THE EFFECT OF TOOTH AGE ON THE FRACTURE TOUGHNESS OF FOUR DENTAL BONDING SYSTEMS

by

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Abstract

Objective: The purpose of this *in vitro* study was to evaluate the effect of dentin age on the fracture toughness (K_{IC}) of dental bonding systems using the notchless triangular prism (NTP) specimen K_{IC} test. **Methods:** 80 human molars and pre-molars, 40 of them from patients not older than 45 years ("young") and 40 from patients not younger than 65 years ("old"), were wet ground on 600-grit silicon carbide abrasive paper to obtain a 4x4x4x4mm triangular prisms with the labial surface exposed for bonding. Within each of the two groups, the specimens were randomly assigned to four subgroups (n=10) according to the bonding system to be used for composite bonding: Adper Prompt L-pop (3M ESPE), Clearfil SE (Kuraray), Prime & Bond NT (Dentsply) and Scotch Bond MP (3M ESPE). The composite, Z-100 (3M ESPE), was bonded to each of the treated surfaces as per the manufacturer's instructions to obtain a 4x4x4x8mm dentin-composite NTP specimens. The specimens were stored in water for 24h at 37°C and tested to determine K_{IC}. The data was analyzed using Levene's test for normality, two-way univariate analysis of variance (ANOVA), and Bonferroni tests for multiple means comparisons. A significance level of 0.05 was used for all tests. **Results:** While Adper Prompt L-pop and Clearfil SE achieved the highest K_{IC} values for both age groups, Prime & Bond NT showed the lowest values. SEM observations of fractured samples revealed differences in fracture path. No statistically significant differences were observed between age groups. **Conclusions:** The K_{IC} values of the four dental bonding systems were not affected by age. Furthermore, statistically significant differences were observed between the K_{IC} values of the four bonding systems, regardless of the age of the dentin.

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Dedication

In memorian

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Chapter 1: Introduction

Root surface caries is a phenomenon of growing significance.[1, 2] Since the prevalence of root caries increases with age, it will probably become a widespread problem considering that the population of North America is growing older whereas edentulism and tooth-loss rates have been dramatically decreased.[3, 4]

Projections of the United States population indicate that the median age of the U.S. adults will increase from 33 to 46 years-old[5] and the percentage of the people aged 65 years and older will virtually double between the year 2000 (12.6%) and 2030 (20%).[6] This age group only amounted to 4% of the population in 1900 and 7% in 1940.[7] In British Columbia, the percentage of the population aged 65 years and older has grown consistently from 1901 (2.2 percent) to 1996 (12.8 percent).[8]

In addition, edentulism has declined as the North American population is retaining more natural teeth for life. The Third National Health and Nutritional Examination Survey illustrated that in a national probability sample of the non-institutionalized U.S. population, almost 57 percent of persons aged 75 years old or older were dentate, with a mean of 16.1 teeth per dentulous person in this age group[9, 10].

The consequences of these tendencies are numerous to the general dentist, particularly with regard to the management of root surface caries.[11] With increasing age, most persons have a progressive and continuing gingival recession with exposure of root surface to the oral environment.[12] The root of the tooth may be more susceptible to mechanical and chemical destruction than coronal enamel, due to the distinctive physical structure and chemical composition of cementum and dentin.[13]

The prevalence of root caries in adults increases markedly with age (Table 1)[14] and when all caries are considered, the aggregate caries increment may be higher in people over 55 years old than in children.[15, 16]

Table 1 Depiction of the prevalence of root surface caries in adults (adapted from Garcia-Godoy et al.[14])

Age	Percentage
18 – 24	0
35 – 44	21
45 – 54	26
55 – 64	38
65 – 74	47
75 +	56

Many epidemiological studies have estimated and compared the prevalence, incidence, risk factors, and the treatment needs of people with root caries[17, 18, 19, 20, 21]; however, compared to enamel caries, there have been limited investigations to evaluate the restorative management of root cavities.

Dental amalgam, cements, and composites have been frequently employed for the restoration of root surface caries (class V restorations). Explicitly recommending an ideal restorative material to be used when managing root caries lesions is extremely difficult, especially since long term data on the clinical performance of these materials is lacking.

In addition, other factors must be taken into consideration when choosing the most appropriate restorative material, such as caries activity, access, moisture control, extension of the lesion, and aesthetics.

Even though dental amalgam has been used frequently to restore root surface lesions, the use of glass ionomer or resin-modified glass ionomer cements (GIC/RMGIC – both being polyalkenoate cements) and composites may be more appropriate in aesthetically prominent teeth. Moreover, while dental amalgam restorations rely mainly on mechanical retention, polyalkenoate cements and composites take advantage of "adhesive dentistry".

The first step towards the era of "adhesive dentistry" started in 1955, when Buonocore[22] proposed the acid etching technique as the preconditioning step for bonding to enamel. Subsequently, the introduction of dental bonding systems challenged and dramatically changed the classic concepts of operative dentistry.

Adhesion to enamel has become a reliable, routinely performed clinical procedure in restorative dentistry. Its application transformed many concepts in dentistry such as caries prevention, cavity preparation, and aesthetics.[23, 24] However, while several new and presumably improved dental bonding systems have become available, generating a bonding system that can successfully interact with dentin is still a challenge since dentin, compared to enamel, has a complex and heterogeneous structure. Furthermore, human dentin undergoes age-related changes that may also affect the effectiveness of existing bonding systems.[25]

Chapter 2: The Tooth Structure

Enamel

Enamel is one of the most important components of a tooth and is unique among all other mineralized tissues of the human body due to its high mineral content.[26]

Enamel is composed by highly organized, tightly packed crystallites that comprise 90% its volume and 95% of its weight. While other mineralized tissues in the human body such as bone and dentin contains approximately 20% organic material by volume, mature enamel has less than 1% by weight, 2% by volume organic matter.

Enamel crystallites are particularly long relative to their thickness and are highly oriented. They normally extend from the underlying dentin towards the surface of the tooth and are ordered into bundles that are called prisms.[27] This exceptional organization and mineralization confer dental enamel outstanding physical properties, making it the hardest tissue in the body.[28]

The different phases that can be observed throughout tooth development are the bud stage, cap stage, bell stage, and crown, or calcification, stage. The formation of enamel, also called amelogenesis, begins during the crown stage through cells called ameloblasts.

The process of amelogenesis is rather complex but can be divided in two distinct stages.[27] The initial stage (secretory stage) is typified by the presence of specific proteins such as alkaline phosphatase and an organic matrix that would create partially mineralized enamel. Subsequently, as the second stage progresses (maturation stage), tooth enamel mineralization is finally accomplished.[26]

During the secretory stage, the ameloblasts cells are characterized by being polarized, columnar cells that release enamel proteins into their vicinity. The released proteins contribute to the formation of an enamel matrix that is then partially mineralized by alkaline phospatase.[26]

Once this first layer is formed, the ameloblast cells move away from the dentin, leaving a cell projection that gets surrounded by the developing enamel (Tomes' processes). Amelogenesis persists in the neighboring ameloblasts, producing a walled area, or pit, that houses a Tomes' process. Mineralization of the matrix located within each pit will ultimately turn into enamel rods, while mineralization of the matrix located in the walls of the pit will turn into interrod enamel.[28]

During the maturation stage, the ameloblasts change from polarized, columnar cells, to striated type of cells. Instead of being responsible for the production of proteins, ameloblasts become responsible for the transportation of proteins, such as amelogenins and tuftelins, used for the final mineralization process. Once the maturation stage is finalized, the enamel has concluded its mineralization.[27, 28]

Dentin

Dentin is a vital, hydrated, and heterogeneous tissue composed of a solid phase surrounding a network of tubules; it lies between enamel and the dental pulp (Figure 1).[29] The structure of dentin has not been fully explained yet; however, the understanding of its nature and complexities can influence the outcome (success or failure) of nearly all procedures in restorative dentistry.[30, 31]

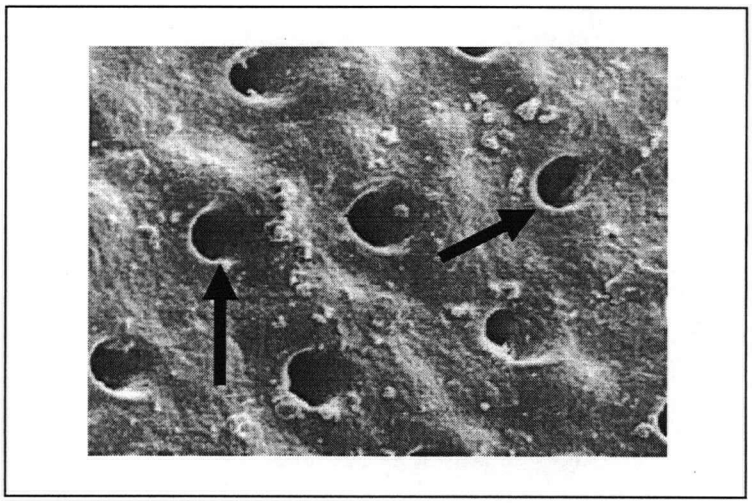


Figure 1 Scanning micrograph of dentin the dentinal tubules (arrows).

The production and deposition of primary dentin occurs throughout tooth development and the amount and form of dentin varies with the size and shape of the tooth. By volume, dentin is composed of approximately 55% mineral content in the form polycrystalline calcium hydroxyapatite [$Ca_{10}(PO_4)_6(OH)_2$], 30% organic component consisting mainly of type I collagen, and 15% fluid.[32, 33]

The distribution of the main components of dentin produces a composite with unique structural features. Embryonically, dentin is the outcome of the relationship between the ectodermal and ectomesenchymal aspects of the tooth germ that provoke the differentiation of odontoblasts, which results in dentinogenesis.[34] Structurally, dentin is composed of mineral crystals filler particles deposited in a matrix of protein fibrils.[35] Approximately 90% of the matrix is type I collagen; the balance consists of a sheath of phosphoproteins surrounding the collagen, and other proteins in small amounts.

Dentin can also be characterized as inter- or peri-tubular[36] whereas situated between the dentinal tubules or surrounding them. The peritubular dentin lacks organic structure and contains mostly apatite crystals forming a well-mineralized sheath that lines the tubule lumen. The intertubular dentin separates the dentinal tubules, a distinct and significant feature of the dentin. Measuring approximately 1 to 3 micrometer in diameter[Error! Bookmark not defined.], the dentinal tubules contain elongated cell bodies, the odontoblastic processes, which spread out from the dental pulp organ to the dentino-enamel junction (DEJ) or cementum. Their average density (number of tubules/unit area) is about 30,000-tubules/sq.mm; however, since the tubules converge towards the pulp chamber, their density and orientation differ with location, which influences hardness[37], permeability[81] and bonding capability (Figure 2).[38, 39, 40, 41, 42] The density of dentinal tubules is less around the DEJ than around the pulp chamber and, therefore, the amount of intertubular dentin available for bonding is higher in superficial than deep dentin.

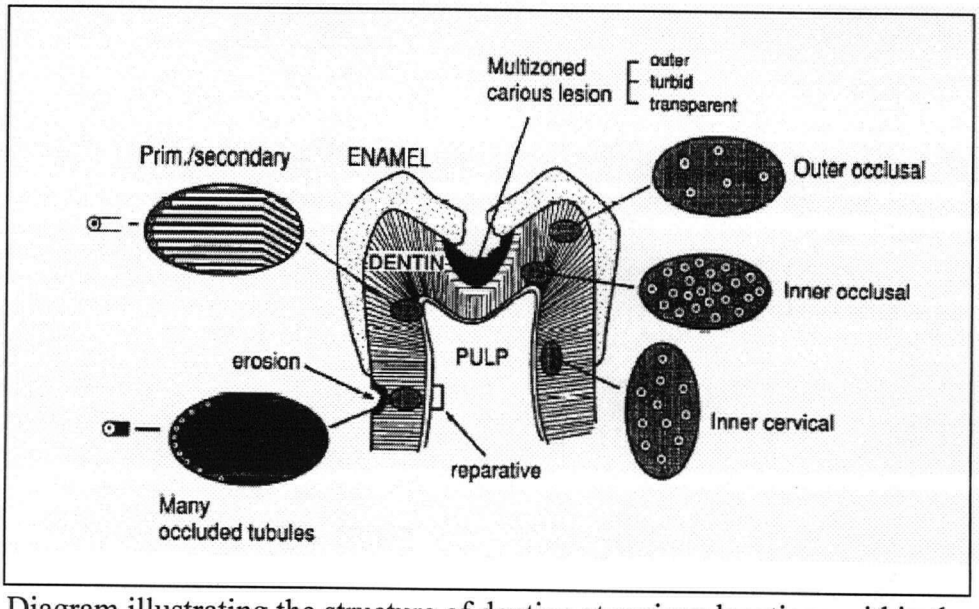


Figure 2 Diagram illustrating the structure of dentine at various locations within the structure of premolar
(from Grayson *et al.*, 1997).[43]

Chapter 3: Adhesion in Dentistry

The great majority of the scientific basis involving tooth preparations was introduced by G.V. Black in the late 1800s.[44] Until mid 1970s, tooth preparations had to be extremely precise and observe specific physical principles of resistance and retention in order to prevent the dislodgement of a restoration due to the forces of mastication.[45, 46]

For instance, an ideal class I preparation would require the parallelism or slight occlusal convergence of two or more opposing external walls, the extension of the outline form to include pits and fissures, thereby placing the margin on relatively smooth, healthy tooth structure, sufficient depth (i.e., 1.5 mm) to result in adequate thickness of the restoration, and the presence of a flat pulpal floor.

Although the precision and the principles of resistance and retention forms proposed by Black are still required for certain types of restorative materials, such as amalgams and cast metal restorations, tooth preparations have changed significantly.

Due to advances and developments in adhesive dentistry, tooth preparation has been simplified and is less specific in form, resulting in a more conservative approach. Composite restorations, for instance, require just the removal of the carious tissue without the need for specific wall designs, undercuts, and/or grooves.[47]

Basic Concepts of Adhesion

In general, adhesion involves an adhesive that upon solidification bonds two substrates, the adherents.[48] The main function of an adhesive is to provide the molecular link between the adherents, therefore, effectively creating a joint and forming two interfaces. Figure 3 depicts schematically a dental adhesion and the related interfaces. In the figure, Adherent 1 illustrates the tooth structure (i.e., enamel/dentin) while Adherent 2

could be any restorative material that needs to be bonded to the tooth structure via an adhesive. If the restorative material was a resin composite, the adhesive would be a dental bonding system.

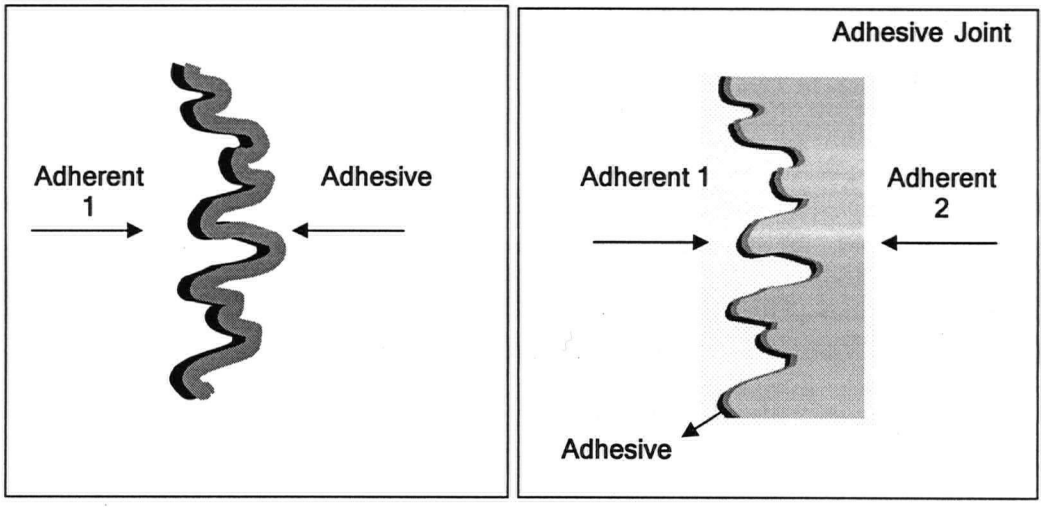


Figure 3 Illustration of the components in an adhesive joint.

