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A microshear test to measure bond strengths of dentin–polymer interfaces

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Abstract

The microbond test, a single fiber shear test, has been adapted to be a microshear test for the measurement of the adhesion of resin-based dental materials to dentin and enamel. The objective of this study is to improve the design of this microshear test so that it can provide accurate and reliable shear bond strength data. In the current design of the microshear test apparatus, the bonding diameters of the specimens have been as small as 0.70 mm. The smaller diameters give researchers the ability to test several bonded specimens on one flat dentin or enamel surface, thus allowing both for the regional mapping of the mineralized surface and the conservation of extracted teeth needed to provide the necessary substrates. The test configuration used in earlier studies has been modified through finite element analysis to address concerns in the test methodology. The results of this study show that the microshear bond test can be a useful tool in helping to understand the complex interactions that occur at the interface between dental composites and dentin and/or enamel surfaces, especially at interfacial sites not amenable to macroshear testing. Published by Elsevier Science Ltd.

Keywords: Bond strength; Shear test; Dentin; Enamel; Microshear test

1. Introduction

Various mechanical test methods, such as macroshear, in-plane shear, tensile, microtensile and flexural, have been used to assess dental adhesion. Although there are no industry standards for dental bond strength testing, there is growing interest in critically assessing the merits of several test methods so that accurate, reliable and user-friendly test methods can be developed [1–3].

Mainly because of its overall simplicity, several variations of shear bond testing have been widely used. Among the advantages are ease of specimen preparation, simple test protocol, and the ability, at least qualitatively, to rank different products according to bond strength values. Challenges to the validity of this adhesion test include the mixed mode loading (both shear and tensile forces are induced during this test), stress concentrations at the point of loading, and the tendency, under certain circumstances, of failure in the dentinal substrate at loads far less than the tensile strength of dentin. This last point is especially troublesome because, as new adhesives yield improved bond strengths, such premature failure caused by the testing geometry would limit the ability of the test to discern real differences among various adhesive systems. Pashley et al. cited this particular issue as a limitation to using conventional shear tests and a reason to consider the microtensile test as a reasonable alternative [4]. Polack recently tried a variation of the Iosipescu shear test to try to overcome the difficulties inherent in the conventional shear test [5]. A finite element analysis model showed that his design imparted more uniform stresses. However, no significant differences were found between the shear bond strengths of the conventional shear test and his test.

Tantbirojn et al. addressed the problems inherent in the conventional shear test by comparing the

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performances of six dentin bonding agents subjected to an interfacial fracture toughness test and the shear bond test [6]. The study suggested that the results obtained from the interfacial fracture toughness test were only marginally different from those obtained by the nominal that the shear test. They expressed concern, however, that shear test may not be able to estimate bond strengths if future generations of bonding systems get appreciably better. For the shear bond test, if dentin pull-out was observed in the failure surface, then the calculated nominal bond strength was no longer based on the cross-sectional area. Thus, they concluded that the bond test could not discriminate between good and very good bonding agents.

Previously, we proposed the use of the microbond test (MBT) to assess the strength and durability of silane-treated glass bonded polymer interfaces [7–10]. By means of the MBT, a wide spectrum of resin systems, coupling agents and fillers can be screened for use in dental composites as well as other applications that depend on the quality of the interface or interphase. We saw an opportunity to modify this test to allow for the testing of the bond between flat dental substrates, such as enamel and dentin, with dental restoratives mediated by intermediary dental adhesives. Our test method allows for the testing of small areas, and this feature permits the regional mapping of substrate surfaces as well as depth profiling of the substrate. Furthermore, the small size of our test specimens permits many tests to be performed on the same substrate (thereby promoting the conservation of extracted teeth) and also easily lends itself to durability studies. Thus, the main objective of this study was to improve the design of our initially developed microshear test so that it can provide accurate and reliable shear bond strength data.

2. Materials and methods

The Experimental section[1] describes the microshear test and the type of adhesion data that can be obtained from its application. Cylinders of dental composite resin are bonded onto prepared enamel or dentin substrates, shear tested at a specified rate, and the interfacial properties calculated (Fig. 1). The data presented come from experiments that will be reported in greater detail in subsequent papers as described below.

