MICROLEAKAGE OF BONDED AMALGAM RESTORATIONS

BY

YOUSRA H. AL-JAZAIRY, B.D.S

A Thesis
Submitted to the faculty of Graduate Studies in Partial Fulfillment of
the Requirements for the Degree of

MASTER of SCIENCE

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DEDICATION

To my Parents
Hussain and Najat

Whose love, values and guidance made me what I am..

To my husband
Salim

and

My lovely children
Sama, Abdul-Rhman, and Hala

For their love, patience and support.

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Foremost among my acknowledgments, I am very grateful to God the most compassionate and most merciful for giving me the will and ability to accomplish my goals, and providing me all helpful people who made this thesis possible.

My years at University of Manitoba/Winnipeg have been particularly rewarding, during which time I feel that I have grown both personally and professionally. No one reached this point in education without the help from several people.

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ABSTRACT

Microleakage at the amalgam restoration-dentin interface has been a major area of concern for many years. Currently, adhesive dentin bonding systems are used to bond and seal amalgam restorations to tooth structure. The purpose of this study was to evaluate quantitatively the effectiveness of two adhesive dentin bonding systems on reducing microleakage at the amalgam-dentin interface in comparison to the presently most used amalgam restoration sealing material Copalite varnish. Cylindrical occlusal cavities, 3 mm diameter and 3 mm depth, were prepared in 24 freshly extracted human third molars. Six samples were randomly assigned to each of the four following groups: unlined amalgam (UN), Copalite varnish (CO), and Amalgambond Plus (AM), All-Bond 2 (AL) adhesive systems. A fluid-under-pressure apparatus connected with the pulp chamber of each tooth was used to evaluate the permeability and marginal leakage of teeth before and after restorations at 16 Psi pressure. Following insertion of the amalgam restorations, microleakage (μl/min/cm H_2O) was measured at 24 hrs, 1 week, and 1, 3, 6 months. Data were analyzed by repeated measures ANOVA and showed a significant difference among the means of the groups ($p<0.0001$). At 24 hours all the restored teeth showed a significant reduction in microleakage compared to the unrestored teeth UN ($p<0.001$), CO ($p<0.001$), AM ($p<0.05$) and AL ($p<0.0001$). At subsequent time intervals UN group showed a significant increase in microleakage ($p<0.01$). AM and AL groups at 3 and 6 months showed significantly lower microleakage than UN ($p<0.01$) and CO ($p<0.05$) groups. In this study both AM and AL adhesive systems are more effective than CO varnish in reducing the microleakage under amalgam restorations. No significant difference was found between the two adhesive systems in vitro at any intervals tested.

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INTRODUCTION

Failure of a dental restoration to seal a cavity in a tooth will "microleakage". Microleakage occurs when oral fluid passes through the interface between the tooth structure and the restorative material. Because the fluid usually contains bacteria, microleakage can result in secondary caries, pulp irritation and even pulp necrosis. Dentin permeability and its moisture content make it difficult for restorative materials to adhere to it, and microleakage frequently occurs. Therefore, optimal sealing between tooth structure and restorations should protect the tooth structure from the effects of microleakage. The studies described in this thesis concern the ability of restorative materials to adhere to the tooth structure and also to seal the interface between them.

The characteristics of an ideal restorative material include the following: (1) biocompatiblity, (2) mechanical stability and durability, (3) resistance to corrosion or chemicals, (4) dimensional stability, (5) minimal conduction of heat and electricity, (6) esthetic acceptance, (7) ease of manipulation, (8) adherence to tissues, (9) tasteless and odorless, (10) cleanable and repairable, (11) cost effective. Although it is desirable for an ideal restorative material to satisfy all these requirements, there presently is no one material that simultaneously fulfills all of them.

For more than a century, silver amalgam has been used in dentistry as a restorative material for posterior teeth, with clinically adequate results. **Dental amalgam** is an alloy formed by combining mercury with an alloy containing silver, tin, copper and sometimes zinc. The popularity of this material is a result of its several distinct advantages such as relatively low cost, ease of manipulation and placement, good wear resistance, low technique sensitivity, acceptable life expectancy and a self sealing ability (Leinfelder, 1993). Because of these advantages, amalgam is still the preferred material for use as a posterior restorative material, a core material for cast restorations, and as a retrograde root filling (Smith et al., 1986). Certain disadvantages, such as a lack of adhesion to the tooth structure, marginal leakage, susceptibility to tarnish and corrosion, and loss of marginal integrity have either restricted its use or limited its success (Ress and Valega, 1985).

Many attempts have been made to prevent the microleakage associated with amalgam restorations. Traditionally, cavity varnish such as Copalite varnish has been used to seal opened dentinal tubules and prevent microleakage. With time, slow dissolution of this varnish layer, results in a greater marginal discrepancy, and leads to further microleakage (Powell and Daines, 1987).

More recently, various substitutes for cavity varnish have been introduced. Zardiackas and Stoner (1983), introduced the concept of bonding amalgam restorations to tooth structure and since then, the use of adhesive resin bonding materials which are also very resistant to dissolution has been advocated. Although the adhesive bonding systems presently available have greatly improved marginal sealing of restorations, they are still not capable of completely eliminating microleakage (Chan et al., 1995; Inai et al., 1995).

Much of today's research on adhesives in dentistry is directed towards achieving a durable bond and a perfect seal between restorative materials and dentin. Although the clinical success of bonding restorative materials to enamel is well documented, the long-term success of bonding to dentin is currently unknown. The development of an ideal dentin bonding agent is still a challenge. Since dentin microstructure is completely different from that of enamel, the adhesion to dentin is weaker, less predictable and the restoration can experience microleakage. In fact, since the bond strength is not uniform across the bonding interface between tooth/restoration, regions of failure can form in an otherwise strongly bonded restoration and microleakage can occur through these areas. However, newer dentin bonding systems are becoming more predictable, and fewer untoward consequences, most often in the form of postoperative discomfort, are being experienced. Dentin bond strengths that approach the bond strength of enamel are being reported (Kanca, 1994).

Several studies have evaluated the effectiveness of some of the current dentin bonding systems as a seal for amalgam restorations. The majority of these were in-vitro studies using qualitative or semi-quantitative methods to assess microleakage and the marginal seal (Cooley et al., 1991; Charlton and Moore, 1992; Charlton et al., 1992; Saiku et al., 1993) . In most cases, Copalite varnish was used as the reference material. These studies lacked the simulation of the intra-oral environment because the teeth were isolated, their surfaces cut and ground, and usually dried with compressed air before restorative bonding was tested. In addition, the dentinal tubules did not contain any fluids during the testing and some methods required sectioning of the tooth structure. Little information is available in the literature on quantitative methods that measure microleakage of bonded amalgams, and few long-term in-vitro studies have been reported.

The purpose of this study was to evaluate quantitatively the effectiveness of adhesive dentin bonding systems on decreasing microleakage at the tooth-amalgam restoration interface.

LITERATURE REVIEW

MICROLEAKAGE

Microleakage is defined as the passage of saliva, fluids, bacteria and bacterial products, soluble ions and molecules through microgaps between prepared tooth surfaces and restorative materials (Kidd, 1976; Litkowski, 1990). Most restorative dental materials fail to adapt perfectly to tooth structure to provide a leak proof marginal seal, resulting in bacterial microleakage.

Figure **1** shows the three possible routes for microleakage, permitting microbial products access to dentinal tubules and underlying pulp (Pashley and Pashley, 1991). These routes are: 1) within or via the smear layer, 2) between the smear layer and the cavity liner or cement, or 3) between the cavity liner or cement and the restorative material. Some adhesive dentin bonding systems have shown promise in sealing and reducing microleakage of amalgam restorations by altering or completely removing the porous, permeable smear layer. As a result, the amalgam can adapt better to cavity walls, and reduce the interfacial space through which fluid may flow (Leelawat et al., 1992). More recently, Sano et al. (1995), described a new pathway for microleakage that can occur within the porous region of the hybrid layer (Sano et al., 1995; Inai et al., 1995), at gingival margins of gap-free composite restorations and is termed "nanoleakage" since it is measurable in nanoliters.

Causes of Microleakage Under Amalgam Restorations

There are many factors that may contribute to microleakage under amalgam restorations. Some are related to the properties of amalgam, material handling during preparation and placement and others relate to the properties and nature of the tooth structure. These factors are described as follows:

(1) Lack of adhesion

Amalgam alloys do not bond to tooth structure and gaps of 2 to 25 µm have been reported between the cavity walls and amalgam restorations (Saltzberg et al., 1976; Ben-Amar and Cardash, 1991). This means that marginal leakage is an on-going phenomenon under amalgam restorations. Figure 2 shows the dye pattern created during a microleakage test. The arrow points to the methylene blue dye which has diffused from the pulp chamber up to the cavosurface margin of an unlined amalgam restoration, clearly delineating the pathway of microleakage.

Amalgam restorations rely on cavity design for mechanical retention. These designs include inclination of walls, slots, occlusal dovetails and grooves or pins. Figure 3 shows mechanical retention created by an occlusal dovetail, and proximal grooves placed along the gingivo-axial line angle. Bonding would eliminate the need for these retentive features of the cavity preparation and would result in improved conservation of tooth structure.

(2) Differences in coefficients of thermal expansion between amalgam and tooth

Since the linear coefficient of thermal expansion of amalgam is about 2 to 3 times that of the tooth (Craig, 1989), thermal changes in the mouth from food or beverages can affect the sealing capacity of restorations. Cooling results in contraction and heating results in expansion of amalgam restorations. Depending on the geometry of the restoration these effects may either close or open the gap between the tooth and restoration, thereby reducing or increasing microleakage.

(3) Dimensional changes and surface texture of amalgam after insertion into cavity

Most modern (high-copper) amalgams exhibit a net contraction when triturated, (i.e. mixing amalgam alloy with mercury), with a mechanical amalgamator. Traditional (low-copper) amalgams, exhibit a net expansion. The classic picture of dimensional change as described by Phillips (1991), is one in which the amalgam undergoes an initial contraction for about 20 minutes from the beginning of trituration and then begins to expand slightly for the next 6 to 8 hours. When the alloy and mercury (Hg) are mixed contraction results as the mercury is absorbed by the alloy particles, and subsequently when the mixture solidifies, the reaction products occupy less volume than the reactants. However, if sufficient mercury is present in the mix, expansion will occur as a result of continuing crystallization and growth of both gamma-1 and gamma-2 phases (these phases are explained later). Therefore, manipulation that results in less mercury in the mix will favor contraction, such as lower mercury/alloy ratios and higher condensation pressure (i.e. packing amalgam alloy into the prepared cavity). Higher condensation pressures will squeeze mercury out of the amalgam, producing a lower mercury/alloy ratio and favoring contraction. Other manipulative procedures that accelerate setting and consumption of mercury also favor contraction, including longer trituration times and use of smaller particle size alloys. Smaller particle size accelerates the consumption of mercury because small particles have a larger surface area per unit mass than larger particles. Therefore, the end result of the dimensional changes could be either a slight final increase or a decrease of the width of the gap between tooth and restoration. Amalgam restorations which exhibit excessive contraction (i.e., 20 μm/cm or more), are more susceptible to marginal leakage than those with little or no contraction, or even a slight expansion (Fanian et al., 1983). The American National Standards Institutes/American Dental Association (ANSI/ADA) Specification No. 1 for dental amalgam alloy limits dimensional change to ± 20 μm/cm, in order to prevent excessive marginal leakage. Expanding amalgam restorations with a smooth surface texture showed less microleakage than those with a coarser texture (Mahler and Nelson, 1983). Coarse texture of an amalgam alloy prevents an intimate adaptation of the alloy particles to the cavity walls, resulting in gap formation and hence increasing microleakage.

(4) Amalgam alloy composition and setting reaction

When amalgam alloy (Ag_3 Sn) or what is known as the **gamma phase** is triturated (mixed) with mercury, metal atoms of silver (Ag), tin (Sn), and copper (Cu) dissolve from the outer surfaces of the particles into the mercury. The atoms then react with the mercury to form new crystal-metal compounds. The most common reaction is between silver and mercury to form the matrix (Ag_2-Hg_3) for the amalgam, and this is known as the **gamma-1 phase**. Gamma-1 phase occupies nearly 40% of the total volume of the amalgam and holds the original alloy particles together. Most of the rest of the volume of amalgam is occupied by unconsumed particles of gamma phase. Other compounds also form and those represent the difference between the low-copper and high-copper amalgams. In the low-copper amalgams, tin reacts with mercury to form tin-mercury (Sn_7-Hg) compounds, which is known as the **gamma-2 phase**. In contrast, in the high-copper amalgams, tin reacts with copper to form copper-tin (Cu_6-Sn_5) compounds or the **eta prime phase**. Since the gamma-2 phase is the weakest component in the amalgam alloy, and the least stable in the corrosive oral environment, the low-copper gamma-2 containing amalgams are more prone to corrosion, marginal breakdown and fracture compared to high-copper amalgam.

Corrosion can be beneficial for the survival of amalgam because the corrosion products, usually metal oxides, can build up over time and eventually plug up the marginal gap, thus effectively sealing the tooth from the oral environment. This is what is called the "self-sealing" ability of amalgam. Therefore, corrosion may actually improve the longevity of the restoration by reducing microleakage and the potential for secondary caries. The metallic ions themselves also may serve as bacteriostatic or bacteriocidal agents (Ferracane, 1995). It has been found that it takes about six months to two years for the corrosion products of high-copper amalgams to seal the gap between the tooth and restoration (Andrews and Hembree, 1978; Gottlieb et al., 1985). Therefore, the need to use liners, sealers, or cements under these restorations is important, particularly in the first few months after the amalgam insertion. The particle shape of amalgam may also influence the degree of microleakage. A spherical alloy is an alloy whose particles are created by means of an atomization process whereby a spray of tiny drops is allowed to solidify in an inert gaseous or liquid environment. Whereas an admixed alloy is an alloy containing a mixture of both spherical particles and lathe-cut particles. The later is made by machining a piece of alloy to create small, irregularly shaped chips of the alloy. The spherical alloy "Tytin" was found to have a greater tendency for marginal leakage than the admixed alloy

"Dispersalloy" (Mahler and Nelson, 1984; Cao et al, 1995).

(5) Technique

Factors that can be controlled by the dentist, such as condensation technique, mercury/alloy ratio, trituration procedure, and selection of an alloy, also govern the quality of an amalgam restoration.

a) Condensation

The specific objectives of condensation (placing the amalgam into the cavity) are 1) to adapt the amalgam intimately to the cavity walls and margins; 2) to develop a uniform, compact mass with minimum porosity within the amalgam; and 3) to reduce excess mercury content by using adequate force on the appropriate condenser. Poor condensation of fresh amalgam toward the cavity walls can result in voids along cavity margins and porosity in the restorative material. This poor condensation contributes to a higher degree of microleakage and inferior mechanical properties of the amalgam restoration. In addition, the presence of porosity in the amalgam restoration produces rough interface surfaces that can promote accumulation of plaque and encourage more corrosion. Mahler and Nelson (1984), found that condensation of amalgam increments directly along or against cavity walls in overlapping thrusts and with the proper amount of condensation force, are two important factors in reducing marginal leakage. Spherical amalgams have little "body" and thus offer only mild resistance to condensation forces. Therefore, lighter condensation pressures (i.e. 8 to 10 pounds) and larger condensers are used to place the amalgam. In addition, spherical amalgams have less mercury content (i.e., 42% to 46%) and hence less or no excess mercury is required to be removed during condensation. Lathe-cut amalgams, containing lath-cut particles on the other hand, have irregular shape particles which tend to lock together during condensation and a higher mercury content, thus require higher condensation forces to compact the particles together and to squeeze out excess mercury.

b) Mercury/alloy ratio

Increasing the mercury content in the low-copper amalgam alloy increases the setting expansion, the proportion of corrodible phases, and decreases the compressive strength of the alloy (O'Brien and Ryge, 1978). Since most operators today are using precapsulated high-copper alloy systems, the variability of mercury content has been largely eliminated.

c) Trituration time

Overtrituration, (mixing amalgam alloy with mercury using amalgamators for a longer period of time than recommended), generally reduces the working time by hastening the setting rate of amalgam, and results in excessive contraction. In contrast, undertrituration may be too short to coat all of the particles adequately with mercury. This produces amalgams that are not cohesive, porous and weak when set, and that have high-setting expansion and increased corrosion (O'Brien and Ryge, 1978). Thus, over or under trituration can produce a weak amalgam alloy that is difficult to condense, with high porosities, and increased microleakage.

d) Delayed condensation

Any delay after trituration allows the amalgam to set partially before being condensed. As a result, the amalgam restoration is more difficult to condense and experiences more microleakage because it is more porous, more prone to corrosion, and is less well adapted to cavity walls.

(6) Masticatory forces

The application of occlusal forces on Class II restored teeth may cause elastic deformation of tooth substance. The facial and lingual cavity walls can flex away from the restoration, resulting in an increase the width of the tooth/restoration gap (Granath and Moller, 1975; Morin et al., 1988). Occlusal forces transmitted from the restoration may create hygroscopic pressure on the fluid in the gap which together with bacteria can be pushed into the dentinal tubules toward the pulp (Pashley, 1985). Repeated flexure forces can ultimately result in fatigue failure causing debonding of bonded restorations (Heymann and Bayne, 1993). This type of failure in cervical Class V composite restorations has been reported to occur as a result of masticatory forces, bruxism or other unfavorable occlusion and tooth flexure.

Consequences of Microleakage

The **consequences of microleakage** are recurrent caries, postoperative sensitivity and pulp pathoses (Phillips, 1965; Ben-Amar and Cardash, 1991). The presence of bacteria and the leakage of fluid resulting from a lack of a seal between the tooth and restoration are believed to be the main factors responsible for inflammatory pulp reactions (Brannstrom, 1986; Cox et al., 1987). Bergenholtz (1977), demonstrated that live bacteria need not be present to cause severe pulpal reactions, only the supernatant of lyophilized bacterial preparations may cause such effect. Bacteria will rapidly colonize any gap present between restorations and tooth structure (Brannstrom and Nyborg, 1973) and the diffusion of their byproducts through dentinal tubules into the pulp can cause pulpal inflammation. In addition, microleakage has been regarded as detrimental to the longevity of dental restorations (Kiyomura, 1987). It may lead to marginal breakdown, dissolution of some cavity liners or cements and tooth discoloration under silver amalgam, and other restorative materials. Therefore, prevention of microleakage along the cavosurface margins of any restoration is a high priority to reduce or prevent such consequences.

Traditionally, amalgam has been retained in cavity preparations by mechanical retention. Recently, adhesive dentin bonding resins have been used under amalgam restorations to supplement the mechanical retention

(Ianzano et al., 1993). These materials have shown promise in reducing marginal microleakage (Hadavi et al., 1993; Moore et al., 1995; Fundingsland and Ario, 1995).

The next section will explain the principles of adhesion and its effect on bonding surfaces together.

ADHESION

The phenomenon of adhesion is involved in many situations in dentistry. It is also the primary method of solving the problem of leakage around dental restorative materials. Therefore, an understanding of the fundamentals associated with adhesion is important.

Principles of adhesion

Adhesion occurs when two dissimilar substances are brought into intimate contact with each other, and the molecules of one substance are attracted to molecules of another. The material or fluid film added to produce adhesion is known as the **adhesive**, and the material to which it is applied is called the **adherend** (Phillips, 1991; Ferracane, 1995). Strong, durable adhesion between two materials can be achieved through chemical or mechanical bonding (see section under types of bonding, page:20).

There are many factors which determine the effectiveness of the adhesion between two surfaces. The most important is probably the degree to which the adhesive wets the surface of the adherend. **Wetting** is characterized by the degree to which an adhesive will spread out on a surface. The angle made between the adhesive and the surface can be measured to give a value of wetting. This angle is called the **contact angle**. Figure **4** shows the contact angle created between liquids and solids surfaces with three different wetting abilities. A zero contact angle formed by the liquid surface at the interface separating the liquid and solid indicates that spontaneous spreading of the liquid has occurred and good wetting has taken place. Values above 90^0 indicate poor wetting. Good wetting promotes capillary penetration (movement of a liquid into a crevice or tube because of capillary attraction) and adhesion, and indicates strong attraction between the liquid and solid surface molecules. Recent adhesive bonding systems incorporate primers to facilitate better wetting of resins onto tooth and produce good adhesion.

Adhesion also depends on achieving **close contact between the adhesive and the adherend** to facilitate molecular attraction that allows either chemical interactions or penetration for micromechanical surface attachment (Beech, 1982; Kinloch, 1987). The attraction is generally negligible when the surface molecules of the attracting substances are separated by distances greater than 0.0007 µm, or 7 Angstroms. Essentially, anything that prevents the two surfaces to come into an intimate contact will compromise adhesion. Contamination of a surface is a deterrent to wetting because it reduces the **surface energy** of the adherend. Atoms and molecules at the surfaces of liquid and solids possess more surface energy than do those in the interior because all their potential chemical bonds are not satisfied (unbonded). The unattached bonds, so confer unstability and bond to other atoms or molecules. Therefore, any contaminant on the surface would interact with the unattached chemical bonds, reduce the surface energy of the adherend and reduce or prevent any wetting by the adhesive. In addition, the adhesive may actually bond to the adherend through the contaminant, resulting in a weak bonds. Since high-energy solids and low-energy liquids encourage good wetting, liquids generally wet high energy solids efficiently (for example, water on metals and oxides). Therefore, to obtain optimal adherence of a material to the tooth surface, it is necessary to clean the surface free of any contaminant such as saliva, plaque or blood, thus increasing the tooth's surface energy. Etching with various acids is generally used to increase the surface energy and the surface area removing the organic contaminants and by producing surface textures that can mechanically interlock with even poorly adherent restorative materials. Similarly, the adhesive has a surface energy of its own that is often referred to as **surface tension**, or the attraction that atoms and molecules on the surface of a liquid have for one another. A high surface tension would inhibit the spreading of an adhesive, which directly affects wetting ability, and therefore, adhesion. An adhesive with a low surface tension and an adherend with a high surface energy will result in good adhesion.

Thickness of the resin adhesive layer has also been shown to affect bond strength. Some recent literature indicates that when bonding is present, an optimum thickness (i.e., 200 µm) of an adhesive layer exists, but at levels above or below this thickness the bond strength is reduced. Davidson et al. (1991), found that the thinner the resin adhesive layer, the faster the stress development in the composite. Due to the lower flow capacity of thin layers, adhesive failure results with composite restorations. On the other hand, thicker adhesive films may include some defects such as porosity and air bubbles that will decrease the bond strength and result in failure of the adhesive restoration. The dimensional change that the adhesive undergoes after it is placed may also dictate whether it remains attached or pulls away from the surface. Most dental adhesives are composed of polymeric materials that shrink during hardening. This shrinkage can pull them away from the tooth surface, compromising the adhesion. Currently, researchers are trying to develop resin materials that do not shrink upon polymerization.