Four essential prerequisites must be fulfilled in order to create adhesion. Firstly, the surface of the adherent(s) must be clean and free of any surface debris or absorbed films of oil or dirt that could prevent an intimate contact between the adhesive and the adherent(s). Secondly, the adhesive must spread onto the adherent(s) so that it wets the surface for intimate contact.[49] Hydrophilic materials do not wet hydrophobic surfaces very well, and *vice versa*. Enamel and dentin are hydrophilic, while most composites are hydrophobic; therefore, the challenge for dental adhesives is to provide acceptable wetting for both of these materials. Thirdly, the adhesive must penetrate into all the surface interstices and develop good adaptation. This process should occur without entrapping air or leaving unfilled surface spaces. Finally, the entire process depends on

the adhesive (and restorative material) becoming fully cured since under-curing allows chemical erosion and/or de-bonding of the bonding system.[50]

Adhesion to Enamel

The era of "adhesive dentistry" started in 1955, when Buonocore[22] proposed the acid etching technique on enamel, after being stimulated by the industrial use of 85% phosphoric acid to assist the adhesion of paints and resins to metallic surfaces. Since then, adhesion to enamel became a routinely performed clinical procedure that transformed many concepts in dentistry such as caries prevention, cavity preparation, and aesthetics.[23, 24]

The acid etching technique requires the application of an acid onto the enamel surface. The acid is used to superficially dissolve the enamel, thus creating a very irregular surface (preconditioning of the enamel).[51]

Since Buonocore's initial use of 85% phosphoric acid to etch the enamel surface, a great variety of phosphoric acid concentration have been employed. Gwinnett et al.[52] have proposed the application of lower concentrations of acid in order to avoid the formation of precipitates that could possibly interfere with the adhesion process. Acid concentration below 27% also appears to produce precipitates (i.e., dicalcium phosphate monohydrate) that cannot be easily removed and could interfere with the adhesion.[52]

Silverstone reported that the use of 30% to 40% phosphoric acid created a very retentive enamel surface. On the other hand, concentrations above 40% have been reported to dissolve less calcium resulting in ill–defined etching pattern.[53] Consequently, most currently phosphoric acid gels concentrations range from 30% to 40%.

Initially, an etching time of 60 seconds was suggested when using 30% to 40% phosphoric acid. However, studies comparing different etching times, using scanning electron microscopy (SEM), have demonstrated that a 15-second etch resulted in similar surface roughness as that provided by a 60-second etch.[54, 55, 56] Moreover, in vitro studies have reported similar bonding strengths values and microleakage for etching times of 15 and 60 seconds.[57]

After the intended etching time, the acid is rinsed away. The irregular surface created is then infiltrated by monomers in primers and bonding agents that become micro-mechanically interlocked with the enamel surface upon polymerization.[58]

Adhesion to Dentin

The adhesion of bonding systems to dentin is also mainly micro-mechanical in nature.[59] Bonding to dentin is accomplished by the penetration of the adhesive into the "filigree" of exposed collagen fibres of an acid-etched dentinal surface, the dentinal tubules, and into the intertubular dentin.[60]

In 1979, Fusayama advocated the application of the acid etching technique to dentin.[61] Acid etching of dentin leads to a superficially demineralised dentin and, consequently, a permeable scaffold of collagen fibers that are surrounded by water. A primer is then applied, which penetrates into the intertubular dentin and the tubules, impregnating the collagen within the intertubular dentin. The final step is the application of a low viscosity resin adhesive. The resin adhesive combines with the primer and they lock onto the surface after curing. This process creates a complex formed by the adhesive and the dentinal tissue called the hybrid layer[60] or the inter-diffusion zone.[62] Several studies have attempted to characterize the physical[63, 64, 65], mechanical[66, 67, 68], chemical[69,

[70, 71], and clinical implication[72, 73, 74] of the hybrid layer (Figure 4). However, many of its aspects are still not yet fully known or understood, such as the influence of the thickness of the hybrid layer on the bonding strength and the existence or not of chemical adhesion between collagen and the adhesive.

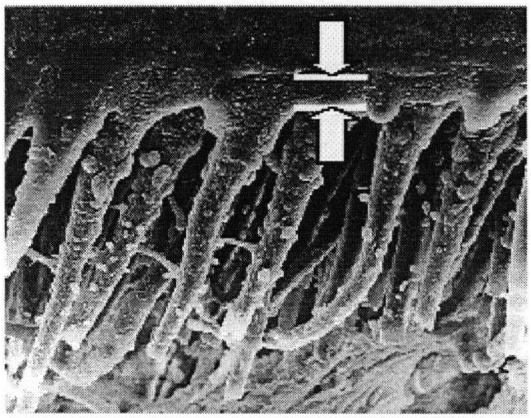


Figure 4 Schematic presentation of the hybrid layer (arrows).

Courtesy of Dr. Jorge Perdigao – School of Dentistry, University of North Carolina. Adapted from Craig, R.G., Powers, J.M., Restorative Dental Materials, 11th edition, 2002, Mosby, Inc.

The Challenges of Dentin Bonding

Adhesion to enamel is a predictable and established procedure, whereas a bonding to dentin is much less reliable. This is due partly to the high organic content of dentin, which, compared to enamel, is much less mineralized and has more water.[75]

Another important factor affecting the consistency of dentin bonding is the presence of the smear layer - a layer of debris created by the instruments used to mechanically cut

the tooth - which is able to avert adhesion of the bonding agents by preventing the material to mechanically interact with dentin.[76, 77]

The disappointing results obtained by the early adhesive systems is related to the fact that the smear layer was retained in order to protect the pulp against possible irritants, resulting in bonding of the adhesive to the surface of the smeared debris[78] as opposed to the dentin.[79]

The SEM micrograph in Figure 5A shows the presence of smear layer on dentin polished with #600 SiC paper, covering the dentinal tubules; Figure 5B shows the surface after treatment with a pre-conditioner, the dentinal tubules being opened and enlarged.

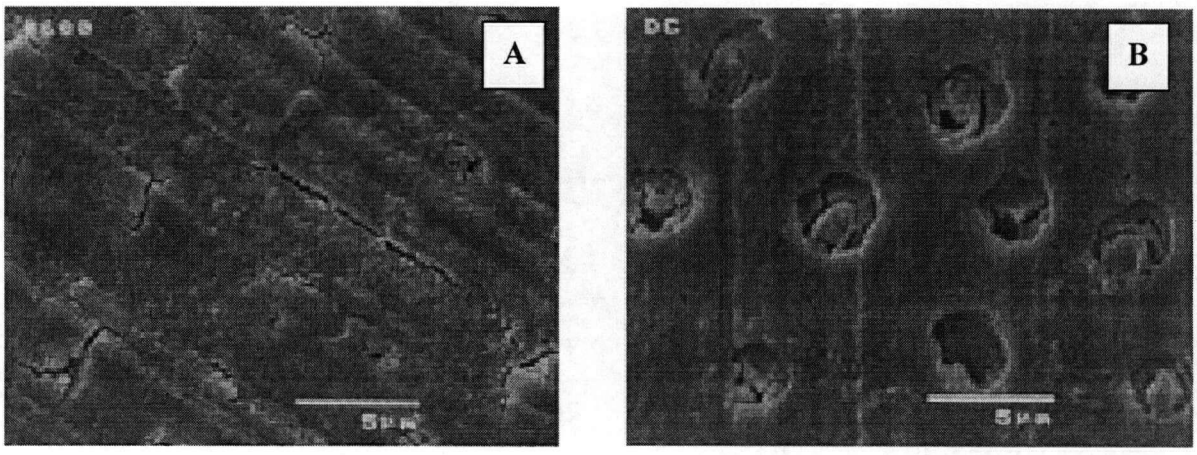


Figure 5 Scanning electron micrographs of dentin sliced perpendicularly to the dentinal tubules. A - polished with #600 SiC paper; B - treated with a dentin conditioner.

Dentin also undergoes changes with advancing age.[27] There is, for instance, an increase in the volume of dentin at the expense of pulp due to the gradual deposition over the years of secondary and tertiary dentin. Dentin that has been affected by caries or has undergone abrasion or erosion may be quite different from unaffected or sound dentin. Aged dentin experiences physiological dentinal sclerosis as part of the natural aging process and reactive sclerosis in response to slowly progressive or mild irritations, such

as mechanical abrasion.[80] Dentinal sclerosis, which occurs in the cervical area of teeth, is a consequence of the obstruction of dentinal tubules by apposition of peritubular dentin and precipitation of mineral crystals. As a consequence, it contains few, if any, patent tubules,[81, 82] has low permeability, and is relatively insensitive to external stimuli.[83, 84] The tubules of sclerotic dentin contain calcified odontoblastic processes and the orifices are occluded by crystalline material, which protect the pulpal structures against trauma from caries or surface abrasion. The intertubular dentin is also hypermineralized.

The effect of dentinal sclerosis on hybrid layer formation by a bonding agent was evaluated by Prati *et al.*[85] who found that sclerotic and old dentin showed a thinner hybrid layer with short resin tags and fewer lateral branches than normal dentin. Adhesive systems do retain resin composites effectively in sensitive, nonsclerotic erosion/abrasion lesions because the dentin in these areas contains open dentin tubules and less mineralized intertubular dentin. Resins flow easily into this type of dentin (after preconditioning) and provide maximum mechanical retention.[86, 87, 88, 85] However, all these structural and morphological transformations result in a dentinal substrate that is less receptive than normal dentin to adhesive treatments. [89, 90, 83]

Dental Bonding Systems

In order to accomplish the four main requirements for bonding described earlier in this chapter, dentin bonding systems comprise three basic steps: (1) **preconditioning** (etching); (2) **priming** (hydrophilic-hydrophobic monomer infiltration of etched surfaces); and (3) **bonding** (hydrophobic monomer connection of primed surfaces to composite restorations). Consequently, dentin bonding systems contain three main components: a **pre-conditioner**, a **primer**, and an **adhesive.**

The **pre-conditioner** could be a weak organic acid (e.g. 10% maleic acid), a stronger inorganic acid (e.g. 30% to 40% phosphoric acid), or a chelating agent (e.g. ethylenediamine tetra-acetic acid [EDTA]). Preconditioners are used to dissolve the enamel surface and/or demineralize the dentin, exposing collagen fibrils, changing its surface permeability by 4 to 9 times.[91]

Current pre-conditioners used in dental bonding systems employ two different means to initiate micromechanical interlocking by either altering or completely removing the smear layer. The smear layer is created by the instruments used to mechanically cut the tooth[92] and is of varying thickness (approximately 5 to 10 microns) and roughness.[93, 94, 95] The total etch technique, attempts to completely remove the smear layer by acid etching and rinsing. The self-etching technique dissolves and incorporates the smear layer into the bonding layer, bypassing the rinsing step associated with the total etch technique.

The **primer** is generally a bi-functional monomer that features a hydrophilic and a hydrophobic part in its molecule and is incorporated in a volatile solvent such as alcohol or acetone. Examples of bifunctional monomers include hydroxyethyl methacrylate (HEMA), *N*-methacryloyl-5-aminosalicylic acid (NMSA), *N*-phenylglycine (NPG), pyromellitic diethylmethacrylate (PMDM), and 4-methacryloxyethyl trimellitate anhydride (4-META).[96]

The main purpose of a primer in an adhesive system is to connect the hydrophilic dentin to the hydrophobic adhesive resin via its bi-functional behavior. Essentially, the hydrophilic aspect of the primer connects to the demineralized dentin and its hydrophobic end bonds to the adhesive resin. [96]

The **adhesive** (bonding resin) is an unfilled or partially filled, low viscosity, chemically or light-curable resin.[97] Typically, the adhesive resin contains predominantly hydrophobic dimethacrylate oligomers (Bis-GMA/TEGMA). The main function of the adhesive resin is to combine on one hand with the hydrophobic component of the monomer present in the primer, creating the hybrid layer and forming resin tags that seal the dentinal tubules, and on the other hand with the composite resin.

Bonding agents may also contain other components such as fluoride, antimicrobial ingredients, and desensitizing agents.

Development of Dental Bonding Systems

The earliest dentin bonding experiments were actually reported in the 1950s. For the next approximately 30 years, dentin bonding development advanced quite slowly. In the early 1980s, a better understanding of the effect of smear layer and hydrophilic monomer systems was achieved. By the late 1980s, dental bonding systems were available that were capable to provide shear bond strengths (SBS) to dentin equivalent to those obtained to enamel (in the range of 20 MPa).

Classification of Dental Bonding Systems

Bonding systems can be classified in several ways.[98, 99, 62] One of the most common classification - the "generational" classification system - is based on the chronological order of their appearance, with five to seven generations of bonding systems, depending on different authors' interpretations.[100, 101] This classification can be quite confusing and difficult to use.

A much simpler classification categorizes bonding systems based on the mechanism of bonding. This classification divides bonding systems into two main categories according to their interaction with the smear layer, as previously mentioned: **Total-etch systems** - bonding systems in which the pre-conditioning step is applied independently and thus completely removes the smear layer and demineralises the underlying dentinal surfaces; and **Self-etch systems** - bonding systems that intend to dissolve the smear layer rather then remove it, demineralise the underlying dentinal surfaces, and simultaneously prime the surface.[102]

Furthermore, this classification subdivides the bonding systems into four subcategories according to the number and combination of the dental adhesive components (preconditioner, primer, and adhesive resin) per bottle: total-etch multi-bottle, total-etch one bottle, self-etching two step, and self-etching one step (Table 2).

Table 2 Classification of bonding systems based on the mechanism of bonding and according to the number and combination of the dental adhesive components per bottle

Bonding System	Acid	Primer	Adhesive Resin
Total-etch multi bottle adhesives	1 bottle	1 bottle	1 bottle
Total-etch one bottle adhesive	1 bottle	1 bottle	
Self-etching two step	1 bottle		1 bottle
Self-etching one step	1 bottle		

Total etch multi-bottle adhesive systems (e.g. ScotchBond MP, All Bond 2, Optibond FL) provide the acid, the primer and the adhesive resin in separate bottles. The bonding process is carried out in three distinct steps. Firstly, the acid gel is applied to the surface for a few (generally 15) seconds. The gel is rinsed and the surface is air dried in order to remove excess water. The primer is then applied and dried gently. The adhesive resin is subsequently applied and light cured. Compared to the other systems, total etch multi-bottle adhesive systems produce the strongest and most durable bonds to dentin.[103] However, since the product is dispensed from several bottles, the bonding process is clinically cumbersome and time consuming.[104]

Total etch one-bottle bonding systems (e.g. Prime & Bond NT, One Step, Single Bond) have the acid in one bottle and the primer and adhesive resin together in another bottle. The bonding process involves two distinct steps: firstly, the etching gel is applied to the surface for a few (generally 15) seconds, followed by rinsing and the surface is air dried in order to remove excess water, cautiously leaving the substrate moist. Subsequently, the combination primer/adhesive resin is applied onto the moist surface and left undisturbed for a few seconds. The surface is air-dried in order to remove the solvent and the water, and the primer-adhesive is light-cured.

Laboratory researches support the use of total-etch one-bottle systems on enamel and dentin and clinical studies up to 3 years show positive results. However, total etch one-bottle systems are very technique sensitive since the quantity of moisture on the substrate highly influences the strength of the adhesion.[105]

The **self-etching primer two-step** bonding systems (e.g. Clearfil SE Bond, Clearfil Liner, Bond 2V) were introduced to the dental marked in an attempt to satisfy the need

for simpler and quicker adhesive procedures for dentists. The process, therefore, was simplified to allow for fewer bonding steps by using a primer containing an acidic monomer in one bottle and an adhesive resin in another. The bonding process is also accomplished in two distinct steps. The acidic primer is applied to the entire substrate surface. After a few seconds, the surface is dried with a mild airflow and the bonding resin is applied. The surface is dried once more with a gentle airflow and subsequently light-cured. It should be highlighted that there is no rinsing involved after the placement of the self-etching primer, a substantial advantage of these systems.

It has been reported that self-etching two-step systems produce less postoperative sensitivity.[106] Clinical studies confirm their effectiveness on the dentin of young adults, but, as of yet, their effectiveness on sclerotic dentin is unknown. They are more prone to cause microleakage in restorations placed on enamel due to insufficient enamel etch and slight degradation of the hybrid layer.[74]

The **self-etching one-step** bonding systems (e.g. Adper Prompt L-Pop, One-Up Bond) are the most recent result of the effort to simplify and reduce the clinical time spent on adhesive procedures even more. The systems are dispensed from a disposable container and the procedure involves only one step, which speeds up the clinical process. The product is applied onto the entire surface with an applicator. The solution is being rubbed against the substrate with moderate pressure. Scrubbing the material onto enamel and dentin is essential in order for the acid to properly alter the surface of the enamel/dentin. Afterwards, a gentle stream of air is used to dry thoroughly the adhesive to a thin layer, which is then light-cured. Self-etching one-step systems do not bond well to unprepared enamel and the strength of the bonds produced is very inconsistent.[107]

Chapter 4: Fracture Toughness

Background

Assessing the bonding strength of restorative materials has been the objective of numerous investigations over the years. Indeed, it dates back to the mid 1950's when Buonocore *et al.*[108] estimated the quantity of force necessary to dislocate composite resin from dentin.

