and 10 MPa[79,121].

The variation in cement lute thickness and accuracy of fit of composite inlays has been described fully in the previous chapter. A large difference in fit between the microfilled and the two hybrid systems was noted. As it has recently been suggested that an increase in the thickness of the cement lute results in increased shrinkage stress generated during curing[121,122]. This increase in stress may be expected to have an adverse effect on the seal of the restoration.

10.3 AIM.

The purpose of this study was to investigate the effect of using unfilled resin or, one of a number of, dentine bonding agents on the marginal microleakage of three proprietary inlay systems. In addition it was hoped to discover if the variation resin composite cement lute thickness, described in the previous chapter, had any bearing on the marginal seal of these restorations.

10.4 MATERIALS AND METHOD.

10.4.1 Cavity Preparation and Inlay Fabrication.

Mesio-occlusal and disto-occlusal cavities were cut in 235 intact, recently extracted human third molar teeth. The teeth were then divided into three groups, one of 75 for microfilled inlays, one of 90 for hybrid A inlays and one of 60 for hybrid B inlays, and stored in water at 5°C until used. Teeth were selected to minimize the variation in inlay volume caused by tooth morphology.

The cavity preparation and inlay fabrication was identical to that in Chapter 9. One important factor in preparation of the standard cavity was that due to the selection of the depths of the conventional cavity outline, each tooth had one box with its margin bounded in enamel while the other had its cervical margin in dentine, this being determined randomly.

10.4.2 Bonding Procedure.

Fifteen teeth were allocated randomly to five groups for the microfilled material, six groups for hybrid A and four groups for the hybrid B. All enamel surfaces were etched using 37% orthophosphoric acid for 60 seconds, washed for 30 seconds, and dried with oil-free compressed air. In one group, of each inlay type, the inlays were cemented using the unfilled resin produced for that system. In the other groups, exposed dentine was treated with dentine bonding agent prior to cementation. All samples were then cemented using the dual curing resin cement produced for each respective system (see Chapter 2 for details of bonding resins and cements).

Details of the dentine bonding systems are shown in Table 10.1. The restored teeth were stored in water at room temperature for one day. The inlays and margins were then polished with Soflex discs (3M Dental Co. St Paul, MN, USA) and replaced in the water for a further six days.

Inlay Type	Bonding Agent Used	Dentine Treatment
Hybrid A	Duo Bond	None; Resin applied to dentine and light cured
	Gluma[a]	0.5% EDTA applied to dentine,washed. 5% gluteraldehyde, 35% HEMA applied, dried. Unfilled resin applied not cured.
	Mirage Bond[b]	2.5% nitric acid,NPG applied,dried. PMDM applied, dried. Duo Bond resin applied,cured.
	Scotchbond Dual Cure[c]	Liquid and resin mixed applied to dentine and cured.
	Scotchbond 2[c]	Maleic acid/HEMA applied,dried. HEMA/Bis-GMA applied, cured.
	Coltene experimental bond	Base & catalyst mixed, applied, dried. Dentine/Enamel bond applied, cured
Hybrid B	Adhesive Bond	None, resin applied to dentine , cured.
	Gluma[a]	Same as in hybrid A except unfilled resin light cured.
	Mirage Bond[b]	Same as in hybrid A
	Denthesive	Aqueous EDTA applied to dentine, washed. Adhesive applied, dried. Unfilled resin applied, cured.
Microfilled	Heliobond	None,resin applied to dentine, cured.
	Gluma[b]	Same as hybrid B.
	Mirage Bond[a]	Same as both hybrid materials.
	Dentin-Protector	Dentine cleaned 3% hydrogen peroxide, washed, dried. Adhesive applied, dried. Unfilled resin applied, cured.
	Syntac	Aqueous, aliphatic dimethacrylate and acetone applied, dried. Aqueous, dimethacrylate and 5% gluteraldehyde applied, dried. Unfilled resin applied, cured.

Table 10.1: Inlay groups bonded, details of dentine treatment and bonding agents used.

a. Bayer Dental, Leverkusen, Germany., b. Chameleon Dental Products, Kansas City, KS, USA., c. 3M Dental Co., St Paul, MN, USA.

10.4.3 Microleakage Assessment.

The samples were thermocycled immediately prior to testing at 5°C, 37°C, 55°C and 37°C for 500 cycles. The dwell time in each bath was 10 seconds.

Following thermocycling, the apices of the teeth were sealed with a cyanoacrylate adhesive and the teeth were coated with two layers of nail varnish leaving a band approximately 1mm wide exposed around the restoration. The teeth were immersed in a 2% aqueous solution of methylene blue dye. After 15 hours they were rinsed thoroughly with tap water and the teeth subsequently sectioned, with a diamond disc, through the centre of the cervical floor of the restorations.

Leakage was assessed blind by two examiners. Each specimen was examined under 6x magnification and the extent of leakage was scored according to the following criteria:

0 = No leakage.

1 = Dye penetration up to half way along floor of box.

2 = Dye penetration to full depth of box.

3 = Dye penetration along axial wall.

4 = Extensive dye penetration towards pulp.

(Examples of each level of leakage are shown in Figures 10.1-10.5)

The microleakage data were analyzed using the Kruskal Wallis test, corrected for ties, to determine if there were any significant difference between the groups. Comparison of pairs of groups was made using the Mann-Whitney U test.

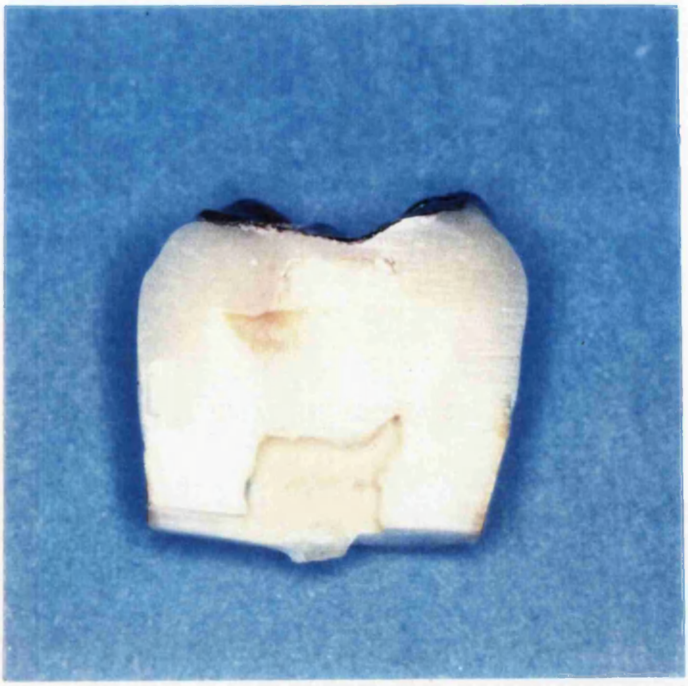


Figure 10.1 Composite resin inlay with leakage score 0 on both enamel and dentine margins.

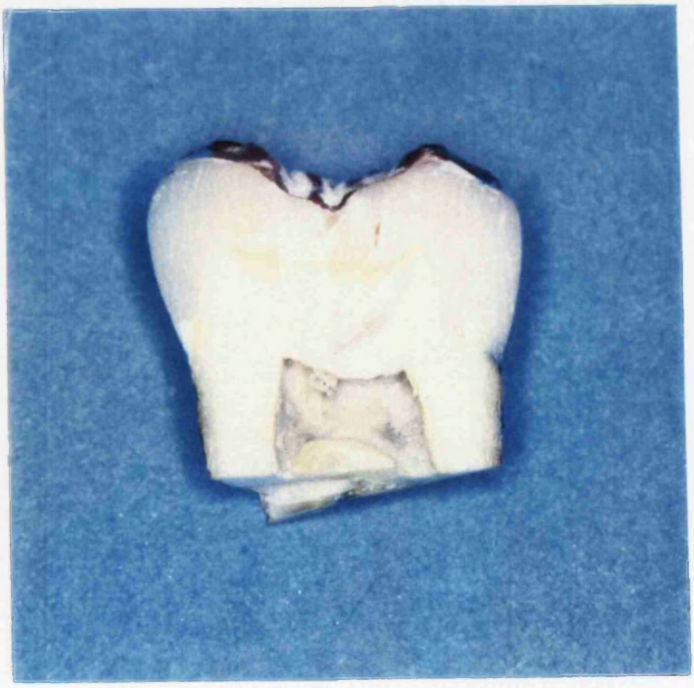


Figure 10.2 Composite resin inlay with leakage score 0 on enamel and 1 on dentine.

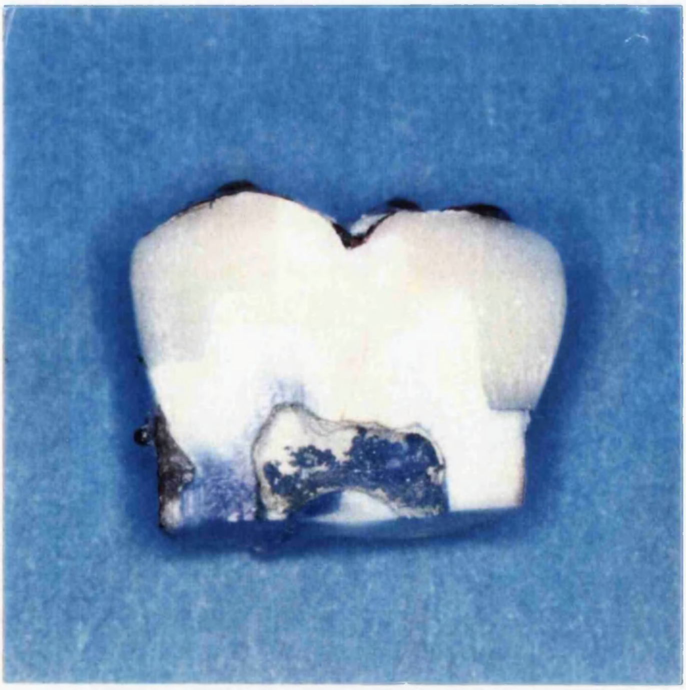


Figure 10.3 Composite resin inlay with leakage score 0 on enamel and 2 on dentine.