Finally, for good spreading the adhesive must have a low viscosity. **Viscosity** is a measure of a liquid's resistance to forces that tend to cause it to flow. The units used to define viscosity are gm/cm-sec or poise, a centipoise is 1/100 of a poise. An adhesive with a high viscosity would have a high resistance to flow and would be less likely to spread. On the other hand, if the viscosity is too low, it may be hard to control the placement of the adhesive. As discussed previously, increased bond strengths result when the adhesive layer has the optimum thickness and when both the adhesive and adherends have high cohesive strengths. **Cohesion** is the force of attraction between the molecules or atoms within a given material.

For effective adhesion the principles outlined above should not be compromised, otherwise adhesion will be weak or might not occur. The following section discusses the types of bonding that can exist between dental restorative materials and tissue surfaces.

TYPES OF BONDING

The two types of mechanisms which bond dental materials to tooth structure or to other dental materials are mechanical and chemical.

Mechanical Bonding

Mechanical bonding results from the physical interlocking of one material with another. Such structural retention may be at a visible or macromechanical level as in the case of screws, grooves, slots, or undercuts (Fig 3), or it may be through more subtle non-visible mechanisms, as in the case of penetration of an adhesive into microscopic or submicroscopic irregularities micro-mechanical crevices and pores in the surface of the adherend. A liquid adhesive penetrates into these surface discrepancies and upon hardening, the multitude of adhesive projections embedded in the adherent surface provide the sites for micromechanical attachment or retention (Phillips, 1991). This mechanism has been commonly used in dentistry with adhesive cements, or adhesive restorative materials.

Chemical Bonding

There are two types of chemical bonding: primary and secondary (Ferracane, 1995). **Primary bonding** results from the interaction of the outer shell electrons of two or more atoms, whereas **secondary bonding** results from the interaction of unlike charges on atoms or molecules. There are three types of primary bond: the **covalent** bond is the result of sharing of electrons between two atoms, while the **metallic** bond is the result of many atoms sharing all of their outer electrons with their neighbors. The strongest and most stable primary bonds are the covalent and metallic bonds. The third type, the **ionic bond**, may also give strong adhesion. Ionic bonds are generated by the interaction of a positive with a negative charge. One atom donates its electron to the other, and both atoms become stable through the linkage. Ionic bonds however are not relied on heavily in dentistry, because they are often unstable in aqueous environments. Water dissociates to form OH^- and H^+ ions. The water molecule forms a **dipole** (a dipole consists of two equal and opposite, for example, positive and negative charges that exist on H^+ and OH^- ions in water molecule). Ideally, a covalent bond between the adhesive molecules and one of the component elements of the of the tooth dentin structure is desirable in order to prolong the tenacity or permanence of the bond.

Secondary bonds or intermolecular bonds are less stable than primary bonds and result from the electrostatic attractions of dipoles. This weaker bond may be said to be more physical than chemical in nature. There are two kinds of secondary bonds, **hydrogen bonds** and **van der Waals forces**. Hydrogen bonds refers to the use of a hydrogen atom as a bridge to link together two electronegative atoms (where a permanent dipole is formed between hydrogen atom and oxygen atom). For example, in water, adjacent water molecules are attracted by hydrogen bonds between the electronegative oxygen atoms. Hydrogen bonding is common in protein molecules and in DNA. Van der Waals forces results from the attraction between an electric dipole in each of the atoms. The dipoles are not permanent ones as in water molecules, but have their origin in the fluctuating positions of the negatively charged electrons about the positively charged nucleus. Secondary bonding plays a major role in wetting and is essential for achieving good micromechanical bonding. For example, it is responsible for the initial wetting and penetration of the adhesive bonding materials into the tooth structure, while strong primary bonding develops later. However, it is very weak, typically having only 1/10 the strength of primary bonding, and may be subsequently disrupted by water.

During the bonding of resin restorative materials to tooth structure both mechanical and chemical bonding may occur. The principles of bonding to tooth structure are explained below.

(A) BONDING TO TOOTH STRUCTURE

(1) Bonding Resins to Enamel

The acid-etch technique was introduced by Buonocore in 1955 and was developed to bond restorative resins to enamel through micromechanical adhesion. Acid-etching removes about 10 µm of the enamel from the tooth surface including the smear layer, and creates a rough, porous layer ranging from 5 to 50 µm deep (Gwinnett and Buonocore, 1965). Etching also increases the wettability and surface area of the enamel substrate. When a low-viscosity resin with surface energy characteristics that cause it to be attracted to the enamel is applied, it flows and penetrates into the microporosities and channels of the etched layer and polymerizes to form a micromechanical interlocking with exposed enamel rods. This unique structure, composed of enamel tags imbedded in resin matrix is often referred to as the "enamel-resin hybrid layer" (Nakabayashi, 1982, 1994). Figure **5** is an illustration of the formation of resin tags in addition to the hybrid layer where a mixture of resin monomer penetrates the etched enamel.

A 60-second etching time has been traditionally used for etching enamel with 30% to 40% phosphoric acid. However, subsequent studies have indicated that etching for 15 seconds provides essentially the same morphological changes as the 60-second etching time (Barkmeier et al., 1986, 1987). The shear bond strengths and marginal microleakage were similar for both 15- and 60-second etching times (Shaffer et al., 1987; Crim and Shay, 1987). Furthermore, lower concentrations 10% to 15% of phosphoric acid have been shown to provide bond strengths similar to those obtained with the 30% to 40% acid (Gwinnett and Kanca, 1992).

Chemical bonding to the inorganic hydroxyapatite (HAP) crystals in enamel may also occur, since enamel contains about 92% volume inorganic structure. Different organic compounds are bonded to the calcium and phosphate ions via ionic bonding (Wilson et al., 1983; Ruyter, 1992). However, micromechanical bonding is the primary mechanism of bonding resins to enamel.

Benefits of Bonding to Enamel

Shear strengths of resin bonded to etched enamel immediately after bonding are typically in the range of 15 to 22 mega pascal (MPa) (Hasegawa et al., 1992; Souza et al., 1993). Such resin bond strengths provide routinely successful retention of restorations, including direct anterior and posterior composite restorations; porcelain and composite resin veneers, inlays, onlays, and porcelain crowns; orthodontic brackets; resin-bonded cast-metal prostheses; and pit and fissure sealants.

Etching reduces microleakage around restoration margins in enamel through the intimate adhesion of the resin to etched tooth structure (Shaffer et al., 1987; Crim and Shay, 1987). Composite resins shrink as they polymerize, and contraction stresses of up to 7 MPa develop within the resin (Bowen et al., 1983; Davidson and De Gee, 1984; Davidson et al., 1984). However, these stresses may be partially compensated by plastic deformation or flow of the composite resin. The magnitude of these stresses varies with the ratio of bonded to unbonded surface of the resin material. The greater the fraction of restoration surface that is bonded, the greater will be the final contraction stresses. In other words, if the ratio of bonded surface to that of unbonded surface is more than 1.0, much greater stresses occur within the material and spontaneous debonding is most likely to occur (Davidson and De Gee, 1984; Davidson et al., 1984; Feilzer et al., 1987). It has been estimated that shear bond strengths of 17 to 20 MPa are required and to resist contraction forces sufficiently to produce gap-free restoration margins (Davidson et al., 1984; Munksgaard et al., 1985). Enamel bond strengths are generally adequate to prevent opening of margins due to polymerization shrinkage.

Composite resin restorations bonded to enamel provide reinforcement of cusps (Swift et al., 1995) which is another important benefit. Cavity preparations weaken tooth structure due to loss of dentin bulk and increase the risk of cuspal fracture. Conventional non-adhesive restorative materials provide little or no reinforcement to the weakened tooth structure.

Although clinical success has been achieved with respect to enamel bonding, bonding to dentin is still a challenge. The next section addresses the factors that affect adhesion to dentin.

(2) Bonding resins to Dentin

Factors Affecting Dentin Adhesion

Adhesion of restorative materials to enamel has become a routine and reliable aspect of modern restorative dentistry, but adhesion to dentin has proved to be more difficult and less predictable. Much of the difficulty in bonding to dentin is the result of the complex microstructure and variable composition of dentin in different areas of the tooth.

(a) Microstructure and Chemical Composition of Dentin

Dentin can be considered as a complex hydrated composite of four elements: **(1)** oriented dentinal tubules containing cell processes surrounded by **(2)** a collar of hypermineralized peritubular dentin, embedded in **(3)** an intertubular matrix consisting largely of collagen fibrils with embedded hydroxyapatite (HAP) crystals, and **(4)** dentinal fluid (Marshall, 1993).

Expressed in volume %, dentin is composed of 50% inorganic (HAP), 30% organic matter (mostly collagen, small quantities of citric acid, insoluble protein, mucopolysaccharide and lipid), and about 20% fluid (Driessens and Verbeeck, 1990). In comparison, enamel is composed of 92% inorganic minerals (HAP), and about 2% organic matter, and 6% water. Thus, the composition of dentin is much different than that of enamel, and one would expect it to behave differently when etched with acid. The presence of water and organic components lower the surface energy of dentin and make bonding with hydrophobic resins essentially impossible. These structural and compositional differences and their affect on adhesion to dentin will be explained in the following section.

(b) Tubular Variation

Dentin is intimately connected with the pulpal tissues, and numerous microscopic tubules transverse through dentin from the pulp to the dentinoenamel junction (DEJ) serving as diffusion channels. These tubules make dentin permeable to many substances and provide access to the pulp. The tubules have irregular walls with many lateral branches and microchannels that connect neighboring tubules (Pashley, 1989a). Normally, they are filled with fluid, odontoblastic processes and may be associated with mineral deposits and recently Marshall (1993), described intratubular deposits of collagen. Presumably, any changes in tubule diameter caused by internal deposits or reduction in the actual size (Bergenholtz et al., 1993) produce large changes in dentin permeability and the rate of fluid flow through tubules. This process of variation in dentin permeability occurs progressively throughout the life of the tooth.

Unlike etched enamel, which provides a relatively consistent surface for bonding agents to adhere to, dentin presents widely different surfaces depending on such factors as the plane and depth of the cut and the distance from the pulp. This creates highly variable bonding surfaces. In order to be effective for dentin an adhesive system must, in essence, be compatible with different structural interfaces.

Pashley (1989a), calculated the fractional areas occupied by tubules, peritubular matrix, and intertubular matrix for occlusal dentin as a function of position. Tubule area and diameter vary from about 22% and 2.5 µm, respectively, near the pulp (deep dentin), to only 1% and 0.8 µm, respectively, at the dentin enamel junction (DEJ) (superficial dentin). The intertubular matrix area reportedly varies from 12% near the pulp to 96% near the DEJ, whereas peritubular dentin decreases from more than 66% near the pulp to 2.9% at the DEJ (Pashley, 1989a). Figure **6** illustrates the ratio of tubular area to solid intertubular dentin as it varies with proximity to the pulp and tooth age (Paul and Scharer, 1993).

Regional variations in dentinal structure and composition are related to other factors besides depth. Regional variation is reflected in the permeability characteristics at different locations within a tooth. For example, the permeability of occlusal dentin is higher over the pulp horns than at the center of the occlusal surface (Pashley et al., 1987) as a consequence of the convergence and increasing diameter of the tubules closer to the pulp, as shown in Figure 7. Similarly, proximal dentin is more permeable than occlusal dentin, and coronal dentin is more permeable than root dentin (Fogel et al., 1988; Pashley and Pashley, 1991).

Adhesion can be affected by these changes in relative surface area. Some dentin bonding systems bond preferentially to intertubular (superficial) dentin rather than peritubular (deep) dentin. Suzuki and Finger (1988), and Prati and Pashley (1992), reported lower bond strengths to deep dentin compared to superficial dentin. Whether the low bond strengths are due to increased wetness or the inability of adhesives to bond to peritubular dentin is unknown. If bonding was solely to the tubules or to the intertubular dentin, we would expect the bond strengths to be directly related to the fraction of either tubules or intertubular dentin composing the bonding surfaces. Since systems such as Superbond that contain 4-methacryloxyethyl trimellitate anhydride (4-META), were found to bond equally well to both superficial and deep dentin (bond strength of 15-18 MPa) in the presence or absence of pulpal pressure (Tao et al., 1991; Prati and Pashley, 1992) resins probably do not derive their adhesiveness solely from penetration of resin into the tubules, but also by interacting with demineralized intertubular dentin. Resin tags will contribute to bond strength only if they are firmly bonded to peritubular dentin. An ideal bonding system would bond to both intertubular and peritubular dentin (Pashley, 1991).

(c) Tubular Fluid

The fluid content of dentin varies from approximately 1% in superficial dentin to 22% in deep dentin (Pashley, 1990). The fluid in the tubules moves in a bi-directional way, via capillary action and pulpal pressure from the oral cavity across dentin to the pulp and vice versa. The intrapulpal pressure is estimated to be approximately 10 to 30 mm Hg (or 15 to 40 cm H_2O) (Andreaus et al., 1989; Vongsavan and Mathews, 1992). Kim et al. (1984), indicated that whenever pulpal blood flow is reduced by, for instance, vasoconstrictors from the use of local anesthetics, pulpal pressure falls. Whereas, during pulpal inflammation, pulpal pressure rise and may be responsible for pulpal pain (Van Hassel, 1971; Tonder and Kvinnsland, 1983). Fluid movement has been shown, at least in-vitro, to severely affect adhesion of some adhesive systems such as Scotchbond 2 (3M Dental) and Tenure (DenMat), however it may not affect All-Bond system (Bisco Dental) (Prati and Pashley, 1992). Unless the adhesive bonding agent can displace or is miscible with the fluid in areas where the dentin is most permeable, it will be difficult to achieve a stable and long-lasting bond (Marshall, 1993). Elhabashy et al. (1993), reported that shear bond strengths of five dentin adhesives including Amalgambond (Parkell) were reduced by 26-46% by bonding to physiologically hydrated dentin. Figure 7 shows dentinal fluid diffusing through the pulpal floor of a Class I occlusal cavity using the intrapulpal fluid pressure technique.

Rapid movement of fluid within the dentinal tubules elicits a sharp well localized pain by activating A-delta pain fibers according to the hydrodynamic theory of dentin sensitivity (Brannstrom et al., 1967). This pain is produced by stimuli that expand or contract dentinal fluid such as hypertonic solutions, thermal changes, physical pressure and blasts of air (Pashley and Pashley, 1991). Hypertonic solutions such as those formed in the mouth by consuming sugars, can osmotically move fluid away from the pulp, through the tubules to the surface (Pashley, 1989b). Fluid also moves toward the pulp whenever castings are seated with cements, or patients bite on an incompressible object that can force fluid through marginal defects around restorations (Pashley, 1989b).

(d) Smear Layer

A smear layer is created whenever dentin is cut or ground with hand or rotary dental instruments and is about 1 to 5 µm thick (Brannstrom, 1982). It consists of debris that presumably reflects the composition of the underlying dentin. Thus, the composition of superficial versus deep smear layers would be expected to be quite different. It has a uniform, amorphous structure which appears to completely occlude the orifices of all dentinal tubules. Smear plugs are projections of the smear layer that extend to variable distances into dentinal tubules when the smear layer is created (Fig. 8).

The smear layer has both advantages and disadvantages in the bonding process (Yap et al., 1994). The main advantages include the reduction of dentin permeability to toxins and oral fluids; the reduction of diffusion (usually inwards) and convection of fluids (outwards by hydrostatic pressure or inwards, for example, while restorations are cemented), the reduction of wetness of cut dentin surfaces, and the prevention of bacterial penetration of dentinal tubules (Olgart et al., 1979; Vojinovic et al., 1973; Michelich et al., 1980). The main disadvantages include its wetness and the harboring of bacteria or their products which may multiply and permeate into the pulp, thereby eliciting an inflammatory reaction. The smear layer may also reduce the adhesion of most bonding systems. It reduces the surface energy which compromises surface wetting, prevents the intimate surface contact between the adhesive and dentin substrate, and prevents resin penetration into dentinal tubules (Gwinnett, 1984; Rees and Jacobsen, 1990). It acts like a separating agent since it is an intrinsically weak structure (5 to 6 MPa cohesive strength) that bonds weakly to the underlying dentin (Tao and Pashley, 1988). Further, the mineral component of the smear layer may dissolve slowly under a leaking restoration or be removed by acid produced by bacteria to leave a gap which in turn increases marginal leakage and possibly secondary caries (Pashley, 1984).

Many recently developed adhesive systems remove the smear layer by using some type of primer or conditioner to increase the surface energy and optimize the bonding to dentin (Sandoval et al., 1989), and this trend is likely to continue. However, the removal of smear layers and plugs increases the permeability of the dentin in-vitro by 5 to 20% (Pashley, 1989a). This should not be a problem if the bond completely seals the dentin/restoration interface. Recent studies have indicated that some currently marketed resin-based dentin bonding systems form strong bonds to moist dentin (Gwinnett, 1992), using acetone-free hydrophilic primers that contain 4-META and hydroxyethyl methacrylate (HEMA) (Amalgambond) or acetone-containing hydrophilic primer solution (All-Bond). Acetone which apparently acts as a water chaser, allows the primer to displace water and to flow over moist surfaces, carrying the hydrophobic resin monomers into the demineralized dentin (Kanca, 1992b). The acetone subsequently evaporates with the water depositing the resin monomer into the collagen fibril matt (the role of acetone will be explained later in detail). Swift and Triolo (1992) and Charlton and Beaty (1994), found no significant difference in bond strengths between dry and wet dentin using hydrophilic ethanol containing polycarboxylic acid and/or (HEMA) monomers free of acetone. Vargas and Swift (1994), found that microleakage of composite restorations bonded with All-Bond 2 or Scotchbond Multi-Purpose (3M Dental) (ethanol containing primer solution), was similar regardless of whether a wet or dry bonding technique was used.

(e) Hypermineralized dentin

Bonding to hypermineralized dentin surfaces such as sclerotic dentin or beneath carious lesions, is even more difficult than bonding to normal dentin. The partial or total obliteration of the tubules and intertubular dentin with mineral deposits may prevent reliable bonding of resins. Perdigao et al. (1994), suggested that increased peritubular dentin and mineral deposits probably prevent adequate etching and resin penetration, resulting in the formation of thin hybrid layers. Van Meerbeek et al. (1994), showed that a limited thickness of hybrid layer (0.5 to 1 μm) was formed at the hypermineralized intertubular dentin with no or only short resin tags present in most dentinal tubules. Nakajima et al. (1995), reported significantly higher tensile bond strengths to normal dentin (26.9 MPa) than to carious dentin (13.0 MPa) and concluded that appropriate mechanical retention should be used in these clinical situations. This is of particular importance if these materials are to be used in the older dentition or in cervical or root restorations.

Although micromechanical bonding of resin materials to enamel has been very successful, adhesion to dentin has continued to be a challenge. Early researchers developed dentin bonding systems that used phosphoric or citric acid to open the dentin tubules to allow the resin to flow 100 μm or more into them for mechanical retention. However, unlike acid etching of enamel, dentin etching produces lower shear bond strength and when used without mechanical retention, debonding often occurs in a very short period of time. Although early reports of the potential for pulpal damage following acid treatment of dentin (Retief et al., 1974) initially discouraged further research on this concepts, current research has produced a multitude of dentin bonding systems based upon a variety of chemical and most recently micromechanical mechanisms for adhesion that appear to have little adverse effect on the pulp.

The following section describes the development of dentin adhesive systems through the years.

DENTIN ADHESIVES

Development of Dentin Adhesives

Over the course of the last two decades, dentin adhesives have undergone considerable evolution. They are often grouped into four generations based on their bonding procedures (Heymann and Bayne, 1993; Leinfelder, 1993). These groups are as follows.

a) First generation dentin adhesives

A tremendous effort was made to identify ways to form chemical bonds between resin adhesives and dentin. Buonocore et al. (1956), introduced the "first-generation" dentin bonding systems consisting of a methacrylate-based primer with a glycerophosphoric acid bifunctional group capable of bonding to the calcium ions of the hydroxyapatite on the dentin surface and capable of bonding to the resin restorative materials by means of an addition polymerization reaction between C=C double bonds. Bowen (1965), synthesized N-phenylglycine glycidyl methacrylate (NPG-GMA), a "surface-active comonomer" which promoted chemical bonds between the dentin and resin restorations. Examples of dentinal adhesives of this generation included Cervident adhesive (SS White, Holmdel, NJ), and Cosmic Bond adhesive (De Trey, Weybridge, Surrey, England). Palakav adhesive (Kulzer, Irvine, CA) is another system in the first generation but was designed to facilitate chemical bonding to the amino-acids in the collagen structure and used tri-n-butyl borane (TBB) as a co-catalyst.

Although improvements in the bond strengths to dry dentin substrate were achieved, they were usually only 2 to 3 MPa and could not be duplicated under moist conditions, because they were further diminished by hydrolysis. The results of clinical trials were poor and there was no improvement in marginal leakage when compared to conventional unfilled enamel bonding resins (Barkmeier and Cooley, 1991).

b) Second generation dentin adhesives

The "second-generation" dentin bonding agents were also based on the calcium-phosphate ionic bond. The bisphenol-glycidyle methacrylate (Bis-GMA) molecule was substituted for the methacrylate, and an unfilled resin was added as a diluent to the Bis-GMA resin. Most of these agents were halophosphorous esters of unfilled resins such as Bis-GMA, and others were based on polyurethane. Examples of these materials included Scotchbond (3M, St Paul, MN), Clearfil (Kurary, Osaka, Japan), Bondlite (Kerr), Prisma Universal Bond (L.D. Caulk, Milford, DE), and Dentin Bonding Agent (Johnson & Johnson, East Windsor, NJ). Dentin Adhesit (Vivadent Schaan, Liechtenstein), is also considered a second generation product, but its chemistry was based on an isocynate monomer. This monomer is capable of forming van der Waal's bonds to the —OH, —COOH and —NH_2 groups located on collagen.