Conventionally, the bond strength of restorative materials has been evaluated by tensile or shear bond tests. The drawback of these traditional methods is that they show unequally distributed forces along the resin-composite-dentin interface (Figure 6).[109] Furthermore, the distribution of stresses along the interface is dependent on the geometrical structure of the test, the loading configuration, and the mechanical properties of the materials.[109] Another frequent criticism of these conventional tests is their reliability, since great discrepancies in test results have been observed.[110]

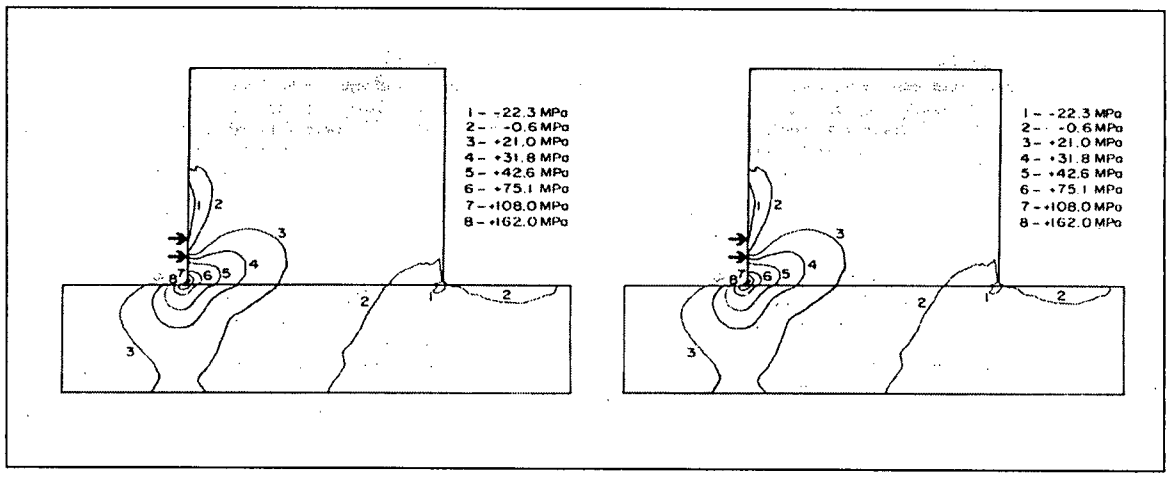


Figure 6 Unequally distribution of forces along the resin-composite-dentin interface in tensile (A) and shear (B) bond tests (from van Noort et al.).[109]

Alternatively, fracture toughness (K_{IC}) tests based on the theory of fracture mechanics have been applied to a number of problems in dentistry. Fracture toughness tests address the performance of materials subjected to stresses and strains in the presence of cracks and flaws, which are normally present in materials.[111, 112] Typically, cracks and flaws weaken a material over time to a point where a catastrophic fracture occurs even below the material's yield strength, which is the stress level where a material yields or permanently deforms.[113]

Principles

Values of K_{IC} are typically reported as the critical stress intensity coefficient (K_{IC}) or stain energy release rate (G_{IC}) which specify the critical intensity of stress or energy after which a crack or flaw of a critical dimension will propagate.[114] The initial point in both analysis is a through-thickness elliptical crack of length 2a (where "a" is one-half the major axis of the ellipse) present in the centre of a solid body subjected to tensile stress (Figure 7).

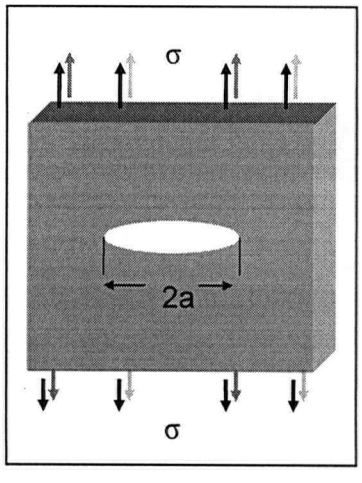
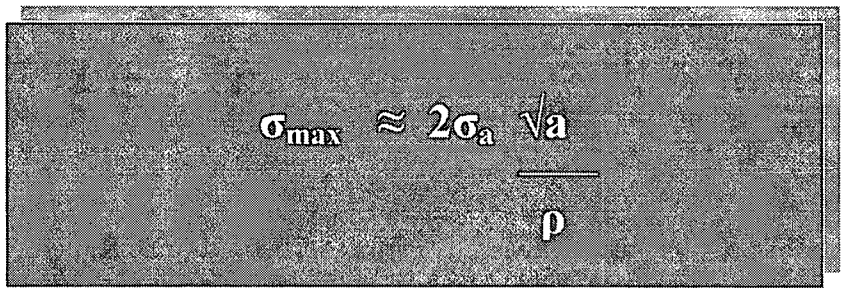


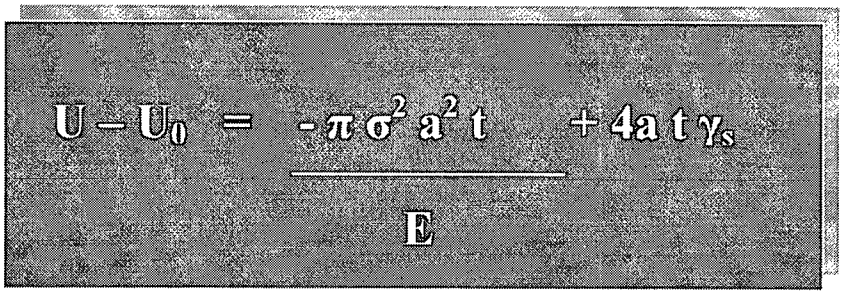
Figure 7 A through-thickness crack of length 2a present in a solid body that is subjected to uniform tensile stress (σ).

Inglis,[115] in 1913, suggested that the solid body subjected to tension does not observe stress uniformly. The maximum stress (σ_{max}) in this situation is present at the two crack-tips on both side of the ellipse and is associated with the applied stress (σ_a), one-half crack length (a), and the crack tip radius (ρ) by the formula:



The term "$2\sigma_a \sqrt{a}/ \rho$" is also known as the stress concentration factor (k_t,) and depicts the influence of the crack geometry on the values of stresses present at the crack tip.[116] The longer the long axis of the ellipse in relation to its minor axis and the longer the crack length, the higher the stress concentration will be at the crack tip.

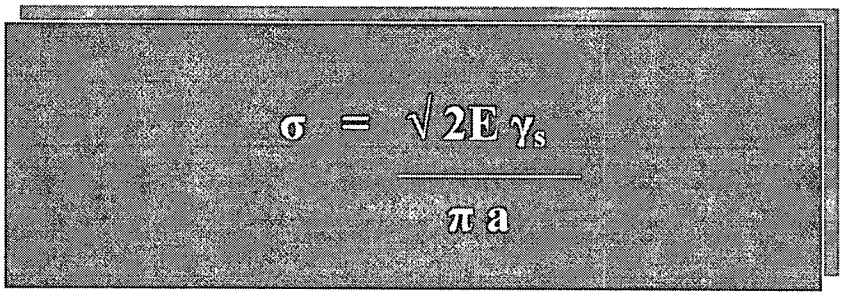
Griffith,[117] in 1920, was the first to propose an equation portraying the relationship between the applied nominal stress and the length of the crack length at fracture, i.e. when it becomes energetically favourable for a crack to grow. He analyzed fractures based on the energy balance between the energy applied to expand the crack and the surface energy resulting from the crack developing. His analysis is summarized by the following equation:



$$U - U_0 = \frac{-\pi \sigma^2 a^2 t}{E} + 4a t \gamma_s$$

Where U = Potential energy of body with crack; U_0 = Potential energy of body without crack; σ = applied stress; a = one-half crack length; t = thickness; E = Modulus of elasticity; γ_s = specific surface energy.

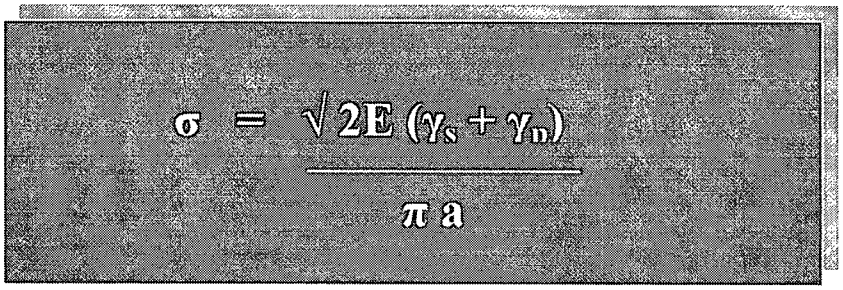
By discriminating the energy difference ($U - U_0$) in relation to the length of the crack, Griffith generated the following equation relating stress to an existing crack length at equilibrium:



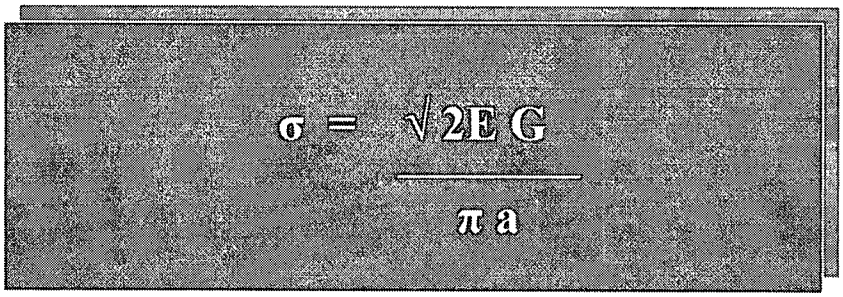
$$\sigma = \frac{\sqrt{2E\gamma_s}}{\pi a}$$

Griffith's analysis, while extremely significant, was based on tests performed on elastic materials in the presence of a very sharp crack ignoring ductile materials in its consideration and was not appropriate for plastically deforming materials such as polymers and metals. Orowan and Irwin[118, 119] determined that there was also a certain

energy from plastic deformation that had to be added to the strain energy originally considered by Griffith. Therefore, the original expression was modified to include the energy of plastic deformation (γ_p):



Irwin also proposed the term dU/da for the amount of elastic energy needed to increase a crack by an amount "da", also expressed as G, a term that corresponds to the elastic energy release rate ($J/m^{½}$),[119] which is then added to the previous equation:



From the two previous equations, it can be noted that the elastic energy release rate (G) is in reality describing the sum of the specific surface energy (γ_s) and the plastic deformation energy (γ_p) and can be expressed as:

$$G = 2(\gamma_s + \gamma_p)$$

As strain increases in the material, a catastrophic fracture occurs when a point of instability is achieved as G reaches the critical value G_c, which can be estimated by:

$$G_C = \frac{(P_{max})^2}{2} \frac{dC}{da}$$

Where P_{max} is the maximum load recorded during test and dC/da is the rate of change of compliance with respect to crack length and depends on specimen geometry.

Irwin also developed an alternative approach to describe crack initiation and propagation based on an analysis of the stress - identified as the "K" approach.[120, 116] By applying the coordinate system depicted on Figure 8, the "K" approach defines the degree to which the stress is concentrated at the crack tip by using a three-dimensional analysis of the stresses at the crack tip and two straightforward parameters: radius (r) and angle (θ), according to the formulae:

$$\sigma_Y = \frac{K}{\sqrt{2\pi r}} \cos\frac{\theta}{2}\left(1 + \sin\frac{\theta}{2}\sin\frac{3\theta}{2}\right)$$

$$\sigma_x = \frac{K}{\sqrt{2\pi r}} \cos\frac{\theta}{2}\left(1 + \sin\frac{\theta}{2}\sin\frac{3\theta}{2}\right)$$

$$\sigma_{xY} = \frac{K}{\sqrt{2\pi r}} \cos\frac{\theta}{2}\left(1 + \sin\frac{\theta}{2}\sin\frac{3\theta}{2}\right)$$

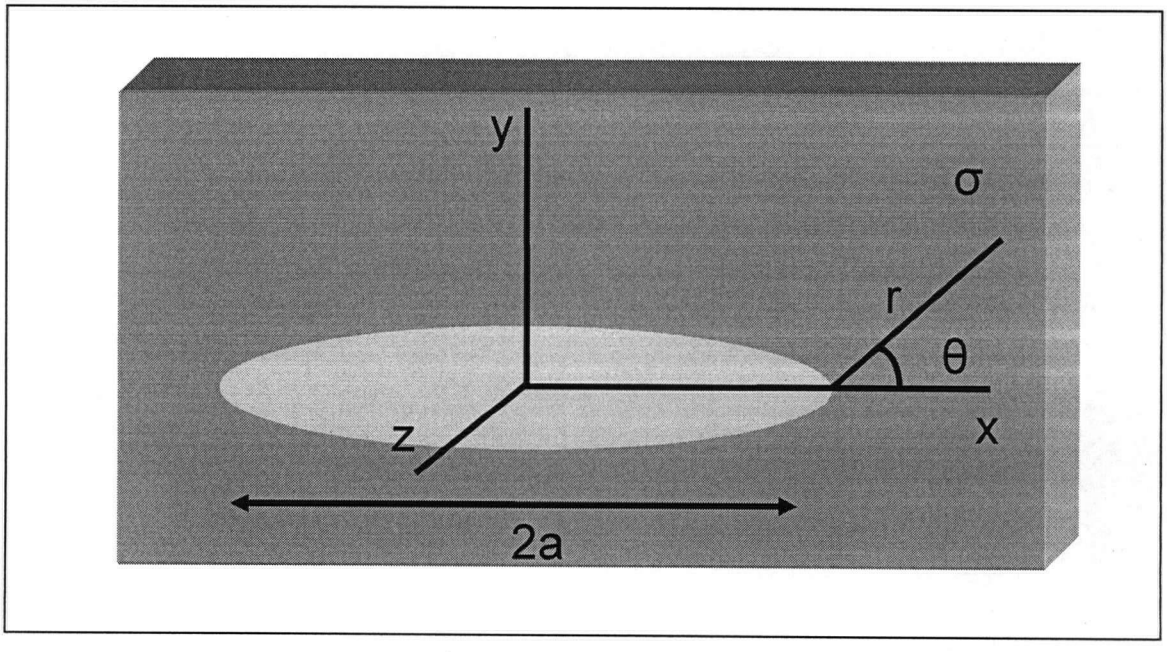


Figure 8 Quantification of the stresses present at a crack tip and its vicinity by using a coordinate system approach with two parameters: radius (r) and angle (θ).

Chevron Notch Fracture Toughness Test

A commonly used method to evaluate the fracture toughness of materials is the chevron notch short rod (CNSR) fracture toughness test developed by Barker[121, 122, 123] by which a tensile force is applied onto a cylindrical-type specimen with a chevron notch (Figure 9).

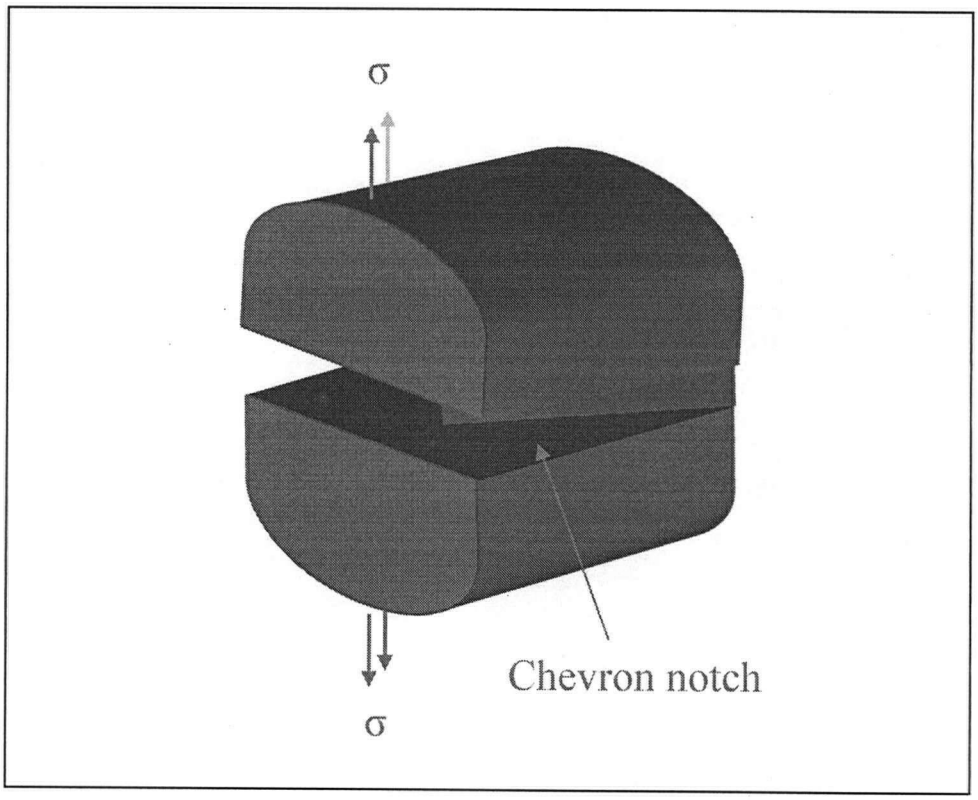
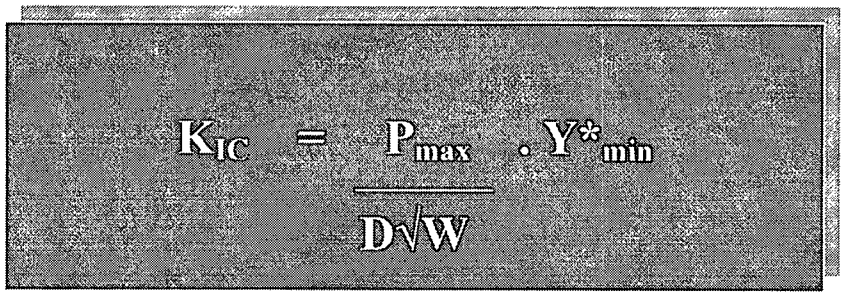


Figure 9 A chevron notch short rod specimen subjected to a uniform stress (σ).

