The Bond Strength of Particulate-Filler Composite to Differently Oriented Fiber-Reinforced Composite Substrate

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Purpose: **The primary failure mode of fiber-reinforced composite (FRC) materials used intraorally is delamination or debonding of particulate filler composite (PFC), the esthetic veneer, from the underlying FRC framework. The objective of the current study was to evaluate the effect of unidirectional fiber orientation and load direction on the shear bond strength of PFC to FRC.**

Materials and Methods: **Unidirectional E-glass FRC was used as an adhesion substrate for the PFC. E-glass FRCs were oriented in three ways—Group A: in the plane perpendicular to the bonding surface; Group B: along the bonding surface longitudinal to the load; and Group C: along the bonding surface, transverse to the load. The FRC substrates were ground flat with 1200 grit. The PFC adherend was bonded to FRC using an intermediate resin. Twelve specimens for each group were water stored (37°C) for 3 days before a shear bond strength test was conducted.**

Results: **A one-way analysis of variance showed that the direction of the applied load to the fiber direction had a significant effect on the bond strength values ($p < 0.001$). A Weibull analysis produced values (characteristic strength and Weibull modulus) of Group A (46.5 MPa, 12.1), Group B (40.6 MPa, 4.6), and Group C (27.6 MPa, 3.5).**

Significance: **The highest shear bond strength values and Weibull modulus were obtained when the fibers were oriented perpendicular to the bonding surface, exposing the fiber ends to the PFC. Interface strategies between hybrid composite layers may be developed to exploit anisotropic behavior in the adherence between FRC and PFC.**

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INDEX WORDS: fiber-reinforced composite, bond strength, anisotropy, Weibull, composite

DURING THE LAST FEW DECADES, the development of adhesive dentistry has changed the concepts of restorative dentistry and directed considerable attention to minimally invasive, tissue-saving treatment alternatives.[1,2] Particulate filler composite resins (PFC) provide successful, durable intracoronal restorations; however, PFCs are reported not to have enough flexural strength to replace a missing tooth.[3] The combination of PFC with fiber-reinforced composites (FRC) for intraoral use has shown to have significant benefits in terms of mechanical properties.[4] Additionally, the possibility of direct chairside application,[5-7] and the ability to bond to tooth structure[8] make FRC an attractive candidate for dental applications.

Most traditional dental materials are isotropic, showing similar physical and mechanical behavior in all directions. Dentine and enamel are, however, naturally anisotropic materials.[9,10] Similarly, FRCs have properties that change from isotropic to anisotropic depending on the fiber orientation in the matrix. Some reports in the dental literature are available concerning anisotropic behavior of FRCs regarding flexural strength,[11] modulus of elasticity,[4] and thermal expansion.[12]

In all applications of FRC, there is a need to cover the FRC framework with a PFC. In contrast

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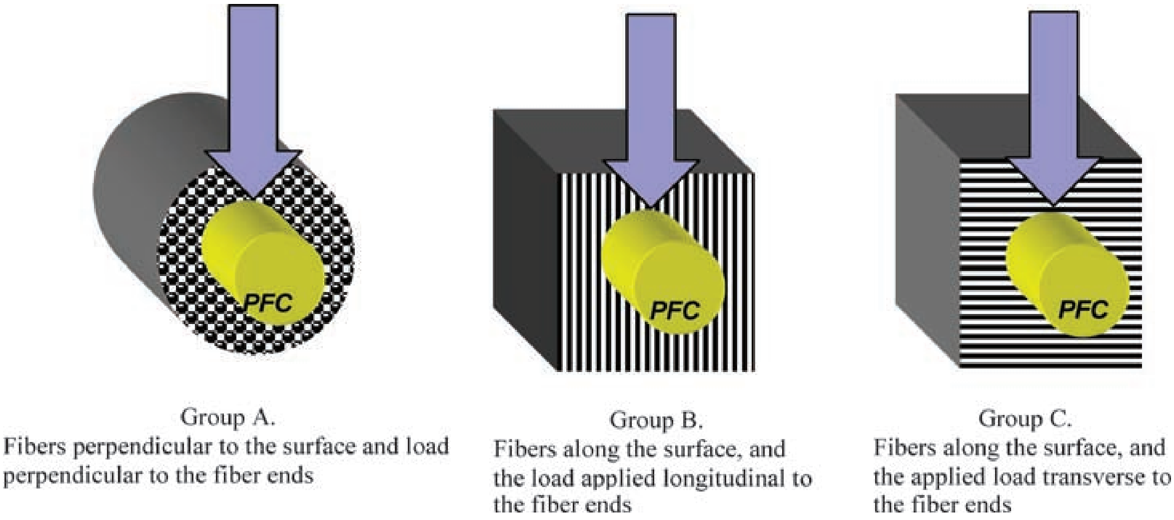
to the in vitro studies showing good adhesion between FRC framework and PFC,[13,14] the clinical failures reported in the literature were mostly delamination or debonding of veneering PFC from the underlying FRC framework.[15] Therefore, adequate bonding of the PFC to the FRC substrate is critical for the longevity of the restoration.