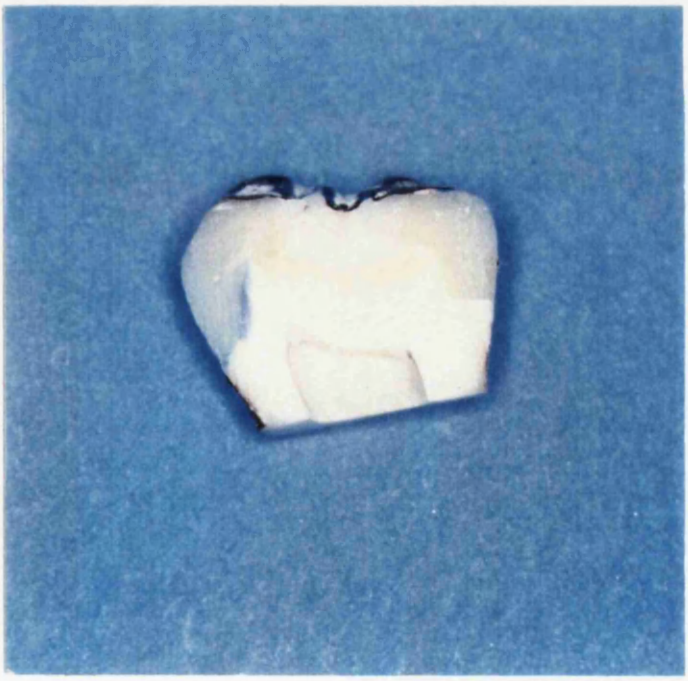


Figure 10.4 Composite resin inlay with leakage score 0 on enamel and 3 on dentine.

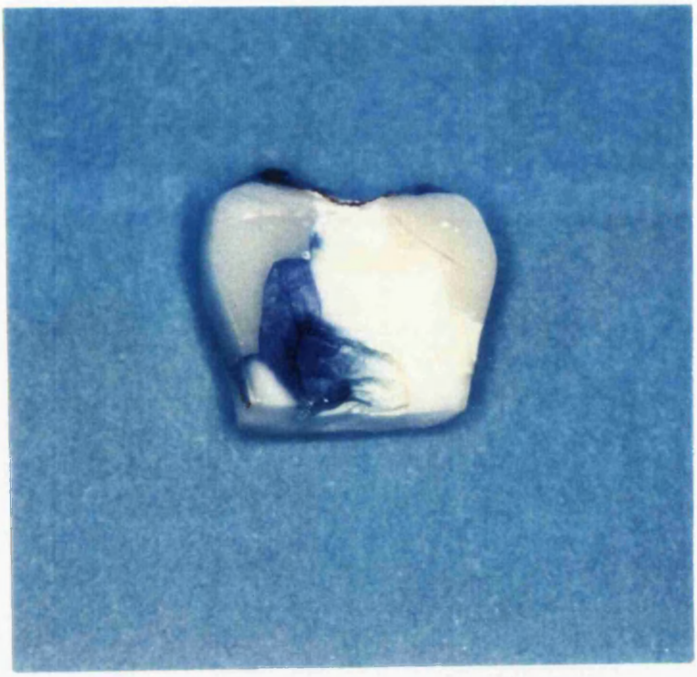


Figure 10.5 Composite resin inlay with leakage score 0 on enamel and 4 on dentine.

10.5 RESULTS.

Marginal leakage in enamel is shown in Table 10.2. For margins bounded in enamel there was little marginal leakage with any of the systems studied. There were no significant differences between any of the systems ($p<0.05$).

Inlay Type	Bonding Agent	Marginal leakage score				
		0	1	2	3	4
Hybrid A	Duo Bond	11	0	2	0	2
	Mirage Bond	12	2	0	0	0
	Gluma	13	1	0	0	0
	Scotchbond Dual Cure	10	4	0	0	1
	Scotchbond 2	11	2	1	0	0
	Coltene experimental bond	11	2	2	0	0
Hybrid B	Adhesive Bond	12	3	0	0	0
	Gluma	13	2	0	0	0
	Mirage Bond	14	1	0	0	0
	Denthesive	11	3	0	0	1
Microfilled	Heliobond	13	2	0	0	1
	Gluma	9	2	2	1	1
	Mirage Bond	14	1	0	0	0
	Dentin-Protector	13	2	0	0	0
	Syntac	12	3	0	0	0

Table 10.2: Leakage scores in enamel

Table 10.3 shows the frequency of leakage scores in dentine for each group.

Inlay Type	Bonding Agent	Marginal leakage score				
		0	1	2	3	4
Hybrid A	Duo Bond	1	1	2	0	12
	Mirage Bond	10	3	1	0	0
	Gluma	5	2	1	1	5
	Scotchbond Dual Cure	1	2	1	1	9
	Scotchbond 2	4	2	2	1	6
	Coltene experimental bond	5	2	4	2	2
Hybrid B	Adhesive Bond	0	2	2	3	8
	Gluma	8	3	2	0	2
	Mirage Bond	10	3	2	0	0
	Denthesive	8	4	1	1	1
Microfilled	Heliobond	2	1	0	2	10
	Dentin-Protector	1	2	1	1	10
	Gluma	7	6	2	0	0
	Mirage Bond	9	4	2	0	0
	Syntac	5	6	1	2	1

Table 10.3: Leakage scores in dentine.

All the third generation dentine bonding agents tested were significantly better at reducing leakage than unfilled resin at the dentine margin ($p<0.05$) in the microfilled and hybrid B systems. With hybrid A only Mirage Bond and Coltene experimental bond were significantly better at reducing microleakage than unfilled resin alone ($p<0.05$). The second generation (Dentin Adhesit) material used with the microfilled system was no better at reducing microleakage than unfilled resin alone. There was no significant difference between any of the third generation dentine bonding agents used ($p<0.05$) within inlay groups except with hybrid A where Mirage Bond proved the most effective ($p<0.05$). There was no difference in leakage between inlay systems when the same bonding agent was used with each. This would indicate that there is no difference in leakage relative to the thickness of the composite resin lute (see Chapter 9 for lute variation between inlay types).

10.6 DISCUSSION.

The fact that there was little leakage in enamel but severe leakage on dentine margins when no dentine bonding agent was used is in agreement with previous studies[106,112]. The good bond strength of resin composite to etched enamel is well documented, and appears to be sufficiently high to oppose the stresses imposed upon it during this experiment. Only 7 out of 210 inlays showed moderate to severe leakage in enamel. No significant difference occurred between any of the systems used on enamel. As all groups used 37% phosphoric acid gel etchant and were bonded with unfilled resin this is exactly as would be expected. The dentine bond however is known to be more fragile and liable to breakdown due to polymerisation of the resin composite cement and the subsequent thermal stresses placed upon it during thermocycling.

It would seem logical that a thin layer of composite would contract less than a thicker layer and place less stress on the tooth/restoration interface, a particularly important factor when bonding to dentine. However, from these results it appears that in many cases even with a dentine bonding agent the bond strength is still inadequate. The report of Feilzer *et al.*[266] in which it was suggested that reduction in thickness of a resin layer restricted its ability to flow and dissipate its polymerisation contraction; hence the volumetric contraction tended to a linear contraction at right angles to the restoration, would seem to be important. Even if forces of only 4-10 MPa are to be expected[79,121] these are enough to debond the older second generation materials such as Scotchbond Dual cure and Dentin Adhesit. These second generation bonding agents do not remove the smear layer and have been shown to have low bond strength and sealing ability in previous studies[348,349]. Dentin Adhesit for instance has been superseded recently by Syntac bond which proved significantly more effective in this study ($p<0.05$).

All the other agents used in this study remove or alter the smear layer in some way, and have been shown to have significantly higher bond strengths than earlier materials. The fact that these materials reduced leakage on a dentine margin is in accordance with previous work[112,113]. The result with Gluma, when used with hybrid A, is different from the other groups; it being significantly less effective than Mirage Bond. This may be due to curing of the unfilled resin on the dentine surface prior to inlay cementation in both hybrid B and microfilled systems and not in hybrid A. This adaptation in the manufacturers instructions came about during the course of the experiment, and was adopted, as it has been shown to be an important factor in decreasing microleakage[350]. The results for Scotchbond 2 with hybrid A were rather disappointing as it was significantly no better than unfilled resin at reducing microleakage. Nonetheless, it was as effective as any of the other bonding agents used in any of the inlay groups apart from Mirage Bond in hybrid A. It would therefore seem possible that the lack of difference observed was due, at least in part, to the large standard deviations common in experiments of this type.

The dentine bonding agents recommended by the manufacturer for use with their own systems performed reasonably reducing leakage significantly ($p<0.05$) when compared with unfilled resin alone. However, they did not perform better than the other agents tested, hence no particular recommendation can be made for a specific bonding agent to be used with a particular inlay system. In fact although supposedly "new" both dentine bonding agents for the hybrid systems were very similar in composition to existing agents. Coltene Experimental Bonding agent, recently marketed as ART bond, has a very similar formulation to Scotchbond 2 both being basically a maleic acid primer and a HEMA resin. Denthesive and Gluma are also very similar with EDTA as dentine treatment and HEMA as primer. As might be expected there was no significant difference, in leakage, between these "new" materials and the older established products.

From the differences in lute thickness it might have been expected that the microfilled system would show more leakage than the hybrid system. However, this was not the case when the same dentine bonding agent was considered for all three systems. Although it has been suggested that the contraction force is greater in inlays with a greater lute thickness this increase in force may not be sufficient to disrupt the adhesive bond to dentine. It may be that the dentine bonding agents tested with the three systems (Mirage Bond and Gluma) were strong enough to counteract these forces, even in the system with the thicker lute. If the increase in force of contraction is only in the order of, from 7.4MPa to 10.2MPa, as has been suggested[121], then recorded bond strength of an efficient third generation dentine bonding agent should be able to counteract this force and reduce microleakage, as found in this study. This is supported by the work of Peutzfelt and Asmussen[118] who found gap free inlays when Gluma dentine bonding agent was used even when the cement lute was 400μm thick.

It has also been shown that microfilled resins undergo hygroscopic expansion when stored in water, to a greater extent than hybrid materials[279]. This expansion may help counteract any increase in polymerisation shrinkage and go some way to explain the lack of difference observed between the two inlay types. Furthermore, the lower flexural modulus of the microfilled inlay material and it's composite resin cement relative to the hybrid materials (Chapters 3 and 4) may allow the contraction forces to be dissipated by flexion of the material to a greater extent than in the hybrid inlays.

Even if the increase in lute thickness does not have an adverse effect on the inlay's sealing ability a thick lute should still to be avoided due to the potential for wear described in Chapter 9.

10.7 CONCLUSIONS.

This in vitro study showed:

1. Composite resin inlays leak extensively when placed on a dentine margin.
2. When placed on an enamel margin little leakage should be anticipated.
3. The use of a third generation dentine bonding agent will reduce, but not eliminate, this leakage.
4. Despite the increase in mean luting cement thickness, from 88μm in hybrid B to 282μm in the microfil, the seal of the inlay was not adversely affected.

Chapter 11. Final Discussion and Conclusions.