The clinical performance of these bonding agents was relatively poor, and shear dentinal bond strengths of only 2 to 6 MPa were reported. A major reason for the poor performance of these bonding agents is the fact that they bond to the smear layer rather than to the underlying dentin. Hence, their bond strength is limited by the cohesive strength of the smear layer or by adhesion of the smear layer to the dentin below (Tao et al., 1988). These bond strengths were not great enough to counteract contraction stress generated by shrinking composites, which is estimated to exceed 20 MPa in some cavities. Furthermore, some studies indicate that bonds between phosphonate esters and dentin may be also hydrolyzed by immersion in water. Therefore, composite resin tends to separate from dentin, forming gaps at restoration margins and increasing the microleakage at margins in dentin or cementum (Swift et al., 1995).

c) Third generation dentin adhesives

The third generation adhesive systems used a primer or dentin conditioning step in conjunction with a hydrophilic resin adhesive. These conditioners or acidic primers attempt to modify or remove the smear layer to allow resin penetration into the underlying dentin for micromechanical retention. In contrast to the conditioner, the primer leaves behind a monomer residue that promotes easier wetting with the adhesive. Shear dentinal bond strengths of agents such as Scotchbond 2, Tenure, Gluma (Bayer/Miles), Prisma Universal Bond 3 (Caulk/Dentsply), Syntac (Ivoclar

Vivadent), and XR Bond (Kerr) usually are greater than those of the second generation agents and can approach (i. e., 10 to 12 MPa) (Leinfelder, 1993) the typical bond strengths of resin to etched enamel. However, their performance still unpredictable, even in laboratory studies. Bond strength variations occur not only among different studies, but also within studies (Swift et al., 1995). The application process remained too complicated and pulp protection from acid conditioners was still recommended in deep preparations. These dentinal adhesives are generally more effective than their predecessors in reducing microleakage at dentinal and cementum margins, although they do not completely eliminate marginal leakage (Swift et al., 1995).

d) Fourth generation dentin adhesives

The concept of the "total etch" technique or simultaneously etching the enamel and dentin with phosphoric acid was first proposed by Fusayama et al., in 1979. Because at that time, acids, particularly phosphoric acid, were considered a cause of irreversible pulpal pathology (Retief et al., 1974), dentin etching had been discouraged. However, the results of recent studies (Cox, 1992) suggested that the role of bacteria and microleakage (Brannstrom and Nyborg, 1971) resulting from a lack of a seal between the restoration and tooth structure is a more acceptable explanation for the pathological findings in the pulp (Cox et al., 1987). Recently, the total-etching technique has been accepted and widely used in the current dentin bonding systems. Although many different types of conditioners, primers, and adhesive resins are used, the bonding mechanisms of the various dentin adhesive systems are similar (Van Meerbeek et al., 1992; Perdigao and Swift, 1994). This bonding mechanism which is primarily micromechanical bonding and is explained in more detail in the following section.

Techniques and Mechanisms of Current Dentin Adhesives

The principles for the use of a typical fourth generation system involves a three step procedure. Step one (conditioning) involves the application of an acid etchant such as either 10% or 37% phosphoric acid, nitric acid, citric acid, maleic acid or EDTA (ethylene-diaminetetraacetic acid). Acid etching removes the smear layer, opens the dentinal tubules, increases dentin permeability, and demineralizes approximately 10 µm of intertubular and peritubular dentin (Leinfelder, 1994), to form a collagen fibril matt structure. The depth of demineralization is affected by various factors, including the pH, concentration, viscosity, and application time of the etchant (Van Meerbeek et al., 1992; Pashley, 1992). Removal of HAP crystals leaves a collagen meshwork that can collapse and shrink because of the loss of the inorganic support that has been replaced by water (Van Meerbeek et al., 1992; Inokoshi et al., 1993). However, recent studies reported that after acid-etching the dentin, a thin layer (0.1 to 0.3 µm) of amorphous-insoluble collagen (collagen smear layer) remains at the top of the fibril matt (Pashley et al., 1993a). This weak layer may obscure the underlying porosity of the etched dentin, affecting the resin infiltration and interfering with the bonding, especially when the surface is air dried (Cobb et al., 1995). Air drying results in the shrinkage and collapse of the collagen fibrils. This reduces the inter-fibril space, and decreases the ability of the resin to infiltrate the collagen fibril matt.

Step two involves the application of a separate primer to the dentin surface. The primer is a hydrophilic resin-containing solution specially formulated to wet the dentin surface, to infiltrate around collagen network and into any open tubules, raising the collapsed fibrils almost to their original level. It also increases the surface energy, and hence the wettability, of the dentin surface. Primer molecules such as HEMA, pyromellitic diethylmethacrylate (PM-DM) and biphenyl dimethacrylate (BP-DM) and 4-META contain two functional groups; a hydrophilic group that has an affinity for the dentinal substrate and, a hydrophobic (methacrylate) group that has an affinity for the resin. These resins either have, or subsequently form, hydroxyl groups which allows them to wet the moist dentin and give them some potential to bond to the tooth via the calcium ions in HAP crystals. Some other systems dissolve hydrophobic resins in acetone to create hydrophilic solutions.

The third step involves the application of the adhesive material onto the primed dentin. The adhesive is essentially an unfilled resin similar in composition to the resin in composite, except that hydrophilic molecules have been added such as 4-META copolymerized with methylmethacrylate (MMA), or Bis-GMA, or urethane dimethacrylate (UDMA) with HEMA. The resin penetrates and impregnates the collagen fibrils copolymerizing with both the primer on one side; to form an interlocking network of collagen and resin, and the restorative composite resin on the other side. The **resin-dentin interface** region that is called the "resin-reinforced zone", "resin-infiltrated layer", or "hybrid layer", was first described by Nakabayashi et al. in 1982, and has emerged as the principal bonding mechanism for virtually all the current dentin adhesive systems (Inokoshi et al., 1990; Suzuki et al., 1991; Spencer et al., 1992; Erickson, 1992; Van Meerbeek et al., 1993; Nakabayashi, 1993; Yap et al., 1994). In addition, it has been shown with 4-META that in the presence of water the anhydride radical is hydrolyzed to form a polycarboxylic acid which reacted by chelation to the calcium ions in HAP tooth substrate. Figure **9-A** shows resin impregnated dentinal tubules and Figure **9-B** shows resin impregnated collagen fibril matt. Currently, micromechanical bonding is emphasized over chemical bonding, and any benefits of chemical bonding appear secondary both to good wetting and to intimate micromechanical bonding. Examples of the fourth generation dentin adhesives include, All-Bond 2, Amalgambond Plus, Scotchbond Multi-Purpose Plus (3M), Clearfil Liner Bond (Kuraray/J Morita), Impreva Bond (Shofu Dental), and OptiBond (Kerr). The first two materials, because of their relation to this study, will be discussed in further detail later.

The **hybrid layer** of collagened resin may be 3 to 10 μm thick, depending on the system used (Tay et al., 1995; Nara et al., 1995). It plays a dual role; first, it completely seals the dentinal tubules and prevents the penetration of fluids and microorganisms into the pulpal tissue via the tubules. Secondly, it prevents the outward flow of the odontoblastic fluids to the surface of the preparation, thereby eliminating or substantially reducing postoperative sensitivity. Further, it is insoluble in acid (Inokoshi et al., 1990; Wang and Nakabayashi, 1991; Van Meerbeek et al., 1993; Tay et al., 1994; Cox and Suzuki, 1994) and if the seal is well maintained, recurrent caries and pulp pathoses could be prevented for long time (Nakabayashi et al., 1991). Bonding agents that form a hybrid layer generally have higher bond strengths than those not forming hybrid layer (Gwinnett and Kanca, 1992; Nakabayashi et al., 1991). The results of a recent in-vitro study by Gwinnett (1993), indicated that hybridization of the dentinal tubules and intertubular dentin accounted for a substantial proportion of the bond of resin to dentin.

Variables Affecting the Formation of the Hybrid Layer Formation

To achieve good bonding to the resin/dentin hybrid layer, the following requirements should be fulfilled. First, the materials used for acid-etching dentin must remove the smear layer along with smear plugs, demineralize the underlying dentin to a certain depth, without denaturing the collagen fibrils or damaging the pulp irreversibly. Denatured collagen (due to the loss of the supporting inorganic HAP crystals) has been shown to shrink or collapse quite easily which in effect decreases the porosity and penetrability of collagen network (Van Meerbeek et al., 1992). However, ferric ions in Amalgambond dentin conditioner (10% citric acid and 3% ferric chloride; 10-3 solution) can effectively stabilize collagen and prevent its denaturation and collapse, thereby increasing the permeability of the dentin substrate. Further desiccation of the conditioned demineralized dentin reduces its volume by 65% but this shrinkage can be restored to its original shape after re-immersion in water (Carvalho et al., 1995). According to the manufacturers instructions, All-Bond 2 technique requires that the dentin surface be kept moist after conditioning and this method is termed as "wet bonding technology". Several studies have indicated that shear bond strengths of All-Bond 2 exceed the typical enamel bond strength of 20 MPa, particularly when dentin is left visibly moist after etching (Gwinnett, 1992; Kanca, 1992a). Perdigao et al., (1993) showed that both All-Bond and

Amalgambond systems bonded preferentially to moist dentin. Some what lower bond strengths have been reported in other studies (Retief et al., 1992; Souza et al., 1993). In addition, 4-META systems appear to be less affected than are most other adhesives by dentinal depth and pulpal pressure. Similarly, All-Bond 2 system has been shown not to be affected by water for up to one year (Nakabayashi et al., 1992a; Tjan et al., 1992).

The second requirement is that the primer must contain hydrophilic resins (Nakabayashi and Takarada, 1992), or resins dissolved in a hydrophilic solvent (Kanca, 1991; Gwinnett, 1992), so that they will easily wet the moist dentin surface, and infiltrate both the collagen network and any open tubules. These primers must also copolymerize with the resin bonding agent by means of free radical C=C double bond breaking. Amalgambond primer is 35% HEMA in water which acts as a wetting agent, improves the permeability of demineralized dentin, even if it had been collapsed, and increases the diffusion and penetration of 4-META resin monomer. Hence, HEMA may be essential not only for the formation of the hybrid layer formation but also for a gap-free dentin-resin interface (Nakabayashi and Takarada, 1992; Nakabayashi et al., 1992b). In the case of All-Bond 2, the primer is composed of aromatic carboxylic acid monomers, [2% N-tolyglycine-glycidyl methacrylate (NTG-GMA) and 16% BP-DM] that are dissolved in acetone solution. The acetone, as explained earlier, displaces water and deposits the resin monomers into the demineralized dentin. This occurs when the acetone/resin primer mixes with water, the vapor pressure of the water is raised, and the water becomes volatile. In addition, the surface tension of the water is reduced, allowing rapid spreading of the primer to wherever the water was present. In effect the acetone and water vaporize from the substrate depositing the resin intimately along dentinal surfaces and deeply into dentin tubules (Kanca and Gwinnett, 1994). Advantages of acetone are: 1) it creates an acetone vapor cloud over resin, preventing air inhibition of the resin during both air thinning or air drying step that is required to evaporate the acetone solvent from within the fibril matt or dentinal tubules, this improves the rate of polymerization of this thin resin layer, and 2) if acetone has been used as solvent for a hydrophobic monomer, then once the acetone has evaporated, the resin is non-water soluble. In contrast, water soluble resins such as HEMA, may be softened by the ingress of dentinal fluid causing a decrease in bond strength with time.

The third requirement is that conditions such as acid type and concentration, and conditioning time must be carefully controlled. Adhesion is compromised if the depth of demineralization exceeds the depth of monomer penetration (Erickson, 1992; Pashley et al., 1992; Van Meerbeek et al., 1992). If dentin is decalcified so deeply that the decalcified zone cannot be thoroughly impregnated by resin, a fragile non-impregnated, decalcified collagen layer may remain resulting in a weak hybrid layer that will degrade over time (Nakabayashi et al., 1992a; Van Meerbeek et al., 1992). Superficial decalcification of the dentin will not allow sufficient resin interpenetration to create a strong bond. The depth of demineralization has been reported to be about 2 μm with Amalgambond dentin conditioner (10-3) (Fukushima and Horibe, 1990; Van Meerbeek et al., 1992; Nakabayashi et al., 1992a), while it was 7.5 μm with All-Bond 2 (10% phosphoric acid) (Van Meerbeek et al., 1992). According to Van Meerbeek et al. (1992) and Nakabayashi et al. (1992a), the depth of the hybrid layer for 4-META systems was about 1.5 to 2 μm, whereas for All-Bond 2 was 2.5 to 5 μm (Van Meerbeek et al., 1992; Jacobsen and Finger, 1993). However, a more recent in-vivo study by Walshaw and McComb (1994), revealed a 5 to 8 μm hybrid layer with All-Bond 2 system. This indicates that when used correctly both adhesive systems are able to penetrate and encapsulate the entire depth of the decalcified dentin, resulting in a maximum adaptation with a gap-free attachment at resin-dentin interface.

The fourth requirement is that once the resin monomer has penetrated into the demineralized substrate to its fullest extent, it must be able to polymerize to its full depth in a wet environment. It must polymerize in such a way that it seals the dentinal tubules and prevents microleakage. Amalgambond system has a unique tri-N-butyl borane (TBB) catalyst. Once contact is made with O_2 and H_2O the polymerization of 4-META/MMA resin monomer is initiated. The polymerization shrinkage is directed towards the initiation point of the reaction which in this case is towards the moist dentinal substrate (Nakabayashi et al., 1992b). In case of All-Bond 2, the polymerization of the BP-DM primer molecule is initiated by the NPG-GMA molecule, and because the layer is very thin the direction of the polymerization is towards the dentin surface.

If these four conditions can be achieved, an effective hybrid layer is formed, and both a good bond and seal are formed.

The following section discusses the characteristics of the dentin-resin interface according to recent findings.

Characteristics of dentin-resin interface

The micromorphology of the resin-tissue interface is now being thoroughly investigated using scanning and transmission electron microscopy (SEM and TEM) (Van Meerbeek et al., 1992; 1993; Inokoshi et al., 1993; Tay et al., 1994; Kanca and Gwinnett, 1994). The relationship between hybridization and bond strength has been documented (Gwinnett and Kanca, 1992). Gwinnett (1993), indicated that resin infiltration into tubular and intertubular dentin can contribute approximately one third of the shear bond strength of the All-Bond 2 system. Of utmost importance is the diffusion of the resin through the entire thickness of the demineralized dentin. SEM and TEM examinations of an in-vivo study (Tay et al., 1994), investigated the interaction between All-Bond 2 system and the dentin interface, and indicated the presence of two morphologically discrete zones that have the potential for sealing and preserving the biological and morphological integrity of the dento-pulpal complex. As shown in figure **10**, the outer zone consisted of a solid resin plug surrounded by a circumferential cuff of resin-impregnated dentin. The inner zone consisted of a hollow resin sheath with resin globules lying along the internal surfaces of the hollow resin string, closely adapting to the odontoblast process. An in-vivo and in-vitro SEM examination by Kanca and Gwinnett (1994), showed string-like resinous structures of varying lengths, densities, and distributions originated from the filling surface of the restorations into the dentin substrate. They indicated that the All-Bond system has the potential to produce gap-free restorations both in-vivo and in-vitro. Results of a more recent in-vitro study using the laser scanning confocal microscope, revealed that the Amalgambond adhesive system is capable of both effectively sealing the interface between the amalgam restoration and tooth, and penetrating into dentinal tubules (Li et al., 1995). However, Sano et al. (1995), evaluated the leakage pathway within the hybrid layer of four dentin adhesive systems using SEM and found that even when a gap was not evident, all systems including All-Bond 2 and Superbond C&B leaked within this layer.

Generally shear bond strengths to dentin for some of the fourth generation dentin adhesives such as All-Bond 2 and Amalgambond adhesives, with composite restorations, are often close to or greater than 20 MPa (Hasegawa et al., 1992; Triolo and Swift, 1992; Kanca and Gwinnett, 1994). These adhesive systems, however, in in-vitro studies show that they still do not completely eliminate microleakage at the dentin margins (Sano et al., 1995; Inai et al., 1995). Holtan et al. (1993), found no significant correlation between the mean bond strengths (Holtan and others, 1992), and the mean microleakage values obtained from four dentin bonding systems. Although some of these materials found to approximate the 20 MPa bond strength, all of them demonstrated varying degrees of microleakage. This suggests that having a dentinal bond strengths of 20 MPa or more may not result in gap free resin restorations (Holtan et al., 1993; Kanca, 1994).

Although the hybridization process was originally designed for use in conjunction with composite resin restorations, there is a growing trend toward using this process for amalgam restorations as well. Many clinicians no longer use cavity bases or liners on a routine basis unless the preparation is quite deep. Furthermore, because the hybridization process completely seals all of the dentinal tubules, it also eliminates the need for cavity varnishes (Leinfelder, 1994).

The previous sections discussed the mechanism of bonding between adhesive resins and tooth structure. The following is a discussion of the mechanism of bonding between resins and metals including bonding to amalgam alloys.

(B) METAL-RESIN BONDING

The use of adhesion is expanding to virtually all aspects of restorative dentistry, including the use of resin bonding to affix dental materials to metallic surfaces. Metal-resin bonding can be classified as either mechanical or chemical or a combination of both.

Mechanical Bonding

Mechanical bonding is subdivided into the following categories (Albers, 1991): a) macromechanical bonding created on the cast metal surface, which relies on visible undercuts, usually with perforated metals, mesh patterns, or pitted metal (using the lost salt or lost porcelain technique) (McConnell, 1994), and b) micromechanical bonding, which employs microscopic porosities on the substrate, like sandblasting or chemical and electrolytic etching (McConnell, 1994). Bond strengths between 15 MPa and 20 MPa have been reported using these techniques (Albers, 1991).

Chemical Bonding

There are two major types of chemical bonding, **(1) interfacial bonding** which uses an intermediate interface like pyrogenic silanization, tribiochemical coating and electroplating (e.g. Silicoating and Rocatec) fused to the metal surface and, **(2) adhesive** which employs chemically attached adhesives such as Amalgambond, All-Bond 2, or Panavia, containing coupling agents to attach resin to metal (Albers, 1991).

(1) Interfacial bonding

a) Pyrogenic silanization

Pyrogenic silanization is a process which heat fuses a thin intermediate bondable glass interface (silicate layer) to sandblasted metal. A silane primer is applied followed by a resin coating. The silica particles are mechanically and possibly chemically bonded to the sandblasted surface. Exposure to water results in loss of some of the bond strengths presumably due to the loss of the initial chemical bonds. An example of this is Silicoating system by Kulzer (Kulzer Slicoater, Irvine CA).

b) Tribiochemical coating

Tribiochemical coating attaches silica glass particles mechanically to the metal by sandblasting. The force of impact during sandblasting imbeds the particles into the metal surface to create a mechanical bond. Thereafter a silane is added to form a copolymerized chemical bond between the ceramic silicate layer and the resin. This systems were developed primarily to bond resin veneering materials to metal frameworks and they have also been used to bond metal restorations to etched enamel preparations. An example of this is Rocatec system (ESPE/Primer)

Bonding resins to ceramic

The surface of glass, being ionic in nature, readily adsorbs water, forming a well-bonded surface layer. As a consequence of this, when glass is mixed with resin to produce a composite the resin will not wet the surface of the glass and the tow are poorly bonded. This has the effect of producing a very weak composite because the glass is not able to take on a load-bearing role and acts merely as a space filler. Therefore, silane coupling agents are used to displace the adsorbed water and provide a strong chemical link between the oxide groups {ceramic contains silica (SiO_2) as a major ingredient in addition to other oxides} on the glass surface and the polymer molecules of the resin (Van Noort, 1994). Acid etching with hydrofluoric acid before silanization process, etches the glass fillers to create a micromechanically retentive surface and enhances the density of --OH groups that are available on the surface. It also removes any absorbed species other than water. Silane coupling agents have the general formula [$R - Si- X_3$] where R represents an organo-functional group and the X units are hydrolysable groups bonded to the silane.

$$R - Si- X_3 + 3\,H_2O \longrightarrow R -Si(OH)_3 + 3\,HX$$

These trihydroxy-silanols are able to compete with water on the surface of the glass by forming hydrogen bonds with the hydroxyl groups on the glass surface. When the silane coated glass is dried, the water is removed and a condensation reaction occurs between the silanol and the surface. Once this bond is formed, it is no longer susceptible to hydrolysis. The organo-functional group, R, reacts with the resin, and forms a strong bond to it. Using coupling agents therefore, produces a strong, water resistant bond.

c) Electroplating

Electroplating is mostly a conditioning procedure before using adhesive cements. The metal is electroplated with tin and oxidized to form a tightly adherent layer of tin oxide crystals. The resin then mechanically bonds to the rough oxide surface. Some chemical bonding may also occur between the tin oxide and the resin. Some adhesive resins like All-Bond do not require tin-plating of nobel alloys to obtain high laboratory bond strengths (Albers, 1991). Tin can be added through melting a tin solder placed onto the metal or more commonly by electrolytic deposition.

(2) Adhesive Bonding

These materials are either based on 4-META (Superbond, Amalgambond), 4-META in addition to silane (Fusion), or NTG-GMA/BP-DM (All-Bond 2), or on modified phosphate containing resins (Panavia Ex) systems. The bonding mechanism is not known but it may be as a result of both chemical (ionic and hydrogen bonds), and micromechanical bonding. Chemical bonding occurs between the metallic ions present on the metal surface as oxides and the carboxyl groups in the resin (Tanaka et al, 1988). Figure **11** shows the postulated reaction mechanism for 4-META and All-Bond 2 bonding to the metal oxides on the metal surfaces. Micromechanical bonding occurs to an air-blasted, tin-plated, or etched cast metal surfaces, resulting in a higher bond strength. Bond strengths in excess of 20 MPa are claimed for bonding to base and nobel metal alloys.

However, there is a controversy regarding the stability of the 4-META system in water over time. Kiyomura (1987), Takarada et al. (1990), and Albers (1991), reported that the initial high bond strength sformed with 4-META systems decreased with time, which may be due to hydrolysis of the initial chemical bonds or hydrolysis of HEMA as a result of water absorption. Gendusa (1992), on the other hand, claimed that this was a myth. Even though there is slight reduction in bond strength it does not indicate degradation due to hydrolysis but it may be due to a breakdown at the metal-resin interface as a result of the differences in coefficient of thermal expansion and contraction of the metal alloy and the resins as discussed earlier in the paper. Although successful bonding occurred between resins and cast metal alloys, bonding to amalgam is still a new treatment modality.

Amalgam Bonding to Tooth Structure

Several laboratory studies have shown that Amalgambond and All-Bond 2 dentin adhesive systems can be also used to bond amalgam to dentin. Although the exact bonding mechanism at the amalgam-resin interface is not known, it may consist of both micromechanical and chemical bonding. Micromechanical bonding occurs when amalgam is condensed into a cavity preparation before polymerization of the adhesive resin occurs. The unset amalgam invaginates and surrounds the resin, and amalgam crystals lock onto it as the alloy sets (Covey and Moon, 1991; Gendusa, 1992). Chemical bonding (ionic and hydrogen bonds) may also be present between the resin and the ion oxides on the amalgam alloy surfaces (Zardiackas and Stoner, 1983; Eakle et al., 1992) similar to that with cast metals. However, it is believed that much of the bond strength of the resin to amalgam is micromechanical (Gendusa, 1992; McConnell, 1994) and any benefits of chemical bonding appear secondary both to good wetting and to intimate micromechanical bonding. Although strong ionic bonds (more than 20 MPa) have been reported to form between resin cements and the oxide layer present on cast metal surfaces that are sandblasted, electrolytically etched, or tin plated (Tanaka et al., 1988; McConnell, 1994), this does not seem to be the case with amalgam alloy, despite the fact that amalgam alloy contains approximately 27% tin which is readily oxidizable. The reported weak bond, ranging from near zero to 6 MPa or more (Ratananakin and Denehy, 1995;

Hadavi et al., 1994; Vargas et el., 1994) with amalgam alloy, may be due to either an insufficient or an unsuitable metal oxide layer forming between the resin and amalgam alloy. In order for good chemical bonding to occur, the metal substrate must develop a thin oxide layer which will bond strongly to the metal, be cohesively strong. Other conditions for good wetting of metal by the resin must be met (see earlier section on wetting, page:16). In any case where any of these requirements is compromised, a weak bond or no bond will occur between resins and amalgam alloy.