[1] Certain commercial materials and equipment are identified in this study for adequate definition of the experimental procedure. In no instance does such identification imply recommendation or endorsement by the National Institute of Standards and Technology or the National Institutes of Health nor does it imply that the materials or instruments are necessarily the best available for this purpose.

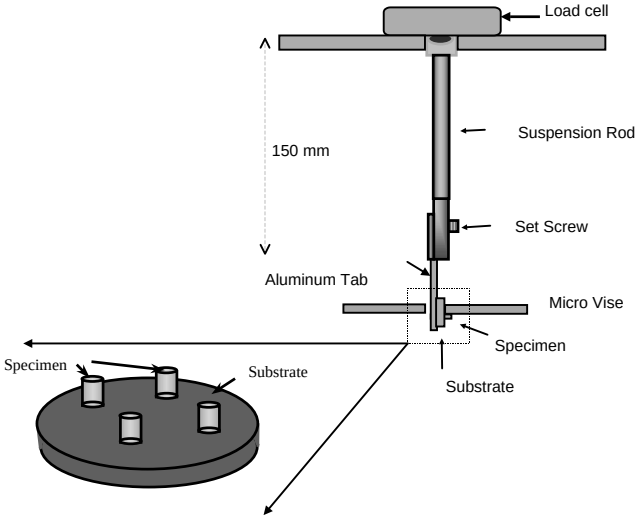

Fig. 1. Modified microbond test apparatus.

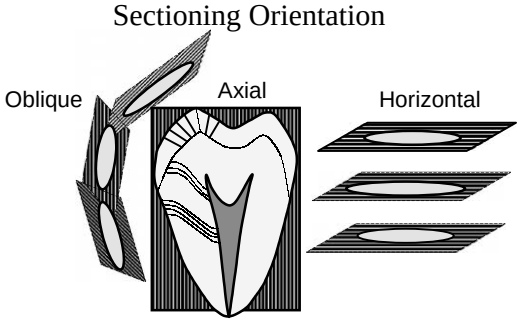

Fig. 2. Sectioning orientation for dental substrates.

2.1. Tooth slices

Tooth slices were obtained from human molars that were stored under refrigeration in a preservative solution containing 0.2 g sodium azide in 100 mL distilled water. Two enamel regions and three dentin regions were chosen as substrates for the microshear bond test. The selected enamel regions were from the cuspal and middle coronal regions, the coronal regions were cuspal, cervical (near the dentin–enamel junction (DEJ)) and from the root region that was approximately 3 mm below the DEJ. The depth of the each tooth region sectioned for bond testing was also controlled; the enamel regions were obtained from near the center between the DEJ and outer surface, and dentin regions were from the outer third of the dentin. Each tooth region was then sectioned in one of three ways, horizontally, axially and obliquely (Fig. 2). Because each of the five regions of tooth structure was directionally sectioned three ways, a total of 15 types of slices were examined. Each slice, approximately 1.0 mm thick, was obtained by cutting with a slowly

rotating diamond blade (Struers Minitom, Struers, Copenhagen, Denmark) under a flow of distilled water. The slices were bonded to aluminum tabs by first air abrading the aluminum with 50 μm aluminum oxide powder, applying a layer of cyanoacrylate adhesive, and then placing the slice down on the aluminum. The exposed tooth surface was then resurfaced under water with 320 grit SiC paper.