The aims of this study as detailed in Chapter 1 were as follows:

1. Assessment of the basic physical properties of three composite resins for use in the inlay technique both with, and where appropriate, without a post curing regime. Assessment was to continue for at least two months to assess possible annealing and degradative changes in the materials.
2. Assessment of fracture toughness of the three materials, with and without post curing, over a 2 month period.
3. Assessment of physical properties of the composite resin cements supplied with each system both with and without light curing.
4. Assessment of the degree of methacrylate conversion of each material with and without post-curing.
5. Measurement of bond strength of inlay to composite resin inlay and investigation of the bonding mechanism with particular reference to the potential for micromechanical bonding.
6. Measurement of accuracy of fit of inlays of different types.
7. Assessment of leakage of inlays in enamel and dentine. Further investigations into the influence of various dentine bonding agents and accuracy of fit on the leakage of the respective systems.

In general these aims have been met throughout this study.

Confirmation was provided that it is possible to improve some of the physical properties of composite resin filling materials by subjecting them to a secondary cure with heat and light. However, which properties can be improved, and by how much, would appear to be dependant on the material and the post curing regime adopted. A long (20 minute), low temperature (60°C) cycle would appear to be as effective as a shorter (7 minute) higher temperature (120°C) post cure. Decreasing the curing cycle duration of the lower temperature process, however, will decrease the post curing effect. It is necessary, therefore, to ensure that shortening of the prescribed post curing cycle does not occur, if the maximum benefit of this process is to be achieved. Furthermore, one post curing procedure, using the Translux lightbox, cannot be recommended as it fails to produce a suitable curing regime to produce any significant improvements in physical properties. Light would not appear to be important in producing a post curing effect, in the systems tested. The light sources employed would appear to be acting solely as heating elements in the curing chamber. No significant change was noted in either light cured or post cured materials following storage at 37°C in water for periods of up to two months. This would suggest firstly, no annealing process with resultant improvement in physical properties is to be expected in light cured composite resins and, secondly, no degradation of post cured composites, relative to light cured materials, is to be expected over this period.

The curing of a microfilled composite resin by heat and pressure resulted in a material with very high compressive and diametral tensile strengths. It seems likely from this and other sections in this study that this effect was mainly due to the pressure decreasing the porosity within the material.

However, what effect the improvements in physical properties may have on clinical performance of these materials remains open to speculation. McCabe and Kägi[99] have suggested that no appreciable effect would be expected from the changes observed. While this may be the case with compressive and diametral tensile strengths, where generally no changes were seen between light cured and post cured composites, the improvements in flexural strength, flexural modulus and hardness cannot be overlooked. Improved flexural properties may be important in durability of restoration margins where bending of the restoration edge, as described by Rees and Jacobsen[121] will be important. This improved durability of the restoration margin is supported *in vivo* by Wendt and Leinfelder[141,142] where there was a decreased incidence of marginal breakdown of heat cured composite resin inlays relative to light cure only composite resin inlays over a three year period.

The increase in Knoop hardness observed following post curing could be expected to improve wear resistance[351]. This proved to be the case *in vitro* where Wendt [20] noted an increase in wear resistance of over 70% following post curing. However, correlation between *in vitro* testing and *in vivo* findings is generally poor. Despite claims to the contrary[352] even the best *in vitro* testing apparatus, with the most lifelike oral simulation, fails to achieve 85% correlation with clinical findings[353]. Moreover, the laboratory system used by Wendt[20] was never suggested for use as a predictor of *in vivo* wear, only being designed for *in vitro* comparison of dental materials[354]. Nonetheless, the lack of improvement in wear resistance, observed clinically[141,142], is rather surprising. However, bearing in mind that, to date, only one study has been published describing clinical wear of a post cured composite resin inlay, and this study did not use materials suggested for this purpose, further studies are required to assess whether the improvements in physical properties observed in this, and other[18-20,94-100], studies do translate to any observable clinical advantage.

The assessment of fracture toughness showed no advantage was obtained by using a post curing process, both light cured and post cured materials having similar values, while the heat pressure cured material achieved comparable K_{IC} to literature values for similar microfilled composite resins. This suggests that in addition to the higher modulus of the resin matrix following post curing, which would lead to a higher value of K_{IC}, some balancing effect must be occurring to decrease the resistance to crack propagation, probably as a result of some internal stress concentration. This evidence together with that from Chapter 3 would suggest that annealing of composites does not occur either at mouth temperature nor at the raised temperatures which occur during post curing. If relaxation of internal stresses does occur at 120°C then a "low stress" material may occur at this high temperature. However subsequent cooling, to oral temperatures, will inevitably result in build up of stress within the material due to the differential in expansion coefficients between filler and matrix. Composite resins do not behave like metals and description of an annealing effect is probably inappropriate.

Further investigation of composite structure, possibly using Nuclear Magnetic Resonance Imaging[355], would be required to discover exactly what occurs to the internal structure of a composite resin during, and following heat treatment.

Increase in methacrylate conversion of the composite resin materials would appear to be the mechanism by which post curing leads to an improvement in physical properties. It seems likely that heating of the composite resin material results in increased molecular mobility allowing more C=C groups to react. This was confirmed by the recorded increases in methacrylate conversion of the two hybrid materials studied (7% and 12% increases being recorded for hybrid A and hybrid B respectively). It has been suggested[78] that it is necessary to heat the composite to above the glass transition temperature (100°C) to allow increased mobility and subsequent greater degree of cure to occur. Consequently, the increase in methacrylate conversion which occurs following curing in the Unilux AC appears difficult to explain, as this oven only heats to around 60°C. However, Dionysopoulos and Watts[104] have shown that significant amounts of molecular motion will occur at lower temperatures in light cured composite resins. Therefore it seems likely that provided the curing cycle is lengthened post curing at lower temperatures (>50°C) should be possible. This hypothesis was borne out when, as mentioned earlier, no improvement in physical properties was seen when the low temperature post curing cycle is shortened.

Curing using a heat and pressure regime did not result in any greater degree of cure than heat and light curing, despite suggestions to the contrary[126].

The suggestion that composite resin inlays lack sufficient C=C bonds to allow bonding to composite resin cement appears inaccurate. Bond strength testing showed adequate bonds could be obtained with both hybrid systems, however, the microfilled system failed to bond adequately. As all three inlay types had similar methacrylate conversion this could not be the cause of bond failure. SEM and confocal light microscopy suggested that the differences in bonding ability were due to differences in the morphology of the fitting surface. Both hybrid systems exhibited significant porosity and surface roughness which allowed resin penetration and micromechanical adhesion to occur. The microfilled material exhibited no porosity and little surface roughness, despite sand blasting. This resulted in poor micromechanical bonding which, in turn, probably led to poor bond strength. This is a serious failing in this system and requires further investigation. However, at present it would be difficult to recommend the use of this heat\pressure cured microfilled system.

The accuracy of fit of the three composite inlay systems was assessed using a tooth sectioning technique. The two hybrid systems showed lute thicknesses of 100μm and 89μm for hybrids A and B respectively. The microfilled system exhibited significantly poorer fit with a lute thickness of more than double the other systems. It seems likely that the single increment build up technique and the sandblasting of the fitting surface were responsible for this inaccuracy. This poor fit may be of significant clinical consequence and again, make it difficult to recommend the use of this system.

Leakage of composite resin inlays occurs to the same extent as in any other resin or resin bonded restoration. Leakage is low on enamel bonded margins but severe where a margin is in dentine. The use of some dentine bonding agents proved successful in reducing leakage with the more modern agents which alter or remove the smear layer proving most successful. Interestingly the leakage of the microfilled inlays was similar to that or the hybrid materials despite the discrepancy in fit; which may have been expected to increase the stress on the tooth\inlay bond. This would suggest that potential for wear of the thicker composite resin lute in this system may be of more consequence than the perceived difficulty in producing an adequate seal.

Work is currently under way assessing the potential for increased wear with regard to increased thickness of luting agent.

Finally the investigations into the composite resin luting cements clearly outlined the hazards of relying on the self curing mechanism of dual curing resins. The significantly poorer physical properties obtained when chemical curing occurred alone, combined with the increases in time required when curing through inlay materials suggest the need to reappraise curing regimes when using these materials. Only through significant lengthening of light exposure, by three or four times, can the ultimate properties of these materials be realised. To call these materials dual curing is not, strictly, untrue but may be considered at least misleading.

REFERENCES.

1. Roulet J.F. The problems associated with substituting composite resins for amalgam: a status report on posterior composites. *J Dent* 1988;16:101-113

2. Molin C. Amalgam - Fact and fiction. *Scand J Dent Res* 1992;100:66-73

3. Simonsen R.J. New materials on the horizon. *J Am Dent Assoc* 1991;122:25-31

4. Combe E.C. Cements based on ion leachable glass. In: *Notes on Dental Materials* 4th Edition. Edinburgh: Churchill Livingston. 1981;113-121

5.Paffenbarger G.C. Dental cements, direct filling resins, composite and adhesive restorative materials. *J Biomed Mater Res* 1972;6:636-639

6. Smith D.C. Posterior composite dental restorative materials: materials development. In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:

7. McLean J.W. and Short J.G. Composite anterior filling materials. *Br Dent J* 1969;127:9-18

8. Bowen R.L., Barton J.A.Jr and Mullineaux A.L. Composite restorative materials. In: *Dental Materials Research, Nat Bur Stand Specials Publ. 345,* Gaithensburg Md: National Bureau of Standards 1972:93-100

9. Bowen R.L. Dental filling material comprising vinyl silane treated fused silica and a binder consisting of the reaction product of Bisphenol and Glycidyl Acetate. U.S. Patent No. 3066112 Nov 1962.

10. Ruyter I.E. and Osyaed H. Composites for use in posterior teeth: Composition and conversion. *J Biomed Mat Res* 1987;21:11-23

11. Glen J.F. Composition and properties of unfilled and composite restorative materials. In: Smith D.C. and Williams D.F. Eds *Biocompatability of dental materials* Bosca Raton, Florida CRC Press 1982:98-130

12. Davidson C.L. Conflicting interests with posterior use of composite materials. In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:61-66

13. Ruyter I.E. Monomer systems and polymerisation. In: Vanherle G. and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:109-135

14. Peutzfeldt A. and Asmussen E. Influence of Carboxylic Anhydrides on selected mechanical properties of heat cured resin composites. *J Dent Res* 1991;70:1537-1541

15. Peutzfeldt A. and Asmussen E. Influence of Aldehydes on selected mechanical properties of resin composites. *J Dent Res* 1992;71:1522-1524

16. Peutzfeldt A. and Asmussen E. Influence of Ketones on selected mechanical properties of resin composites. *J Dent Res* 1992;71:1847-1850

17. Asmussen E. Restorative resins: hardness and strength vs. quantity of remaining double bonds. *Scand J Dent Res* 1982;90:484-489

18. Bausche J.R., De Lange C. and Davidson C.L. The influence of temperature on some physical properties of dental composites. *J Oral Rehab* 1981;8:309-317.

19. Wendt S.L. The effect of heat used as a secondary cure upon the physical properties of three composite resins. I. Diametral tensile strength, compressive strength and marginal dimensional stability. *Quint Int* 1987;18:265-271.