Benefits of Resin Bonding Amalgam

The use of adhesive liners to bond amalgam to tooth structure may provide several benefits. **First**, adhesive resin liners reduce leakage around amalgam restorations more effectively than do traditional cavity varnishes (Varga et al., 1986; Shimizu et al., 1987; Pashley et al., 1991; Cooley et al., 1991; Edgren and Denehy, 1992; Charlton et al., 1992; Souza et al., 1993; Saiku et al., 1993; Moore et al., 1995; Fundingsland and Ario, 1995; Cao et al., 1995). The reduced leakage could result in less recurrent caries at restoration margins (Torii et al., 1989; Chan et al., 1995). The reduced leakage is probably the result of marginal sealing and reduced permeability produced by the adhesive resin, in addition to the seal provided by amalgam itself (Pashley et al., 1988; Hansen et al., 1992). **Second**, the amount of force required to dislodge an amalgam restoration is increased by bonding, thus reducing the need for retentive devices such as dovetails, grooves, and pins (Staninec, 1989; Ianzano et al., 1993; Eakle et al., 1994). **Third**, teeth restored with resin bonded amalgam restorations may be more resistant to fracture than are teeth restored in the conventional way (Eakle et al., 1992; Ianzano et al., 1993). **Fourth** and finally, clinical experience indicates that the use of dentinal adhesives as amalgam liners reduces the incidence of postoperative sensitivity (Masaka, 1991). Although previous studies seem promising with bonded amalgam restorations, no long-term in-vivo studies are available on any of the currently used resin adhesives.

Because bonded amalgam restorations are a new treatment modality, there is no documented clinical evidence of their durability or superiority over conventional amalgam restorations. One potential problem, according to Charlton et al. (1991), is the weakening of the restoration caused by the incorporation of resin into the amalgam. They indicated that Panavia EX adhesive resin reduced the compressive strength of Tytin amalgam, and they recommended that it to be applied only in a thin layer. However, the Amalgambond system did not have an adverse affect on the compressive strength of restorations (Charlton et al., 1991).

Bond Strength Measurements

Laboratory studies are often used to evaluate dental bonding resin systems and are used as screening tests before proceeding with long-term and expensive clinical studies that will determine the clinical efficacy of the bonding system. Laboratory tests, such as measurement of bond strength and microleakage studies, will continue to be necessary for the development and initial evaluation of adhesive restorative materials. Regarding bond strength measurements, three research studies have reported a high shear bond strength of 13.0 MPa for amalgam bonded to dentin with All-Bond 2 (Souza et al., 1993) and 12 MPa with Amalgambond Plus (HPA powder) (Vargas et al., 1994; Ratananakin and Denehy 1995). The bond strengths of amalgam are generally lower (less than 10 MPa) than those of composite resin (15-20 MPa) (Cooley et al., 1991; Hasegawa et al., 1992; Pashley et al., 1991; Covey and Moon, 1991; Miller et al., 1992; Chang et al., 1992; Bagley et al., 1994; Eakle et al., 1994; Diefenderfer and Reinhardt, 1995; Larson et al., 1995). These results suggest that dentin bonding adhesives are not sufficient to serve as the major retention for amalgam restorations. According to Diefenderfer and Reinhardt (1995), shear bond strengths of amalgam to dentin appear to be affected by both amalgam particle shape and adhesive resin. Tytin (spherical) amalgam and filled resins produced higher shear bond strengths than Dispersalloy (admixed) and unfilled resins. The mode of failure is predominantly adhesive, between resin and amalgam or resin mixed with remnants of amalgam left on dentin surface (Miller et al., 1992; Nakabayashi et al., 1992b; Diefenderfer and Reinhardt, 1995). Although fracture occurs between the amalgam and resin interface, the dentin surface in almost all the cases remains sealed with resins (Saiku et al., 1993; Jacobsen and Finger, 1993), indicating a potential advantage for the use of these adhesives as liners under amalgam restorations protecting pulp tissues even if the amalgam/tooth bond should fail.

Microleakage Evaluation

Many studies have been carried out to demonstrate the microleakage associated with materials and, to improve the marginal seal. These laboratory tests include the use of dye penetration, chemical and radioactive tracers, bacterial penetration and artificial caries, neutron activation analysis, and electrical current investigations (Trowbridge, 1987; Taylor and Lynch, 1992). Direct visualization of marginal gap, using light microscopy or scanning electron microscopy (SEM), and clinical assessment of biological effects of recurrent caries, pulp pathology, post operative sensitivity are other methods used to demonstrate microleakage (Trowbridge, 1987; Taylor and Lynch, 1992). Unfortunately, most of these methods are either qualitative or semiquantitative, and do not simulate the oral environment (i.e. hydration state of dentin, physiological pulpal pressure, and body temperature). Some of these methods require destruction of the sample during processing while others do not permit observation of the specific location of the leakage. Several reports however, described quantitative techniques that can determine the efficacy of lining materials to reduce marginal leakage and/or dentin permeability (Trowbridge, 1987; Taylor and Lynch, 1992). Although the air-pressure method permits quantitation of microleakage in-vitro, nondestructively, and in a longitudinal manner (Granath and Svensson, 1970), this technique does not qualify the information obtained other than to define the smallest path of leakage which gives no detail of the true leakage pattern. Further, it is difficult to photograph the location of microleakage because specimens are studied under a fluid. Terkla et al. (1987), developed a laboratory system that simulates the physiological and clinical conditions of a prepared human tooth in order to determine the sealing efficacies of restorative dental materials. Although this system is a very sensitive method for measuring fluid movement, it is also sensitive to any changes in temperature or pressure and the time required to make triplicate measurements is extensive (Pashley 1989b). Fortunately, Derkson et al. (1986), developed a technique that permits the sequential, quantitative-volumetric measurements of microleakage and dentin permeability under pressure. Additional advantages of this later technique are: **1)** it is a simple method, **2)** measurements are reproducible with a given specimen which reduces inter-tooth variations, **3)** quadruplicate measurements can be done in a short time (i.e. within 30 minutes) on the same specimen, **4)** it allows for photographic records of microleakage areas by incorporating dyes in the fluid used, **5)** measurements under physiological pressure can be done if required, and finally, **6)** the fluid flow measurements can be made repeatedly on the same samples in longitudinal studies. This is impossible to do in studies using the dye diffusion method. For these reasons we decided to utilize the fluid pressure technique to measure microleakage of amalgam bonded restorations in this study.

Study Objectives

The purpose of this study was to evaluate longitudinally: **(1)** the effectiveness of two adhesive dentin bonding systems on reducing microleakage at the tooth-amalgam restoration interface quantitatively; and **(2)** to compare these adhesive systems to the use of Copalite varnish. The dentin bonding systems Amalgambond Plus and All-Bond 2, were chosen for this study because of their increasing clinical use at this time.

MATERIALS AND METHODS

Tooth Selection and Preparation

Twenty four extracted, unerupted, non-carious human third molars were used within one month after extraction.. Soderholm, (1991), reported that the maximum time that an extracted tooth can be stored is up to six months, since no major changes occurs to the tooth structure within this period of time. Teeth were cleaned of hard and soft tissue accretions and then stored at room temperature in a glass container in isotonic saline solution (pH 7.4) containing 0.2% sodium azide (Mallinkrodt Organic Reagent Inc., Paris, KY.) as an antimicrobial agent. All teeth were then sterilized in a steam autoclave (Amsco Eagle Services 200, American Sterilizer CO, Erie, PA, USA) at 122 ^{o}C at 15 Psi, for 30 minutes without dry cycle and then stored in the same sterile glass container at room temperature. Sterilization was performed to assure the protection of the investigator from potential pathogens in extracted teeth (Pashley et al., 1993b).

The specimens were prepared in the manner described by Pashley and Depew (1986). Crown segments were prepared by sectioning the roots from the crowns at a level 2 mm below the cemento-enamel junction (CEJ) using a high speed, diamond saw (Brownwill WVR Scientific St. Louis, Mo. USA) at

3450 RPM with water coolant. The coronal pulp tissues were removed from the pulp chamber with cotton pliers, being careful to avoid creating a smear layer on the predentin layer on the inner surface of the pulp chamber. The prepared crown segments were then cemented with cyanoacrylate cement, (Zapit, DVA dental Ventures of America, Inc, Anaheim Hills, CA. 92808) to 2 x 2 x 0.3 cm pieces of plexiglas pierced in the center by 18-gauge stainless steel tubes. The stainless steel tubes were also cemented in place with cyanoacrylate cement. Crown segments were placed on the pieces of plexiglas with the pulp chambers centered over the tubes. The pulp chambers and the tubes were both filled with isotonic saline solution at all times.

Experimental Groups

This study was designed with four experimental groups. Six samples were randomly assigned to each group.

Group **1** : Unlined Dispersalloy amalgam restorations (Caulk Dentsply, Canada Ltd., Ontario L4L 4A3 Lot No. 940923) served as the control group.

Group **2** : Copalite varnish liner (Cooley and Cooley, Ltd., Houston, Texas distributed by Denco Toronto, Canada, No. 749,471) plus Dispersalloy amalgam.

Group **3** : All-Bond 2 Adhesive Dentin Bonding System (Bisco Inc, Itasca, IL, USA, 60143, Lot No. 129033) plus Dispersalloy amalgam.

Group **4** : Amalgambond Plus Adhesive Dentin Bonding System (Parkell, Biomaterials, Farmingdale, NY, USA made in Japan Stock No. S370) plus Dispersalloy amalgam.

Materials used in group 2, 3, and 4 are intended to be cavity liners to control microleakage around amalgam restorations.

Experimental Method

The crown segments were connected to a fluid under pressure apparatus described by Pashley and Depew (1986) and Derkson et al. (1986), and illustrated in (Fig. **12-A**). The apparatus consists of a nitrogen pressure tank (**A**) which supplied the hydrostatic pressure gradient to move the isotonic saline solution containing 0.2% sodium azide and 0.2% methylene blue dye (SIGMA chemical Co, St. Louis, MO, USA (63178) from the pressure reservoir (**B**) (Presto, National Presto Industries, Inc, Eau Claire, WI, 54701, working at 16 Psi), through a micropipette (**C**) (Microcaps, Drummond Scientific Co., Broomall, PA) (32 mm in length and 5 µl in volume), into the pulp chamber of the crown segment (**D**), and across the dentin up to the cavo-surface margins of the restoration. Methylene blue dye was added to the fluid to assist in the visualization and photographic recording of the microleakage pattern. Figure **12-B**, is a photograph of the apparatus. The micropipette, containing a tiny air bubble, was positioned over a millimeter scale (Fig. **12-B**, bottom). The position of the air bubble in the micropipette was adjusted using a microsyringe (**E**) (Gilmont Instruments Inc, Great Neck, NY. 2 ml) and the movement of the air bubble was measured in mm/min. Hydrostatic pressure was controlled and kept constant at 16 Psi throughout the experiment by the regulator (**F**) on the nitrogen tank.

Prior to making cavity preparations, the system was pressurized to determine if there was any movement of the air bubble inside the micropipette indicating fluid movement that may be caused by a leak. If any fluid movement occurred, all connections were remade until a zero fluid flow was reached and hence zero permeability.

Cylindrical occlusal cavities were prepared well into dentin using a carbide tapered fissure bur No. 169 followed by a plain fissure bur No. 57 in a high speed water cooled handpiece. A new bur was used for every six samples. The cavity dimensions were 3 mm in diameter and approximately 3.5 to 4 mm deep and were measured by a periodontal probe (Immunity stainless steal probe Michigan "0" Williams Marking). Permeability of the dentin of each specimen was measured following the cavity preparation.

Copalite Treatment

Copalite varnish is composed of a natural gum, such as copal (fossil resin), rosin, rosin esters, or synthetic phenolic resin with their individual solvents such as chloroform, acetone, or ether. The solvent evaporates, leaving a film that serves as a barrier against microleakage and protects the underlying tooth structure. The Copalite treatment involved two applications of the varnish to the cavity walls using a fine brush in a manner described by Ben-Amar et al. (1993). The first layer was applied and immediately dried for 3 seconds with a stream of oil-free compressed air. After 30 seconds, a second layer was applied and dried for 3 seconds. After another 30 seconds, the amalgam was condensed into the cavities. The application of two thin layers (i.e., less than 30 μm) of Copalite varnish has been found to be more effective than one heavy layer (Craig et al., 1992). This is because when the first film dries, small pinholes usually develop and a second layer will fill most of these pores and thereby produces a more continuous coating. Thicker films on the other hand, will not be able to dry completely and with time oral fluids will dissolve out moist resin and result in a gap between the restoration and the tooth surface.

All-Bond 2 Bonding Agent Treatment

Treatment with All-Bond 2 involved three applications as described by the manufacturer. First, All-Etch (10% phosphoric acid) was applied to enamel and dentin cavity walls for 15 seconds with agitating the acid on the enamel using a fine brush. The cavity walls were then rinsed with distilled water for 30 seconds and kept wet. The specimen was air dried for one second just to remove the excess water and leave the dentin surface moist. Then, equal amounts of Primer (A) and Primer (B) were mixed and applied in five consecutive coats onto the specimen or until a glossy surface was obtained. Primer (A) is N-toludene glycine-glycidyl methacrylate (2% NTG-GMA) in an acetone solvent, and Primer (B) is biphenyl dimethacrylate (16% BP-DM) in an acetone solvent. The entire dentin surface was air dried for 5 to 6 seconds to complete the evaporation of solvent and then light cured for 20 seconds. Third, an equal amount of Dentin-Enamel Bonding resin and Pre-Bond resin were mixed and applied in a thin layer onto the entire cavity surface and lightly air-thinned to avoid pooling. The Dentin-Enamel Bonding resin is bisphenol A diglycidyl methacrylate (Bis-GMA), hydroxyethyl methacrylate (HEMA), while the Pre-Bond resin is benzoyle peroxide (BPO) and BIS-GMA. Amalgam was then condensed immediately into the cavities.

Amalgambond Plus Bonding Agent Treatment

Amalgambond Plus also involved three applications. First, the activator, a solution of (10% citric acid and 3% ferric chloride) (10-3 solution), was applied to dentin cavity walls for 10 seconds with agitation by brush on the enamel surfaces. The dentin and enamel surfaces were washed thoroughly for 30 seconds with distilled water and air dried for 10 seconds. Second, the adhesive agent, (an aqueous solution of 35% HEMA in water), was brushed onto the dentin surfaces (it is not necessary to apply adhesive agent to etched enamel surfaces, but there is no harm if some overruns onto enamel), left undisturbed for 30 seconds, and blown gently to a thin layer. Third, the base (2 drops) and catalyst (1 drop) were mixed and brushed onto the entire cavity surface. The base solution is a mixture of HEMA, 4-methacryloxyethyl trimellitate anhydride (5% 4-META) and methyl methacrylate (MMA), while the catalyst is tri-n-butyl borane (TBB). The amalgam was then condensed immediately before the adhesive resin dried.

Amalgam Restorations

The specimens were restored with precapsulated Dispersalloy (Caulk Dentsply). Dispersalloy is an admixed non gamma 2 amalgam which was triturated in an amalgamator (Caulk Dentsply) for 12 seconds at medium speed. Amalgam was manually condensed in small increments until the cavity preparation was slightly overfilled. Lateral and vertical condensation with overlapping thrusts was employed, to ensure optimum adaptation of amalgam to the cavity walls, using an amalgam plugger (1/2 15/25 HuFriedy). The amalgam restorations were surfaced with a ball burnisher (BB D2 18 HuFriedy) prior to and after carving. The surface was carved with a discoid-cleoid carver (4/5 HuFriedy). Final surfacing was completed within 5 to 7 minutes from the end of trituration. A fresh mix of amalgam was used for each restoration. All the teeth were kept for 30 minutes at room temperature after amalgam condensation, then returned to the saline solution storage medium at 37 °C. All restorative procedures and specimen storage were done at a zero pulpal pressure, while connected to the apparatus to ensure dentin hydration. According to Kim et al. (1984), local anesthetics that contain vasoconstrictors would reduce the pulpal tissue pressure to values near zero, therefore, bonding procedure during zero pulpal pressure would be relevant in this study. The adhesive systems used in this study were applied according to the manufacturer's instructions.

MEASUREMENTS

Cavity Permeability Measurements

Dentin permeability is directly related to the fluid flow rates across the dentin via the dentinal tubules. It was measured and quantified from the movement of the air bubble (mm/min) in the micropipette and was expressed in (μl/min). Dentin permeability was quantified using the following formula (adapted from Pashley and Depew, 1986):

$$\text{Dentin permeability } (\mu l/min) = \frac{\text{volume of micropipette } (\mu l) \times \text{movement of air bubble } (mm/min)}{\text{Length of micropipette } (mm)}$$

The first permeability measurement was made with enamel intact prior to tooth preparation. This was done to check for abnormally high permeability in the tooth due to cracks or other defects, and also for leaks in the system. Defective teeth were discarded.

The second permeability measurement was made after the cavity was prepared to check for any pulp exposure. Teeth that showed any pulp exposure or high dentin permeability were discarded from the experiment.

The third measurement was made after acid etching the entire cavity with 37% phosphoric acid for 30 seconds in order to remove the smear layer created by cavity preparation and to maximize dentin permeability (Pashley et al., 1993b). This value was then assigned as the maximum dentin permeability (i.e. 100%). Due to variations in dentin permeability between different teeth, any subsequent change in dentin permeability was then expressed as a percentage change from the maximum permeability of the etched specimen. Therefore, each tooth served as its own control.

The fourth measurement was made after the creation of a new standardized smear layer on the dentin surface using No. 57 plain fissure bur in a low speed handpiece for one minute but only removing minimal dentin. This measurement was then assigned as the minimum dentin permeability for each sample.

Quadruplicate measurements of dentin permeability were taken for each treatment stage to assure the replicability and the accuracy of the readings. For each test condition the tooth was connected to the apparatus for at least 15 minutes, measurements were taken every two minutes and only the last four consistent readings were used.

For both Amalgambond Plus and All-Bond 2 treatment groups, an additional measurement was taken after treating the dentin surface with an etchant supplied by the manufacture. If this value was higher than the maximum value (i.e. after acid etching with 37% phosphoric acid), it was then considered as the new maximum value of that specimen.

Microleakage Measurements

Microleakage is directly related to the fluid flow rate between the cavity walls and restorative materials. It was measured in ($\mu l/min/cm\ H_20$) and expressed as an apparent reduction in the maximum dentin permeability of each specimen as a result of the treatment provided. Microleakage was measured after amalgam insertion at the following time intervals; at 24 hours, one week, and at one, three and six months. Microleakage was measured using this formula (adapted from Pashley and Depew, 1986):

$$\text{Microleakage } (\mu l/min/cm\ H_20) = \frac{\text{dentin permeability } (\mu l)}{\text{applied pressure (cm of } H_20)}$$

The applied pressure was 16 Psi. To convert from Psi to cm of water the following equation was used (CRC Handbook, 1988),

1 Psi = 70.3070 cm of H_20.

16 Psi = 1124.912 cm of H_20.

Statistical Analysis

The microleakage values for each group of materials were compared longitudinally using a repeated measures ANOVA. Least square means contrasts were used for pair wise comparisons with Bonferroui adjustment p-values ($p < 0.05$). A statistical package, SAS was employed for this purpose (SAS institute Inc. Cary, North Carolina). The statistical analysis was performed by the Biostatistical Consultant Unit, Faculty of Medicine, University of Manitoba.

RESULTS

The maximum dentin permeability of the empty cavities was determined after removal of the smear layer with 37% phosphoric acid for both unlined and copalite restorations, with 10-3% citric acid-ferric chloride solution for Amalgambond Plus and 10% phosphoric acid for All-Bond 2 restorations. This value was assigned as 100%. Table **I** represents the mean values of dentin permeability and microleakage ($\times 10^{-4}$) and the standard deviation (SD) for the four tested groups. The Amalgambond Plus group showed the highest permeability (25.41×10^{-4} µl/min/cm H_2O) which was not significantly different compared to the other groups (table **I**).

Table **II** represents the dentin permeability and microleakage as a percentage of maximum and the standard error of the mean (SEM). Creation of new smear layers reduced the permeability by 75.2% to 84.9% to values ranging from 15.1% to 24.8% of the maximum permeability of those samples (Table **II**). This value was called "minimum permeability". All materials showed similar permeability reduction except Amalgambond Plus group which had lower permeability than the other tested groups but was not significantly different (Fig. **13**).

Twenty-four hours after insertion of the amalgam restorations the apparent permeability of the restored cavities was greatly reduced compared with the unrestored cavities (minimum) the unlined teeth ($p < 0.001$), Copalite ($p < 0.001$), Amalgambond Plus ($p < 0.05$), and All-Bond 2 ($p < 0.0001$). Microleakage was reduced to values between 8.1% to 2.5% for the unlined and the All-Bond groups, respectively (table **II**). The unlined amalgam group showed statistically higher microleakage than the other groups ($p < 0.01$) (Fig. **14**).

At one week Amalgambond Plus and All-Bond 2 restorations had statistically lower microleakage than those obtained from unlined amalgam ($p < 0.01$) and Copalite varnish restorations ($p < 0.05$) (Fig. **15**).

At one month most groups showed an increase in microleakage. Amalgambond Plus and All-Bond 2 had statistically significantly lower microleakage than the unlined group ($p < 0.01$) (Fig. **16**). Although the values for Copalite was lower than the others, this difference was not statistically significant from All-Bond 2 and Amalgambond Plus. It appears that Copalite permeability is less than that of Amalgambond Plus and similar to All-Bond 2. At one month the unlined group was significantly higher in microleakage than that at one week ($P < 0.01$) (Table. **II**).

At three months Amalgambond Plus and All-Bond 2 groups exhibited a significant reduction in microleakage compared to both unlined ($p < 0.01$)

and Copalite groups ($p < 0.05$) (Fig. **17**). The unlined amalgam group was statistically higher in microleakage over that measured at twenty-four hours ($p < 0.01$).

At six months Amalgambond Plus and All-Bond 2 were significantly lower in microleakage than both the unlined ($p < 0.01$) and the Copalite groups ($p < 0.05$) (Fig. **18**). The unlined group had a significant increase in microleakage over that of Copalite group, and was also significantly higher than those obtained at twenty-four hours, one week, one month and is higher than that of the freshly created smear layer ($P < 0.01$).

Figure **19** and figure **20**, represent changes in microleakage and dentin permeability for the seven tested conditions. The unlined group shows an increase trend in microleakage over time periods. Whereas all the other groups remain steady around 3.5 % of maximum.

