

Microtensile bond strength of fiber-reinforced composite with semi-interpenetrating polymer matrix to dentin using various bonding systems

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This study investigated the microtensile bond strength (μ TBS) of fiber-reinforced composite (FRC) to dentin using various adhesive systems. Forty eight (n=8/group) human molars were flattened to expose dentin. A layer of preimpregnated unidirectional FRC (everStick) was applied on the dentin surface after treatment with either a single-step self-etching adhesive, two-step self-etching system, or a conventional three-step adhesive system. For the control, particulate filler composite (PFC) (Filtek Z250) layering without FRC was used. After 24-hour water storage at 37°C, the specimens were sectioned, further water-stored at 37°C for 30 days and then tested. Data were analyzed using ANOVA and Tukey's test, and reliability was analyzed with Weibull distribution. μ TBS values differed significantly according to the adhesive material used (p<0.05). Single-step self-etching adhesive showed the lowest bond reliability and μ TBS values with both FRC and PFC, whereas conventional three-step and two-step self-etching systems showed higher bond reliability and μ TBS with both materials.

Key words: Fiber-reinforced composite, Microtensile bond strength, semi-IPN matrix,

Received Apr 1, 2008; Accepted Jun 10, 2008

INTRODUCTION

For the past decade, the continuous development of fiber-reinforced composites (FRC) has seen an increased clinical use in a wide range of applications—such as periodontal splints¹, minimally invasive fixed partial dentures², cusp-replacing restorations³, and orthodontic appliances⁴. Due to the high-quality needs of dental FRCs, preimpregnation of fibers with a resin system by the manufacturers has proven to be important⁵. Preimpregnation is based on either photopolymerizable dimethacrylate monomers or a combination of dimethacrylate monomer resin and linear polymer, whereby the latter forms a semi-interpenetrating polymer network (semi-IPN) after polymerization. In principle, the semi-IPN is formed from a linear polymer such as polymethylmethacrylate (PMMA), which is partially or totally dissolved by bi- or multifunctional monomers⁶. Semi-IPN matrices are highly viscous compared to the dimethacrylate system, thereby improving both the handling properties and bonding properties of the FRC after it is polymerized^{5,6}.

Today, a wide array of adhesive systems is available—ranging from multi-step etch-and-rinse systems (which entail separate applications of etchant, primer, and adhesive) to one-bottle self-etch non-rinse systems. The latter type combines the etchant, primer, and adhesive into one bottle, as well

as the acidic monomers and acetone/ethanol as solvents in the same bottle⁷⁻⁹.

Polymerization of the primer and adhesive monomers ensures cohesive strength within the restored structure. On this premise, the compatibility of different materials at the adhesive interface plays an important role to the longevity of a restoration. For direct chairside application of FRCs, the fibers are applied after the adhesive treatment of the bonding surface. Therefore, there belies a possibility that highly solvent-containing bonding systems might yield different bond strengths with semi-IPN matrices as compared to dimethacrylate matrix-based particular filler composites (PFC).

In view of the above concern, the aim of this *in vitro* study was to evaluate the microtensile bond strength (μ TBS) of a semi-IPN matrix of FRC to dentin substrate using various adhesive systems and then compare it against PFC. The null hypothesis tested was that different bonding systems will show comparable bond strengths with both semi-IPN matrix of FRC and PFC.

MATERIALS AND METHODS

Tooth specimens

Forty-eight (n=8 per test group) extracted, sound human molars free of visible caries obtained from 18- to 25-year-old donors were used within one month after extraction. Upon collection, adhering soft tissues and blood were removed under running water

and the teeth were stored in a 0.5% chloramin-T solution at 4°C until use.

Materials used

As for the adhesive systems, PFC and FRC materials used in this study, their brand names, manufacturers, batch numbers, compositions, and application procedures are shown in Table 1.