The traditional laboratory testing methodology used to characterize the adhesion between materials is based on shear bond strength measurements. Shear bond strength tests have been shown to be sensitive to many parameters.[16-18] Consequently, the results achieved between different test centers and testing devices are not always comparable. The specimen design and specimen preparation variables such as the height of the adherend material,[19,20] the thickness of the intermediate resin layer,[16,17] the polymerization conditions,[21] and the cross-sectional bonding area[22] have been reported to influence the shear bond strength values achieved. Despite these documented limitations,[16,17,23,24] the shear bond strength test has remained the standard method routinely used to evaluate the adhesion properties of dental materials to tooth substrate. Some studies have concluded that the tooth substrate anisotropy, specifically the orientation of the enamel prisms to the applied load direction, influences the bond strength values.[10,25] This finding raises the hypothesis that, like enamel, the anisotropic nature of FRC as a substrate may result in differences in the bond strength. There appear to be no reports available in the dental literature about the effect of anisotropy of an FRC substrate on shear bond strength measurements.

As a result, the aim of the current study was to evaluate the shear bond strength and fracture probability of PFC to FRC as a function of direction of load to the fiber orientation.

Materials and Methods

A commercially available FRC with continuous unidirectional silanated E-glass fibers, approximately 15 μm in diameter, preimpregnated with a polymer-monomer matrix of poly(methylmethacrylate) (PMMA)-bisphenol - A-glycidyl dimethacrylate (bisGMA) (everStick, Stick Tech, Turku, Finland) was used as the FRC substrate for this study. The FRC substrate was first placed in a cavity prepared in an acrylic resin block with different directions. The fiber directions of the substrate were—(A) in the plane perpendicular to the bonding surface; (B) along the bonding surface longitudinal to the load; and (C) along the bonding surface, transverse to the load (Fig 1). Following the initial light polymerization for 40 seconds, the FRC substrates were polymerized in a light curing oven (LicuLite, Dentsply DeTrey GmbH, Dreieich, Germany) for 15 minutes. After polymerization, the substrates were stored in water at 37 ± 1°C for 1 week, and at the end of the storage time, the substrates were wet ground flat using 1200 grit Federation of European Producers of Abrasives [FEPA] silicon carbide grinding paper. After grinding, the surface of the substrate was treated with an adhesion primer



Group A.
Fibers perpendicular to the surface and load perpendicular to the fiber ends

Group B.
Fibers along the surface, and the load applied longitudinal to the fiber ends

Group C.
Fibers along the surface, and the applied load transverse to the fiber ends

Figure 1. Orientation of the fibers; direction of the applied load is indicated by the arrow. PFC refers to the adherend particulate filler composite at the substrate surface.