Although fluid seepage was seen around the cavosurface margins of unlined amalgam restorations at three and six months time intervals, no apparent fluid could be seen with the other specimens in the remaining groups (Fig. **2**).

Discussion

A primary objective in the placement of restorations is the development and maintenance of an intimate adaptation between the restorative material and tooth structure that will retain its integrity over time. Improper adaptation of the restoration to the tooth structure results in the presence of a fluid-filled space at the interface of the tooth and the restoration. This space allows the passage of fluids, bacteria, and chemical substances to the pulp via dentinal tubules that could result in marginal staining, secondary caries formation, pulp irritation and necrosis. In the case of dental amalgam, leakage can occur at the interface between the cavity walls and the restoration because amalgam does not adhere to tooth structure. In addition, leakage can occur within or through the smear layer covering dentin surfaces, or between the smear layer and cavity liner, or between cavity liner and amalgam restorative material. More recently the use of resin liners under amalgam restorations has shown promise in sealing and reducing microleakage compared to the traditional Copalite varnish. However, a new leakage pathway that progresses through the porous, unprotected, demineralized collagen layer created in conjunction with utilization of dentin bonding materials has been described (Sano et al., 1995).

The method used in this study, fluid under pressure, demonstrated that the magnitude of dentin permeability reduction provided a quantitative, longitudinal measure of the amount of microleakage between dentin cavity walls and the restorative materials. A major advantage is that the method does not require the destruction of the samples and this allows a longitudinal study to be done. The method involves measurement of fluid flow across dentin before cavity preparation (fluid flow is zero), after cavity preparation and acid etching with 37% phosphoric acid (this value is assigned a value of 100% permeability or 'microleakage'), and after recreation of standardized smear layers. Finally, after insertion of restoration into the cavity, the fluid flow was remeasured at various timed intervals for the duration of the study. An ideal restorative material would reduce the apparent permeability and microleakage of the cavity to zero. This study demonstrated that removing the smear layers with a combination of citric acid and ferric chloride (10-3% solution) supplied with Amalgambond Plus or 10% phosphoric acid supplied with All-Bond 2 produced the highest dentin permeability (Table **I**, Fig. **20**). However, this additional treatment increased the permeability above the 100% (37% phosphoric acid) value and was due, in part, to the possibility that the dentin was made thinner when further cavity preparation recreated the standard smear layer. According to Fogel et al. (1988), as dentin is made thinner (less remaining dentin thickness) (RDT), the dentin permeability increases. Ten seconds of the 10-3% solution and 15 seconds of the 10% phosphoric acid treatments increased dentin permeability to 173% and 89.5%

respectively of the 37% phosphoric acid treatment value. The use of such solutions and duration of application is recommended by the manufacturer.

The smear layers created in this study reduced the dentin permeability of all groups by 75.2% to 84.9% of the maximum (Fig. **13**), demonstrating how effective the smear layer is in reducing dentin permeability. Amalgambond Plus-treated samples, however, exhibited slightly lower permeability (15.1% which was not statistically significant) than the other samples. This was due to the fact that Amalgambond Plus conditioners produced the highest maximum permeability to begin with. Pashley and others (1991), reported a similar permeability which was 23% of maximum. In that study they used a different etchant (0.5 M EDTA, pH 7.4) and measured dentin permeability by hydraulic conductance i.e., using a known surface area. Although it has been found that the smear layer decreases dentin permeability by blocking the dentinal tubules (Pashley and Depew, 1986), it hinders the sealing of amalgams (Jodaikin and Austin, 1981) and increases the microleakage around these restorations (Pashley and Depew, 1986). Acid-etching dentin removes the smear layer and allows adhesive resins to bond to dentin structure. Thus, the amalgam in conjunction with the cavity liner can adapt better to cavity walls and reduce the interfacial space between the cavity walls and restorative materials through which fluid may flow (Pashley and Depew, 1986; Leelawat et al., 1992).

Twenty-four hours after insertion of amalgam restorations into the prepared cavities, the apparent permeability or microleakage of all groups was reduced greatly to values ranging between 8.1% to 2.4% of the maximum (Table **II**, Fig. **19**). This is an indication that the tested materials had sealed the dentin well and permitted little microleakage of fluid under pressure from the dentin around the material to the cavosurface margins. All of the three lined groups (Copalite, Amalgambond Plus and All-Bond 2) were significantly lower in microleakage than the control group (unlined amalgam), which was restored in the absence of any liners or varnishes. There was no significant difference between the three lined groups (Fig. **14**).

It was expected that unlined amalgam restorations would show a slow decrease in permeability with time, as a result of corrosion of the amalgam by the saline solution in which they were stored. Instead, there was a general increase in microleakage of unlined amalgams (41% of the maximum) (Fig. **18**), this is similar to the results reported by Derkson at al., (1986). This increase could have been caused by the slow dissolution of the smear layers, as a result of fluid leakage (Pashley, 1990). Another possibility is the presence of bacteria, whose acidogenicity and metabolic activity could create a gap (1 to 2 µm) between the varnish and the original location of the smear layer (Pashley, 1990; Prati et al., 1994). In this study the teeth were stored in an isotonic-saline solution containing sodium azide, therefore, it is unlikely that the result was due to any bacterial action. However, it is unknown whether the sodium azide solution has any effect on the solubility of corrosion products of amalgam, if any, which is mainly copper oxides in high-copper amalgams. Most likely, the high-copper, dispersed-phase amalgam used in this study did not corrode sufficiently in six months to occlude the spaces responsible for microleakage. Fayyad and Ball (1984), showed that in-vitro corrosive treatment improved the marginal seal of the traditional low-copper amalgam, but had no effect on the marginal seal of high-copper amalgam. Andrews and Hembree (1980), reported that leakage around low-copper amalgams subsided faster than amalgam with high-copper alloy. It is well known that deposition of corrosive products in the amalgam-tooth interface can reduce or block the leakage of low-copper amalgam alloys with time (Andrews and Hembree, 1975). Jodaikin and Grossman (1984), reported that amalgam restorations stored in 1% NaCl leaked at ten days but not at one year later. Gottlieb et al. (1985), indicated that all high-copper amalgams leak initially for up to one and a half years, but at two years, there is considerable reduction in leakage, comparable to low-copper amalgams. Therefore, although high-copper amalgams have better physical properties, corrosion characteristics, and better marginal integrity in clinical trials than low-copper amalgams, they are much less to seal the space between the amalgam and the tooth to minimize the initial microleakage and during the life of the restoration.

Generally, Copalite treatment in this study produced a highly significant reduction in microleakage around the amalgam compared to unlined amalgams (4.63% of the maximum) (fig. **18**). This is consistent with the findings of Pashley and Depew (1986) and Derkson et al. (1986). Cavity varnishes are known to seal the dentinal tubules, reduce the initial microleakage and its consequences under newly condensed amalgam restorations (Derkson et al., 1986; Pashley and Depew, 1986) and also at one year (Murray et al., 1983). Although our results showed a decrease in microleakage at one month, it was not statistically different from those at the other time intervals. Results at six months revealed a higher microleakage than that at twenty four hours which was not statistically significant (Fig. **19**). This suggests that two changes may be occurring simultaneously; first, dissolution of the underlying smear layer and second, dissolution of the Copalite varnish. Both changes may have increased the space between the cavity walls and amalgam restoration and hence increased microleakage. Powell and Daines (1987), indicated that Copalite varnish is subject to solubility in oral fluids. Pashley et al. (1985), reported that treatment of the smear layer with two layers (10 µm) of Copalite resulted in a significantly greater reduction in permeability (approximately 50%), which was statistically better than a single layer (5 µm) as measured by the radioactive isotope technique. In this study the application of two layers of Copalite varnish was utilized to assure the maximum benefit of this treatment.

There is a relation between the properties of Copalite varnish and the direction of fluid movement that can influence measurements of dentin permeability. Although Copalite varnish reduces dentin permeability and microleakage, it does not bond to tooth structure or to amalgam restorations (Ben-Amar et al., 1993), its residue is hydrophobic and tends to lie on top of the cavity surfaces much like a gasket (Pashley et al., 1985). In fact, these properties may be responsible for the influence of the direction of fluid movement on dentin permeability. Filtering fluid from the pulp towards the cavosurface margins of the restoration tends to lift the cavity varnish off the dentin, which increases dentin permeability. Such tendencies may also exist clinically if pulpal tissue pressures are normal or elevated by inflammation. Pulpal tissue pressure would tend to slowly raise the varnish from the dentin if there were microscopic spaces between the restoration and the cavity walls. Fluid may also be filtered from the enamel-restoration gap to the pulp. This may occur in response to masticatory forces, where the fluid forces the varnish against the dentin, which improves the sealing properties during the time the force is applied (Pashley et al., 1985; Pashley and Depew, 1986; Derkson et al., 1986). Copalite varnish is usually applied on a dentin surface that is covered with a smear layer, therefore, an interfacial space will be always present through which fluid may flow, thus, increasing the space and microleakage between the cavity walls and restorative materials. In this study the force was only in one direction from the pulp towards the cavosurface margins of the restoration, and this may explain the increase in leakage with Copalite-treatment group.

The use of dentin-bonding resins should resist microleakage more than Copalite varnish and remain insoluble. In fact, bonding resin not only seals the space between amalgam-tooth interface but also increases the adherence of amalgam to tooth surfaces. The results of this study showed that when cavities were treated with Amalgambond Plus and All-Bond 2 resin liners, microleakage around amalgams was significantly reduced compared to both unlined and Copalite-lined treatment groups at almost all test intervals after the insertion of restorations. However, this was not the case at twenty-four hours and one month when compared to Copalite (Fig. **14**, Fig. **16**). There was no significant difference between both resin groups at all time periods studied. In this in-vitro study bonded amalgam restorations sealed dentin effectively (microleakage values near zero) and controlled microleakage better than Copalite restorations for up to six months. Several microleakage studies reported the same findings when comparing Copalite to resin-lined amalgam restorations (Charlton and Moore, 1992; Charlton et al., 1992). Saiku et al. (1993), in an in-vitro study showed that Amalgambond resin-liner reduced microleakage significantly more than Copalite varnish or unlined amalgam restorations. Their SEM evaluation found internal cavity surfaces to be sealed by the resin liner with separations and apparent microleakage occurring at the Amalgambond-amalgam interface (Saiku et al., 1993). Cao et al. (1995) and Fundingsland et al. (1995), reported similar microleakage reduction with both All-Bond 2 and Amalgambond Plus for amalgam restorations. Moore et al. (1995), found that 4-META was able to seal dentin and reduce microleakage effectively compared to Copalite at one week and six months. Therefore the results of this study are consistent with similar studies utilizing different methodologies.

There are some important concerns that emerge regarding these resin liners. **First**, how long could these resin liners control microleakage under amalgam restorations? **Second**, will they be still more effective in reducing microleakage than Copalite varnish after a longer period when they are used under clinical conditions? Moore et al. (1995) found that there was no significant difference between 4-META system and Copalite at one year. This was caused by an increase in average leakage of 4-META and decrease of average leakage in the Copalite group. However, it is not clear if this is the same with Amalgambond Plus system although they share the same adhesive monomer, i.e., 4-META. **Third**, although bonded amalgam restorations in this study reduced effectively microleakage and showed a potential advantage over Copalite varnish, they did not completely eliminate microleakage. This small amount of microleakage might permit oral fluids, bacteria or their products to irritate the pulp; causing postoperative pain or hypersensitivity, and allowing secondary caries to develop. Previous studies had identified this problem (Derkson et al., 1986; Saiku et al., 1993; Moore et al., 1995), and this is confirmed in the present study. However, the extent of microleakage that a human-vital tooth can tolerate before jeopardizing pulpal health is unknown. In other words, the clinical significance of a 97% seal versus a 100% seal is not known. Since this study used higher pressure compared to the normal physiologic pulpal pressure, and since less leakage occurs in-vivo than in-vitro (Barnes et al., 1993), the minute amount of microleakage detected in our study may actually not exist clinically. Pashley and Pashley (1991), described how the pulpal circulation contributes to the health of the pulp in-vivo, by supplying nutritients and by removing toxic materials that diffuse across dentin via the microcirculation. They explain that there is a delicate balance involving the rate at which bacterial products diffuse around microgaps between restorative materials and dentin, the rate these materials permeate across dentin and the rate they are removed via pulpal circulation. All restored teeth exhibit some degree of microleakage, and if the great majority of these vital teeth were asymptomatic, this would suggest that the delicate balance between the rate of exogenous materials permeating from the dentin to the pulp was maintained by the removal of these materials by the pulpal circulation. Another possibility is that dentinal tubules may become occluded by precipitation of minerals, or plasma proteins such as fibrinogen.

In this study, it is apparent that at six months there was very little difference in the reduction of microleakage between Copalite (88%) and the bonding resins (92% for Amalgambond Plus) and (94% for All-Bond 2) even though the nature of the interface between Copalite and tooth structure and between bonded resin and tooth is so different. Figure **21**, illustrates the differences in interfacial space between amalgam restoration and tooth structure lined with either Copalite varnish or resin bonding agents. The Copalite varnish does not bond to the tooth structure or amalgam restoration and it is usually applied onto cavity walls covered with smear layer. In contrast, resin liners bond to both the tooth structure and amalgam restoration mechanically and possibly chemically (as explained earlier) and are usually applied on etched tooth surfaces (i.e., no smear layer). Because of the enormous structural differences, we would have expected that the microleakage of bonded resin restorations would be very low, compared to Copalite restorations. Although sterilizing the teeth showed no significant effect on the dentin permeability or bond strength, it may produce biochemical changes in dentin (Pashley et al., 1993b). Further, its effect on microleakage and the sealing ability of resin bonded restoration is not documented. We would expect, however, that sterilizing the teeth under such a high temperature (121 ^{0}C would denature the protein molecules and result in a mass of collapsed collagen fibrils. This might prevent any subsequent bonding and resin infiltration into the demineralized dentin with the result that the effect of the Copalite treatment would not be so different from that of resins. This may explain the small difference in microleakage reduction that exists between these treatments.

Since the level of microleakage that causes pulp hypersensitivity is not known we do not know whether 6% less leakage with Copalite when compared to resin-lined restorations is clinically significant. Clinically, it might be significant, since this may reduce hypersensitivity associated with the placement of new amalgam restorations. Perhaps, under the conditions of this study, the bonding resins simply create a better gasket, and true bonding is non-existent, i.e., fluid can still diffuse along collagen fibrils/resin interface and right through resin seal.

The results of this in-vitro study can be used to predict the clinical (in-vivo) performance of the Amalgambond Plus and All-Bond 2 dentin adhesive systems, however, there are always other factors that may contribute and influence the results of an in-vitro testing. Factors such as, pulpal pressure, thermal changes, masticatory forces, exposure to chemicals in oral environment, and type and duration of microleakage testing methods. The effect of those factors was explained earlier in the literature review. The method utilized in this study has attempted to simulate the clinical conditions to some extent by keeping the dentin structure hydrated at all time, by preparing the tooth to create a cavity geometry more representative of the clinical situation, and by storing the teeth at 37 ^{o}C.

Microleakage at the margins of amalgam restorations was found with and without a cavity liners. The corrosion products of dental amalgam at the tooth-restoration interface will not provide an effective protection against microleakage. However, an effective sealing agent could prevent early hypersensitivity or the formation of secondary caries at the tooth-restoration interface. Future research should be directed at evaluating the efficacy of dentin bonding systems to reduce microleakage and dentin permeability in clinical conditions for long-terms.

Conclusions

The results of this in-vitro study indicate the following:

1 - Microleakage was significantly reduced ($p<0.01$) and ($p<0.05$) when Amalgambond Plus and All-Bond 2 were used as liners in comparison to either Copalite varnish or no liner under amalgam restorations.

2 - No significant difference was found between the two dentin bonding systems at all time periods studied at (24 hours, 1 week, 1, 3 and 6 months

3 - The use of Amalgambond Plus or All-Bond 2 bonding systems has the potential to reduce dentin hypersensitivity, secondary caries, and pulpal irritation under well-adapted amalgam restorations compared to a Copalite varnish seal.

FIGURES

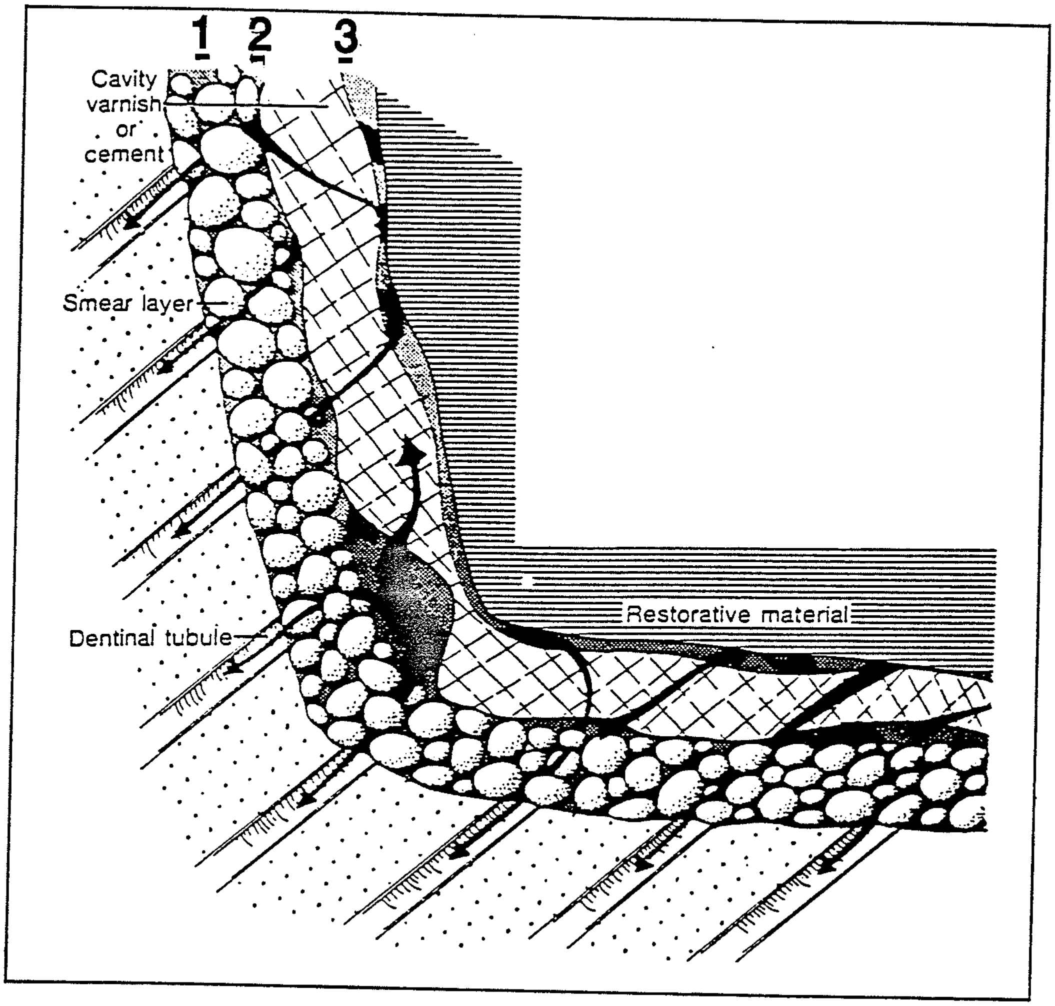


Figure 1. **Different routes for microleakage 1) within or via smear layer, 2) between the smear layer and the cavity liner or cement, 3) between the cavity liner and the restorative material** (Adapted from Pashley DH (1984). The smear layer: Physiological considerations. Oper Dent (Suppl 3):13-29).

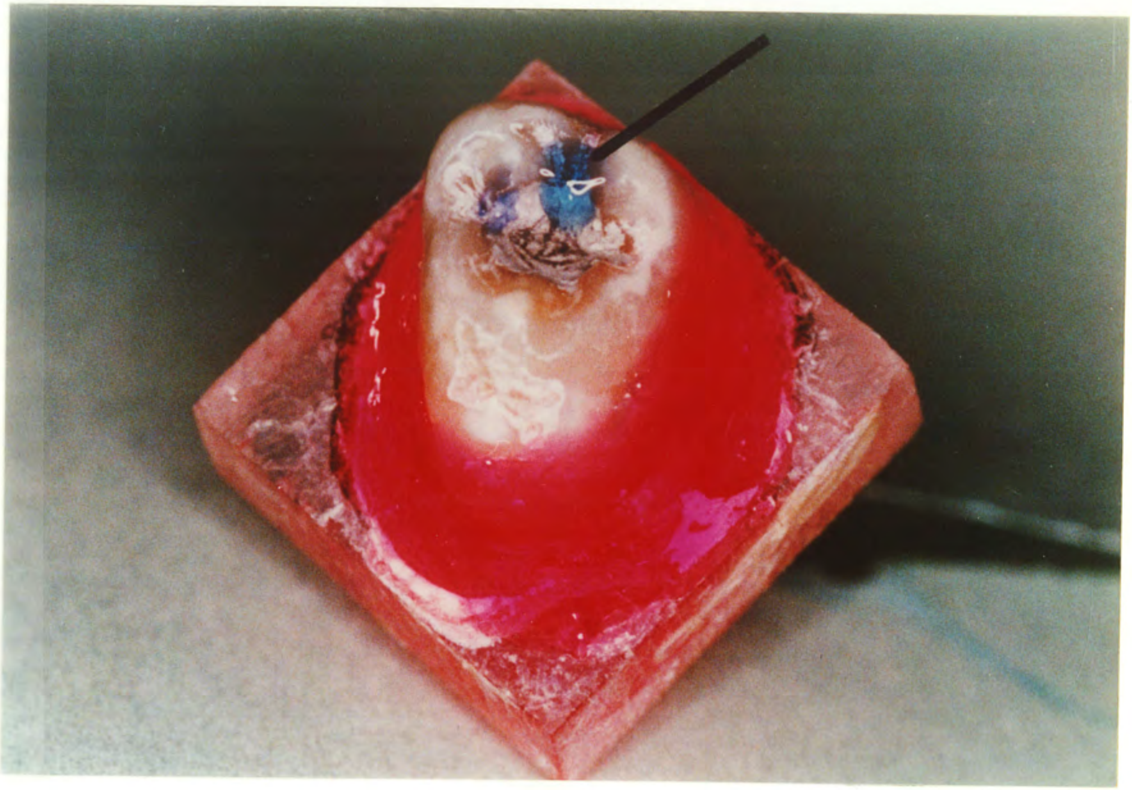


Figure 2. Microleakage around an unlined amalgam restoration margin. The arrow points to the blue dye solution that has seeped across the restoration/tooth interface to the restoration margin.