Specimen preparation

Occlusal enamel was removed perpendicular to the

long axis of the tooth to expose a flat dentin surface using a slow-speed diamond saw (Ernst Leitz GmbH, Wetzlar 1600, Germany) under water lubrication. A standard smear layer was produced within the limits of superficial dentin by wet-grinding the dentin surface with 1200-grit (Federation of European Producers of Abrasives) silicon carbide abrasive paper (Struers SiC) at 300 rpm, using an automatic grinding machine (Rotopol-11, Struers, Copenhagen, Denmark) at a force of 20 N. The dentin surface was then thoroughly washed with water and gently dried

Table 1 Materials used in this study

Material	Lot No	Manufacturer	Type	Composition	Application
ScotchBond Multi-purpose	MP 20000907	3M, St. Paul, MN, USA	Multi-step etch and rinse	A: HEMA, polyalkeneonic acid polymer, water B: HEMA, bis-GMA, tertiary amine	15 s etching, rinsing. Primer application: gently drying 5 s. Bonding application: 10 s light curing.
Clearfil SE Bond	CE 41156	Kuraray Co., LTD, Osaka, JAPAN	Two-step Self etch	Primer:10-MDP, HEMA, Hydrophilic DMA, tertiary amine, water Bonding :10-MDP, HEMA bis-GMA, 39.7% vol silanated colloidal silica	Primer application to tooth surface and leave in place 20 s. Dry with mild air. Bonding application: 10 s light curing.
i BOND	IB 010049	Heraus Kulzer, Hanau, GERMANY	All-in-one Self etch	UDMA, 4-MET, gluteraldehyde, acetone,water, stabilizer	Applied in 3 layers and massaged into the prepared tooth structure for 30 s. Gentle air drying. 20 s light curing.
everStick	EV 2040823-ES-108	StickTech, Turku, Finland	Preimpregnated E-Glass fiber	PMMA,Bis-GMA, E-glass 60 vol%	40 s light curing
Filtek Z250	ZO 20011110	3M, St.Paul, MN, USA	Particulate filler composite	bis-GMA, UDMA, bis-EMA 60 vol% filler	40 s light curing per increment

10-MDP = 10-methacryloyloxydecyl dihydrogen phosphate

4-MET = 4-methacryloxyethyl trimellitic acid

DMA = Dimethacrylate

Bis-GMA = Bisphenol A-glycidyl dimethacrylate

TEGDMA = Triethylene glycol dimethacrylate

UDMA = Urethane dimethacrylate

Bis-EMA = Bisphenol A polyethylene glycol diether dimethacrylate

PMMA = Polymethylmethacrylate

HEMA = Hydroxyethylemethacrylate

E-glass = Electrical-glass fibers

with an air spray, leaving the surface visibly moist.

Adhesive materials (Table 1) were applied to the flat dentin surface according to manufacturers' recommendations. To simulate direct chairside application, a layer (0.5 mm thickness) of preimpregnated unidirectional FRC (everStick) was pressed to the treated dentin surface with a silicon instrument (Refix D, StickTech, Turku, Finland) and light-cured for 40 seconds (Optilux 501, Kerr, USA). PFC (Filtek Z250) of shade A3.5 was applied on the FRC layer incrementally to a height of 5 mm. Each layer of 1 to 1.5 mm was light-cured for 40 seconds. As a control, PFC was applied to treated dentin surfaces without an intermediate FRC layer.

Microtensile bond strength test

After storage at 37°C water for 24 hours, the specimens were sectioned into 1.0×1.0 mm composite-dentin beams and further water-stored at 37°C for 30 days. After 30 days of water storage, μ TBS test was carried out using a microtensile test apparatus (Microtensile Tester, Bisco, USA) at a crosshead speed of 1.0 mm/min. Before testing, the cross-sectional area of each beam was precisely measured with a digital micrometer (Mitutoyo Corp., Tokyo, Japan; accuracy: ± 0.002 mm) and its ends were glued with a cyanoacrylate adhesive (Zapit, DVA, Corona, CA, USA). Microtensile bond strength was expressed in MPa, by dividing the tensile force (N) at fracture with the bonded surface area (mm²). Any specimens that failed prior to testing were discarded, and a record of the number of pre-test failures (failures that occurred during the cutting and gluing procedures) was kept.

After microtensile testing, the debonded surfaces were examined under a stereomicroscope (Wild M3B, Heerburg, Switzerland) at $\times 40$ magnification and the locations of failure were recorded. Failure modes were classified as adhesive, cohesive, or mixed failure.