20. Wendt S.L. The effect of heat used as a secondary cure upon the physical properties of three composite resins. Wear, hardness and colour stability. *Quint Int* 1987;18:351-356.

21. Cook W.D., Beech D.R. and Tyas M.J. Resin based restorative materials - a review. *Aust Dent J* 1984;29:291-295

22. Ferracane J.L. Using posterior composites appropriately. *J Amer Dent Assoc* 1992;123:53-58.

23. Lutz F. and Phillips R.W. A classification and evaluation of composite resin systems. *J Prosthet Dent* 1983;50:480-488

24. McCabe J.F. Developments in composite resins. *Br Dent J* 1984;157:440-444

25. Soderholm K-J.M. Filler systems and resin interface. In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:139-159

26. Bowen R.L. Compatibility of various materials with oral tissues I: The components in composite restorations. *J Dent Res* 1979;58:1493-1503

27. Willems G., Lambrechts., Braem M., Celis J.P. and Vanherle G. A classification of dental composites according to their morphological and mechanical characteristics. *Dent Mater* 1992;8:310-319

28. Fraunhofer J.A. and Curtis P.The physical and mechanical properties of anterior and posterior composite restorative materials. *Dent Mater* 1989;5:365-368.

29. Vanherle G., Lambrechts P. and Braem M. Overview of the clinical requirements for posterior composites. In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:21-40

30. Lambrechts P., Ameye C. and Vanherle G. Conventional and Microfilled composite resins part II: Chip fractures. *J Prosthet Dent* 1982;48:527-538

31. Willems G., Noack M.J., Inokoshi S., Lambrechts P., Van Meerbeck B., Braem M., Roulet J.F. and Vanherle G. Radiopacity of composites compared with human enamel and dentine. *J Dent* 1991;19:362-265

32. Mazer R.B. and Leinfelder K.F. Evaluating a microfill posterior composite resin: a five year study. *J Amer Dent Assoc* 1992;**123**:33-38

33. Craig R.G. Composition and properties of composite resins. *Dent Clin North Am* 1981;25:219-239

34. Bowen R.L. and Reed L.E. Semi-porous reinforcing fillers for composite resins I. Preparation of provisional glass formulations. *J Dent Res* 1976;55:738-747

35. Bowen R.L. and Reed L.E. Semi-porous reinforcing fillers for composite resins II. Heat treatment and etching characteristics. *J Dent Res* 1976;55:748-756

36. Ehrnford L. Dental composites reinforced with microporous sintered glassfiber networks. *Swed Dent J* 1983;supplement 18

37. Soderholm K-J.M. Degradation of glass fillers in experimental composites. *J Dent Res* 1981;60:1867-1875

38. Soderholm K-J.M. and Roberts Influence of water exposure on the tensile strength of composites. *J Dent Res* 1990;69:1812-1816

39. Brauer G.M. Initiator-accelerator systems for acrylic resin and composites. In: Eds Geblein C.G. and Koblitz F.F. *Polymer science and technology.* Vol14 Biomedical and dental applications of polymers. Plenum Press New York and London 1981;395-409

40. McCabe J.F. The influence of porosity on the properties of composite resins. *J Dent Res* 1984;63:532 Abs. 16

41. Lee H.L., Orlowski J.A. and Rogers B.J. Comparison of Ultraviolet and self curing polymers in preventive, restorative and orthodontic dentistry. *Int Dent J* 1976;26:134-171

42. Birdsell D.C., Bannon P.J., Webb P.B. Harmful effects of near Ultraviolet radiation used for polymerisation of sealant and composite resin. *J Am Dent Assoc* 1977;94:311-314

43. Mills L.F., Lytle C.D., Anderson F.A. *et al.* A review of biological effects and potential risks associated with ultraviolet radiation as used in dentistry. Rockville Ma. *US Dept of Health and education and welfare, Public health service, Food and drug administration, Bureau of radiological health.* 1975

44. Taira M., Urabe H., Hirose T., Wakasa K. and Yamaki M. Analysis of photoinitiators in visible light cured dental composite resins. *J Dent Res* 1988;67:24-28

45. Watts D.C. Kinetic mechanisms of visible light cured composite resins. In: Ed Watts D.C. *Setting mechanisms of dental materials*, Chapter 4, Dental Materials Group Symposium, IADR Glasgow 1992.

46. Davis L.G., Baker W.T. and Cox E.A. Optical hazards of blue light curing units: preliminary results. *Br Dent J* 1985;159:259-262

47. Ellingson O.L., Landry R.L. and Bostram R.G. An evaluation of optical radiation emissions from dental visible polymerisation devices. *J Am Dent Assoc* 1986;112:67-70

48. Mosely H., Strang R. and MacDonald I. Evaluation of the risk associated with the use of blue light polymerising sources. *J Dent* 1987;15:12-15

49. Ruyter I.E. and Oysaed H. Conversion in different depth of ultraviolet and visible light activated composite materials. *Acta Odontol Scand* 1982;40:179-192

50.Cook W.D. Spectral distributions of dental photopolymerization sources. *J Dent Res* 1982;61:1436-1438

51. Cook W.D. and Standish P.M. Cure of resin-based restorative materials II: White light photopolymerised resins. *Aust Dent J* 1983;28:307-311

52. Watts D.C., Amer O. and Combe E.C. Characteristics of visible light activated composite systems. *Br Dent J* 1984;156:209-215

53. Rees J.S. and Jacobson P.H. The current status of composite materials and adhesive systems. Part 5, Clinical techniques for direct placement. *Restorative Dent* 1990;6:4-7

54. Feilzer A.J., De Gee A.J. and Davidson C.L. Curing contraction of composites and glass ionomer cements. *J Prosthet Dent* 1988;59:297-300

55. Rees J.S. and Jacobsen P.H. Polymerisation shrinkage of composite resins. *Dent Mater* 1989;5:41-44

56. Goldman M. Polymerisation shrinkage of polymeric restorative materials. *Aust Dent J* 1983;28:156-161

57. Davidson C.L., De Gee A.J. and Feilzer A. The competition between the composite-dentine bond and the polymerisation stress. *J Dent Res* 1984;63:1396-1399

58. Jorgensen K.D., Asmussen E. and Shimokobe H. Enamel damages caused by contracting restorative resins. *J Dent Res* 1975;83:120-122

59. Jensen M. E. and Chan D.C.N. Polymerisation and microleakage. In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:243-262

60. McCulloch A.J. and Smith B.G.N. *In Vitro* studies on cuspal movements produced by adhesive restorative materials. *Br Dent J* 1986;161:405-409

61. Buonocore M.G. A simple method of increasing the adhesion of acrylic filling materials to enamel surfaces. *J Dent Res* 1955;34:849-853

62. McCabe J. Adhesive restorative materials. In: *Applied dental materials* Seventh Edition, Blackwell Scientific Publications, London 1990:157-172

63. Mejare B., Mejare I. and Edwardson S. Bacteria beneath composite restorations: A culturing and histobacteriological study. *Acta Odontol Scand* 1979;37:267-275

64. Brown R.M. and Tobias R.S. Microbial microleakage and pulpal inflammation: a review. *Endod Dent Traumatol* 1986;2:177-183

65. Bater J.F., Stafford G.D. and Harrison A. Masticatory function - A review of movement of the mandible, rate of chewing and forces developed in chewing. *J Oral Rehab* 1975;2:349-361

66. Mitchem J.C. The use and abuse of aesthetics in posterior teeth. *Int Dent J* 1988;38:119-125

67. Leinfelder K.F. Posterior composite resins. *J Amer Dent Assoc* 1988(special edition):21E-26E

68. Lutz F., Phillips R.W., Roulet J.F. and Setcos J.C. *In vivo* and *in vitro* wear of potential posterior composites. *J Dent Res* 1984;63:914-920

69. Lambrechts P., Vaylsteke M., Vanherle G. and Davidson C.L. Quantitative evaluation of the wear resistance of posterior dental restoratives: A new 3-D measuring technique. *J Dent* 1984;12:252-267

70. Jacobsen P.H. The influence of dental materials on conservative dentistry. *Dental Update* 1980;7:285-291

71. Jacobsen P.H. The restoration of class II cavities by polymeric materials. *J Dent* 1984;12:47-52

72. Rukmo M. and Wilson N.H.F. A comparison of methods for contouring the occlusal surfaces of posterior composite restorations. *J Dent* 1985;13:109-122

73. Suzuki M., Jordan J.E. and Boksman I. Posterior composite resin restorations - clinical considerations In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:455-464

74. Lutz F., Imfeld T., Barbakow F. and Iselin W. Optimising the marginal adaptation of MOD composite restorations. In: Vanherle G and Smith D.C. Eds *International symposium on resin based posterior filling materials.* Utrecht:Peter Szulc, 1985:405-409

75. Lutz F., Kreji I., Luescher B. and Oldenburg T.R. Improved proximal margin adaptation of class II composite resin restorations by use of light-reflecting wedges. *Quint Int* 1986;17:659-664

76. de Goes M.F., Rubbi E., Baffa O. and Panzeri H. Optical transmittance of reflecting wedges. *Amer J Dent* 1992;4:78-80

77. Chung K and Greener E.H. Degree of conversion of seven visible light cured posterior composites. *J Oral Rehab* 1988;15:555-560

78. Eliades G.C., Vougiouklakis G.J. and Caputo A.A. Degree of double bond conversion in light cured composites. *Dent Mater* 1987;3:19-25

79. Watts D.C. Composite inlay systems: material properties and design. *J Dent* 1990;18:69-70

80. Christenson G.J. A look at the state of the art of tooth coloured inlays and onlays. *J Amer Dent Assoc* 1992;123:66-68

81. James D.F. An aesthetic inlay technique for posterior teeth. *Quint Int* 1983;14:725-731

82. Jackson R.D. and Ferguson R.W. An esthetic, bonded inlay/onlay technique for posterior teeth. *Quint Int* 1990;21:7-12

83. Hussey D.L. Direct hybrid composite inlays. *Restorative Dent* 1988;4:28-31

84. Beiderman J.D. Direct composite resin inlay. *J Prosthet Dent* 1989;62:249-253

85. Ramanathan G and White G.E. A direct composite onlay technique for primary molars using a light/heat curing system. *J Clin Paed Dent* 1990;15:13-16

86. Jordan R.E. and Suzuki M. Direct hybrid composite inlay technique. *Esthet Dent* 1989;1:57-62

87. Donly K.J. Composite resin inlays: A single appointment technique. *Am J Dent* 1990;3:181-184

88. Cassin A.M. and Pearson G.J. Microleakage studies comparing a one visit indirect composite inlay system and a direct composite restorative technique. *J Oral Rehab* 1992;19:265-270

89. Martin F.E. The composite resin inlay as an alternative to amalgam. *New Zealand Dent J* 1991;87:43-45

90. Reid J.S., Saunders W.P. and Baidas K.M. Marginal fit and microleakage of indirect inlay systems. *Am J Dent* 1993;6:81-84

91. Plasmans P.J.J.M., van't Hof M.A. and Creygers N.H.J. Fabrication times for indirect composite resin restorations. *J Dent* 1992;20:27-32

92. Rees J.S. and Jacobson P.H. The current status of composite materials and adhesive systems. Part 6: Techniques for indirect placement. *Restorative Dent* 1991;4:21-23

93. Wilson M.A. and Norman R.D. An investigation into the incidence of voids in indirect composite inlays formed using different packing techniques. *J Dent* 1991;19:296-300

94. Cook W. D. and Johannson M. The influence of post curing on the fracture properties of photo-cured dimethacrylate based dental composite resin. *J Biomed Mater Res* 1987;21:979-989

95. Asmussen E. and Peutzfeldt A. Mechanical properties of heat treated resins for use in the inlay/onlay technique. *Scand J Dent Res* 1990;98:564-567.