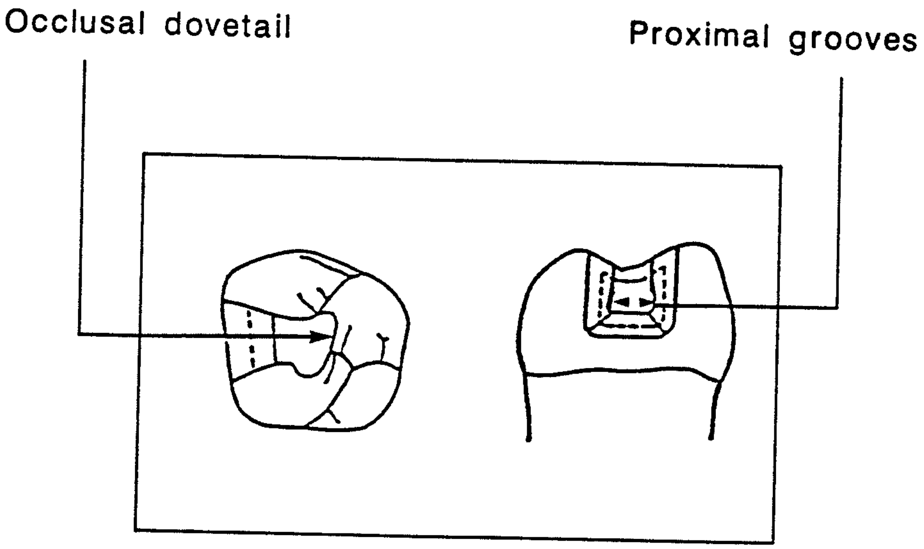


Figure 3. Two types of mechanical retention used under an amalgam restoration.

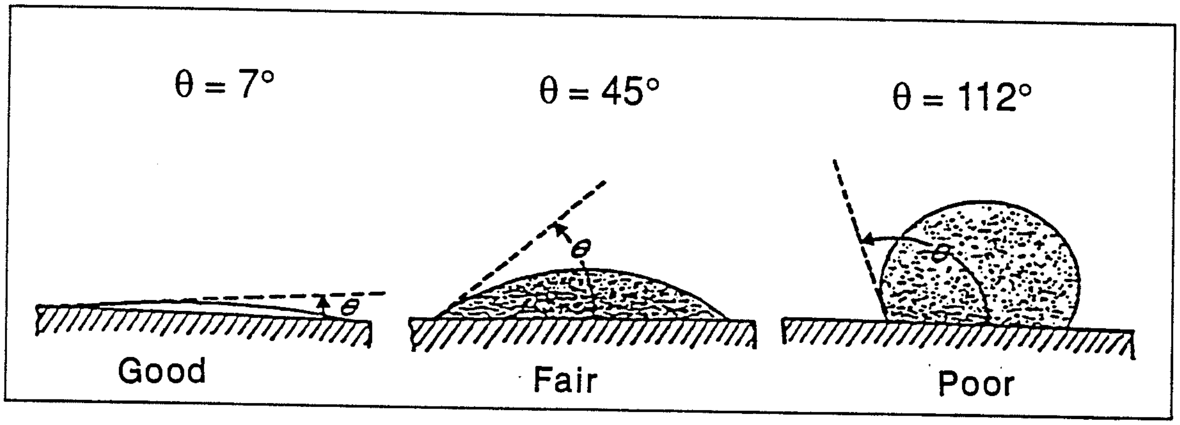


Figure 4. Different contact angles formed by three different liquids. Lower contact angle is an indication of better fluid spreading and wetting (Adapted from Phillips RW (1991). Science of dental materials. 9th Ed. Toronto: W.B. Saunders Co. :26).

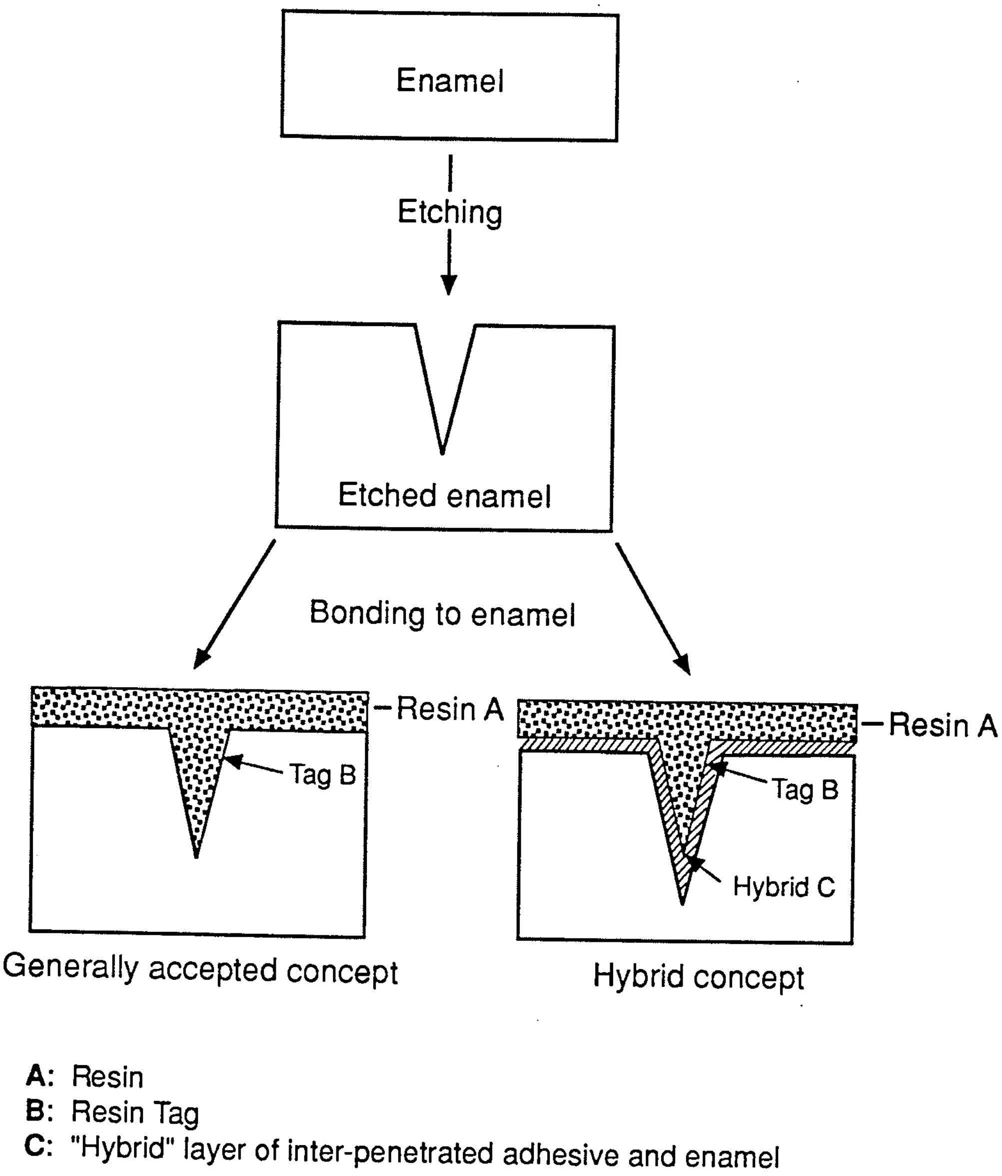


Figure 5. Enamel-resin hybrid layer formation (Adapted from Nakabayashi N, et al. (1991). Hybrid layer as a dentin-bonding mechanism. J Esthet Dent 3:133-138).

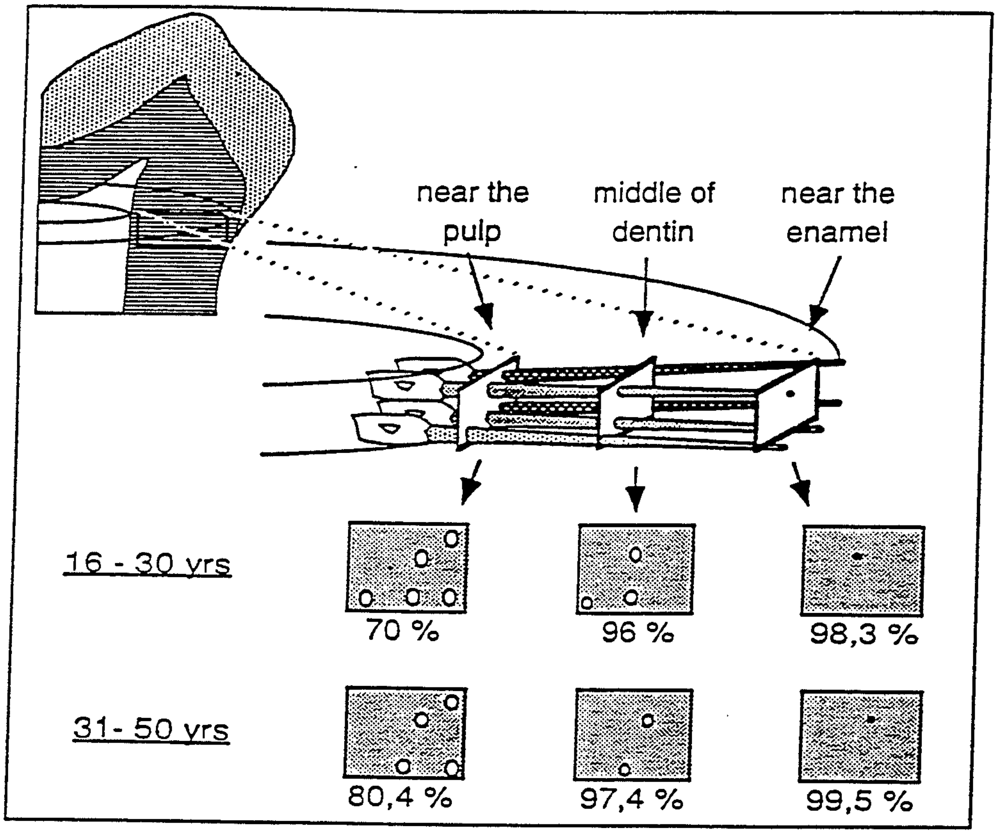


Figure 6. Tubular density variation in dentin in different age groups (Adapted from Paule SJ, et al. (1993). Factors in dentin bonding part I. J Esthet Dent 5:5-9).

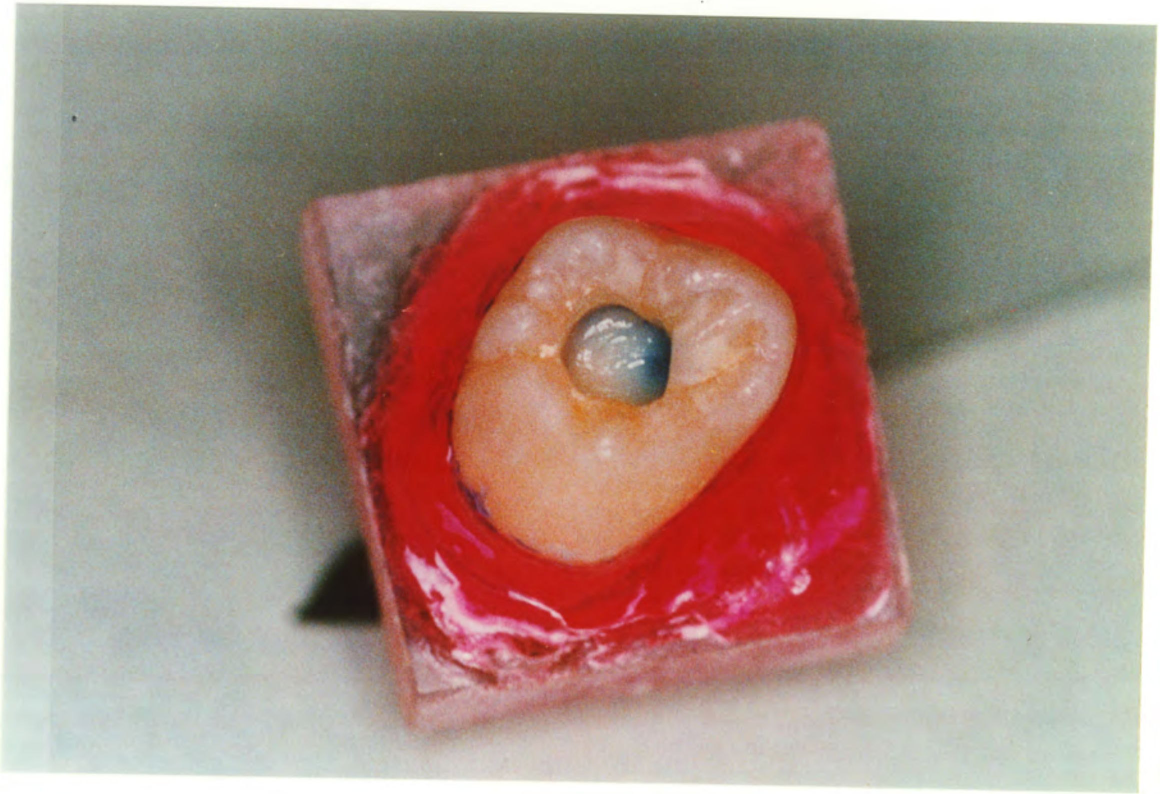


Figure 7. Dentinal fluid diffusing through the pulpal floor of a Class-I occlusal cavity.

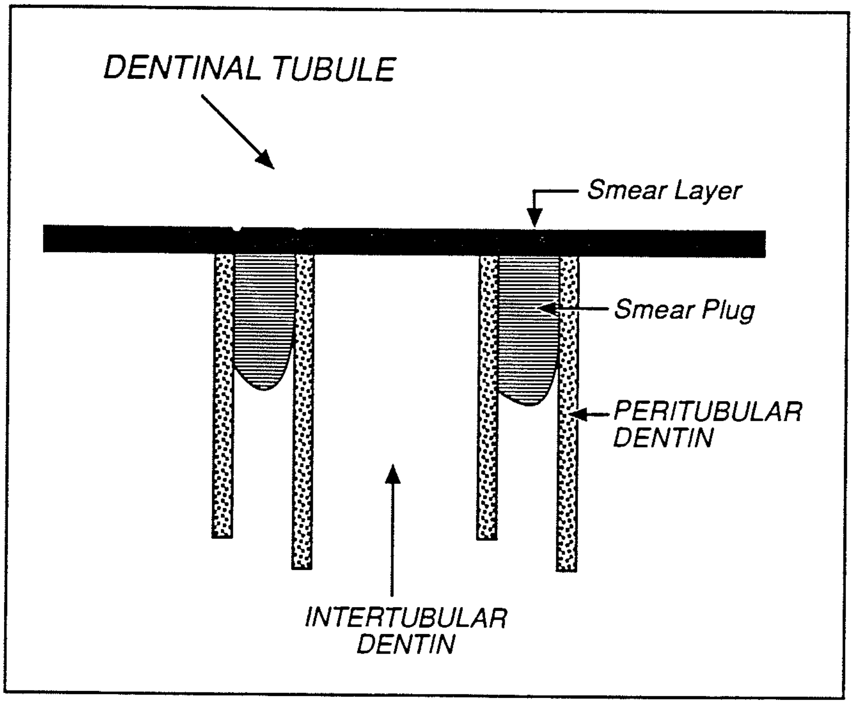


Figure 8. Smear layer and smear plugs occluding dentin surface (Adapted from Heymann HO, et al. (1993). Current concepts in dentin bonding. JADA 124:27-36).

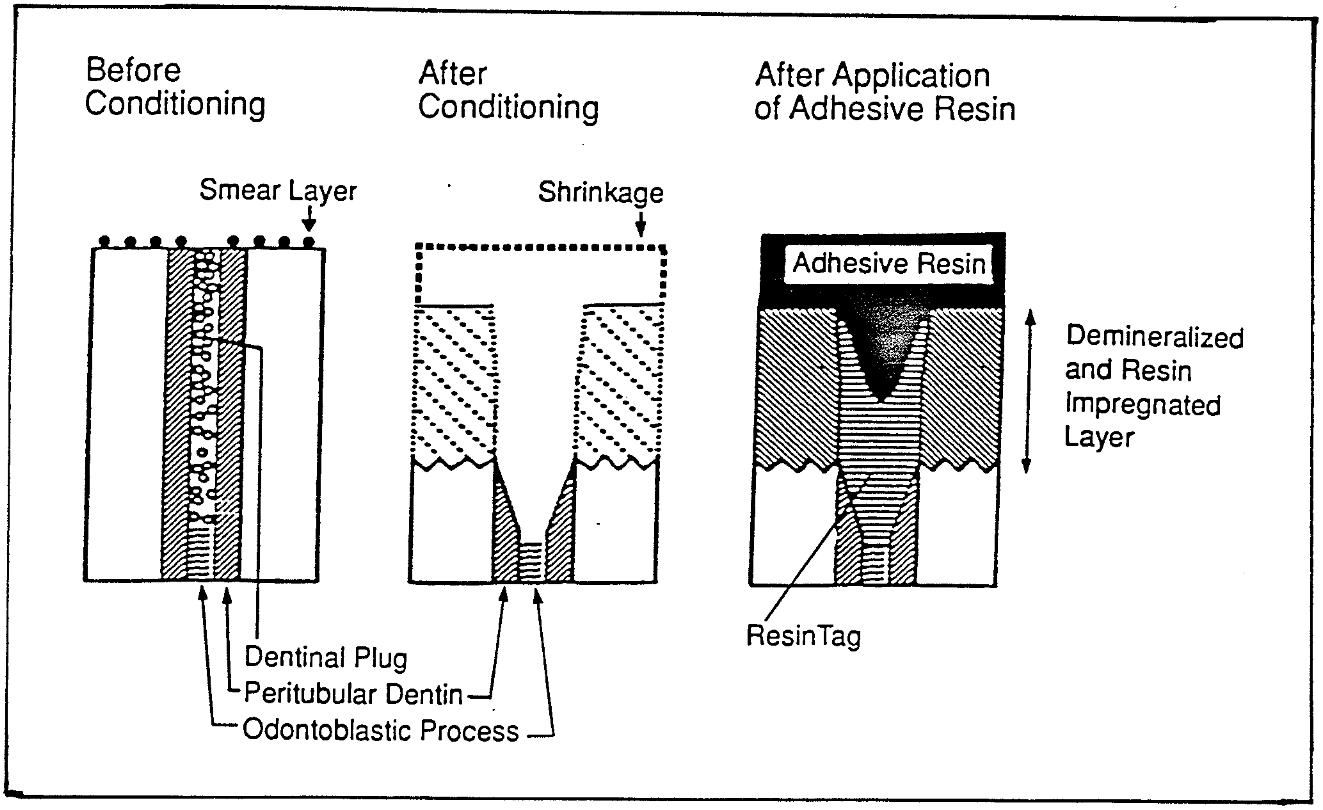


Figure 9-A. Resin impregnated dentinal tubules (Adapted from Pashley DH, et al. (1993). Permeability of dentin to adhesive agents. Quint Int 24:618-631).

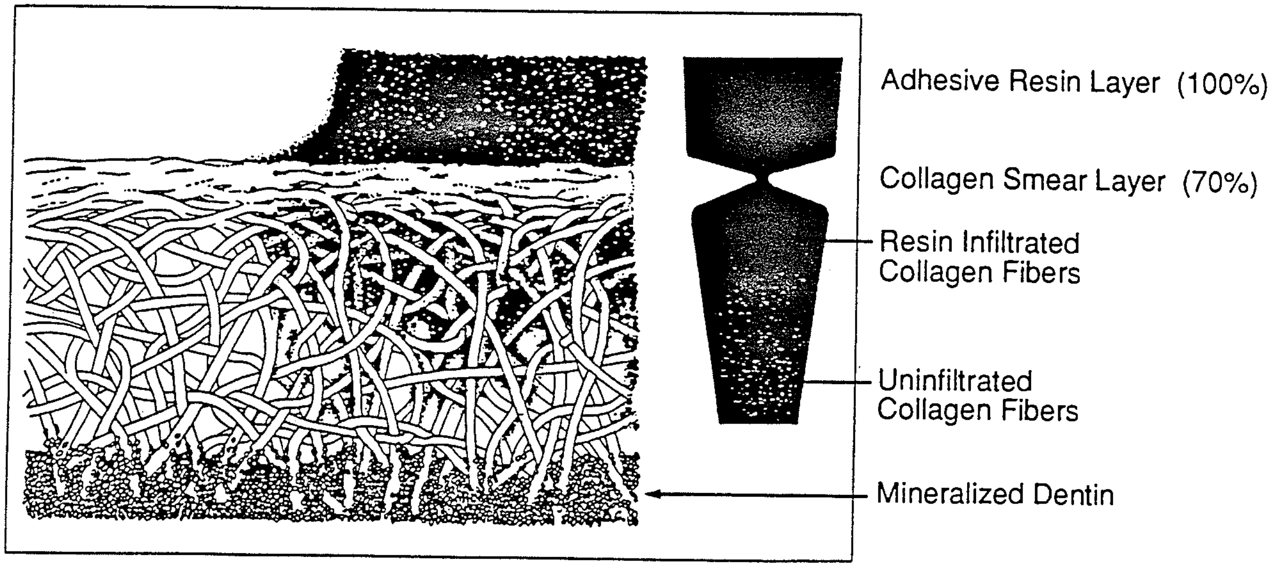


Figure 9-B. Resin impregnated collagen fibril matt (Adapted from Pashley DH, et al. (1993). Permeability of dentin to adhesive agents. Quint Int 24:618-631).

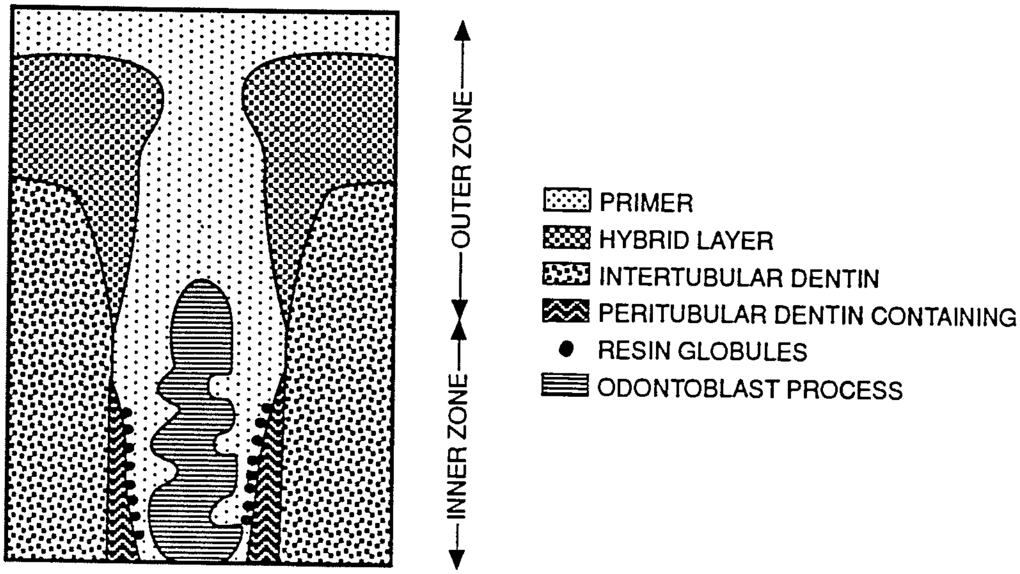
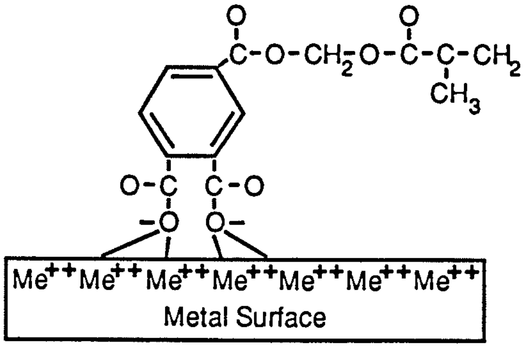


Figure 10. Schematic representation of the resin-dentin interface following the application of All-Bond 2 system (Adapted from Tay FR, et al. (1994). Structural evidence of a sealed tissue interface with a total-etch wet-bonding technique in vivo. J Dent Res 73:629-636).