The CNSR fracture toughness test employs the maximum load at fracture (P_{max}) to calculate the fracture toughness (K_{IC}) using the expression below proposed by Barker and adopted by the American Society for Testing and Materials (ASTM) standard E1304-89:



$$K_{IC} = \frac{P_{max}}{D\sqrt{W}} \cdot Y^*_{min}$$

Where P_{max} = maximum load recorded at fracture; D = specimen diameter; W = specimen width; Y^*_{min} = the minimum dimensionless stress intensity coefficient, a specimen configuration related constant

Barker[121, 122] tested the validity of this approach by confirming that the fracture toughness of fused quartz, siltstone rock, aluminium alloy and poly-methylmethacrylate established by his test, agreed with K_{IC} values for the same materials tested by other methods. Koblitz *et al.*[124] found similar K_{IC} values for poly-methylmethacrylate and a dental composite resin (Adaptic™, J&J New Jersey, USA) when applying the short-rod fracture test on specimens with dimensions similar to those of a dental restoration.

Since then, the chevron notch short rod test has been used to identify the fracture toughness of a variety of dental materials. However, very few studies using this method have been conducted to investigate the fracture toughness of the interface between a composite resin and dentin[125, 59] since the specimens are complex and expensive to make and the testing process is difficult to control as the cut of the chevron notch may result in specimen fracture during cutting in the case of very low fracture toughness materials.

Notchless Triangular Prism Specimen Fracture Toughness Test

Ruse *et al.*[126] developed a method for determining the fracture toughness of materials and adhesive interfaces in 1996. While the theoretical concepts behind the notchless triangular prism (NTP) and the specimen geometry were developed based on the standardized CNSR fracture toughness test, the difficulties involving the sample making process were overcome.

Essentially, the test uses a 4 x 4 x 4 x 8 mm specimen (Figure 10) griped by two stainless steel holders (Figure 11). The new test eliminates the need to create a chevron notch by using spacers or blades in order to separate the two stainless steel halves producing a gap similar to the gap in the chevron notch short rod test (Figure 12). The final specimen (NTP specimen secured by the specimen holder) is equivalent to the CNSR test specimen (Figure 13).

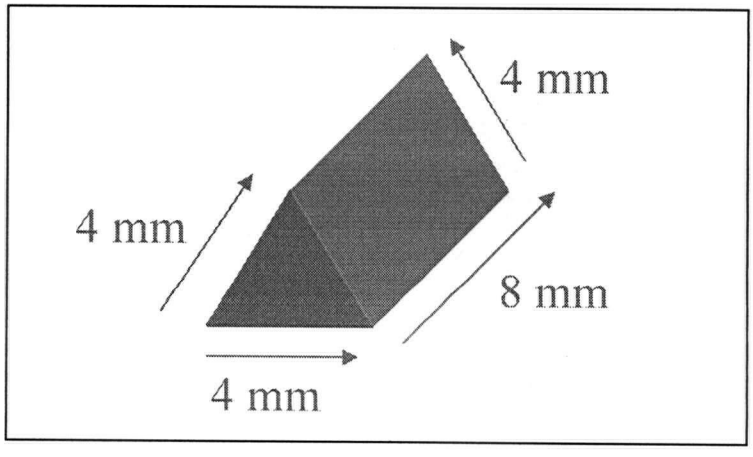


Figure 10 Schematic representation of the Notchless Triangular Prism (NTP) specimen.

A.

B

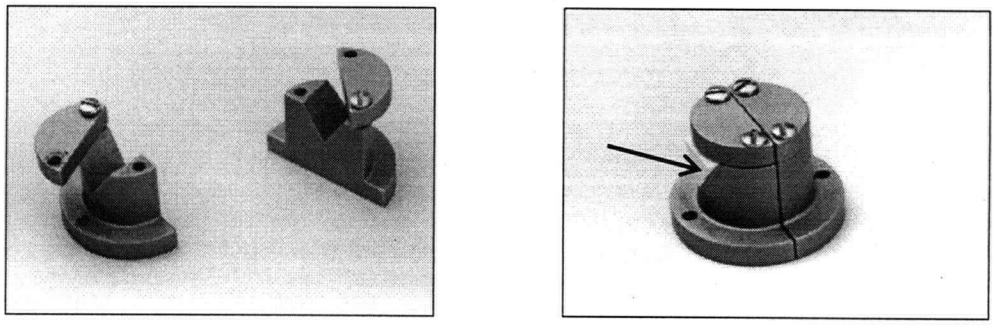


Figure 11 Metal jigs used in the testing of the notchless triangular prism (A). The NTP specimen (arrow) being secured by the two testing jigs (B).

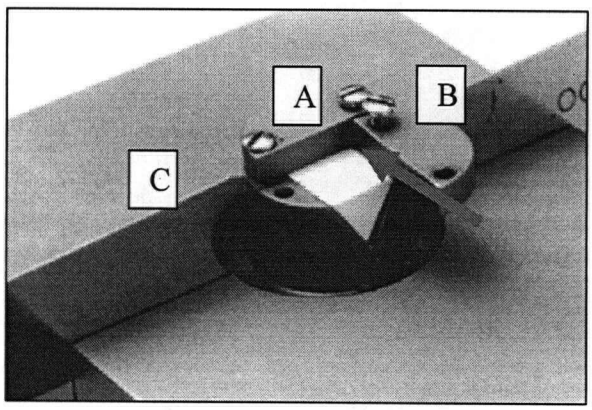


Figure 12 The process of placing the specimen into the metal jigs. A. First jig containing half of the specimen. B. Second jig containing the other half of the specimen identified by the arrow. C. 200 micron thick spacer on the surface of the dentin before mounting the second jig.

A.

B.

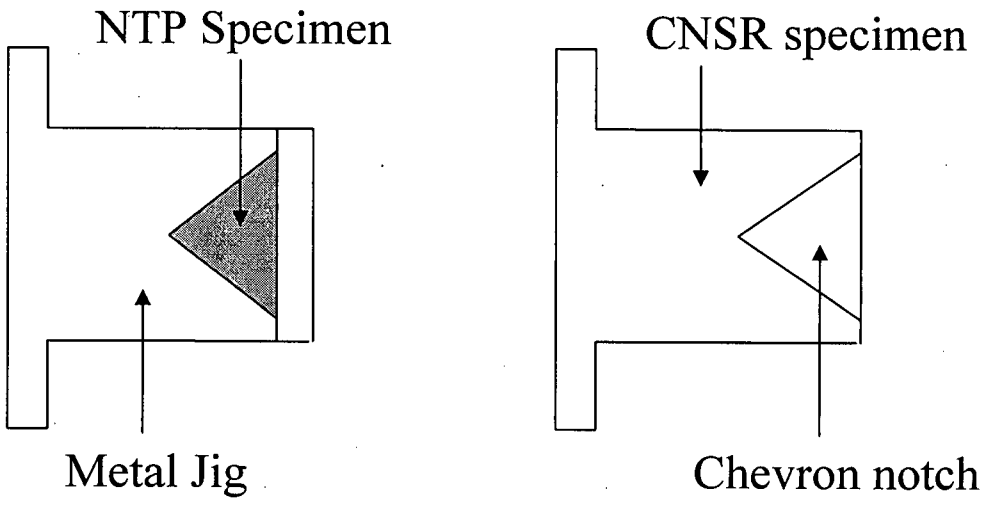


Figure 13 Schematic representation depicting the position of the NTP specimen in the holder (A) and comparing with the configuration of a CSNR specimen (B).

Validity of the Notchless Triangular Prism Test

A valid fracture toughness test distributes the stress evenly along the interfaces. According to Brown and Strawley,[116] in order for a fracture toughness test to fulfill this premise, the thickness of the specimen (D) should be determined by the formula:

$$D \geq 2.5 \{ K_{IC} / \sigma_{ys} \}^2$$

Where K_{IC} = stress intensity coefficient determined by the test; σ_{ys} = the 0.2% offset yield strength of the material in the direction of loading.

The last part of this equation ($\{K_{IC}/\sigma_{ys}\}^2$) approximates the size of the plastic (permanent deformation) zone, consequently the test is influenced strongly by the relationship between the general dimensions of the plastic zone and the thickness of the specimen. If the thickness of the specimen is greater than the thickness of the plastic zone, the K_{IC} value will correspond to the sum of all the stresses focused at the crack tip.[120]

While Iwamoto and Ruse[127] assessed the fracture toughness of dentin and found that K_{IC} values vary from 1-2 MPa·m$^{1/2}$according to the orientation of dentinal tubules, Mowafi and Watts reported values greater than 3.[128]. Since dentin has a 0.2% yield strength value of 200 MPa·m$^{1/2}$, the minimum specimen dimension required for a fracture toughness test is 0.25mm, which is reasonably well below the 8mm thickness of the NTP specimen.

Fracture toughness of dental composites varies between 1.18 and 1.95 MPa·m$^{1/2}$. [129] Since the yield strength values of these materials vary between 35-80 MPa/m$^{1/2}$, the minimal dimensions of a specimen required for valid test should be 1.48mm[130].

Although this dimensional principle can be applied to assess the validity of the chevron notch and NTP fracture toughness test for homogeneous materials, we do not know how it applies to a non-homogeneous surface, since the principles established by Brown and Strawley were based on stresses applied to homogeneous materials and thus the matter requires additional investigations.

Finite element analysis (FEA) was also used to validate the NTP fracture toughness test.[126] The NTP specimen and the specimen holder modeled using the solid modeling module of I-DEAS software (Structural Dynamics Research Corporation,

Milford, OH). The stress distribution demonstrated tensile stresses concentrated in the area equivalent to the tip of the chevron notch.

In addition to validating the NTP fracture test through finite element analysis, Ruse and coworkers compared fracture toughness test results for selected dental materials to values found on the literature and reported a good correlation.[126]

Accuracy of the Notchless Triangular Prism Test

The NTP test and the chevron notch test use the same standardized formula to calculate K_{IC} The estimation of K_{IC} values using the NTP test requires simply the peak load needed to fracture the specimen. [123, 126] (Figure 14).

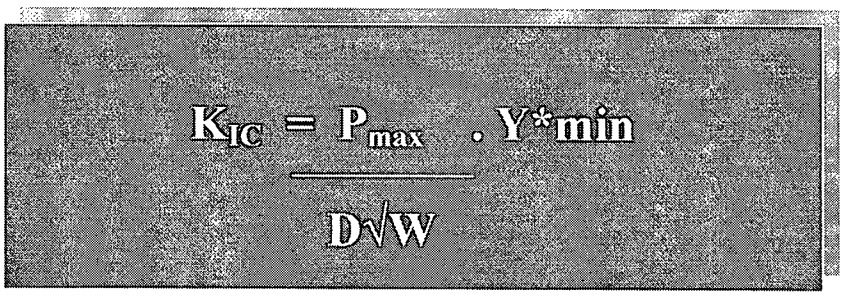


Figure 14 Formula used to calculate the fracture toughness. Where P_{max} = maximum force recorded during loading; D = specimen diameter; W = specimen width; Y*min = minimum dimensionless stress intensity factor.

There are, however, some reservations about the closeness of the calculated K_{IC} to the actual value because the value established for the dimensionless stress intensity coefficient (Y*min) may introduce a systematic error. Y*min, is a constant whose value alters according to the specimen's geometrical structure. The value established for Y*min in the NTP was obtained by Ruse *et al.*[126] through linear and logarithmic extrapolation of values proposed by Bubsey *et al.*.[123] The uncertainty of this extrapolation is estimated at

less than 10%[126]; therefore, the K_{IC} value obtained using the NTP test should be very close to the true K_{IC} value.[126]

Precision of the Notchless Triangular Prism Test

Ruse *et al.*[126] compared the fracture toughness of poly-methylmethacrylate obtained by the NTP test with those obtained using three different chevron notch geometries (Table1). The results demonstrated that both tests were uniformly precise in repeated measurements of the same materials, and that they revealed the accuracy of the NTP test as all conditions apart from the test method were alike.

Table 3 K_{IC} (in MPa·m$^{1/2}$) of poly-methylmethacrylate obtained by NTP and CNSR tests

Sample	W/D	α_0	K_{IC}
CNSR*	1.23	0.27	1.15 ± .15
CNSR*	1.63	0.518	1.08 ± .13
CNSR*	1.75	0.552	1.15 ± .12
NTP•	0.88	0.5	1.03 ± .15

• NTP = Notchless triangular prism test; * CNSR = Chevron notch short rod test
W/D = length to diameter ratio; α_0 = crack length to specimen length ratio; ; K_{IC} = critical stress intensity factor (fracture toughness)

The problem of comparing the precision of both techniques (NTP *vs* CNSR) for evaluating bonded interfaces is due to the scarcity of data comparing the tests under the same conditions. Tam and Pilliar,[125] using the chevron notch short rod test, reported K_{IC} values of 0.34 ± 2.1 MPa·m$^{1/2}$ from Scotchbond MP (3M ESPE, St. Paul, MN, USA), whereas Ruse and Feduik[131] reported values of 0.50 ± 1.4 MPa·m$^{1/2}$ from the same

material. Further tests are needed, therefore, to determine the fracture toughness of various bonding agents under equal conditions using both the NTP and CNSR tests.

Relatively few studies have reported on the fracture toughness of interfaces between two dissimilar materials, and in particular composite-dentin. Moreover, there have been no studies to date that have investigated the fracture toughness of the interface between different bonding agents and young and old dentin.

Chapter 5: Bonding Resin Composite to Old and Young Dentin

Purpose and Objectives

A variety of studies have explored and addressed the issue of root caries focusing mostly on the problems of pathologenesis[132, 13] and microbiology,[133] prevalence and incidence,[17, 134, 19] and estimating risk factors and treatment needs[20, 21] However, very few studies have examined and compared the behaviour of different bonding agents used for restoring primary root lesions in young and old dentin,[135, 136] and the studies that have been performed are about 10 years old. Since then, several new and presumably improved adhesive systems have become available. Therefore, the aim of this study was to evaluate the effect of the age of dentin on the bond strength of four different adhesive systems by comparing their performance on dentin obtained from "young" (not older than 45 years) and "old" (not younger than 65 years) patients.

Hypotheses

Since the physiologic and morphologic transformations that dentin undergoes as a consequence of aging may produce a dentinal substrate that shows different behavior in relation to adhesion of bonding agents [83, 89, 90] the following hypothesis was tested:

H_0: Fracture toughness of the dentin/restorative material interface is not affected by the age of the tooth.

H_a: Fracture toughness of the dentin/restorative material interface is affected by the age of the tooth.

Moreover, since each adhesive system has distinct chemical, mechanical and adhesive properties, the following hypothesis was tested as well:

H_0: The behaviour of the different adhesive systems is the same in relation to old and young dentin.

H_a: The behaviour of the different adhesive systems is different in relation to old and young dentin.

Materials and Methods

Forty teeth from patients not older than 45 years ("young teeth") and forty teeth from patients not younger than 65 years ("old teeth") were collected in Vancouver. The teeth were cleaned gently of all soft gingival and periodontal tissues with a periodontal hand instrument. They were then stored in tap water at 4°C for up to six months, in accordance with the guidance on testing adhesion to teeth described in ISO/DTS 11405.[137] Both the "young" and "old" teeth were assigned randomly to four experimental groups each according to the adhesive system used.

The interfacial K_{IC} of the four bonding systems to young and old dentin (Table 4) was determined using the notchless triangular prism (NTP) fracture toughness test, introduced by Ruse *et al.*[126] based on the standardized Chevron notch short rod (CNSR) test (ASTM E1304-89) developed by Barker.[138, 123]

Table 4 Bonding agents used to test the bond-strength to young and old dentin

Material	Classification	Manufacturer
Scotchbond MP	Total etch multi-bottle	3M ESPE, St. Paul, MN, USA
Prime & Bond NT	Total etch one bottle	Dentisply, York, PA, USA
Clearfil SE	Self-etching two step	Kuraray Co., Osaka, Japan
Adper Prompt L-Pop	Self-etching one step	3M ESPE, St. Paul, MN, USA

Specimen Preparation

The dentinal segment of the specimen

Prior to specimen preparation, the buccal surface of teeth was chosen to be used for bonding; consequently, the dentinal aspects of the specimen were prepared in such a fashion that the long axis of the specimen was oriented in the bucco-lingual direction revealing the buccal dentin for bonding. Equilateral triangles were drawn on the buccal and lingual aspects of each tooth, with the base oriented towards the occlusal plane (Figure 15). Subsequently, the roots as well as the majority of the occlusal enamel were eliminated with 240 grit SiC sandpaper (Buehler) mounted on a wet grinding machine (Buehler). The outline of the drawn triangle was then used to help place two cuts at approximately 60° to the occlusal surface. These cuts produced a roughly triangular prism that was oriented in the direction previously described.