96. Covey D.A., Tahaney S.R. and Davenport J.M. Mechanical properties of heat-treated composite resin restorative materials. *J Prosthet Dent* 1992;68:458-461.

97. Gregory W.A., Berry S., Duke E. and Dennison J.B. Physical properties and repair bond strength of direct and indirect composite resins. *J Prosthet Dent* 1992;68:406-411

98. Peutzfeldt A. and Asmussen E. Mechanical properties of three composite resins for the inlay/onlay technique. *J Prosthet Dent* 1991;66:322-324.

99. McCabe J.F. and Kägi S. Mechanical properties of a composite inlay material following post curing. *Br Dent J* 1991;171:246-248

100. Ferracane J.L. and Codon J.R. Post-cure treatments for composites: properties and fractography. *Dent Mater* 1992;8:290-295

101. Lloyd C.H. The fracture toughness of dental composites II. The environment and temperature dependence of the stress intensification factor (K_{IC}). *J Oral Rehab* 1982;9:133-138

102. Ferracane J.L. and Marker V.A. Solvent degradation and reduced fracture toughness in aged composites. *J Dent Res* 1992;71:13-19

103. Uctasli S., Wilson H.J. and Zaimoglu L. Variables affecting the fracture toughness of resin-based inlay/onlay systems. *J Oral Rehab* 1993;20:423-431

104. Dionysopoulos P. and Watts D.C. Dynamic mechanical properties of an inlay composite. *J Dent* 1989;17:140-144

105. Robinson P.B., Moore B.K. and Swartz M.L. Comparison of microleakage in direct and indirect composite resin restorations *in vitro. Oper Dent* 1987;12:113-116

106. Hasegawa E.A., Boyer D.B. and Chan D.C.N. Microleakage of indirect composite inlays. *Dent Mater* 1989;5:388-391

107. Douglas W.H., Fields R.P. and Fundingsland J. A comparison between the microleakage of direct and indirect composite restorative systems. *J Dent* 1989;17:184-188

108 Wendt S.L. Microleakage and cusp fracture resistance of heat treated composite resin inlays. *Am J Dent* 1991;4:10-14

109. Sheth P.J., Jensen M.E. and Sheth J.J. Comparative evaluation of three resin inlay techniques: microleakage studies. *Quint Int* 1989;20:831-836

110. Yanikoglu F. and Schrer W. Comparison of microleakage between direct placement techniques and direct inlay techniques. *J Marmara Univ Dent Fac* 1990;1:40-46

111. Llena Puy Ma. C., Forner Navarro L., Faus Llacer V.J. and Ferrandez A. Composite resin inlays: a study of marginal adaptation. *Quint Int* 1993;24:429-433

112. Shortall A.C., Baylis R.L., Baylis M.A. and Grundy J.R. Marginal seal comparisons between resin-bonded class II porcelain inlays, posterior composite restorations and direct composite resin inlays. *Int J Prosthodont* 1989;2:217-223

113. Shortall A.C and Baylis R.L. Microleakage around direct composite inlays. *J Dent* 1991;19:307-311

114. Karaagaclioglu L., Zaimoglu A. and Akoren A.C. Microleakage of indirect inlays placed on different kinds of glass ionomer cement linings. *J Oral Rehab* 1992;19:457-469

115. Line A., McIntyre N.S and Davidson R.D. Studies on the adhesion of glass ionomer to dentine. *J Dent Res* 1992;71:1836-1841

116. Taggart S.E. and Pearson G.J. The effect of etching time on glass ionomer cement. *Restorative Dent* 1988; :43-47

117. Reeves G.W., Lentz D.L., O'Hara J.W., McDaniel M.D. and Tolbert W.E. Comparison of marginal adaptation between direct and indirect composites. *Oper Dent* 1992;17:210-214

118. Peutzfeldt A. and Asmussen E. A comparison of accuracy in seating and gap formation for three inlay/onlay techniques. *Oper Dent* 1990;15:129-135

119. Ariyaratnam M., Wilson M.A., Wilson N.H.F. and Watts D.C. Variation in the thickness of the composite lute with an indirect composite inlay system. *Restorative Dent* 1990;6:16-18

120. Qualtrough A.J.E., Piddock V. and Kypreou V. A comparison of two *in vitro* methods for assessing the fitting accuracy of composite inlays. *Br Dent J* 1993;174:450-454

121. Rees J.S. and Jacobsen P.H. Stresses generated by luting resins during cementation of composite and ceramic inlays. *J Oral Rehab* 1992;19:115-122

122. Kunzelman K.H., Hickel R., Meister C. and Petschelt A.Curing contraction in thin bonding composite resin layers. In: *Proceedings of the symposium on computerised restorations: State of the art of the CEREC-Method.* Quintessence Publishing 1991;577-589

123. Swift E.J.Jr., Brodeur C., Cvitko E. and Pires J.A.F. Treatment of composite surfaces for indirect bonding. *Dent Mater* 1992;8:193-196

124. Ariyaratnam M.T, Watts D.C. and Wilson M.A. Composite inlay\cement bond strengths in shear and flexure. *J Dent Res* 1990;69:989(Abs no.274)

125. Causton B.E. Repair of abraded composite fillings. *Br Dent J* 1974;139:286-288.

126. Ruyter I.E. Types of resin-based inlay materials and their properties. *Int Dent J* 1992;42:139-144

127. Tam L.E. and McComb D. Shear bond strengths of resin luting cements to laboratory-made composite resin veneers. *J Prosthet Dent* 1991;66:314-321

128. Peutzfeldt A. and Asmussen E. Retention of composite inlays in enamel dentin cavities. *Dent Mater* 1991;7:11-14

129. Wendt S.L. Time as a factor in the heat curing of composite resins. *Quint Int* 1989;20:259-263

130. De Gee A.J., Pallav P. Werner A. and Davidson C.L. Annealing as a mechanism of increasing wear resistance of composites. *Dent Mater* 1990;6:266-270

131. Burgoyne A.R., Nicholls J.I. and Brudvik J.S. In vitro two-body wear of inlay-onlay composite resin restoratives. *J Prosthet Dent* 1991;65:206-214

132. Smalley W.M. and Nicholls J.I. In vitro two body wear of polymeric veneering materials. *J Prosthet Dent* 1986;56:175-181

133. McCabe J.F. and Smith B.H. A method for measuring the wear of restorative materials *in vitro. Br Dent J* 1981;151:123-126

134. Rueggeberg F.A., Harvey D.K. and Evans A.L. Colour changes in post-cure heat-treated resin composites. *Am J Dent* 1991;4:171-176

135. Burke F.J.T., Watts D.C., Wilson N.H.F. and Wilson M.A. Current status and rationale for composite inlays and onlays. *Brit Dent J* 1991;170:269-273

136. Wassell R.W., Walls A.W.G. and McCabe J.F. Cavity convergence angles for direct composite inlays. *J Dent* 1992;20:294-297

137. Lopes L.M.P., Leitao J.G.M. and Douglas W.H. Effect of a new resin inlay\onlay restorative material on cuspal reinforcement. *Quint Int* 1991;22:641-645

138. Bishop B.M. A heat and pressure cured composite inlay system: a clinical evaluation. *Aust Prosthodont J* 1989;3:35-41

139. Cvar J.F. and Ryge G *Criteria for the clinical evaluation of dental restorative materials* San Francisco US Public Health Service Publication N. 790-244 USPGO 1971

140. Bessing C. and Lundqvist P. A 1-year clinical examination of indirect composite resin inlays:a preliminary report. *Quint int* 1991;22:153-157

141. Wendt S.L. and Leinfelder K.F. The clinical evaluation of heat-treated composite resin inlays. *J Amer Dent Assoc* 1990;123:177-181

142. Wendt S.L. and Leinfelder K.F. The clinical evaluation of heat-treated composite resin inlays; 3-year results. *Amer J Dent* 1992;5:258-262

143. Norman R.D. and Wilson N.H.F. 5-year findings of a multiclinical trial for a posterior composite. *J Dent* 1991;19:153-159

144. Leinfelder K.F., Taylor D.F. and Barkmeier W.W. Quantitative wear measurement of posterior composite resins. *Dent Mater* 1986;2:198-201

145. Jacobsen P.H. and Rees J.S. Luting agents for ceramic and polymeric inlays and onlays. *Int Dent J* 1992;42:145-149

146. Strang R., McCrosson J., Muirhead G.M. and Richardson S.A. The setting of visible-light-cured resins beneath etched porcelain veneers. *Brit Dent J* 1987;163:149-151

147. Breeding L.C., Dixon D.L., and Caughman W.F. The curing potential of light activated composite resin luting agents. *J Prosthet Dent* 1991;65:512-518

148. Hasegawa E.A., Boyer D.B. and Chan D.C.N. Hardening of dual cured cements under composite resin inlays. *J Prosthet Dent* 1991;66:187-192

149. Rueggenburg F.A. and Caughman W.F. The influence of light exposure on polymerisation of dual-cure resin cements. *Oper Dent* 1993;18:48-55

150. Zuellig-Singer R., Kreji I. and Lutz F. Effect of cement-curing modes on dentine bonding of inlays. *J Dent Res* 1992;71:1842-1846

151.Tyas M.J. Correlation between fracture properties and clinical performance of composite resins in Class IV cavities. *Aust Dent J* 1990;35:46-49

152. Leinfelder K.F., Isenberg B.P. and Essig M.E. A new method for generating ceramic restorations. CAD-CAM system. *J Am Dent Assoc* 1989;118:703-707

153. Van Meerbeck B. Inokoshi S., Willens G., Noak M.J., Braem M., Lambrechts P., Roulet J.F. and Vanherle G. Marginal adaptation of four tooth coloured inlay systems *in vivo. J Dent Res* 1992;20;18-26

154. Friedman J. Variability of lamp characteristics in dental curing lights. *J Esthet Dent* 1989;6:189-190

155. Jorgenson K D. Restorative resins: abrasion vs. mechanical properties. *Scand J Dent Res* 1980;88:557-568.

156. Harrison A. and Draught R.A. Abrasive wear, tensile strength and hardness of dental resins - is there a relationship? *J Prosthet Dent* 1976;36:395-399

157. Ruyter I.E. Methacrylate-based polymeric dental materials: conversion and related properties. Summary and review. *Acta Odontol Scand* 1982;40:359-376.