Statistical analysis

Data were analyzed with two-way analysis of variance (ANOVA) for the following factors: bonding agent and restorative material. For interactions

between the factors, one-way ANOVA and Tukey's multiple comparison *post hoc* analysis were performed at a significance level of $p < 0.05$ with a statistical software, SPSS 14.0 (Statistical Package for Social Sciences, SPSS Inc., Chicago, IL, USA).

The μ TBS values of all the test groups were ranked in an ascending order, and Weibull analysis was performed to calculate the cumulative fracture probability (P_f) as a function of bond strength¹⁰⁻¹² using a software (Weibull++ software, ReliaSoft Corp., Tucson, Arizona, USA). The basic form of Weibull distribution is shown as follows:

$$P_f = 1 - \exp(-((\sigma - \sigma_\mu) / \sigma_0)^m)$$

where the constant m (Weibull modulus) determines the slope of the distribution function and which characterizes the spread of failure data with respect to σ (stress or load axis). Characteristic strength (σ_0) is the stress level at which 63% of the specimens fail, and σ_u is the theoretical failure stress at which the failure probability approaches zero and is known as the threshold stress.

The stress at 5% failure probability was calculated for each group, and failure probability curves were examined in an attempt to assess the distribution of microtensile bond strengths.

RESULTS

Figure 1 summarizes the mean μ TBS and standard deviation values obtained in this study. Two-factor ANOVA showed significant differences among the groups according to the adhesive material used ($p < 0.05$) (Fig. 1). The interactions between the factors were also significant ($p < 0.05$). Among the FRC groups, highest mean μ TBS (28.3 \pm 8.41 MPa) was obtained with conventional three-step system, followed by two-step self-etching primer system (27.9 \pm 8.59 MPa). For the PFC groups, mean μ TBS with the use of two-step self-etching primer system (34.5 \pm 11.9 MPa) was significantly higher than conventional three-step system and single-step self-etching adhesive system ($p < 0.05$). Both FRC and PFC groups showed the lowest μ TBS values

Table 2 Results for characteristic strength (σ_0) (MPa), Weibull modulus (m), and bond strengths (MPa) for 5% probability of failure ($\sigma_{5\%}$) of different bonding systems. For abbreviations, see Table 1

Adhesive	Material	σ_0	m	$\sigma_{5\%}$
IB	PFC	16.5	2.53	5.0
	FRC	17.1	2.26	4.6
CE	PFC	38.5	2.58	12.2
	FRC	30.7	3.50	13.1
MP	PFC	30.9	4.16	15.1
	FRC	28.3	5.48	16.5

(15.2±7.3, 15.1±9.1 MPa) by use of single-step self-etching adhesive.

Table 2 presents the Weibull analysis results. As seen in the table, characteristic strength (σ) (MPa), Weibull modulus (m), and bond strength for 5% probability of failure ($\sigma_{5\%}$) for different bonding agents were different. Conventional three-step system (Fig. 2) and two-step self-etching primer system (Fig.3) showed higher characteristic strengths, whereas single-step self-etching adhesive showed lowest characteristic strength with or without FRC (Fig. 4).

The main failure type for single-step self-etching adhesive groups, including the pre-test failures, was

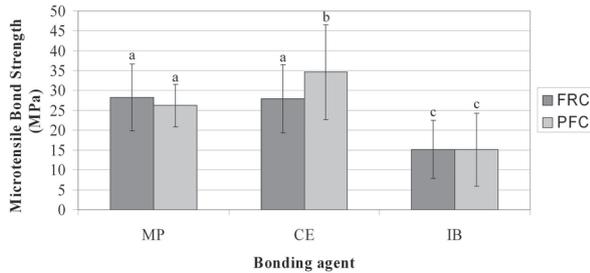


Fig. 1 Means and standard deviations of μ TBS of different test groups. For abbreviations, see Table 1. Vertical lines represent standard deviations. Groups with similar letters represent statistically similar subgroups (ANOVA, $p < 0.05$).

adhesive in nature with both PFC and FRC (Table 3). Conventional three-step system and two-step self-etching primer system showed fewer pre-test failures. The percentage of mixed or cohesive failures increased in FRC groups (Table 3).