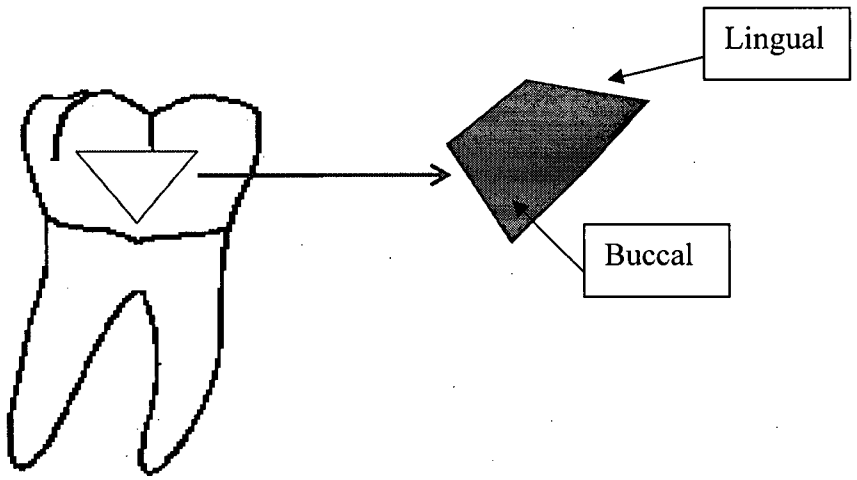


Figure 15 Equilateral triangle drawn on teeth, which oriented the shape of the dentinal aspect of the specimen.

Subsequently, the triangular prism was placed into a custom-made holder and the long axes of the prism were continually rotated on the grinding machine in order to reduce the dimensions uniformly. Once the proportions of the triangular prism were close to the required design (≈ 4.5 x 4.5 x 4.5 mm), 600 grit SiC sand paper (Buehler) was used until all three sides of the prism were 4mm in length. The dentinal prisms were stored in tap water at 37°C prior to bonding.

Bonding[1]

Scotchbond MP

The etching gel was applied to the dentin surface for 15 seconds, after which the surface was rinsed with water and dried with compressed air, as in a clinical context, for two seconds to remove excess water. The primer was applied to the surface, which was dried again with air for five seconds; the resin was applied and light cured for 10 seconds (Coltolux 4, Coltene).

Prime & Bond NT

The etching gel was applied to the surface for 15 seconds, after which the surface was rinsed with water and dried with compressed air, as in a clinical context, for two seconds to remove excess water. The bonding agent was applied to the surface of the specimen and left undisturbed for 30 seconds. The surface was then dried with an airflow for a few seconds before light curing for 20 seconds (Coltolux 4, Coltene).

Clearfil SE Bond

The self etching primer was applied to the entire dentinal surface. After 20 seconds, the surface was dried with an airflow and the bonding resin was applied. The surface was

1 All bonding systems were applied according to the manufacture specifications.

dried again with an airflow and subsequently light cured for 10 seconds (Coltolux 4, Coltene).

Adper Prompt L pop

The self etching primer adhesive was applied to the entire surface and the applicator containing the solution was rubbed against the dentin with moderate finger pressure for 15 seconds, as requested by the manufacture. Afterwards, the adhesive was dried with an airflow to a thin layer, which was light cured for 10 seconds (Coltolux 4, Coltene).

Composite Build-Up

A reverse impression mould of a 4 x 4 x 4 x 8 mm triangular prism (Figure 16) was made to enable the packing of the composite restorative material (Z 100 - 3M ESPE, St. Paul, MN, USA) onto the bonding agent treated dentin surface. Bonding agent treated dentin specimens were placed in the reverse mould and the composite resin was added in small increments and light cured for 40 seconds (Coltolux 4, Coltene). The final specimen was an 8mm long triangular prism with equilateral (4mmx4mmx4mm) sides consisting of dentin and composite resin linked by a thin layer of adhesive (Figure 17). Completed specimens were stored in tap water at 37°C for 1 week to allow water sorption by the composite resin to counter the effects of polymerization shrinkage.[139] Final preparation of the dentin/composite specimens was achieved by grinding under water with 600 grit sandpaper.

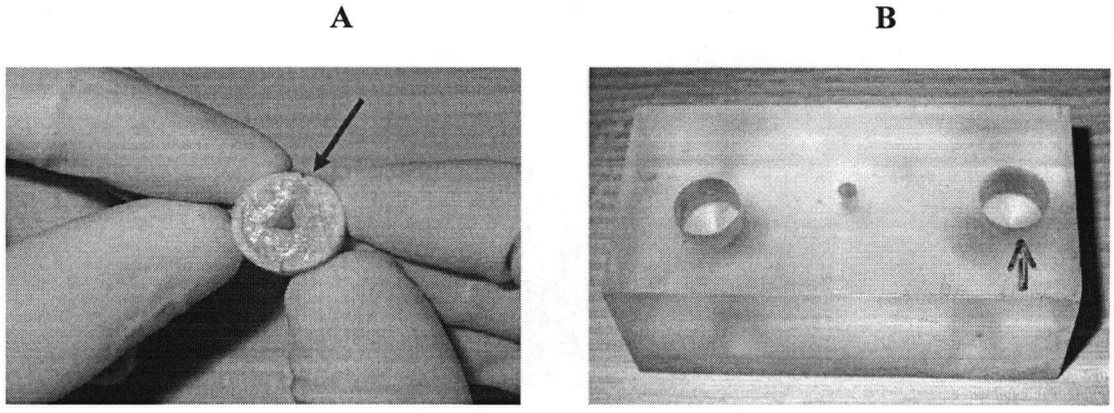


Figure 16 A. The reverse impression mould (arrow) of a triangular prism that was built to pack the restorative composite into the bonding agent. B. Base that was used to secure the reverse impression mould (arrow), which helped the process of packing the composite resin.

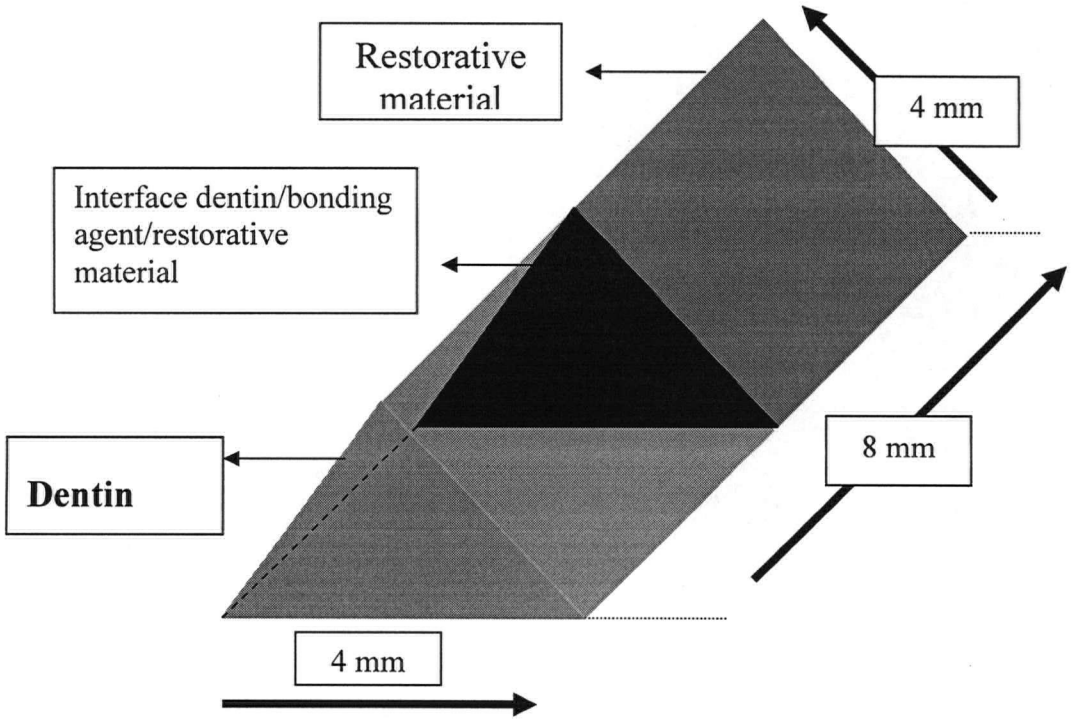


Figure 17 Shape and dimensions of the resin composite-dentin specimens for the NTP K_{IC} test.

Fracture Toughness Testing

Each specimen was mounted in the NTP specimen holder with the crack tip located in deep (pulpal) dentin. Initially, the dentinal half of the specimen was secured in the holder (Figure 28) with the adhesive interface located about 100 μm outside the testing jig. The procedure was conducted while viewing under 16-fold magnification stereomicroscope.

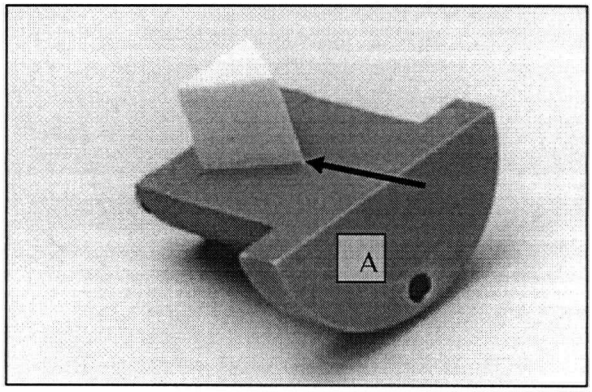


Figure 18 Final specimen being secured in one half of a notchless triangular prism testing jig (A) with the adhesive interface located about 100 μm outside the testing jig (arrow).

An approximately 100 μm deep cut was placed at the interface under 16-fold magnification to act as a crack initiator, and a 200 μm metal spacer was held against the dentin before mounting the second half of the specimen holder. The spacer held the two jigs 200 μm apart (Figure 19) with the initiating crack in the middle of the space.

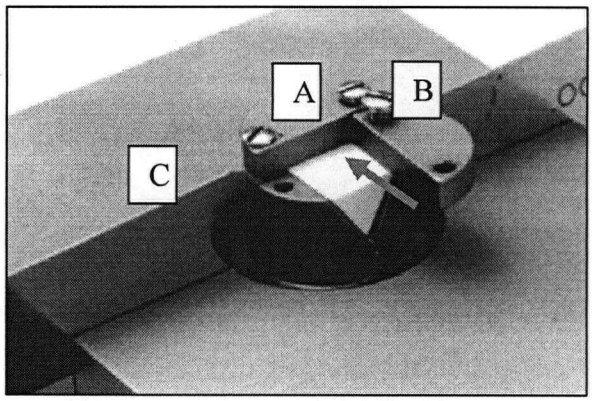


Figure 19 The process of placing the specimen into the metal jigs. A. First half of the specimen holder containing half of the specimen. B. Second half of the specimen holder containing the other half of the specimen, identified by the arrow. C. 200 μm thick spacer on the surface of the dentin before mounting the second jig.

Finally, the second half of the specimen holder was fastened creating a complex - metal jigs / final specimen (Figure 20) that was then mounted onto an Instron 4301 Universal Testing Machine (Instron Canada, Canada). The samples were loaded in tension with a cross-head speed of 0.1mm/min (Figure 21). The maximum load recorded at fracture was used to calculate the critical stress intensity factor (K_{IC}).

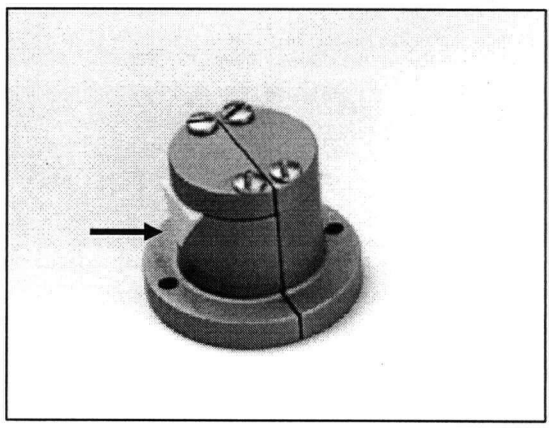


Figure 20 Complex of metal jigs (specimen holder) and dentin/ bonding agent/composite resin specimen (arrow) that is set to be tested.

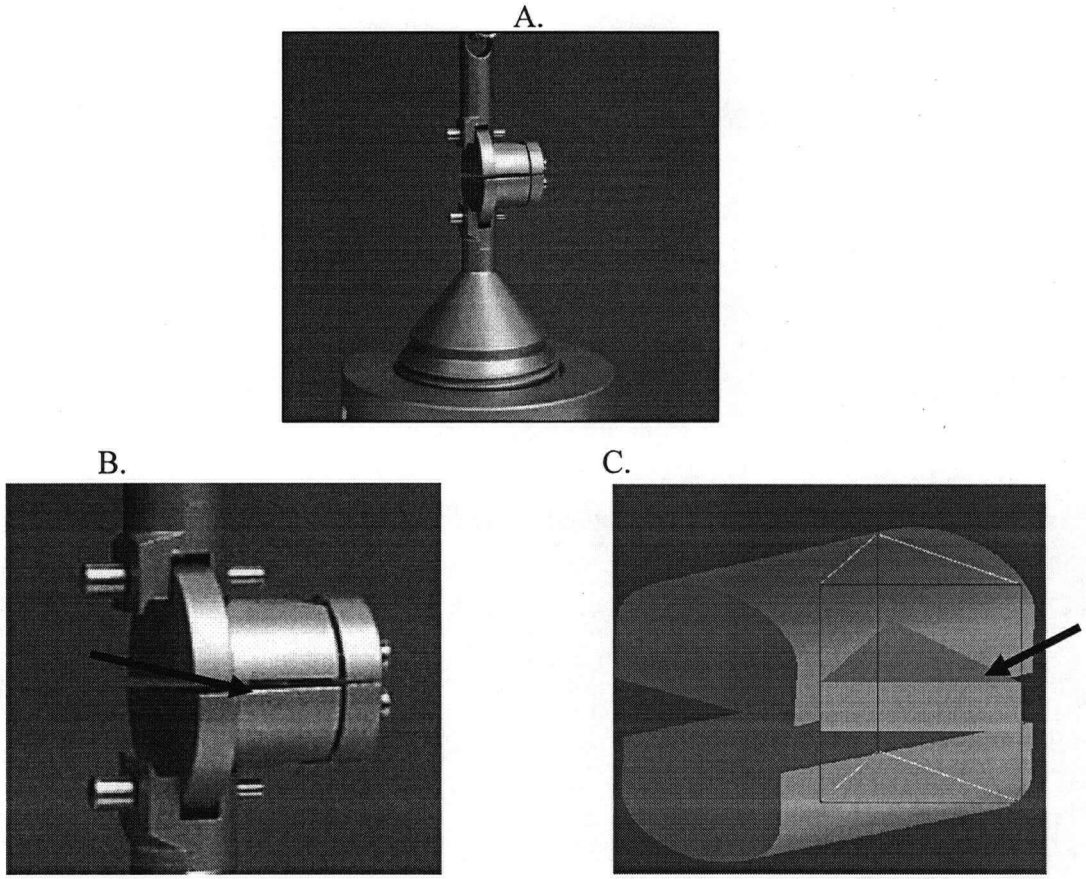


Figure 21 A. An assembled jig with test specimen mounted on an Instron 4301 Universal Testing Machine (Instron Canada, Canada). B. Closer view of the assembled jig (arrow) C. Schematic view of the assembled jig showing the position of the triangular prism (arrow) in relation to jig itself.

Scanning Electron Microscope Observations

Following the fracture toughness testing, the samples that were considered to be representative of each group, based on the determined K_{IC} means, were chosen for scanning electron microscope characterization. The dentinal bonded surface of the fractured specimens was "sputter coated" with gold (Hummer VI, Technics) and examined with a scanning electron microscope (StereoScan 260, Cambridge). Photographs were taken at x25, x50, x500, and in some cases at x1500, and compared for surfaces characteristics.

Statistical Analysis

SPSS statistical software was used to analyze the data. The general linear model (GLM - Type III Sum of Squares) was employed to carry out a two-factor univariate analysis of variance (ANOVA) on the effect of age and adhesive system. The Bonferroni multiple means comparison test was used for multiple mean analysis. A significance level of 0.05 was used for all tests.[140]

Some common statistical procedures assume that variances across samples are equal and Levene's test was used to examine this assumption.

Chapter 6: Scanning Electron Microscopy

One of the most extensively used equipment in science, the scanning electron microscope (SEM) allows for the study of the morphology and composition of organic and inorganic specimens. Although a microscopic characterization is difficult to translate into numeric data, images from the SEM yield qualitative information about a specimen.[141] In fracture toughness testing, for instance, analyses of the images obtained by SEM can establish fracture patterns by determining if the fracture occurred cohesively, adhesively, or if a mixed pattern occurred.