A)

Superbond (4-META)



B)

All-Bond Metal Primer (BP-DM)

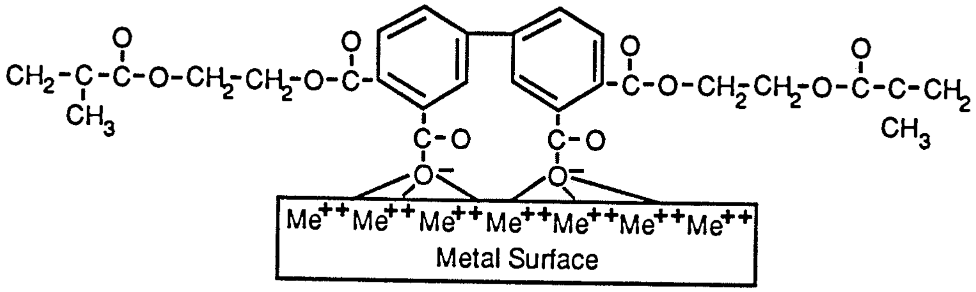


Figure 11. **Postulated reaction mechanism for A) 4-META and B) All-Bond bonding to the metal oxides on the metal surfaces** (Adapted from Albers HF (1991). Metal resin bonding. Adept Report 2:25-40).

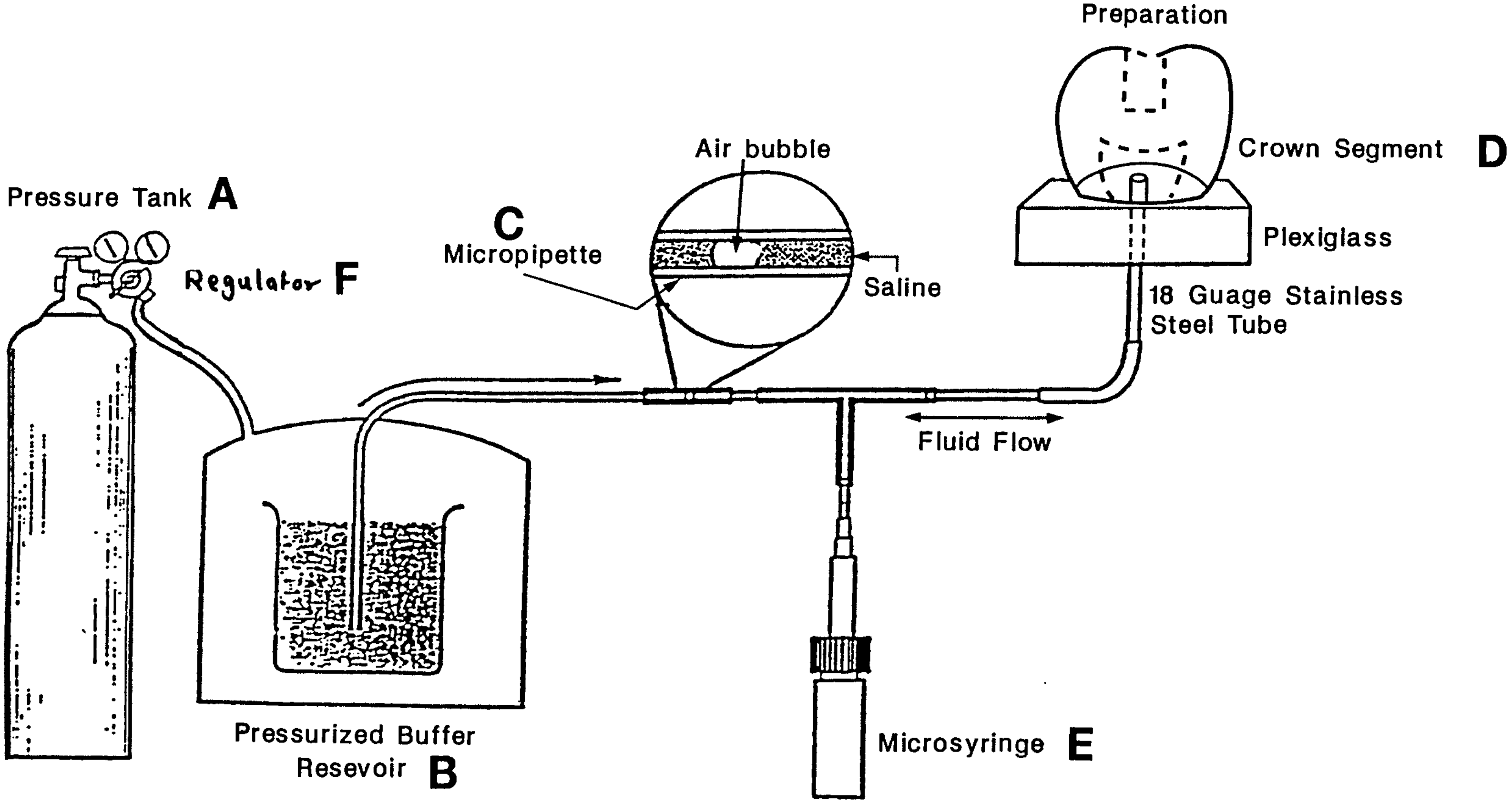


Figure 12-A. Diagram of the apparatus used to measure the dentin permeability and microleakage. The movement of the air bubble toward the chamber was used to determine the rate of fluid movement across the dentin to the cavosurface margin of the restoration.

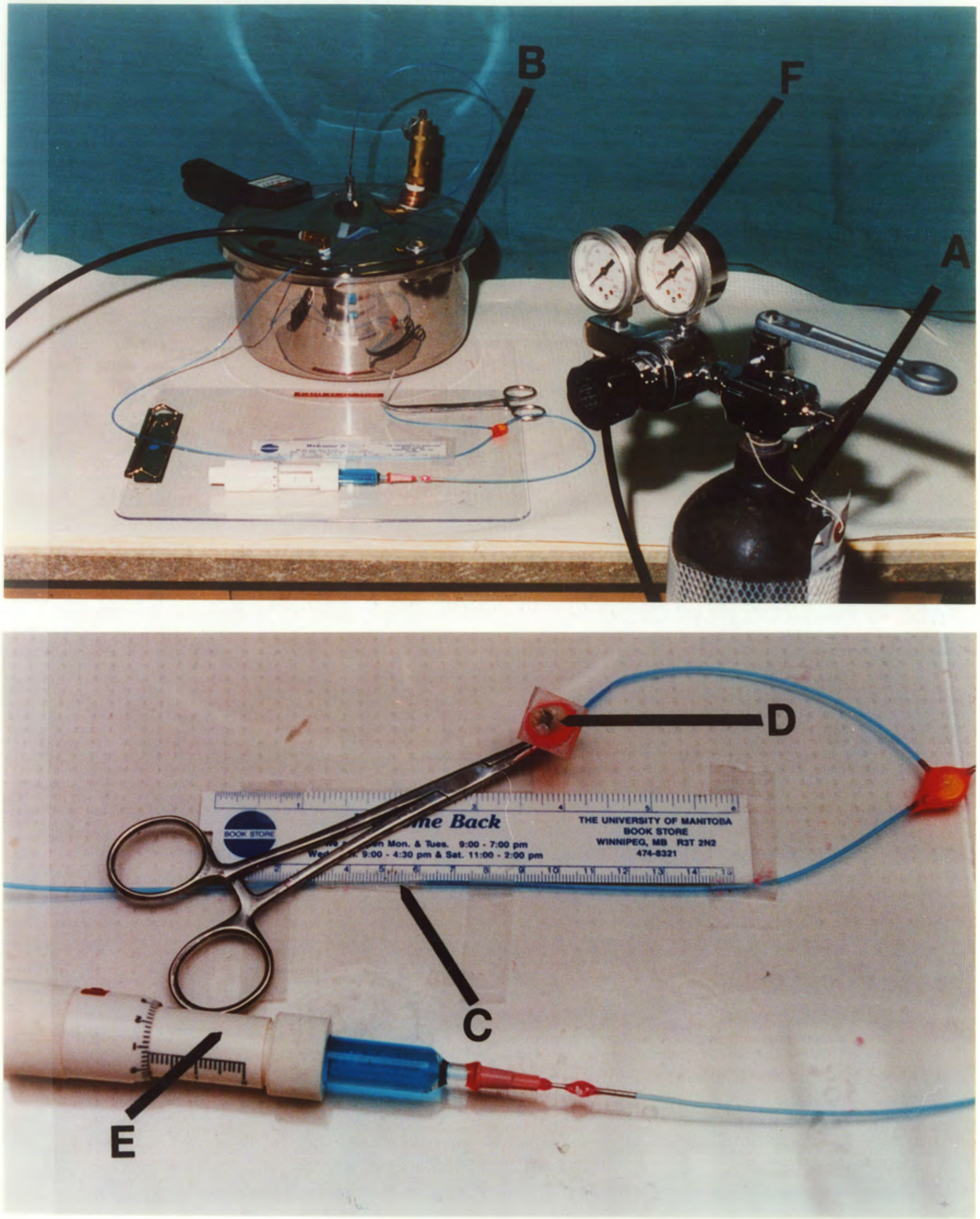


Figure 12-B. Photographs of the apparatus utilized in this study with corresponding reference points.