2.2. Bonded sample preparation

A phosphoric acid gel (K-etchant gel, Kuraray) or self-etching primer (Clearfil Liner Bond 2V Primer, Kuraray) was applied to the enamel or dentin surface for 30 s. In the case of the K-etchant gel, the H_3PO_4 was rinsed with distilled water and air dried. The excess self-etching primer was removed with a stream of air. One of the resin-bonding agents was then applied, air-thinned and light cured by 30 s irradiation. Acid etching or priming was followed by application of Clearfil Photo-Bond or Clearfil Liner Bond 2V Bond Liquid A, respectively. Subsequently, a low-viscosity resin composite (Clearfil Protect Liner-F, Kuraray) was filled into an iris cut from microbore Tygon tubing (TYG-030, Small Parts Inc., Miami Lakes, FL) with an internal diameter approximately 0.7 mm and a height of 0.4 mm. The iris was held firmly on the surface using a double-sided tape to prevent the resin from seeping away from the defined area at the base. After this step, the resin composite was photo-irradiated for 60 s, and because the Tygon tubing was clear, the resin could be thoroughly cured through the iris. In this manner, very small cylinders of resin composite, approximately 0.7 mm in diameter and 0.4 mm in height, were bonded to the surface at five or six locations of each enamel or dentin sectioned slice. Subsequently, the Tygon tubing was removed. The specimens were then stored in water at 37°C for 24 h until they were tested. After storage, all the samples were checked under an optical microscope (30 ×) for bonding defects. The samples that showed apparent interfacial gap formation, bubble inclusion, or other obvious defects were excluded from this study. The composite–substrate bonds were subsequently broken by micro-shear testing as described below.

2.3. Microshear testing

The test device was suspended on a rod that was connected to a load cell (PG5001, Mettler-Toledo Inc., Switzerland) (Fig. 1). The microvise of the microshear apparatus was positioned just above the cylinder of resin composite to be tested and then the moveable blade was closed to where the vise was almost touching the tooth surface (approximately 0.02 mm or less). The vise blade opposing the tooth side did not touch the aluminum tab and was kept approximately 0.3–0.5 mm from the tab so as not to introduce frictional forces. The position of the vise and the diameter of each sample were measured on the monitor screen by macrovideo photography.

During the test, the microvise sheared the cylinder from the tooth at the rate of 0.5 mm/min set through an actuator (Newport 850A, Newport Co, Fountain Valley, CA) [11]. The change in load as a function of time was recorded on a computer and the shear strength was calculated from the equation $\tau = F/(\pi R^2)$, where τ was the interfacial shear strength, F was the load at failure and R was the radius of the resin cylinder [12]. The data were statistically analyzed using parametric one-way ANOVA and the Student's t-test at the confidence level of 95%.

3. Results

Mean shear bond strength values and standard deviations in MPa, obtained from the microshear bond test, are shown in Table 1. The shear bond strength was dependent on the bonding materials, the region of tooth structure and the orientation employed in sectioning tooth structure. Enamel with its prismatic, rod-like apatitic morphology and dentin with its array of dentin tubules, are complex, anisotropic mineralized tissues. This anisotropy is especially pronounced in the case of the highly mineralized enamel (mass fraction of 95% hydroxyapatite) and depends upon the orientation of the enamel rods. Dentin has a more heterogeneous structure than enamel, consisting of fibrillar collagen interdispersed with a crystalline apatitic mineral (similar to the hydroxyapatite found in enamel), and also exhibits tubular variation especially with depth. Dentin thus presents a more complex substrate than enamel for bonding.

There was a significant difference among the mean composite–enamel bond strength values of the conventional non-priming adhesive system (K-etchant gel and Clearfil PhotoBond) when applied to the various enamel surfaces (one-way ANOVA, $F = 60.8$; df = 5, 34; $P < 0.05$). Phosphoric acid etching produced mean bond strength values of 26–31 MPa to this surface, if the enamel prisms were ground and oriented horizontal or obliquely. The bond strengths decreased to 13–14 MPa if the enamel rods were oriented axially to the bonded surface (Student's t-test, $P < 0.05$). Differences of bond strength values between the cuspal and middle coronal regions were observed only with the horizontal sections (Student's t-test, $P < 0.05$).