Essentially, SEM is able to produce high resolution images of the morphology of a specimen, with vast depth of field at extremely low or extremely high magnification by scanning an electron beam across the specimen.[142] The beam of electrons is formed at the top of the SEM by heating of a metallic filament. Subsequently, the electrons follow a vertical path through the column of the microscope passing through electromagnetic lenses, which have the function of focusing and orientating the beam down towards the specimen. Once the electron beam contacts the specimen, electrons are scattered around and subsequently collected by detectors, which in turn translate them into a signal that is sent to a screen producing an image (Figure 22).[143]

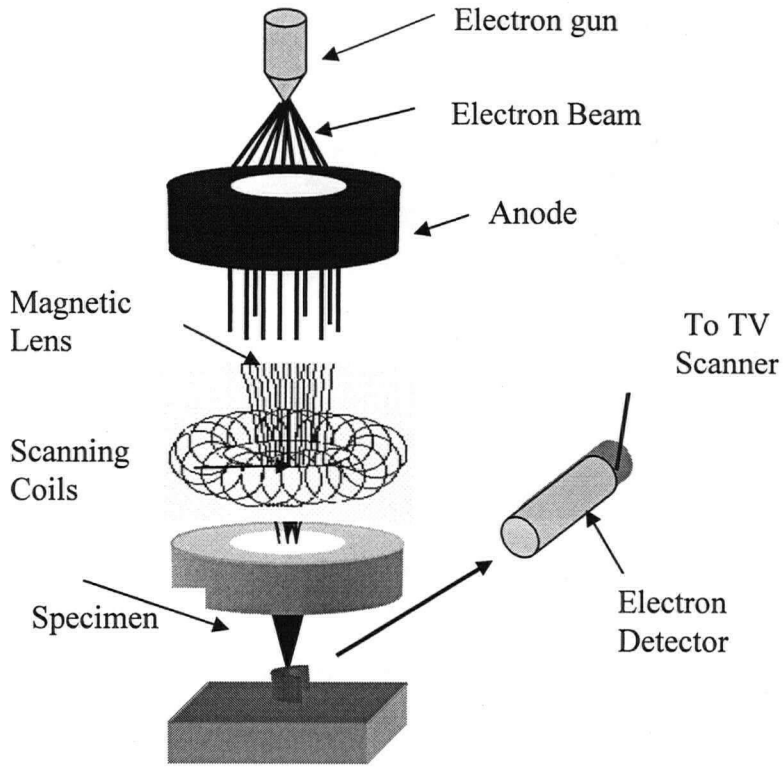


Figure 22 Schematic representation of the scanning electron microscope.

An extremely important condition necessary for producing an SEM image is that both the column and the sample must be under vacuum.[144] There are many reasons for requiring a vacuum environment. Firstly, the SEM filament would be rapidly burnt out in the presence of air. Secondly, the electrons would collide with air molecules inside the column which would prevent them from reaching the sample. Finally, gas molecules could react with the sample altering the surface and as a result, the quality of the image.

Another critical condition for producing an SEM image is that the specimens be electrically conductive, since the electron scanning microscope uses an electron beam. [145] Metal specimens easily fulfill this requirement since all metals are conductive materials;

however, it could be a challenge to analyse non-conductive samples such as ceramics and plastics.[145] This challenge is overcome by covering a non-conductive specimen with a very thin layer of a conductive material such as gold, by employing a sputter coater.

The sputter coater is a device that uses argon gas and a small electric field.[146] The prepared specimen is firstly placed inside a small chamber. Subsequently, air is removed from the chamber and argon gas is then introduced. An electric field is then used in order to cause an electron to be removed from the argon atoms transforming the atoms into argon ions with a positive charge. This process is extremely important since Argon ions are now attracted to a negatively charged piece of gold foil present in the system. The Argon ions act in essence like sand in a sandblaster that knocks gold atoms out of the surface of the foil, which will then settle onto the surface of the specimen producing a gold coating as a result. [147]

Chapter 7: Results

A factorial experimental design was applied due to the desire to investigate the effect of two independent factors on a dependent variable. Consequently, a two-factor univariate ANOVA was used in order to evaluate the effect of two levels of age and four levels of adhesive systems (the independent variables) on the fracture toughness (the dependent variable). The results of the interfacial fracture toughness tests are summarized in Table 5.

Table 5 Interfacial fracture toughness (K_{IC}) in $MPa \cdot m^{1/2}$ (Mean ± SD) of dental bonding agents bonded to young (≤45 years) and old dentin (≥65 years).

Dental Bonding Agent	Age	K_{IC} ± standard deviation	Coefficient of Variance
Adper Prompt L-Pop (APLP)	65	.87 ± .35	40
	45	.82 ± .25	30
Clearfil SE (CSE)	65	.99 ± .23	25
	45	.74 ± .16	21
Prime & Bond NT (PBNT)	65	.24 ± .19	79
	45	.22 ± .16	72
Scotchbond MP (SBMP)	65	.52 ± .27	52
	45	.67 ± .35	52

Firstly, a Levene's test for homogeneity was performed in order to compare the variances of the samples. The test depicted that the variance was significant for all the

data (although close to non-significance – 0.042) but not significant for the data without PBNT (0.112). (Tables 6 and 7)

Since a large variation was observed in the results obtained for PBNT when compared with the other adhesive bonding agents (a variation that may not be explained by the natural variation of the samples alone), it was decided to run the statistical analysis twice: with and without the results obtained for PBNT.

Table 6 Levene's test of equality of error variances for all data.

F	df1	df2	Sig.
2.223	7	72	.042

Table 7 Levene's test of equality of error variances excluding PBNT.

F	df1	df2	Sig.
1.886	5	54	.112

A univariate (K_{IC}) analysis with two factors (age and dentin bonding agent) was performed including all the results and was then repeated excluding PBNT. The summary tables are presented in Table 8 (all the data) and Table 9 (without PBNT).

Primarily, the analysis did not indicate a statistically significant difference based on age. However, the analysis indicated a statistically significant difference in fracture toughness relative to the bonding agents, regardless the age of the sample. The dental bonding system – age interaction term was not statistically significant.

Adper Prompt L-Pop achieved the best mean values of fracture toughness. Nevertheless, no statistically significant difference was observed between Adper Prompt L-Pop and Clearfil SE Bond and both bonding agents were statistically stronger than Scotchbond MP. When added into the statistical analysis, Prime & Bond NT was the weakest bonding agent studied ((PBNT) < SBMP < PLP = CSE).

Table 8 Univariate ANOVA for all data.

Source	Type III Sum of Squares	df	Mean square	F	Sig.
Dental bonding systems	5.296	3	1.765	27.267	.000
Age	.041	1	.041	.631	.430
Dental bonding system * Age	.395	3	.132	2.031	.115
Error	4.661	72	.065		

Tabl 9 Univariate ANOVA without PBNT

Source	Type III Sum of Squares	df	Mean square	F	Sig.
Dental bonding agents	.918	2	.459	6.023	.004
Age	.043	1	.043	.558	.458
Dental bonding system * Age	.391	2	.195	2.562	.087
Error	4.118	54	.076		

Subsequently, since F was significant, a Scheffé test for multiple means comparison was performed in order to evaluate which specific cell mean differs from which other specific cell mean. Table 10 reflect the results of Scheffé test with all data while table 11 depicts the result of the test without Prime and Bond NT.

Table 10 Scheffé test for multiple means comparison with all data.

Dental Bonding System	N.	Subset		
		1	2	3
Prime & Bond NT	20	.2291		
Scotchbond MP	20		.5949	
Clearfil SE	20			.8450
Adper Prompt L – Pop	20			.8681
Sig.		1.00	1.00	.994

Table 11 Scheffé test for multiple means comparison without PBNT.

Dental Bonding System	N.	Subset		
		1	2	3
Scotchbond MP	20		.5949	
Clearfil SE	20			.8450
Adper Prompt L – Pop	20			.8681
Sig.		1.00	1.00	.966

Scanning Electron Microscopic Analysis

SEM observations of the fracture surfaces showed two different patterns of failure. Examined fracture surfaces of samples bonded with PBNT showed signs of adhesive failure within the adhesive layer, showing opened dentinal tubules that were not impregnated by the adhesive resin. In contrast, bonded interfaces with APLP, CSEB and SBMP exhibited predominantly a cohesive type of failure, including cohesive failure of the adhesive layer and cohesive failure of the composite resin. (Figures 23-30).

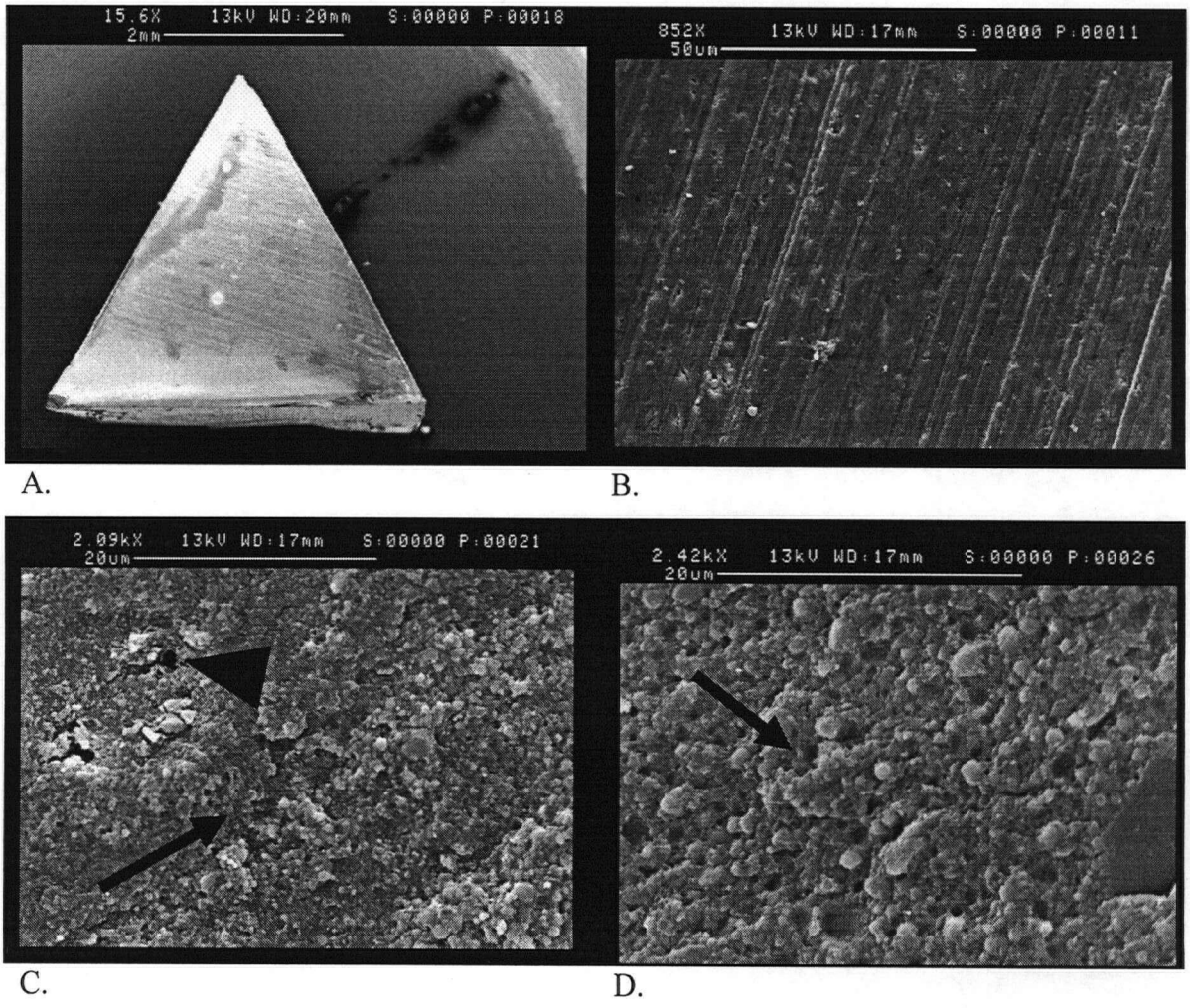


Figure 23 Adper Prompt L-Pop (young, ≤45 years old) A) Broad view of the dentin fracture surface. B) Higher magnification (x852) of the sample in A. C) and D) Higher magnification showing resin adhesive (arrows) and open of dentinal tubules (pointer).

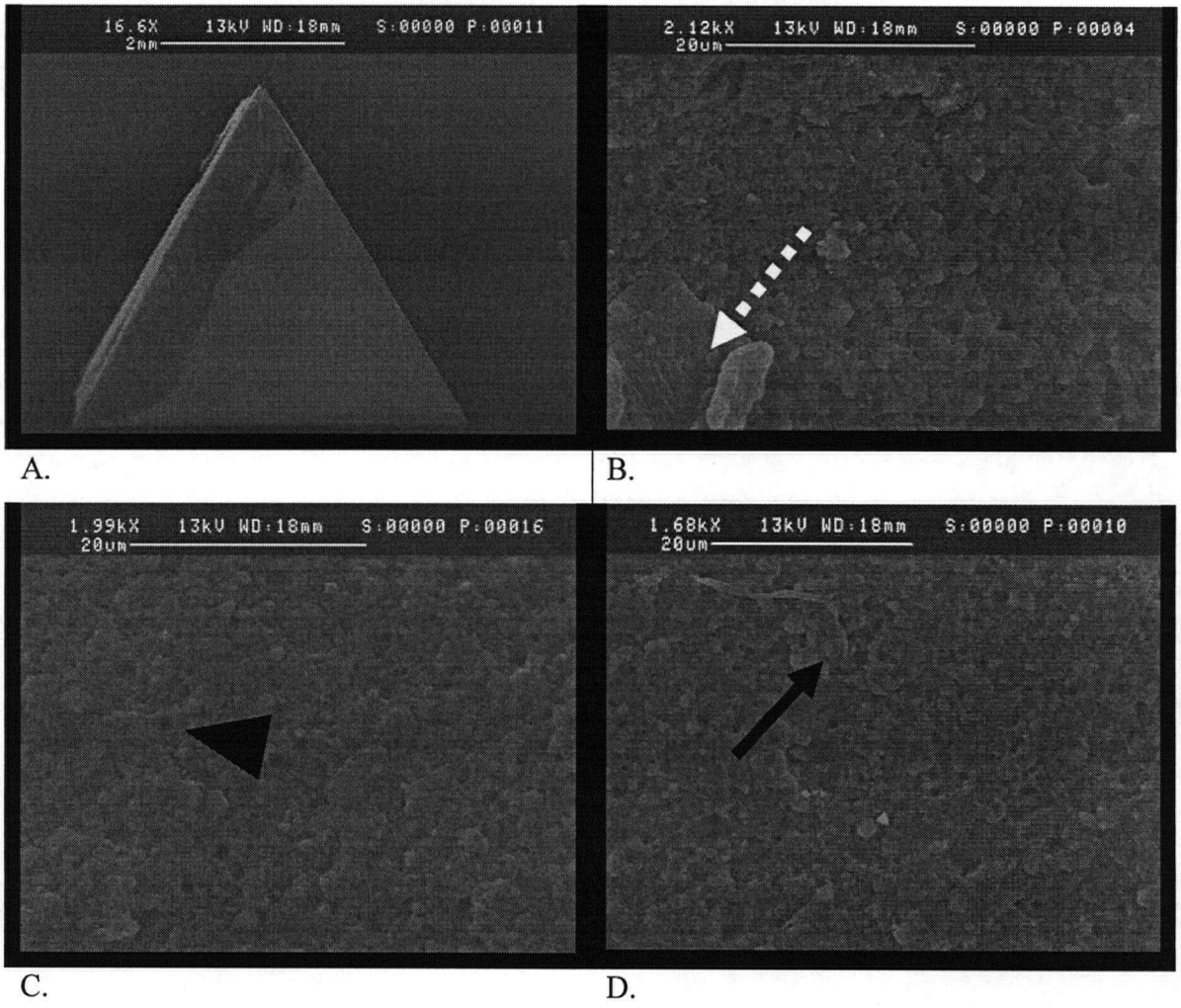


Figure 24 Adper Prompt L-pop (old, ≥65 years old) A) Broad view of the dentin fracture surface. B), C) and D) Higher magnification observation of the sample seeing in A. showing the resin adhesive (dark arrow), the opening of dentinal tubules (pointer), and a fraction of composite resin (white, dashed arrow).