158. Asmussen E. Factors affecting the quantity of remaining double bonds in restorative resin polymers. *Scand J Dent Res* 1982;90:490-496

159. Vankerckhoven H., Lambrechts P., Van Beylen M., Davidson C.L. and Vanherle G. Unreacted methacrylate groups on the surface of composite resins. *J Dent Res* 1982;61:791-795

160. Feilzer A.J., De Gee A.J. and Davidson C.L. Quantitative determination of stress reduction by flow in composite restorations. *Dent Mater* 1990;6:167-171.

161. International Standard ISO 4049: Dentistry - Resin-based filling materials. Twelfth edition. *International Organisation for Standardisation*; 1988

162. Gjerdet N.R. and Hegdahl T. Porosity of resin filling materials *Acta Odontol Scand* 1978;36:303-306

163. Koliniotou-Kubia E. and Jacobsen P.The effect of irradiation time on the physical properties of light-cured resins. *Clin Mater* 1990;6:21-28

164. Reinhard K.J. and Smolka R. Kunststoffe in seitenzahn bereich - fullung oder inlay? *Dtsch Zahnärztl Z* 1988;43:909-913.

165. Chan K.C. and Boyer D.B. Curing light activated cement through porcelain *J Dent Res* 1989;68:476-480

166. Cook W. and Thomasz F. A photocured composite resin with apparent infinite depth of cure. *Aust Dent J* 1983;28:243-246

167. Skinner E.W. *Science of Dental Materials* 3rd edition, Philidelphia, W.B. Saunders 1946:140-141

168. Shillingburg H.T., Jacobi R. and Brackett S.E. Biomechanical principles of preparations. In: *Fundamentals of tooth preparation for cast metal and porcelain restorations.* Quintessence Publishing Co. Inc. Chicago 1987 13-44

169. British standard. Specification for dental glass ionomer cement. BS 6039 section 6.3 Film thickness *British Standards Institution, London.* 1981;2-3169.

170. Young K.C., Hussey M., Gillespie F.C. and Stephen K.W. The performance of u.v. lights to polymerise fissure sealants *J Oral Rehab* 1977;4:181-191

171. Main C., Cummings A., Moseley H., Stephen K.W. and Gillespie F.C. An assessment of new dental ultraviolet sources and u.v. polymerised fissure sealants *J Oral Rehab* 1983;10:215-227

172. McCabe J.F. Resin based filling materials In: *Applied dental materials* Seventh edition, Blackwell scientific publications, Oxford. 145-156

173. Inokoshi S., Willems G., Van Meerbeck B. Lambrechts P., Braem M. and Vanherle G. Dual-cure luting composites. Part 1: filler particle distribution *J Oral Rehab* 1993;20:133-146

174. Jacobsen P.H. Extension of working time of light activated composite materials *Br Dent J* 1986;160:162-165

175. Strang R., MacDonald I., O'Hagan S., Murray J. and Stephen K.W. Variation in performance of curing light units by determination of composite resin setting time *Br Dent J* 1987;162:63-65

176. Goldman M. Fracture properties of composite and glass ionomer. *J Biomed Mater Res* 1985;19:771-783

177. Pilliar R.M., Vowles R. and Williams D.F. The effect of environmental aging on the fracture toughness of dental composites. *J Dent Res* 1987;66:722-726

178. Lloyd C.H. and Ianetta R.V. The fracture toughness of dental composites I. The development of strength and toughness. *J Oral Rehab* 1982;9:55-66

179. Morena R., Lockwood P.E. and Fairhurst C.W. Fracture toughness of commercial dental porcelains. *Dent Mater* 1986;2:58-62

180. Griffiths A.A. The phenomena of rupture and flow in solids. *Proc Trans Royal Soc* 1920 A221:163

181. Causton B.E. Fracture mechanics of dental poly(methyl methacrylate). *J Dent Res* 1975;339-343

182. Stafford G.D., Causton B.E., Hugget R. and Hanley R.W. Study of the effect of environment upon the fracture toughness of PMMA. *J Dent Res* 1979;58:1234 Abs 53

183. Lloyd C.H. and Craigton J. Mechanical properties of injected PMMA. *J Dent Res* 1980;59:1810

184. Stafford G.D. and Causton B.E. Fracture toughness of denture base acrylic. *J Dent Res* 1980;59;1842 Abs 205

185. El Mowafy O.M. and Watts D.C. Fracture toughness of human dentine. *J Dent Res* 1986;65:677-681

186. Hassan R., Caputo A.A. and Bunshah R.F. Fracture toughness of human enamel. *J Dent Res* 1981;60:820-827

187. Lloyd C.H. and Anderson J.N. The strength and toughness of Calcium Hydroxide preparations. *J Oral Rehab* 1980;7:155-165

188. Hill R.G. and Labok S.A. The influence of polyacrylic acid molecular weight on the fracture of Zinc Polycarboxylate cements. *J Mater Sci* 1991;26:67-74

189. Lloyd C.H. and Mitchell L. The fracture toughness of tooth coloured restorative materials. *J Oral Rehab* 1984;11:257-272

190. Lloyd C.H. and Adamson M. The development of fracture toughness and fracture strength in posterior restorative materials. *Dent Mater* 1987;3:225-231

191. Roberts J.C., Powers J.M. and Craig R.G. The fracture toughness and critical strain energy release rate of dental amalgam. *J Mater Sci* 1978;13:965-972

192. Lloyd C.H. and Adamson M. The fracture toughness of amalgam. *J Oral Rehab* 1985;12:59-68

193. Cruickshanks-Boyd D.W. and Lock W.R. Fracture toughness of dental amalgams. *Biomater* 1983;4:234-242

194. Hassan R., Vaidanathan T.K. and Schulman A. Fracture toughness determination of dental amalgams through microindentation. *J Biomed Mater Res* 1986;20:135-142

195. Kvam K. Fracture toughness determination of ceramic and resin-based dental composites. *Biomater* 1992;13:101-104

196. Rosensteil S.F. and Porter S.S. Apparent fracture toughness of dental porcelain with a metal substructure. *Dent Mater* 1988;4:187-190

197. Wiley D.R. and Waters N.E. A fracture mechanics approach to ceramo-metal bond evaluation. *J Mater Sci* 1989;24:2337-2342

198. Herman M., Rottenegger R., Tinschet J. and Marx R. The effect of corrosive environment on the porcelain-to-metal bond - A fracture mechanics approach. *Dent Mater* 1992;8:2-6

199. Davis D.M. and Waters N.E. An investigation into the crack behaviour of tooth-coloured particulate filled dental restorative materials. *J Mater Sci* 1987;22:3508-3516

200. Davis D.M. and Waters N.E. An investigation into the crack behaviour of a particulate-filled Bis-GMA resin. *J Dent Res* 1986;66:1128-1133

201. Pilliar R.M., Smith D.C. and Maric B. Fracture toughness of dental composites determined using the short rod fracture toughness test. *J Dent Res* 1986;65:1308-1314

202. Ferracane J.L., Antonio R.C. and Matsumoto H. Variables affecting the fracture toughness of dental composites. *J Dent Res* 1987;66:1140-1145

203. Lange F.F. The interaction of a crack front with a second phase dispersion. *Philosophical Magazine* 1970;22:983-988

204. Broutman L.J. and Sahu S. The effect of the interfacial bonding on the toughness of glass filled polymers. *Mater Sci Eng* 1971;8:98-107

205. Lange F.F. and Radford K.C. Fracture energy of an epoxy composite system. *J Mater Sci* 1971;6:1197-1206

206. Young R.J. and Beaumont P.W.R. Failure of brittle polymers by slow crack growth - part 2. *J Mater Sci* 1977;10:1343-1350

207. Lloyd C.H. The toughness of dental composites III. The effect of environment upon the stress intensification factor (K_{IC}) after extended storage. *J Oral Rehab* 1982;9:133-138

208. Montes G.G.M. and Draughn R.A. Slow crack propagation in composite restorative materials. *J Biomed Mater Res* 1987;21:629-642

209. Mair L.H. and Vowles R. The effect of thermal cycling on the fracture toughness of seven composite restorative materials. *Dent Mater* 1989;5:23-26

210. Dhuru V.B. and Lloyd C.H. The fracture toughness of repaired composite. *J Oral Rehab* 1985;12:413-421

211. Lloyd C.H. and Dhuru V.B. Effect of a commercial bonding agent upon the fracture toughness (K_{IC}) of repaired heavily filled composite. *Dent Mater* 1985;1:83-85

212. Kovarik R.E. and Ergle J.W. Fracture toughness of posterior composite resins fabricated by incremental packing. *J Prosthet Dent* 1993;69:557-560

213. Ferracane J.L., Hopkins J.K. and Codon J.R. The properties of heat-treated composites after aging. *J Dent Res* 1993;72:Abstract no. 256

214. BS 5447: Methods for plain strain fracture toughness (K_{IC}) testing. British Standard Institution, London 1977

215. Lloyd C.H. The fracture toughness (K_{IC}) of composite filling materials and crack tip radius. Letter to editor. *J Dent Res* 1988;67:883-884

216. Knott J.F. Fundamentals of fracture mechanics. Butterworths, London 1973:132

217. Truong V.T. and Tyas M.J. Prediction of *in vivo* wear in posterior composite resins: a fracture mechanics approach. *Dent Mater* 1988;4:318-327

218. Ferracane J.L. and Greener E.H. The effect of resin formulation on the degree of conversion and mechanical properties of dental composite resins. *J Biomed Mater Sci* 1986;20:121-131

219. Truong V.T., Cock D.J. and Padmanathan N. Fatigue crack propagation in posterior dental composites and prediction of clinical wear. *J Appl Biomat* 1990;1:21-39

220. Yoshimoto S. Study on the fracture toughness of posterior composite resin restorative material. *Kanagawa Shigaku* 1984;19:19-30

221. Watts D.C., McNaughton V. and Grant A.A. The development of surface hardness in visible light cured posterior composites. *J Dent* 1986;14:169-174

222. Smith D.C and Bains M.E.D. The detection and estimation of residual monomer in polymethyl methacrylate. *J Dent Res* 1956;35:16-20

223. Harie K., Otagawa A., Moraoka M. and Mita L. Calorimetric investigation of polymerisation reactions. V. Crosslinked polymerisation of methyl methacrylate with ethylene dimethacrylate. *J Polymer Sci Polymer Chem* 1975;13:445-449

224. Inoue K. and Hayashi I. Residual monomer (Bis-GMA) of composite resins. *J Oral Rehab* 1982;9:493-497

225. Tanaka K., Tiara M., Shintari H., Wakasa K. and Yamaki M. Residual monomers (TEGMA and Bis-GMA) of a set Visible light cured dental composite resin when immersed in water. *J Oral Rehab* 1991;18:353-362

226. Ferracane J.L. Correlation between hardness and degree of conversion during the setting reaction of unfilled dental restorative resins. *Dent Mater* 1985;1:11-14

227. Ruyter E.I. and Svendsen S.A. Remaining methacrylate groups in composite restorative materials. *Acta Odontol Scand* 1977;36:75-82

228. Kemp W. Infrared Spectroscopy In: *Organic Spectroscopy* 3rd Edition MacMillan Educational Ltd. London 1991 p19-57

229. Heigl J.J., Bell M.F. and White A. Applications of infrared spectroscopy to the analysis of liquid hydrocarbons. *Anal Chem* 1947;19:293-298

230. Rueggeberg F.A., Hashinger D.T. and Fairhurst C.W. Calibration of FTIR conversion analysis of contemporary dental resin composites. *Dent Mater* 1990;6:241-249

231. Watts D.C Dental restorative materials In: *Materials science and technology Vol. 14* Weinheim p209-258

231. Nakamichi I., Iwaku U., Fusayama T. Bovine teeth as possible substitutes in the adhesion test. *J Dent Res* 1983;62:1076-1081.