By contrast, for the shear bond test results of the self-etching primer system (Clearfil Liner Bond 2V) to the various enamel surfaces, no significant differences were observed among the groups (one-way ANOVA, $F = 0.606$; df = 5, 34; $P = 0.696$). Mean shear bond strength

Table 1
Microshear bond strength (MPa ± SD)

	Area	Direction	Bonding system	
			Conventional non-priming system K-etchant + Clearfil PhotoBond	Self-etching primer system Clearfil Liner Bond 2V
Enamel	Cusp	Horizontal	31.12 ± 3.38 ($n = 7$)	21.87 ± 3.31 ($n = 7$)
		Axial	13.90 ± 3.11 ($n = 8$)	21.25 ± 2.28 ($n = 7$)
		Oblique	27.78 ± 2.17 ($n = 5$)	22.16 ± 2.38 ($n = 6$)
	Middle	Horizontal	14.22 ± 2.46 ($n = 7$)	20.07 ± 2.89 ($n = 8$)
		Axial	13.29 ± 2.73 ($n = 7$)	20.28 ± 2.40 ($n = 6$)
		Oblique	26.49 ± 1.78 ($n = 6$)	21.17 ± 3.26 ($n = 6$)
Dentin	Cusp	Horizontal	16.08 ± 3.23 ($n = 10$)	26.18 ± 3.68 ($n = 6$)
		Axial	16.05 ± 3.24 ($n = 8$)	23.14 ± 3.21 ($n = 6$)
		Oblique	15.04 ± 2.42 ($n = 5$)	23.79 ± 2.82 ($n = 7$)
	DEJ	Horizontal	15.08 ± 2.01 ($n = 7$)	24.91 ± 1.59 ($n = 6$)
		Axial	15.03 ± 2.73 ($n = 9$)	22.15 ± 2.55 ($n = 7$)
		Oblique	14.47 ± 2.30 ($n = 5$)	22.84 ± 2.21 ($n = 10$)
	Root	Horizontal	12.18 ± 2.90 ($n = 5$)	22.47 ± 2.13 ($n = 5$)
		Axial	13.34 ± 2.31 ($n = 8$)	21.89 ± 3.82 ($n = 8$)
		Oblique	13.01 ± 2.75 ($n = 6$)	22.04 ± 2.62 ($n = 6$)

Numbers in parentheses represent the numbers of samples tested.
SD is the standard deviation of the measurement.

values of 20–22 MPa were obtained from all the enamel surfaces.

When bonding to dentin surfaces, the self-etching primer system exhibited significantly higher bond strength values than those of the conventional system for all of the surfaces (Student's t-test, $P < 0.05$). For the self-etching primer system, the bond strengths ranged from 22 to 26 MPa, while the bond strength for the conventional system ranged from 12 to 16 MPa. With the self-etching primer system, no significant difference was observed among the groups (one-way ANOVA, $F = 1.67$; df = 8, 52; $P = 0.129$). The mean shear bond strength values of both systems tended to be higher toward the cusp, while lower bond strength values were found close to the root dentin. With the conventional non-priming system, there was no significant difference among the test groups (one-way ANOVA, $F = 1.67$; df = 8, 54; $P = 0.127$), except for the horizontal section between the cuspal area and root area (Student's t-test, $P < 0.05$).

Although various patterns of failure were observed among the tested groups, the dentin after the treatment by the conventional bonding system revealed a large area of adhesive failure between the dentin and bonding resin combined with a small zone of dentin failure in most cases. When the dentin surface was treated with the self-etching primer system, a complex failure pattern occurred; cohesive failure mixed with dentin failure, hybrid layer failure, bonding resin failure. In case of enamel surfaces, cohesive failures within the enamel were frequently observed with both bonding systems.

As shown in Fig. 1, the current design of our microshear test calls for line loading on a cylinder of the restorative resin. Preliminary microshear bond strength data is very encouraging. However, there are concerns that the type of loading that we impart to the bonded joint may cause substantial tensile forces to be formed. We ran a finite element analysis routine in an effort to address these concerns and to consider ways for improving this test method.

The finite element analysis (FEA) was performed using the ABAQUS commercial program. Half of the specimen was utilized as shown in Fig. 3 due to the symmetry of the problem. The geometry and material properties used in FEA are as follows. For the enamel, the thickness (L) ranged from 0.5 to 1.0 mm, the radius ($D/2$) was 0.7 mm, the Young's modulus was 84.1 GPa and Poisson's ratio was 0.33. For the composite, the thickness (L) was 0.5 mm, the Young's modulus was 18.3 GPa and Poisson's ratio was 0.31. Twenty-node solid elements were used to model the adhesive bond specimens, with the element sizes continuously decreasing towards the leading edge.