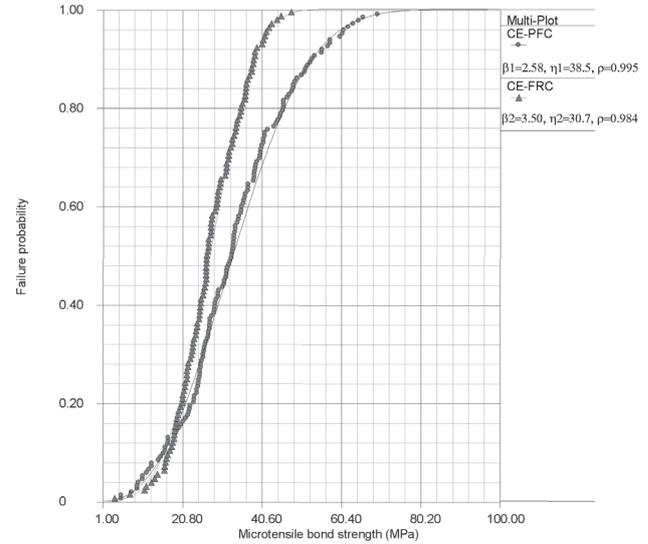


Fig. 3 Failure probability curves of CE with and without FRC at the interface.

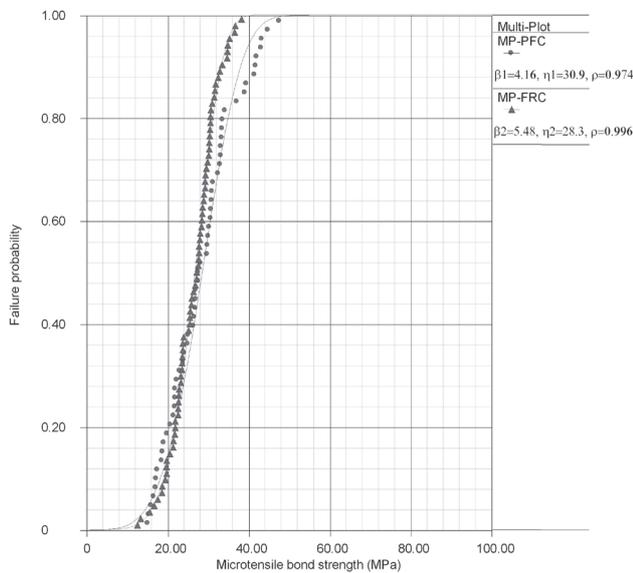


Fig. 2 Failure probability curves of MP with and without FRC at the interface.

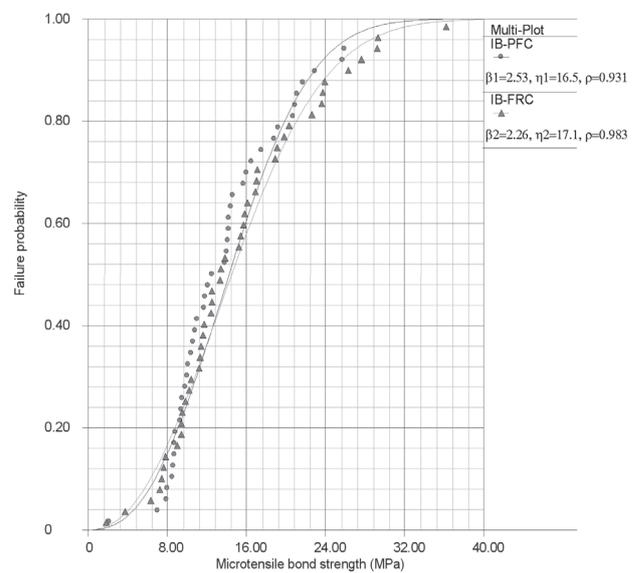


Fig. 4 Failure probability curves of IB with and without FRC at the interface.

Table 3 Failure analysis (%) of the test groups. For abbreviations, see Table 1

Adhesive	Material	Pre-test failures (%)	Adhesive(%)	Cohesive(%)	Mixed(%)
IB	PFC	37	100	0	0
	FRC	40	94	0	6
CE	PFC	0	93	0	7
	FRC	0	45	48	7
MP	PFC	0	97	0	3
	FRC	0	47	48	5

DISCUSSION

This study aimed to compare the performance of three different adhesive systems with a conventional particulate filler composite *versus* a semi-IPN FRC system after one month of water storage. Before microtensile testing, the specimens were aged in water. The water storage of sectioned test beams was considered to be more realistic as the reduced cross-sectional area of the specimens will lead to a direct exposure of the interface to water diffusion.