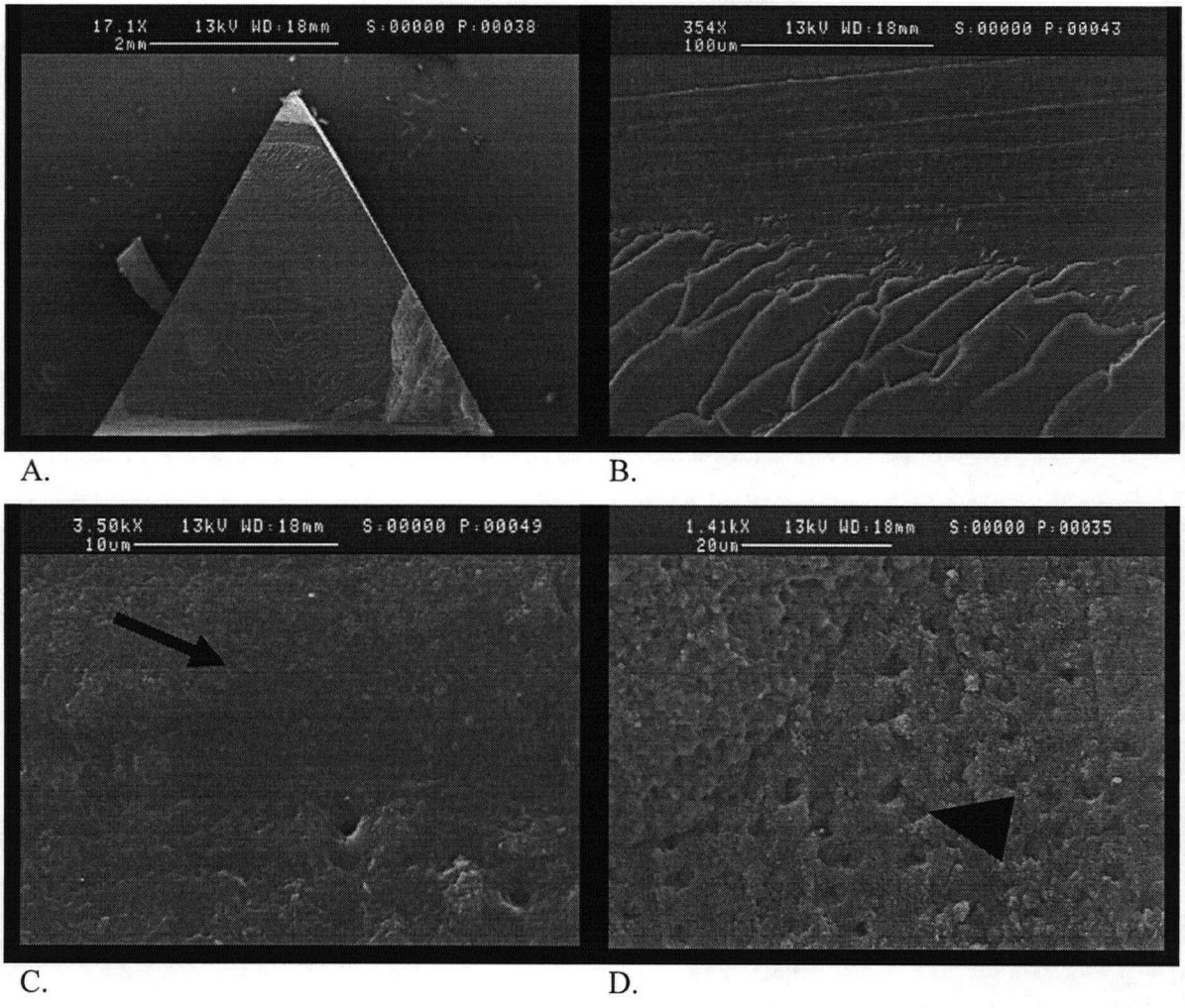
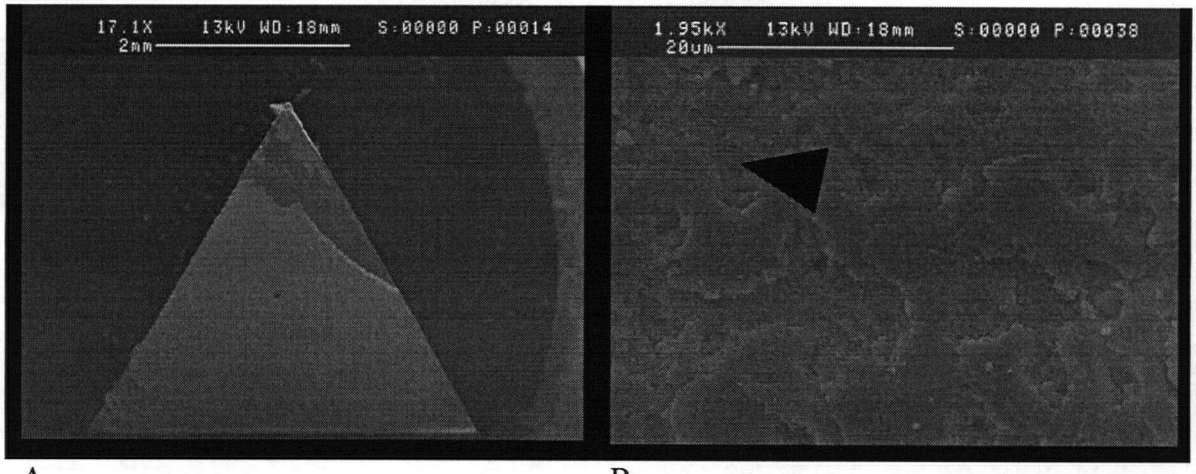
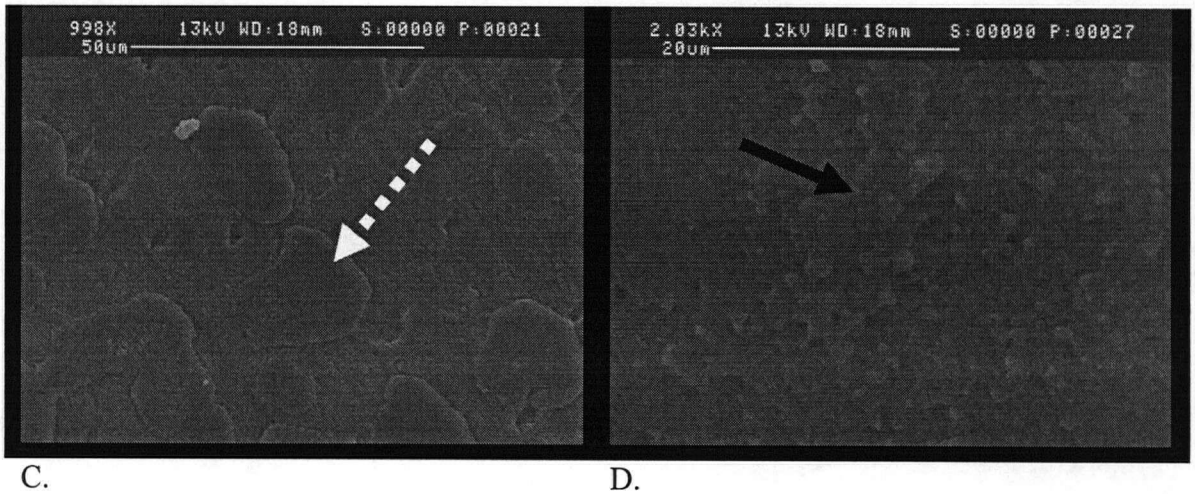


Figure 25 Clearfil SE Bond (young, ≤45 years old) A) Broad view of the dentin fracture surface. B) Higher magnification (x354) observation of the sample seeing in A. C) and D) Higher magnification observation of the sample seeing in A and B showing the resin adhesive (black arrow) and the opening of dentinal tubules (pointer).



A. B.



C. D.

Figure 26 Clearfil SE Bond (old, ≥65 years old) A) Broad view of the dentin fracture surface. B), C) and D) Higher magnification observation of the sample seeing in A. showing the resin adhesive (black arrow), opening of dentinal tubules (pointer) and fractions of composite resin (white, dashed arrow).

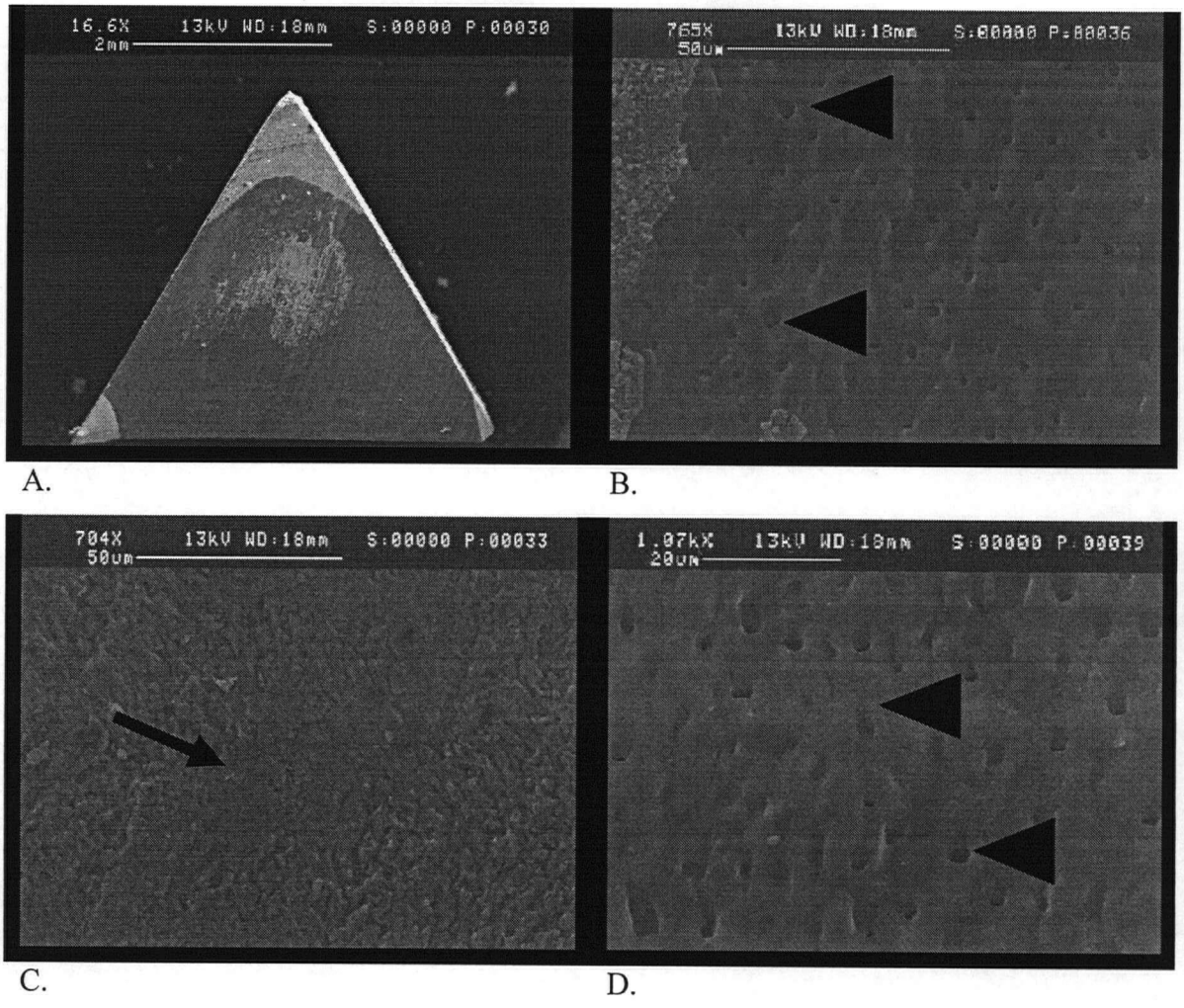


Figure 27 Prime and Bond NT (young, ≤45 years old). A) Broad view of the dentin fracture surface. B) Higher magnification (x765) observation of the sample seeing in A. C) and D) Higher magnification observation of the sample seeing in A and B showing the resin adhesive (black arrow) and the opening of dentinal tubules (pointers), which were much more abundant in this group.

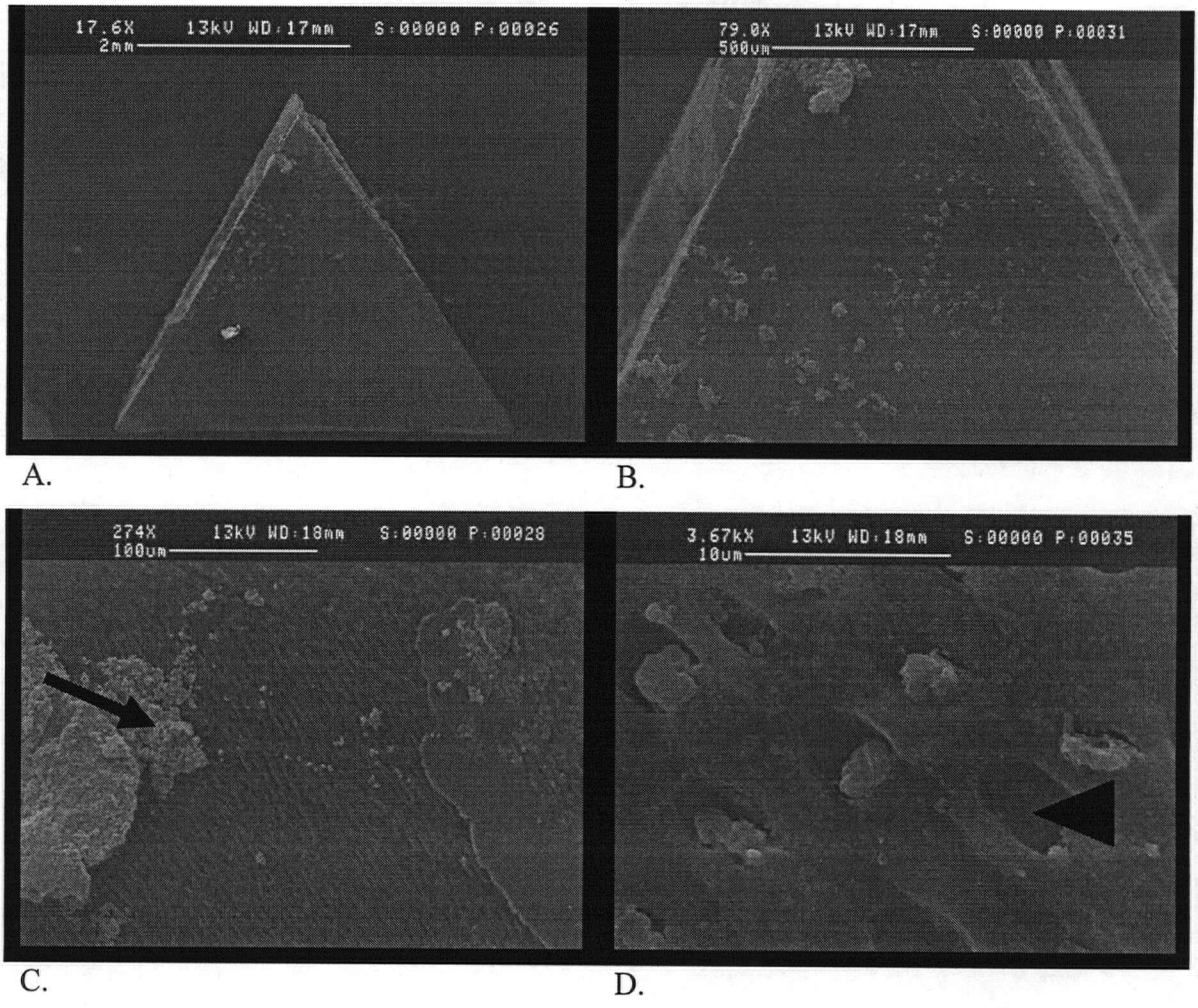


Figure 28 Prime and Bond NT (old, ≥65 years old) A) Broad view of the dentin fracture surface. B) Higher magnification (x79) observation of the sample seeing in A. C) and D) Higher magnification observation of the sample seeing in A. showing the resin adhesive (black arrow) and opening of dentinal tubules (pointer), also more evident in this sample group.