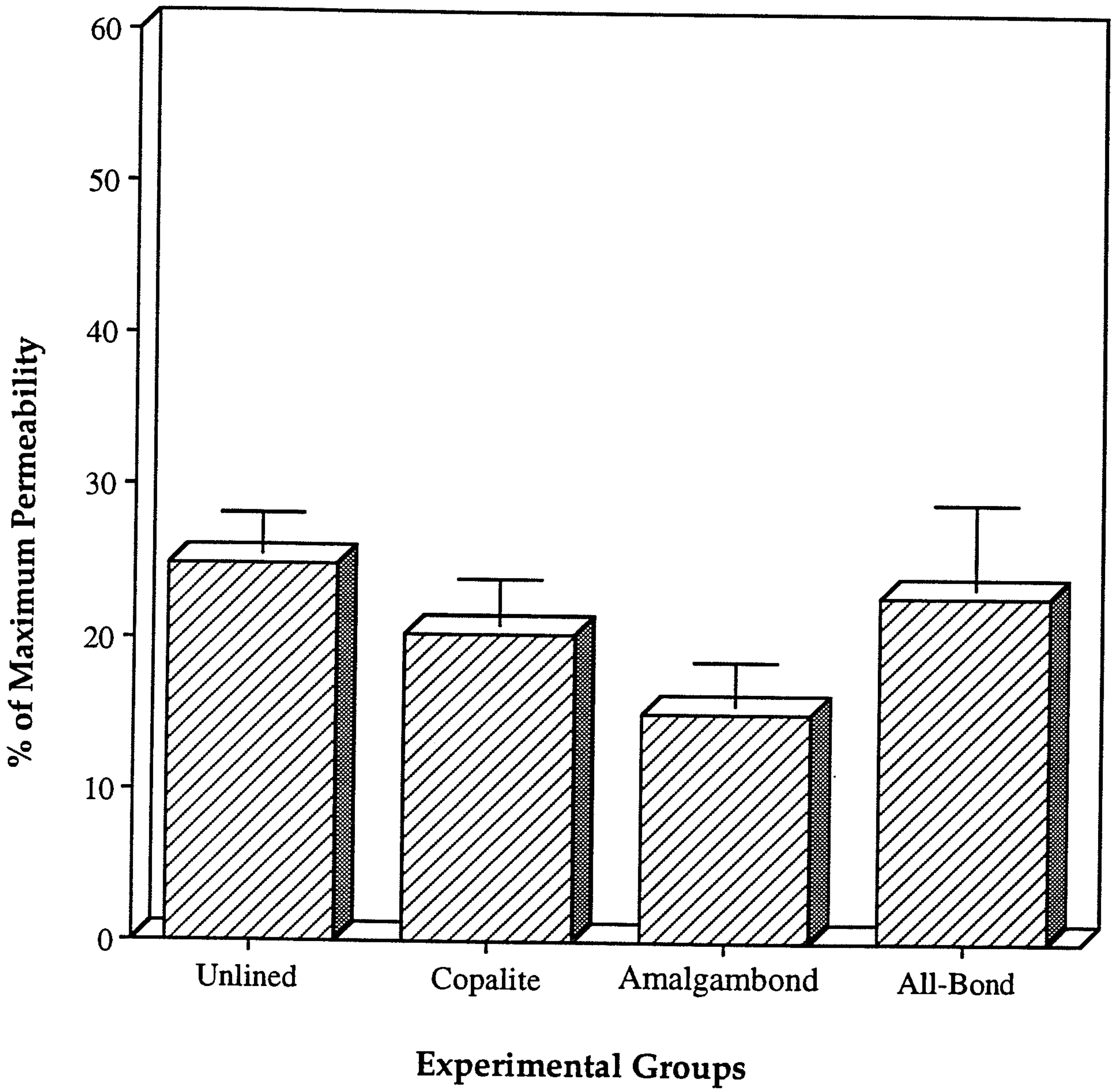


Figure 13. Dentin Permeability With Smear Layer

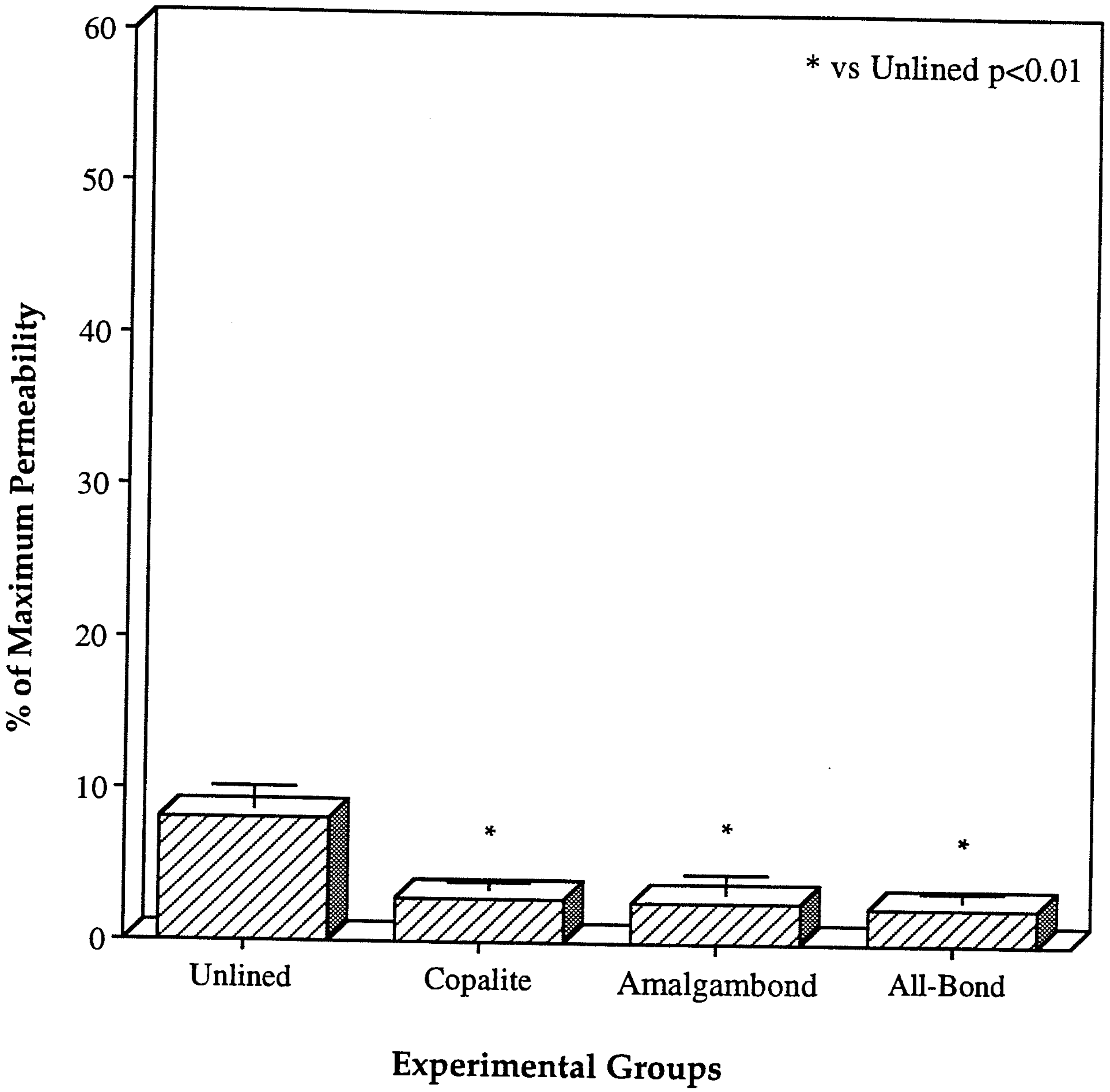


Figure 14. Dentin Permeability At 24 hours

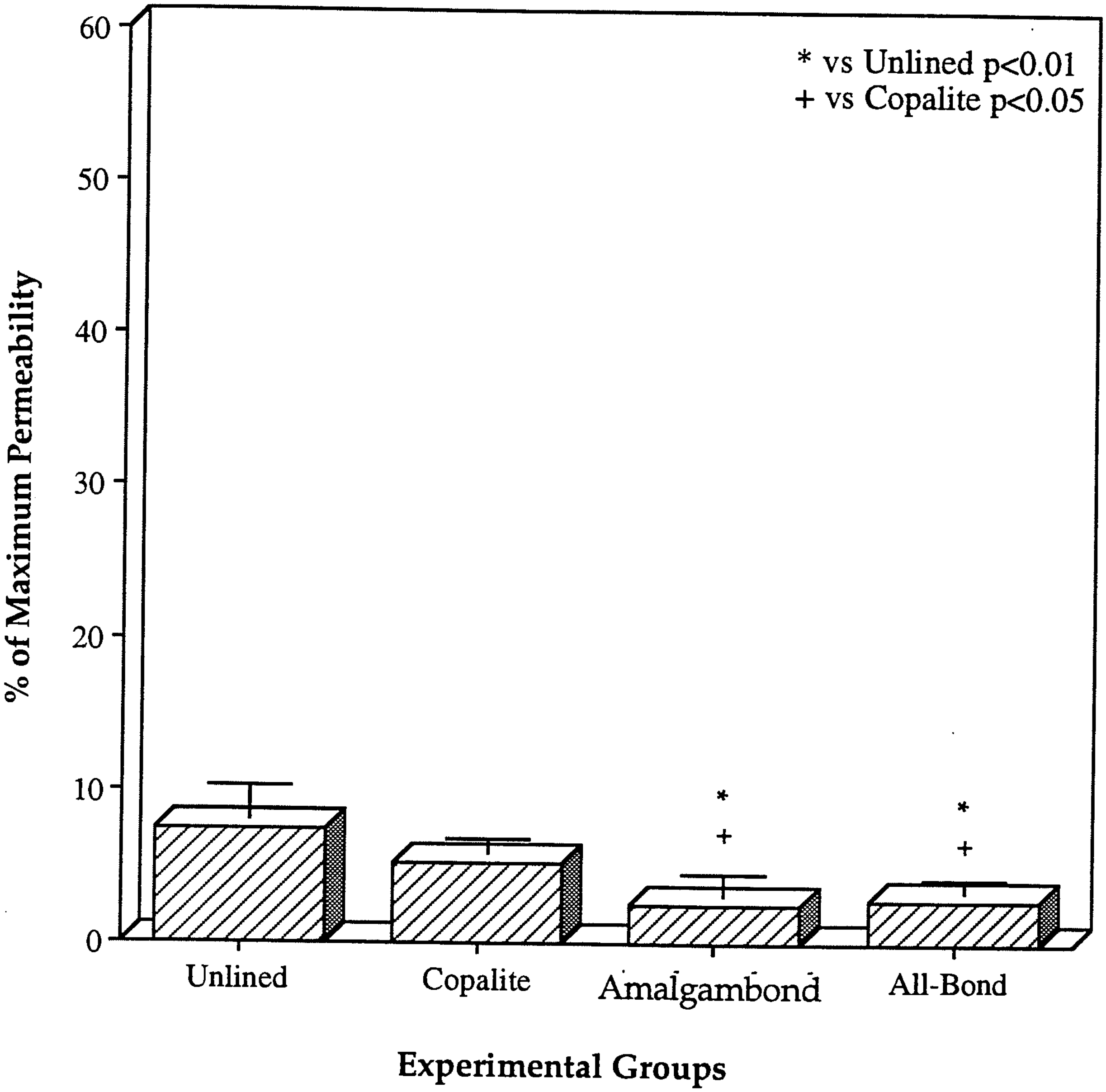


Figure 15. Dentin Permeability At One Week

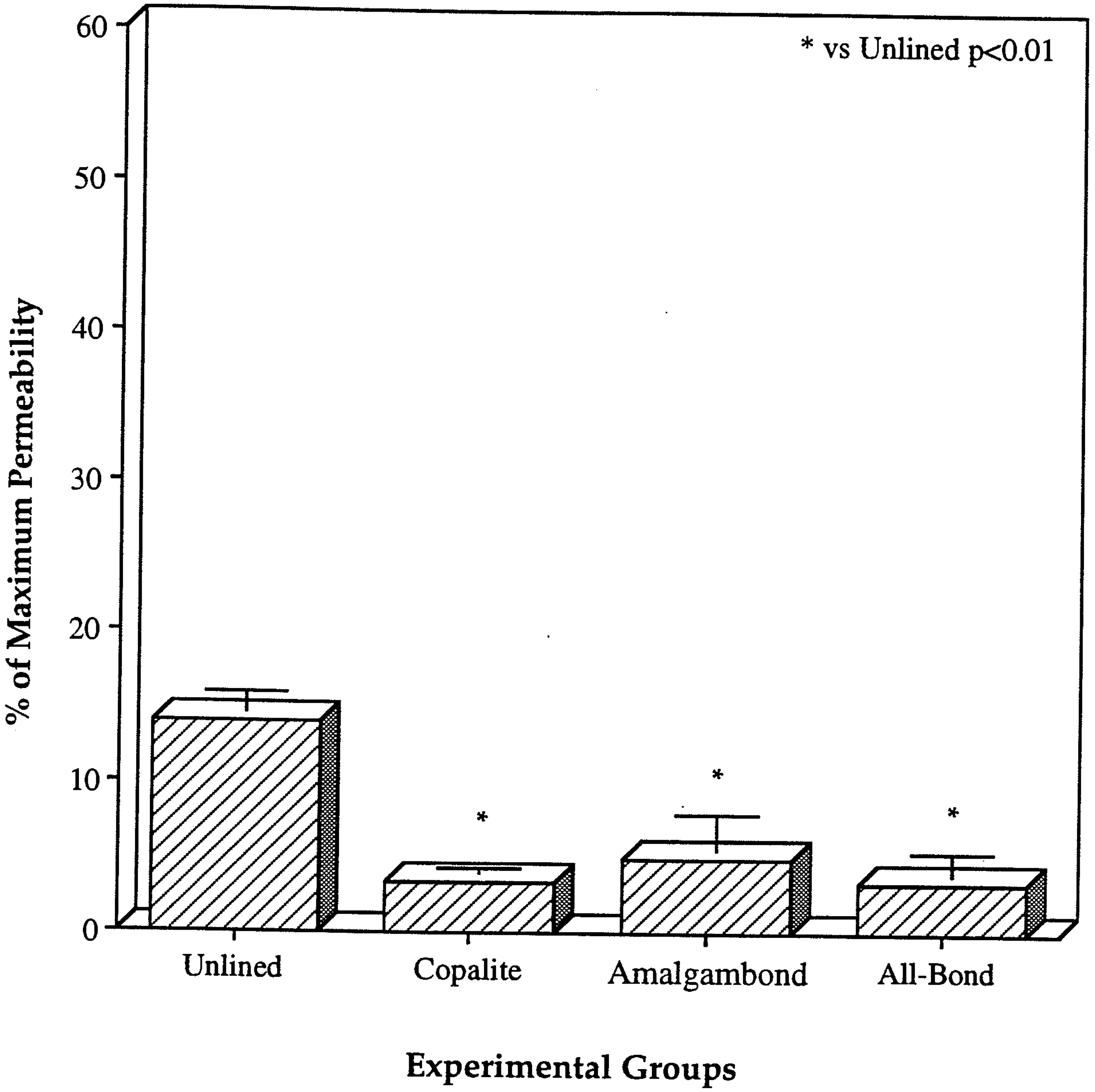


Figure 16. Dentin Permeability At One Month

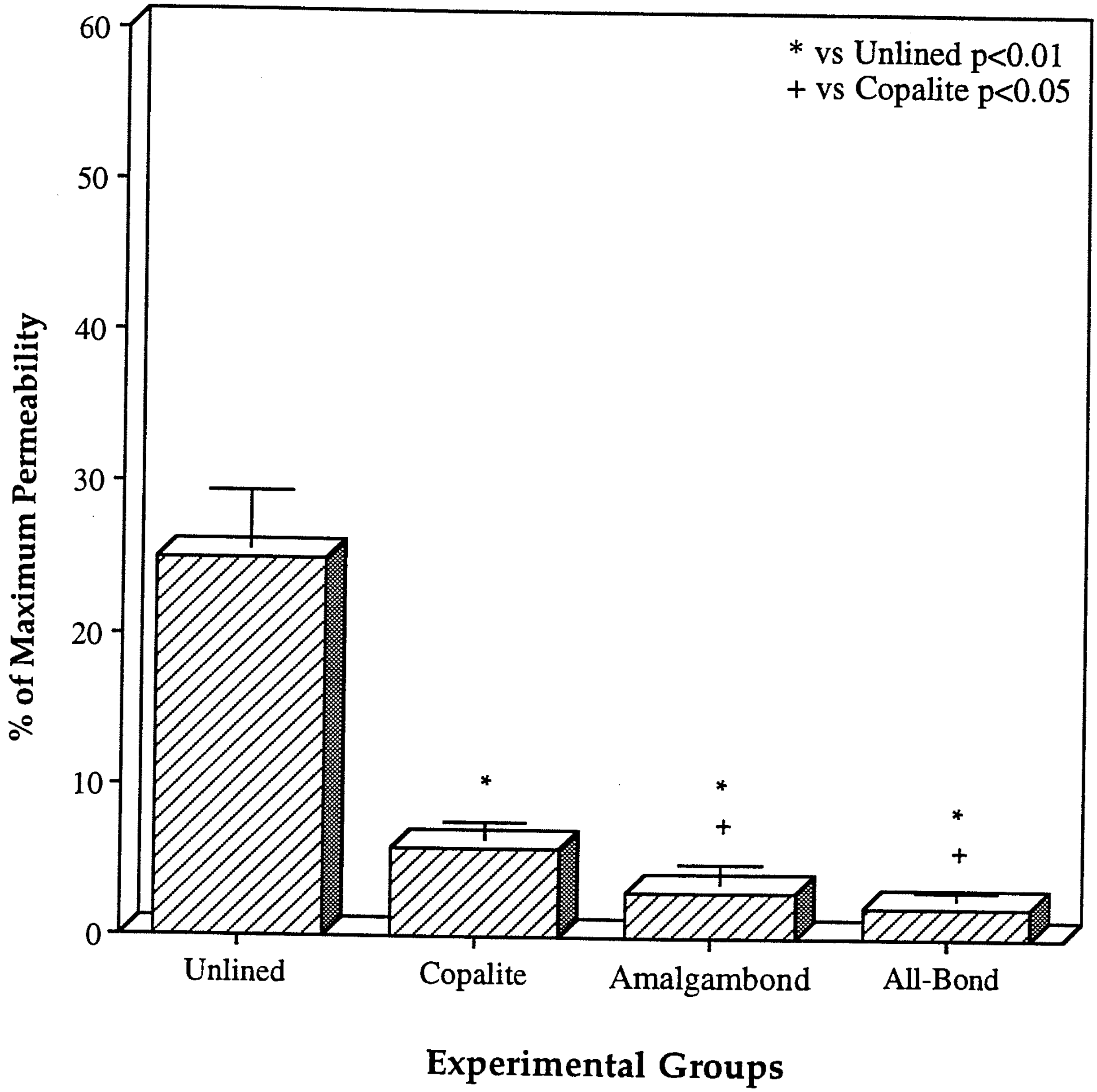


Figure 17. Dentin Permeability At Three Months

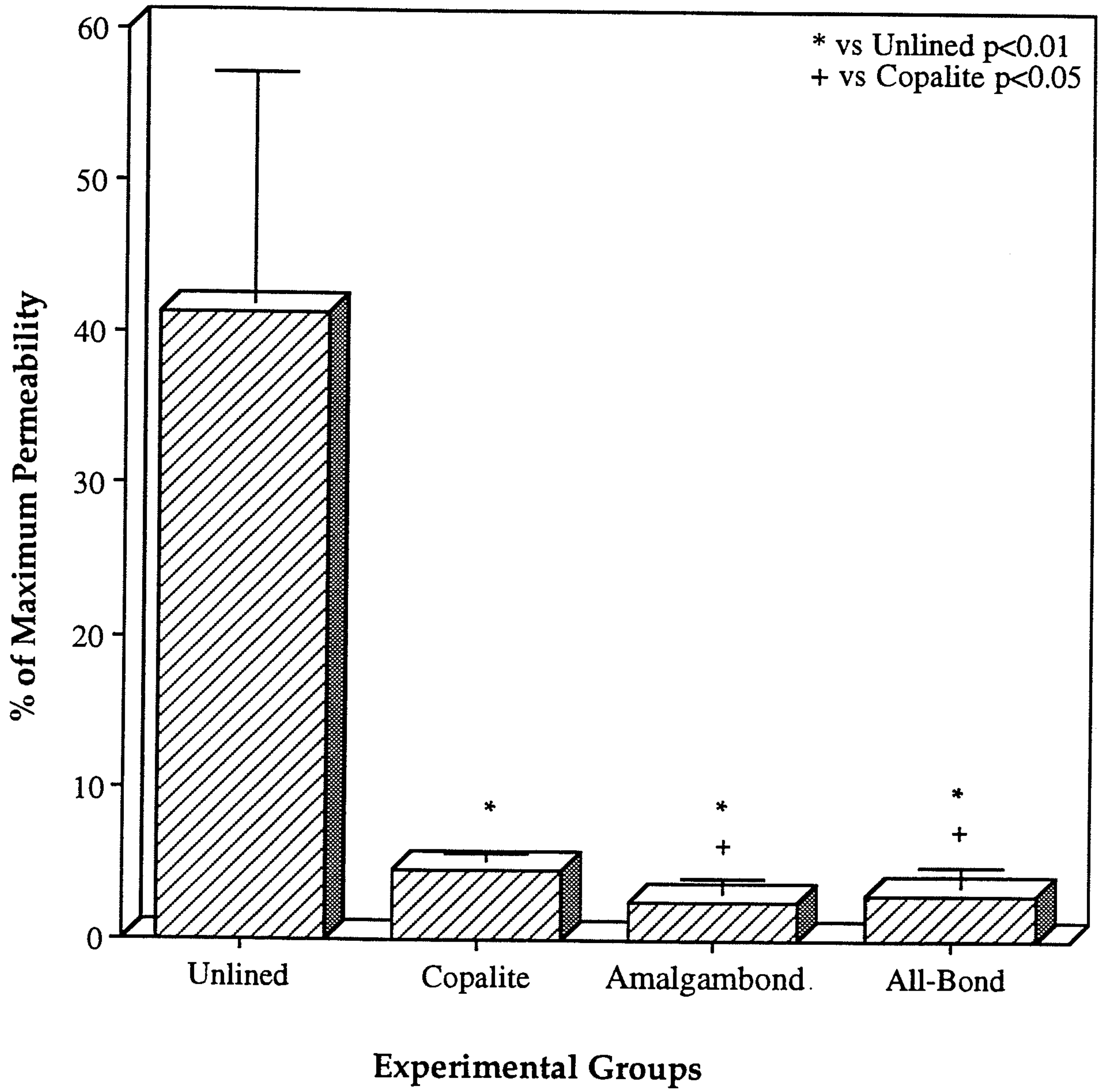


Figure 18. Dentin Permeability At Six Months

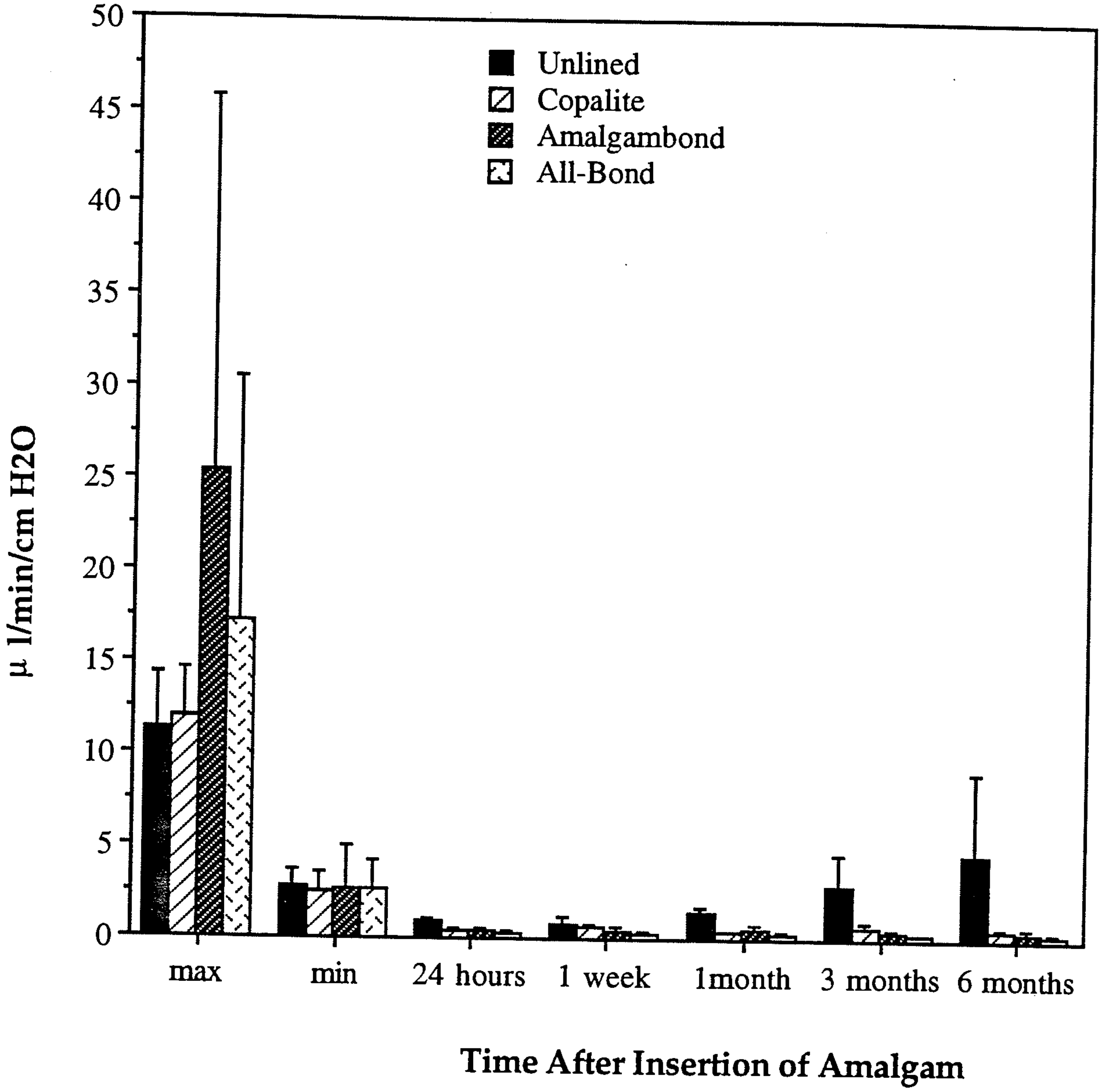


Figure 19. Mean Values of Microleakage at Different Conditions and Times, ±SD.

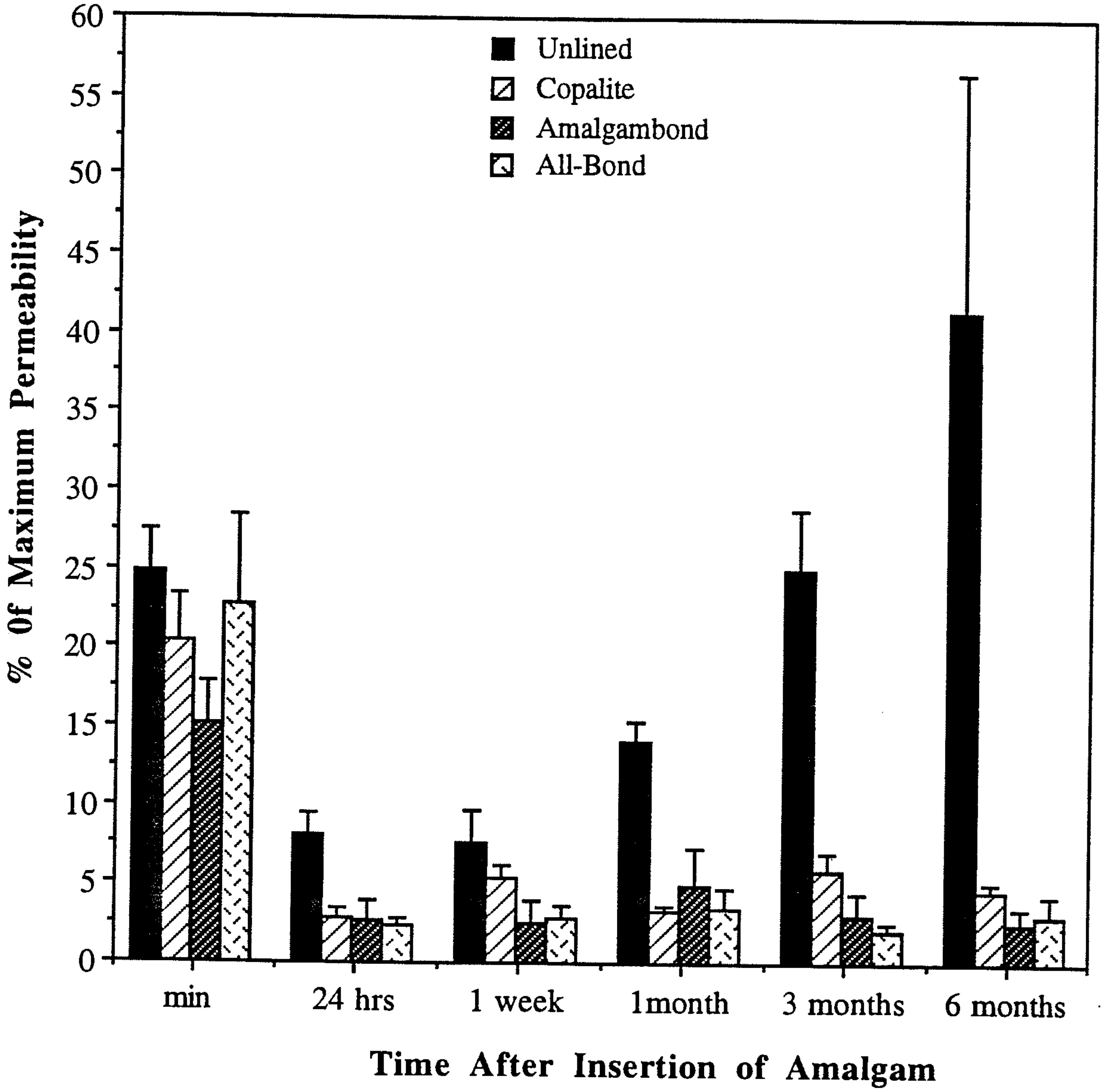


Figure 20. The Effect of Different Cavity Liners on Dentin Permeability at Different Conditions and Times, ± SEM.

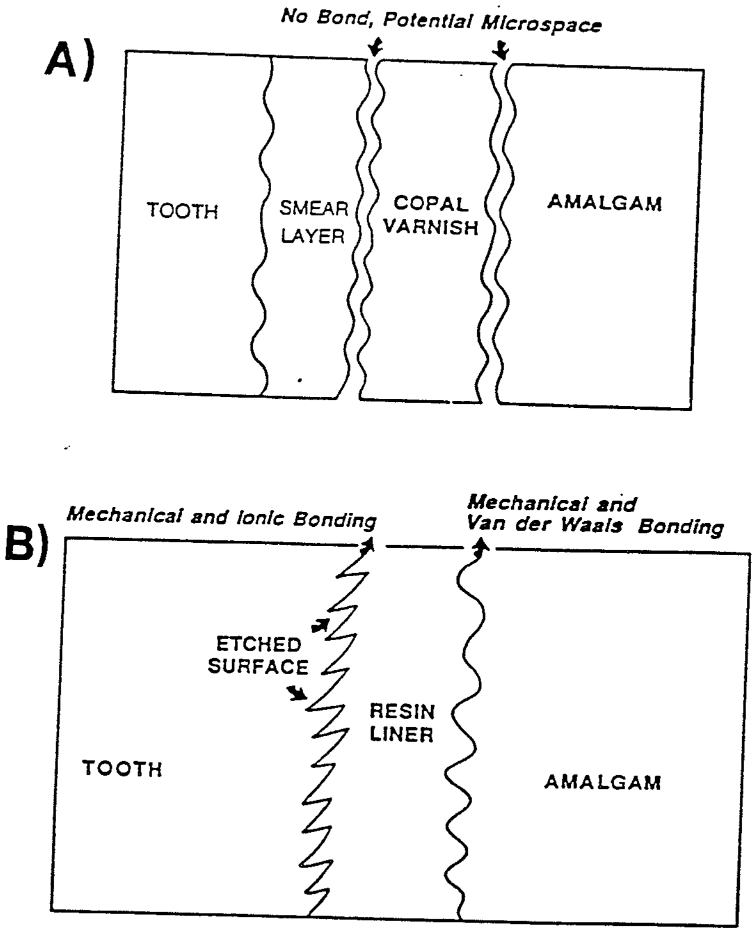


Figure 21. Differences in interfacial space and possible bonding mechanism. A) Copalite varnish-lined restoration. B) Resin-bonded amalgam restoration (Adapted from Staninec M, et al. (1988). Bonding of amalgam to tooth structure: Tensile adhesion and microleakage test. J Prosthet Dent:397-402).

TABLES

Table I. Mean Values of Microleakage, µl/min/cm H_2O, ± SD.

Material	Maximum permeability	Minimum permeability	24 hours after insertion	1 week after insertion	1 month after insertion	3 months after insertion	6 months after insertion
Unlined	11.34 ± 3.06	2.78 ± 0.87	0.86 ± 0.22	0.77 ± 0.35	1.51 ± 0.17	2.91 ± 1.76	4.64 ± 4.47
Copalite	12.02 ± 2.62	2.49 ± 1.06	0.35 ± 0.14	0.62 ± 0.17	0.38 ± 0.07	0.66 ± 0.32	0.56 ± 0.16
Amalgambond Plus	25.41 ± 20.40	2.59 ± 2.38	0.35 ± 0.18	0.43 ± 0.26	0.54 ± 0.20	0.36 ± 0.18	0.37 ± 0.23
All-Bond 2	17.19 ± 13.40	2.60 ± 1.66	0.28 ± 0.06	0.31 ± 0.07	0.32 ± 0.11	0.21 ± 0.09	0.29 ± 0.15

Mean values (x 10^{-4})
N= 6

Table II. The Effect of Different Cavity Liners on Dentin Permeability and Microlerakage at Different Conditions and Times, ± SEM.

Material	Maximum permeability	Minimum permeability	24 hours after insertion	1 week after insertion	1 month after insertion	3 months after insertion	6 months after insertion
Unlined	100	24.80 ± 2.74	8.11 ± 1.34	7.53 ± 2.22	14.10 ± 1.30	25.10 ± 3.80	41.30 ± 15.20
Copalite	100	20.31 ± 3.10	2.91 ± 0.50*	5.34 ± 0.81	3.30 ± 0.41*	5.83 ± 1.23*	4.63 ± 0.40*
Amalgambond Plus	100	15.11 ± 2.82	2.73 ± 1.20*	2.61 ± 1.30*+	4.90 ± 2.41*	3.05 ± 1.32*+	2.46 ± 1.06*+
All-Bond 2	100	22.84 ± 5.64	2.43 ± 0.51*	2.86 ± 0.81*+	3.42 ± 1.41*	2.03 ± 0.58*+	3.03 ± 1.17*+

Mean values
N= 6
*,vs Unlined $p < 0.01$
+, vs Copalite $p < 0.05$

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