Single-step self-etch adhesive systems are composed of aqueous mixtures of hydrophilic and hydrophobic monomers with a view to combining the etching, priming, and bonding steps into a single step. A relatively high concentration of solvent is required, as water is an essential ionization medium to enable the self-etching activity to occur^{13,14}. During some clinical applications, such as root canal post application, it might be difficult for the solvent to evaporate. Against this backdrop, there arose a concern about the bonding efficacy of this highly solvent-containing system with the semi-IPN matrix of FRC, as this type of matrix contains a mixture of linear polymer together with cross-linked dimethacrylates⁶.

Results of the current study showed that the microtensile bond strengths of conventional three-step system and two-step self-etching primer system were significantly higher than that of single-step self-etching adhesive system with or without semi-IPN FRC. The very low bond strengths of single-step self-etching adhesives were noted and addressed in many previous studies¹⁵⁻¹⁹. Reasons for their poor performance were suggested to be due to the weaker cohesive strengths of the adhesives^{18,19} or the compromised polymerization of the adhesives¹⁶, whereby the latter was due to the combination of acidic hydrophilic and hydrophobic monomers into a single step. In addition, the increased permeability of the resin-dentin interfaces created by single-step self-etch adhesive systems may further contribute to their hydrolytic instability after aging in water¹⁶.

Taking all these factors into account, the bond strength results of the current study were thus consistent with those of previous reports showing low bond strength for IB¹⁷. Similarly, a high percentage of pre-test failures were observed for IB, as was thus reported in a previous study¹⁷. These failures were attributed to possibly poor collagen infiltration by IB¹⁵ or the presence of porosities due to monomer-solvent phase separation upon evaporation of acetone²⁰.

Despite the comparable mean bond strengths of IB obtained with PFC and FRC, the higher percentage of pre-test failures in the FRC group confirmed the concern about the bonding efficacy of IB with the semi-IPN matrix of FRC.

Results of Weibull analysis also confirmed this concern. The Weibull modulus (m) gives an indication of the reliability of the bond strength. A higher value indicates a more predictable failure behavior and a more homogeneous substrate-adherend interface. Additionally, with this statistical approach, a more relevant clinical approximation for the risk level of failure can be made. This is because the low stress levels at which the first 5-10% of the specimens failed were considered more clinically relevant to evaluate the reliability of the bond rather than the average strength values or very high values²¹. Despite the slightly higher characteristic strength (17.1 MPa) obtained with IB-FRC group compared with IB-PFC group (16.5 MPa), the m value of IB-FRC was lower (2.26) compared to that of IB-PFC (2.53), thus indicating a less predictable failure behavior. On the other hand, between the two-step self-etch adhesive and three-step conventional bonding system, the latter provided the highest m value (5.48) with FRC, indicating a more predictable bonding behavior. In the same vein, the 5% failure probability was 4.6 MPa for IB-FRC group, being remarkably lower than CE-FRC (13.1 MPa) and MP-FRC (16.5 MPa) groups. On the other hand, both CE and MP showed higher Weibull modulus and 5% failure probability values with FRC than with PFC. These results affirmatively indicated a more predictable bonding with semi-IPN composite

than with dimethacrylate-based composite. It is also noteworthy that these results were consistent with previous reports showing improved bonding properties with the use of semi-IPN FRC materials^{6,22}.

Failure analysis results were also consistent with μ TBS results. The highest percentages of adhesive failure were obtained with the IB group with or without semi-IPN FRC. For both CE and MP groups, the main failure type was cohesive failure inside the FRC material, thus indicating good bonding at both the adhesive-FRC and FRC-PFC interfaces.

In conclusion, results of the present study rejected the null hypothesis that different bonding systems result in comparable bond strengths. In addition, extra consideration and caution should be exercised when using one-step self-etch adhesives with semi-IPN FRC.

ACKNOWLEDGEMENTS

This study was supported by funding from the Academy of Finland. We would also like to thank CDT Jasmina Bijelic and CDT Milla Lähdenperä for their technical assistance during the preparation of the specimens.

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