The results for the finite element analysis are shown in Figs. 4 and 5. Fig. 4 shows the maximum shear stress normalized to the maximum tensile stress at the bonded area, when the length to diameter ratio (L/D) was changed with fixed D at 1.4 mm, and the length of distributed loading was kept as equal to the length of the specimen L. From this model it is evident that the shear stress can be increased relative to the tensile stress by reducing the specimen length to give a maximal shear stress and a minimal tensile stress. The curve points out the limitations of these factors on the conduct of the test. Although the test could be designed so that the shear force was greater than the tensile force, the tensile

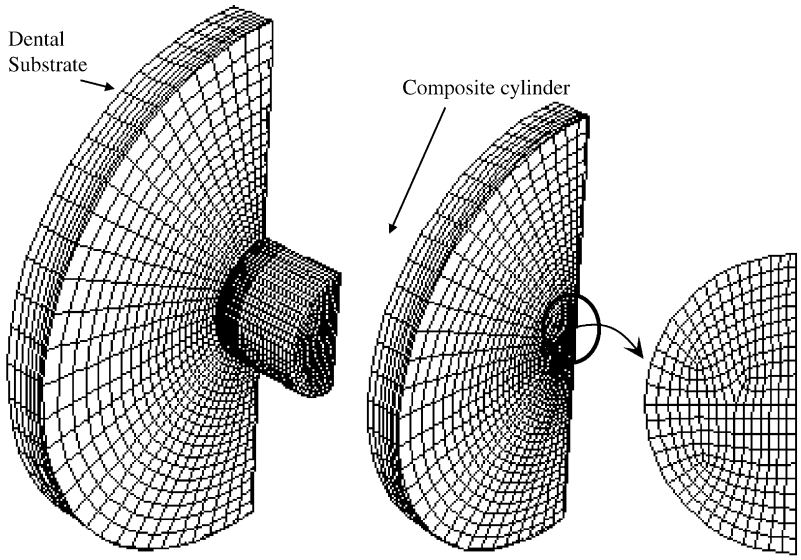

Fig. 3. The finite element model used for the shear bond test. From left to right, the images are of the mesh used showing the dental substrate and composite cylinder, the dental substrate, and the portion of the dental substrate under the composite cylinder.

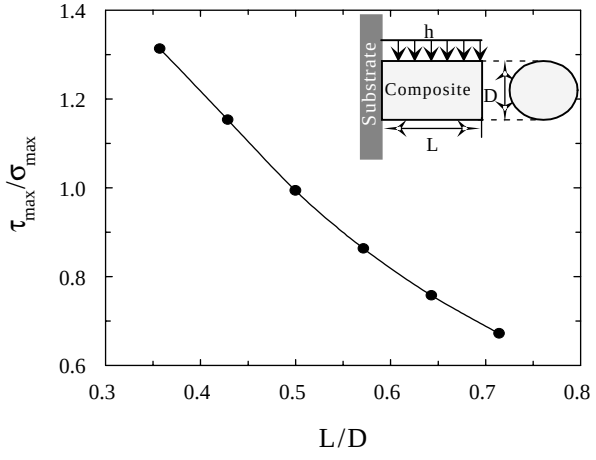

Fig. 4. The shear strength (τ_{max})/tensile strength (σ_{max}) comparison with varying specimen length, L, and diameter, D. h represents the length of the specimen subjected to line loading. In this case, h is held equal to L.