232. Boyer D.B., Chan K.C., Torney D.L. The strength of multilayer and repaired composite resin. *J Prosthet Dent* 1978;39:63-67

233. Chan K.C., Boyer D.B. Repair of conventional and microfilled composite resins. *J Prosthet Dent* 1983;50:345-350

234. Crumpler D.C., Bayne D.C., Sockwell S. Brunson D. and Roberson T.M. Bonding to resurfaced posterior composites. *Dent Mater* 1989;5:417-424

235. Zidan O., Asmussen E., Jorgensen K.D. Correlation between tensile and bond strength of composite resins. *Scand J Dent Res* 1980;88:348-351

236. Watson T.F. Applications of confocal scanning optical microscopy to dentistry. *Brit Dent J* 1991;171:287-291

237. Petran M., Hadravsky M. and Boyde A. The tandem scanning reflected light microscope. *Scanning* 1985;7:97-108

238. Watson T.F. and Boyde A. Confocal light microscopic techniques for examining dental operative procedures and dental materials. A status report for the American Journal of Dentistry. *Am J Dent* 1991;4:193-200

239. Watson T.F., Billington R.W. and Williams J.A. The interfacial region of the tooth/glass ionomer restoration: a confocal optical microscope study. *Am J Dent* 1991;4:303-310

240. Barnes I.E. Replication techniques for the scanning electron microscope 1. History, materials and techniques. *J Dent* 1978;6:327-341

241. Barnes I.E. Replication techniques for the scanning electron microscope 2. Clinical and laboratory procedures: interpretation. *J Dent* 1979;7:25-37

242. Minsky M. Microscopy apparatus. U.S. patent office filed Nov 7, 1957, granted 2 Dec 1961 Patent No. 3,013,467

243. Egger M.D. and Petran M. New reflected light microscope for viewing unstained brain and ganglion cells. *Science* 1967;157:305-307

244. Wilson T. and Sheppard C.J.R. *Theory and practice of scanning optical microscopy.* Oxford, Academic Press, 1984

245. Carlsson K., Danielson P.E., Lenz R., Lilijborg G.A., Majlof L and Ashlund N. Three dimensional microscopy using a confocal laser scanning microscope. *Optics letters* 1985;10:53-55

246. White J.G., Amos W.B. and Fordham M. An evaluation of confocal versus conventional imaging of biological structures for fluorescent light microscopy. *J Cell Biol* 1987;105:41-48

247. Draaijer A. and Houpt P.M. A standard video rate laser scanning reflection and fluorescence microscope (CSLM). *Scanning* 1988;10:139-145

248. Brackenhoff G.J. and Visscher K. Confocal imaging with bilateral scanning and array detectors. *J Microsc* 1992;165:139-146

249. Xiao G.K. and Kino G.S. A real time confocal scanning optical microscope. In: Eds Wilson T.F. and Balk L. *Scanning image technology* Vol 809,107-113. Proc SPIE, 1987

250. Xiao G.K., Corle T.R. and Kino G.S. Real-time confocal scanning optical microscope. *Appl Phys Lett* 1988;53:716-718

251. Boyde A., Xiao G.K., Corle T.R., Watson Y.F. and Kino G.S. An evaluation of unilateral TSM for biological applications. *Scanning* 1990;12:273-279

252. Watson T.F., Petroll W.M., Cavanagh H.D. and Jester J.V. *In Vivo* confocal microscopy in clinical dental research: an initial appraisal. *J Dent* 1992;20:352-358

253. Watson T.F. and Boyde A. The use of fluorescent markers for studying the distribution of a dentine bonding agent between a composite restoration and a tooth. *Clin Mater* 1987;2:45-53

254. Benn D.K. and Watson T.F. Correlation between film position, bitewing shadows, clinical pitfalls and the histological size of approximal lesions. *Quint Int* 1989;20:131-141

255. Watson T.F. A confocal optical microscope study of the morphology of the tooth/restoration interface using Scotchbond 2 dentine adhesive. *J Dent Res* 1989;68:1124-1131

256. Watson T.F. The application of real time confocal microscopy to the study of high speed dental bur\tooth cutting interactions. *J Microsc* 1990;157:51-60

257. Watson T.F. A confocal microscopical study of some factors affecting the adaptation of a light cured glass ionomer to tooth tissue. *J Dent Res* 1990;69:1531-1538

258. Boyde A., Jones S.J., Taylor M.L., Wolfe L.A. and Watson T.F. Fluorescence in the tandem scanning microscope. *J Microsc* 1990;157:39-49

259. Boyde A. Stereoscopic images in confocal (tandem scanning) microscopy. *Science* 1985;230:1270-1272

260. Carlsson K. and Aslund N. Confocal imaging for 3-D digital microscopy. *Applied Optics* 1987;2:3232-3238

261. Pawley J.B. Handbook of Confocal Microscopy, Plenum Press, New York. 1990;75-82

262. Watson T.F. Personal Communication. 1992

263. Nakabayashi N., Kojima and Masuhara E. The promotion of adhesion by the infiltration of monomers into tooth substrates. *J Biomed Mater Res* 1982;16:265-273

264. Gwinnett A.J. and Kanca J.A. Micromorphology of the bonded dentine interface and is relationship to bond strength. *Am J Dent* 1992;5:73-77

265. Roulet J.R. and Noak M.J. Criteria for substituting amalgam with composite resins. *Int Dent J* 1992;41:195-205

266. Feilzer A.J., deGee A.J., Davidson C.L. Increased Wall-to-Wall curing contraction in thin bonded resin layers. *J Dent Res* 1989;68:48-50.

267. Mahler D.B. Letter to editor. *J Dent Res* 1990;69:913

268. Assif D., Antopolski B., Helft M. and Faffe I. Comparison of methods of clinical evaluation of marginal fit of complete cast gold crowns. *J Prosthet Dent* 1985;54:20-24

269. Weyns W. and DeBoever J. Radiographic assessment of the marginal fit of cast restorations. *J Prosthet Dent* 1984;51:485-489

270. Qualtrough A.J.E. and Piddock V. Fitting accuracy of Indirect Restorations: A review of Methods of Assessment. *Eur J Prosthodont Res Dent* 1992;1:57-61

271.Guyer S.E., Lefkowitz W., Malone W.F., Rhoads J.E., Staffanou R.S. and Woody R.D. A comparison of the marginal fit of three cast ceramic crown systems. *J Prosthet Dent* 1985;59:534-542

272. Cooney J.P., Richter W.A. and MacEntee M.I. Evaluation of ceramic margins for metal-ceramic restorations. *J Prosthet Dent* 1984;54:1-5

273. Zena R.B., Zafrulla K. and von Frauhofer Z. Shoulder preparations for collarless metal ceramic crowns: hand planing as opposed to rotary instruments. *J Prosthet Dent* 1989;62:273-277

274. Fransson B., Oilo G. and Gjeitanger R. The fit of Metal ceramic crowns a clinical study.*Dent Mater* 1985;1:197-199

275. Essig M.E., Isenberg B.P. and Leinfelder K.F. An in vivo evaluation of Duo cured cements with CAD-CAM ceramic inlays. *J Dent Res* 1991;68:324 (Abstract 1141)

276. Kidd E.A.M. Microleakage: a review. *J Dent* 1976;4:199-205

277. Hegedahl T. and Gjerdet N.R. Contraction stress of composite resin filling materials. *Acta Odontol Scand* 1977;35:191-195

278. Bowen R.L., Rapson J.E. and Dickson G. Hardening shrinkage and hygroscopic expansion of composite resins. *J Dent Res* 1982;61:654-658

279. Torstenson B. and Brannstrom M. Contraction gap under composite restorations: effect of hygroscopic expansion and thermal stress. *Oper Dent* 1988;13:24-31

280. Jorgensen K.D. and Shimokobe M.D. Deformation of cavities and resin fillings in loaded teeth. *Scand J Dent Res* 1976;84:175-182

281. Ben-Amar A. Microleakage of composite resin restorations. A status report for the American Journal of Dentistry. *Am J Dent* 1989;2:175-180

282. Going R.E., Massler M. and Dute H.L. Marginal penetration of dental restorations as studied by crystal violet dye and I^{131}. *J Am Dent Assoc* 1960;61:285-300

283. Phillips R.W., Gilmore H.W., Swartz M.L. and Schenker S.I. Adaptation of restorations as assessed by Ca^{45}. *J Am Dent Assoc* 1961;62:9-20

284. Loiselle R.J., Goldberg A.F., Gross R.L. and Stuever C.H. Marginal microleakage - An *in vivo* assessment. *J Am Dent Assoc* 1969;78:758-760

285. Going R.E., Myers H.M. and Prussin S.G. Quantitative method for studying microleakage *in vivo* and *in vitro. J Dent Res* 1968;47:1128-1132

286. Harper W.E. The character of the adaptation of amalgam to the walls of cavities attained by present methods of instrumentation and the use of the best known alloys, as indicated by the air pressure test. *Dent Rev* 1912;26:1179-1198

287. Fiasconaro J. and Sherman H. Sealing properties of acrylics. *N Y St Dent J* 1952;18:189-198

288. Pickard H.M. and Gayford J.J. Leakage at the margins of amalgam restorations. *Br Dent J* 1965;119:69-77

289. Fraser C.J. A study of the efficacy of dental fillings. *J Dent Res* 1929;9:507-517

290. Seltzer S. The penetration of micro-organisms between the tooth and direct resin fillings. *J Am Dent Assoc* 1955;51:560-566