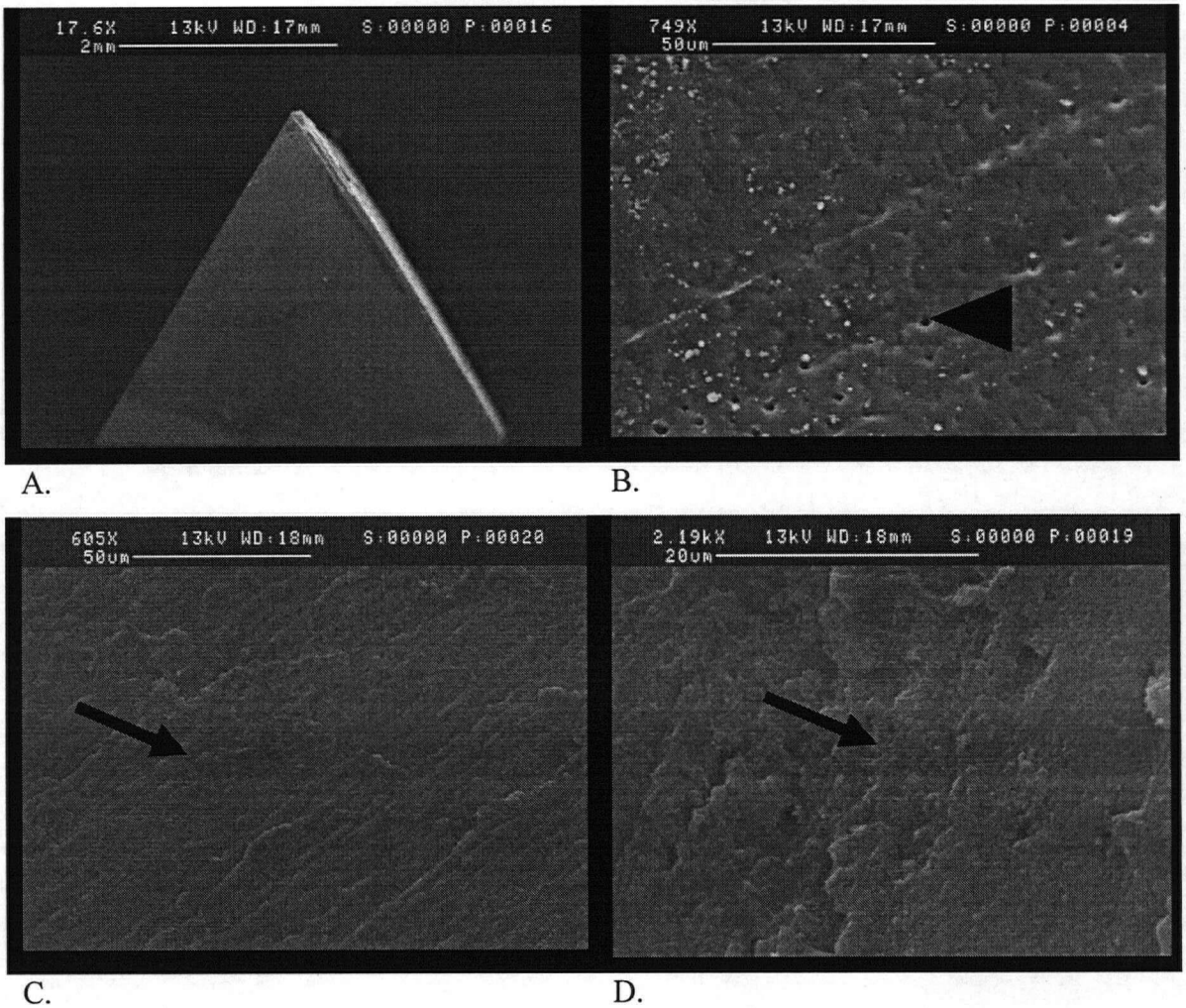


Figure 29 Scotch Bond MP (young, ≤45 years old) A) Broad view of the dentin fracture surface. B) and C) and D) Higher magnification observation of the sample seeing in A. showing the resin adhesive (arrows) and relatively fewer openings of dentinal tubules (pointer) when compared to Prime and Bond NT.

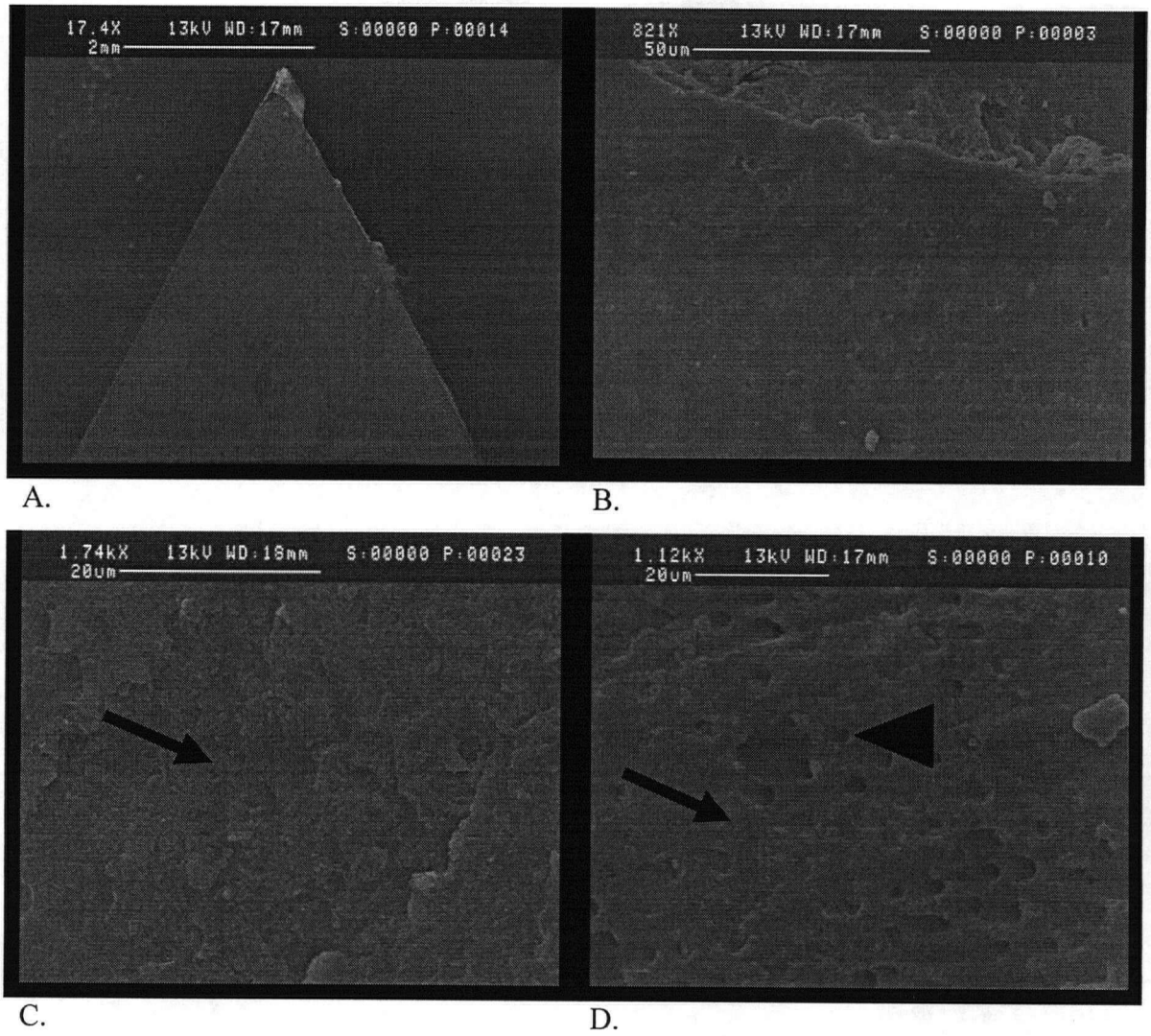


Figure 30 Scotch Bond MP (old, ≥65 years old) A) Broad view of the dentin fracture surface. B) and C) and D) Higher magnification observation of the sample seeing in A. showing the resin adhesive (arrows) and also relatively fewer opening of dentinal tubules (pointer) when compared to Prime and Bond NT.

Chapter 8: Discussion

Dental caries is not a problem that is essentially related to infancy and adolescence any longer. Efficient preventive dentistry efforts are producing a shifting pattern of dental caries amid children and teenagers in North America resulting in adults retaining more of their natural teeth for life.

In addition, projections of the North American population indicate that the percentage of the population aged 65 years and older has grown consistently since the turn of the last century. The consequences of these tendencies are numerous to the general dentist practitioner since the call for restorative procedures is expected to rise in the elderly population, especially in relation to root surface caries.

The results of this thesis, which set out to test the effect of the age of dentin on the fracture toughness of four different dental bonding systems, suggest that the fracture toughness of the dentin/dental bonding system/restorative material interface is not affected by the age of the tooth and that the fracture toughness values of the four different bonding systems tested were different, regardless of the age of the dentin.

Human caries-free molars, premolars, and incisors extracted from patients not older than 45 years and patients not younger than 65 years were used in this experiment. One of the greatest challenges in this study was to obtain teeth, particularly caries- free teeth, amongst the elderly population. Only about 20 percent of all teeth collected for this research were derived from this particular age group.

The teeth were stored in tap water prior to specimen configuration and fracture toughness test. The reason for preferring tap water instead of distilled water, physiologic solutions or any other storage means, was to include electrolytes since saliva contains a

variety of electrolytes including sodium, potassium, magnesium, calcium, bicarbonate, and phosphates. [148]

There are a great variety of bonding systems in the market for the dental practitioner to select from. The rationale for choosing ScotchBond MP, Prime & Bond NT, Clearfil SE Bond, and Adper Prompt L-Pop, the systems tested in this study, was that they were considered to be representative of the four different groups of dental bonding systems, i.e. total etch multi bottle, total etch one bottle, self etch two steps, and self etch one step, respectively.

A rather large variation was noted in the results of some of the data, particularly in the case of Prime & Bond NT. The difference between the lowest and the highest value covered approximately 200%, a variation that may unlikely be explained by the natural variations of the substratum.

Actual bond strengths depend on a variety of factors such as the design of the cavity and the location of the restoration. However, one of the most critical factor influencing bonding strengths of restorative materials is the operator, a plausible cause for such a variation in this study.

Prime & Bond NT resulted in the lowest interfacial fracture toughness values of all four bonding agents tested in both young and old dentin. The explanation for the low values achieved under the conditions in this study may be related to the technique sensitiveness of this adhesive system since the preservation of an ideally moist dentin is crucial.[149]

The volatile solvents featured by primers, such as acetone and alcohol, act as water chasers, subsequently dislocating water and allowing the infiltration of resin

monomers into the underlying collagen scaffold. When the etched dentin is not ideally moist, the interaction between the primer and the etched dentin becomes unfavorable.[150, 151] Excessive drying of the etched dentin surface can cause the collapse of the collagen scaffold. Consequently, interfibrillar spaces once present between collagen fibres disappear reducing the effectiveness of primer penetration and creating microscopic porosities in the hybrid layer.[152, 149]

The presence of porosities and defects within the hybrid layer may encourage fracture initiation and propagation which will as a consequence, diminish the resistance of the adhesion and, therefore, the retention and durability of the restoration. Tam and Pilliar have reported that bonding to "wet" dentin produced higher dentin – composite fracture resistance when compared to bonding to "dry" dentin.[153] Although care was taken to follow the instructions for PBNT, it is possible that "ideal" moisture conditions were not always achieved.

The reason for such low values obtained for PBNT, however, may also be related to the chemical composition of the material. Tam and Pilliar have reported increased internal K_{IC} of a thin adhesive layer versus a thick one.[154] However, filled adhesive resins are more viscous when compared to unfilled adhesive resins and may not simply form a thin adhesive layer.

Walshaw and Tam[155] reported that the filler particles of two dental bonding systems used in their study did not appear to penetrate into the dentinal tubules and that the resin adhesive diffused preferentially, perhaps by capillary action. The authors suggested that the fillers may possibly have been held up by a form of filtration or even by reaction products. Other authors have reported the same fact when using PBNT,

implying that a gathering of the adhesive resin filler particles at the surfaces prevent efficient infiltration of the demineralised dentin". [156, 157]

Tam *et al.* [155, 153] have greatly emphasized, however, that filled adhesive resin improved the properties of the dentin/composite interface in relation to interfacial fracture toughness, while Swift *et al.*[158] have reported statistically similar clinical results between a filled and unfilled adhesive resin after 36 months of a clinical evaluation.

ScotchBond MP was the only bonding agent used in this study that has been evaluated using different methods for determination of fracture toughness values, i.e. the chevron notch short rod (CNSR) test and the notchless triangular prism fracture toughness test (NTP). The results obtained in this study are in agreement with Ruse and Feduik that reported a K_{IC} of .50 ± .14 MPa·m^{½} for ScotchBond MP[131]. Tam and Pilliar[125] evaluated the fracture toughness of the resin composite interface using CNSR fracture toughness test and reported a K_{IC} of .34 ± .21 MPa·m^{½}.

The results of this study produced a rank order of the bonding agents, according to their fracture toughness. This rank order was a little different from what most authors have reported in the literature since in this study self–etching bonding systems generated higher values of fracture toughness when compared to total etching bonding systems. This fact may be explained accorded to Tantbirojn *et al.*[114] that reported considerable discrepancy in ranking order of stronger adhesives when comparing fracture toughness tests with shear bond tests, which unequally distribute forces along the resin-composite-dentin interface and commonly lead to fracture in dentin.

Adper Prompt L-Pop, for instance, demonstrated the highest fracture toughness values amongst the dental bonding systems tested in this study. Although no studies to

date have reported fracture toughness values for this bonding system, values and rank order descriptions after tensile, micro – tensile, and shear bonding tests have been very inconsistent.[107]

While Perdigao *et al.*[159] reported that PBNT achieved bond strengths significantly higher than APLP, Kelsey *et al.*[160] reported no difference between the two dental bonding systems. Atash *et al.*[161] reported higher values of bond strength for APLP when compared to PBNT.

The statistical analysis did not indicate a significant difference based on the age of the dentin samples. As teeth age, sclerosis of the dentin increases and the organic component decreases.[162] The dentinal tubules become narrower due to the deposition of peritubular dentin, while an increasing number of tubules become completely obliterated, reducing dentin permeability to aqueous solutions.[163] Therefore, it is not surprising that dentin of older teeth behave differently to a variety of chemical and physiological stimuli, being a less suitable substratum for dental bonding systems.[83]

Heymann *et al.*[164] observed a tendency toward poorer retention of dentin-bonded class V restorations in older subjects. In addition, Richards *et al* reported a significantly better retention of class V restorations in younger subjects than in older subjects.

Prati *et a.l* [85] have shown that sclerotic, old coronal dentin present thinner resin infiltrated dentin layer with shorter resin tags, apparently due to the inability of the acid conditioners used in the study to demineralise sclerotic, old dentin in 25 seconds. The same effect was demonstrated by Yoshiyama *et al.*[165] in relation to cervical root dentin. Kwong[166] and Tay [167] have also reported the effect of sclerotic dentin from non – carious cervical lesions on bonding strengths of dental bonding systems.

Although the statistical analysis of this study did not indicate a significant difference on fracture toughness values based on the age of the dentin samples, the results are in agreement with Tagami *et al.*[135] who reported similar bond strengths to both young and old dentin for all four adhesive systems tested.

The inability to detect an effect of age on fracture toughness values in this study, does not necessarily suggest that age does not affect interfacial fracture toughness of dental bonding systems. The structure of aged dentin is extremely complex and presents numerous challenges that may reduce the efficacy of dental bonding systems. The lack of statistically significantly differences may suggest that these forms of dentin were exceptionally heterogeneous. It might also be possible that a repeat study with a significantly larger sample size could enable the identification of age-related differences.

The effect of aging on bonding is extremely complex and further investigations should be performed. A long-term clinical trial could potentially allow further investigation.

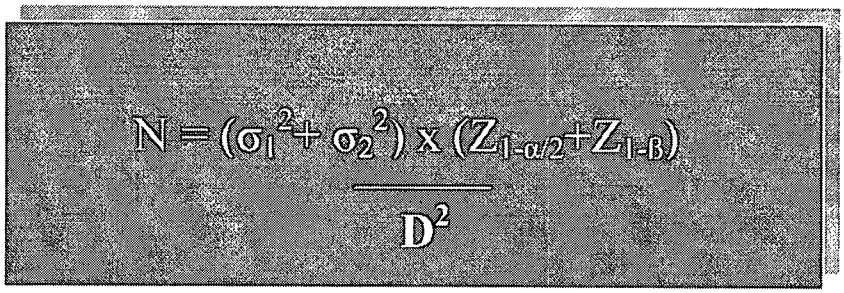
Conclusions

Within the limitations of this study, it was possible to conclude that despite the physiologic and morphologic transformations of dentin as a consequence of aging, the fracture toughness values of the dentin/dental bonding system/restorative material interface were not affected.

Furthermore, since each adhesive system has distinct chemical, mechanical and adhesive properties, different values were of fracture toughness were observed, regardless the age of the dentin. While Adper Prompt L-pop and Clearfil SE achieved the highest fracture toughness values, Prime & Bond NT showed the lowest values.

Future Research

The inability to detect an effect of age on the interfacial fracture toughness of the four dental bonding systems used in this study, does not necessarily preclude the fact that age does not play a role. Since the large natural variation of dentin might well have masked differences due to the age of dentin, a large sample size may be used in a new study and the data obtained in this thesis may be used for power calculation. For instance, for a bonding system with interfacial fracture toughness mean of 0.6 $MPa \cdot m^{1/2}$, based on a coefficient of variance of 30%, the standard deviation would be 0.18 $MPa \cdot m^{1/2}$. In order to identify a medium effect (0.5 standard deviations) with a power of .80 and a confidence level of .95, a researcher would need 64 samples per group according to the formula:[168, 169]



$$N = \frac{(\sigma_1^2 + \sigma_2^2) \times (Z_{1-\alpha/2} + Z_{1-\beta})}{D^2}$$

Where N = Sample size; σ = the standard deviation; Z = normal distribution; α = probability of type I error; β = probability of type II error, and D = clinically relevant difference.

Furthermore, stratification of the sample as well as randomization during the bonding procedure may impact the final results. Statistical differences identified by such changes in methodology might however prove to be of little or no clinical significance.

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