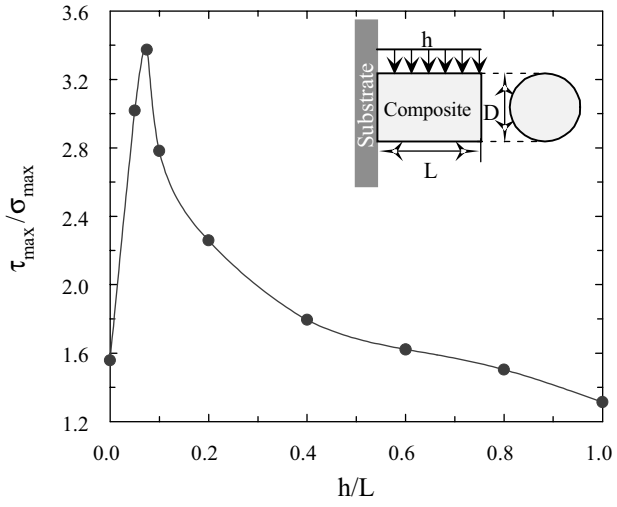

Fig. 5. The shear/tension comparison due to the change of loading length, h (specimen length, L, and specimen diameter, D, fixed). One implication from this picture is that the shear force/tensile force ratio can be maximized if the specimen is loaded over a length 1/10 the length of the specimen (starting at the interface between the substrate and the specimen).

force was still significant. To address this issue, the effect of variations in the application of the load was investigated.

Fig. 5 shows the maximum shear stress normalized to the maximum tensile stress at the bonded area, when the length L was fixed at 0.5 mm and the loading length was changed from 0 to 1 times of the specimen length. From here we can see that the maximum shear stress can be increased to greater than three times relative to the maximum tensile stress by changing the loading condition. Please note that when the loading length (h) becomes small, the loading condition becomes a concentrated load. In such a case, the maximum tensile stress increases due to the interaction between the load and edge.

4. Discussion

The results in Figs. 4 and 5 suggest that, by changing the specimen thickness and loading conditions, the dominant failure mode can be altered between tension and shear depending on the ratio of the tensile strength and shear strength of the joint. It also suggests that the

loading condition is more important to the failure mode. From Fig. 4, if line loading is chosen, then the length of the specimen needs to be small enough to ensure that shear in the predominant mode of failure. Even with shallow specimens, this type of loading will always have a strong tensile component, but this problem can be avoided by following the loading scheme suggested in Fig. 5. If we line load a distance along the specimen of 1/10 the length of the specimen starting at the substrate, the likelihood of shear failure increases significantly. This curve also shows that deviations from this 1/10 of the length can increase the likelihood of tensile failure.

We have found that by following the guidelines shown by finite element analysis, it is possible to control the mode of failure in the microshear test to the point of maximization of the shear component. However, it should be noted that should the bond strength be significantly weaker in tension than in shear, then the tensile failure mode should overwhelm any design attempts to induce the specimen to fail in shear.

As noted in the Introduction, there is much controversy as well as a great deal of research directed at measuring the bond strength between enamel or dentin and dental restoratives. One must exercise care when interpreting the significance of adhesion tests. In the dental materials field, clinical performance does not always parallel laboratory test results. Subsequent to this study, a workshop was held on micro-mechanics measurement technologies for fiber–polymer interfaces [13]. One of the findings from this workshop was that a contributing factor to variability in results came from differences in testing procedures and sample preparation techniques. These findings are also applicable for assessing bond strength between dental substrates and dental restorative materials. Researchers need to understand the limitations of their test technique and must be sure to document how they conducted the test.

We view the tests used to assess the bond strength between dental substrates and dental restoratives to be useful screening tools to discriminate between a multitude of adhesive systems and thus allow researchers to focus on more promising materials for more in-depth studying. That being said, the test results show that the microshear test can be a very versatile and useful test to assess the strength of the bond between mineralized tissue and other relevant dental substrates and polymeric composite restoratives. Because bonded surfaces for the microshear test are very small (approximately $0.4\,mm^2$), we are able to test many specimens on a single surface of sectioned enamel or dentin, resulting in significant conservation of extracted teeth. This new bond test allows regional mapping and depth profiling of tooth structure. Because of the small sizes of the test specimens, equilibrium conditions can rapidly be obtained, which is an important factor in conducting accelerated durability studies of adhesive bonds.

5. Conclusion

Finite element analysis of a previously developed microshear test indicated that a modified test design that would maximize shear forces by a factor of more than three over tensile forces was feasible. The microshear test offers the advantages of facile bond testing for rapid screening of adhesive systems, regional and depth profiling of a variety of substrates for their relative adhesiveness, and conservation of teeth. The microshear test also lends itself to in vitro durability studies and may aid in elucidating adhesion mechanisms.

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