291. Rose E.E., Lal J., Williams N.B. and Falcetti J.P. The screening of materials for adhesion to human tooth structure. *J Dent Res* 1955;34:577-588

292. Mortensen D.W., Boucher N.E. and Ryge G. A method of testing for marginal leakage of dental restorations with bacteria. *J Dent Res* 1965;44:58-63

293. Fayyad M.A. and Ball P.C. Bacterial penetration around amalgam restorations. *J Prosthet Dent* 1987;57:571-574

294. Bauer J.G. and Henson J.L. Microleakage; A measure of the performance of direct filling materials. *Oper Dent* 1984;9:2-9

295. Douglas W.H., Chen C.J. and Craig R.G. Improved Neutron activation analysis of microleakage around a hydrophobic composite restorative. *J Dent Res* 1980;59:1507-1510

296. Jacobsen P.J. and von Fraunhofer J.A. Assessment of microleakage using a conductimetric technique. *J Dent Res* 1975;54:41-48

297. Delivanis P.D. and Chapman K.A. Comparison and reliability of techniques for measuring leakage and marginal penetration. *Oral Surg, Oral Med, Oral Pathol* 1982;53:410-416

298. Mattison G.D. and von Frauhofer J.A. Electrochemical microleakage study of endodontic sealer\cements. *Oral Surg, Oral Med, Oral Pathol* 1983;55:402-407

299. Momoi Y., Iwase H., Nakano Y. *et al.* Gradual increases in marginal leakage of resin composite restorations with thermal stress. *J Dent Res* 1990;69:1659-1663

300. Lim K.C. The microleakage of glass ionomer cement using two methods of moisture protection. *Quint Int* 1987;18:835-839

301. Ellis J.M. and Brown L.R. Application of an *in vitro* cariogenic technique to study the development of carious lesions around dental restorations. *J Dent Res* 1967;46:403-408

302. Hals E. and Nernaes A. Histopathology of *in vitro* caries developing around silver amalgam fillings. *Caries Res* 1972;5:58-77

303.Grieve A.R. The occurrence of secondary caries-like lesions *in vitro.* The effect of a fluoride cavity liner and a cavity varnish. *Br Dent J* 1973;134:530-536

304. Kidd E.A.M. Harrington E. and Grieve A.R. The cavity sealing ability of composite restorations subjected to thermal stress. *J Oral Rehab* 1978;5:279-286

305. Grieve A.R. and Glyn Jones J.C. An *in vitro* study of marginal leakage associated with composite restorations using an acidified agar technique. *J Oral Rehab* 1980;7;215-233

306. Asmussen E. and Jorgensen K.D. A microscopic investigation of the adaptation of some plastic filling materials to dental cavity walls. *Acta Odontol Scand* 1972;30:3-21

307. Asmussen E. Marginal adaptation of restorative resins in acid etched cavities. *Acta Odontol Scand* 1977;35;125-134

308. Shortall A.C. Microleakage, marginal adaptation and composite resin restorations. *Br Dent J* 1982;153:223-227

309. Qvist V. Correlation between marginal adaptation of composite resin restorations and bacterial growth in cavities. *Scand J Dent Res* 1980;88:296-300

310. Powis D.R., Prosser H.J. Shortall A.C. *et al.* Long term monitoring of microleakage of composites part 1: radiochemical diffusion technique. *J Prosthet Dent* 1988;60:304-307

311. Going R.E. Reducing marginal leakage: a review of materials and techniques. *J Am Dent Assoc* 1979;99:646-651

312. Crisp S. and Wilson A.D. Radioactive tracer technique for monitoring of microleakage: an interim report. *J Biomed Mater Res* 1980;14:373-382

313. Herrin H.K. and Shen C. Microleakage of root caries restorations. *Gerodontics* 1985;1:156-159

314. Kornfield B. Study of clinical behaviour of resins in operative dentistry. *J Dent Res* 1953;32:714-715

315. Pintado M.R. and Douglas W.H. The comparison of microleakage between two different dentin bonding resin systems. *Quint Int* 1988;19:905-907

316. Eakle W.S. and Nakamoto D.K. Microleakage in MOD resin composite with three dentin bonding agents. *Dent Mater* 1989;5:361-364

317. Kanca J. The effect on microleakage of four dentin-enamel bonding systems. *Quint Int* 1989;20:359-361

318. Leinfelder K.F., Russell C.M. and Mueninghoff L.A. Use of Ca(OH) for measuring microleakage. *Dent Mater* 1986;2:121-124

319. Douglas W.H. and Zakariasen K.L. Volumetric assessment of apical leakage utilising a spectrophotometric dye-recovery method. *J Dent Res* 1981;60A:627 Abs no 512

320. Rigsby D.F., Retief D.H., Russell C.M. and Denys F.R. Marginal leakage and marginal gap dimensions of three dentinal bonding systems. *Am J Dent* 1990;3:289-294

321. Mandras R.S., Retief D.H. and Russell C.M. Quantitative microleakage of six dentine bonding agents. *Am J Dent* 1993;6:119-122

322. Youngson C.C. A technique for three-dimensional microleakage assessment using tooth sections. *J Dent* 1992;20:231-234

323. Taylor M.J. and Lynch E. Microleakage. *J Dent* 1992;20:3-10

324. Christen A.G. and Mitchell D.F. A fluorescent dye method for demonstrating leakage around dental restorations. *J Dent Res* 1966;45:1485-1492

325. Kempler D., Stark M.M., Leung R.L. and Greenspan J.S. Enamel-composite interface relative to cavosurface configuration, abrasion, and bonding agents. *Oper Dent* 1976;1:137-145

326. Itoh K., Iwaku M. and Fusayama T. Effectiveness of glazing composite resin restorations. *J Prosthet Dent* 1981;45:606-613

327. Chow M.H. Effects of sealant placed over composite resin restorations. *J Prosthet Dent* 1980;44:531-535

328. Raadal M. Microleakage around preventive composite fillings in loaded teeth. *Scand J Dent Res* 1979;87:390-394

329. Stewart G.P., Balda B.A. and Norman R.D. The effect of mechanical loading on marginal integrity of composite restorations. *Dent Mater* 1986;2:151-152

330. Darbyshire P.A., Messer L.B. and Douglas W.H. Microleakage in class II composite restorations bonded to dentin using thermal and load cycling. *J Dent Res* 1988;67:585-587

331. Mandras R.S., Retief D.H. and Russell C.M. The effects of thermal and occlusal stresses on the microleakage of the Scotchbond " dentinal bonding system. *Dent Mater* 1991;7:63-67

332. Hansen E.K. Effect of cavity depth and application technique on marginal adaptation of resins in dentin cavities. *J Dent Res* 1986;65:1319-1321

333. Nelsen R.J., Wolcott R.B. and Paffenbarger G.C. Fluid exchange at the margins of dental restorations. *J Am Dent Assoc* 1952;44:288

334. Coy H.D. Direct resin fillings. *J Am Dent Assoc* 1953;47:532-537

335. Paffenbarger G.C., Nelsen R.J. and Sweeney A.B. Direct and indirect filling resins: a review of some physical and chemical properties. *J Am Dent Assoc* 1953;47:516-524

336. Fitchie J.G., Reeves G.W., Scarbrough A.R. and Hembree J.H. Microleakage of two new dentinal bonding systems. *Quint Int* 1990;21:749-752

337. Fayyad M.A. and Shortall A.C.C. Microleakage of dentine-bonded posterior composite restorations. *J Dent* 1987;15:67-72

338. Gross J.D., Retief D.H. and Bradley E.L. Microleakage of posterior composite restorations. *Dent Mater* 1984;1:7-10

339. Crim G.A. Assessment of microleakage of three dentinal bonding systems. *Quint Int* 1990;21:295-297

340. Zaimoglu A. and Karaagaclioglu L. Microleakage in porcelain laminate veneers. *J Dent*;1991;19:369-372

341. Saunders W.P. and Muirhead J.M. Microleakage of composite restorations with Syntac Bond and Denthesive. *Am J Dent* 1992;5:255-257

342. Longman C.M. and Pearson G.J. Variation in tooth surface temperature in the oral cavity during fluid intake. *Biomaterials* 1987;8:411-414

343. Harper R.H., Schnell R.J., Swartz M.L. and Phillips R.W. *In vivo* measurements of thermal diffusion through restorations of various materials. *J Prosthet Dent* 1980;43:180-185

344. Crim G.A. and Garcia-Godoy F. Microleakage: the effect of storage and cycling duration. *J Prosthet Dent* 1987;57:574-576

345. Crim G.A., Swartz M.L. and Phillips R.W. Comparison of four thermocycling techniques. *J Prosthet Dent* 1985;53:50-53

346. Crim G.A. and Mattingly S.L. Evaluation of two methods for assessing marginal leakage. *J Prosthet Dent* 1981;45:160-163

347. Wendt S.L., McInnes P.M. and Dickson G.I. The effect of thermocycling in microleakage analysis. *Dent Mater* 1992;8:181-184

348. Komatsu M. and Finger W. Dentin bonding agents: Correlation of early bond strength with margin gaps. *Dent Mater* 1986;2:257-262.

349. Sorenson J.A. and Dixit N.V. In vitro shear bond strength of dentin adhesives. *Int Journal Prosthet* 1991;4:117-125

350. Crim G.A. Prepolymerisation of Gluma 4 sealer: effect on bonding. *Am J Dent* 1990;4:25-27.

351. McCabe J.F. Properties used to characterize materials In: *Applied Dental Materials* Seventh Edition, Blackwell Scientific Publications, Oxford, 1990:4-28

352. Finger W. and Theimann J. Correlation between *in vitro* and *in vivo* wear of posterior restorative materials. *Dent Mater* 1987;3:280-286

353. DeLong R. and Douglas W.H. An artificial oral environment for testing dental materials. *IEEE Trans Biomed Eng* 1991;38:339-345

354. Slack F.A. A preliminary method of testing abrasion hardness. *J Amer Dent Assoc* 1949;39:47-50

355. Lloyd C.H. Personal Communication 1993.

Appendix 1: List of abbreviations.

ADA	American Dental Association
BIS-EMA	Bisphenol-ethylmethacrylate
BIS-GMA	Bisphenol-glycidylmethacrylate
BIS-MA	Bisphenol-methacrylate
C	Centigrade
DBA	Dentine Bonding Agent
DTS	Diametrial Tensil Strength
FTIR	Fourier transform Infra-Red
Hz	Hertz
K	Kelvin
m	metre
MOD	Mesial Occlusal Distal
NA	Not Applicable
N	Newtons
Pa	Pascal
PTFE	Poly-tetra-fluoro-ethylene
SEM	Scanning Electron Microscope
TEGMA	Triethylene glycol dimethacrylate
USPHS	United States Public Health Service
U.V.	Ultra Violet
W	Watts