Table 1. Materials Used

Brand	*Code*	*Manufacturer*	*Lot No.*	*Chemical Composition*
EverStick	ES	StickTech, Turku, Finland	201.0723.ES025	PMMA, Bis-GMA, E-glass unidirectional fibers ~65 vol%
Z250	Z25O	3M ESPE, St. Paul, MN	OHR	Bis-GMA,UDMA, Bis-EMA
Clearfil Repair*	CF	Kuraray Co., LTD, Osaka, Japan	41156	A: MDP, HEMA, Water, B: MDP, BisGMA, HEMA, Hydrophobic Dimethacrylate, silanated colloidal silica C: Bis-Phenol-A-polyethoxydimethacrylate, MPTS

*Clearfil bond procedure: A + C mixed and applied to the surface for 5 seconds and dried with mild air flow. B applied to the surface and light cured for 10 seconds.

PMMA = polymethyl methacrylate; Bis-GMA = bisphenol A-glycidyl dimethacrylate; UDMA = urethane dimethacrylate; Bis-EMA = bisphenol A polyethylene glycol diether dimethacrylate; HEMA = hydroxyethylmethacrylate; MDP = 10-methacryloyloxydecyl dihydrogen phosphate; MPTS = 3-methacryloxypropyl trimethoxysilane; A = SE bond primer; B = SE bond adhesive; C = porcelain bond activator.

(Clearfil Repair, Kuraray, Osaka, Japan) in accordance with the manufacturer's recommendations (Table 1). Following the surface treatment, 2-mm increments of PFC Z250 (3M ESPE, St. Paul, MN) was built up onto the FRC substrate at a height of 4.0 mm, using a polyethylene mold with an inner diameter of 3.6 mm, and polymerized with a hand-held light-curing unit (Optilux-501, Kerr, CT) for 40 seconds for each layer. The light intensity was 800 mW/cm^2 measured with the device's internal radiometer. Twelve specimens were prepared for each group, and the specimens were stored in water at 37 ± 1°C for 3 days prior to testing. The shear bond strength test was performed using a universal testing machine (model LRX, Lloyd Instruments, Fareham, UK) at room temperature (23 ± 1°C) and recorded using PC software (Nexygen, Lloyd Instruments Ltd.). The specimens were mounted to get the fibers in the orientation specified for Groups A, B, and C and secured in a mounting jig (Bencor Multi-T shear assembly, Danville Engineering Inc., San Ramon, CA) with the shearing rod against and parallel to the flat prepared bonding sites (Fig 2). A circular edge blade created the shear type load positioned over the FRC-PFC interface at a crosshead speed of 1.0 mm/min.

All fractured surfaces of the specimens were examined visually, and three randomly selected specimens from each group were gold-sputtered and evaluated with a scanning electron microscope (SEM) (JSM-5500, Jeol Ltd, Tokyo, Japan) to determine the failure pattern. In addition, the gold-sputtered substrate surfaces were analyzed prior to bonding using SEM-energy dispersive spectroscopy system to measure the area percentage of the fibers and polymer matrix at the bonding surface by aid of image analysis software (Spirit, PGT, Princeton, UK). The analyses were based on the difference in the elemental composition of the glass fibers compared with that of the polymer matrix.

The data for Groups A-C were analyzed statistically with a one-way analysis of variance (ANOVA) at the 95% significance level ($p < 0.05$) with SPSS (Statistical Package for Statistical Science, SPSS Inc., Chicago, IL) to establish the effect of substrate orientation on the shear bond strength. Additionally the fracture loads of each group were ranked in ascending order and a Weibull analysis was performed to provide a calculation of the cumulative fracture probability (P_f) as a function of the applied load.

A Weibull analysis was carried out using Equation 2 while the mean rank estimated fracture probabilities were calculated from Equation 1:[26]

$$P_{fe} = \frac{n}{N+1} \qquad (1)$$

where N is the total number of specimens in the group, and n was the ranking of the ordered number of the specimens.

Figure 2. The shear bond test setup.

$$P_f = 1 - \exp\left(-((\sigma - \sigma_u)/\sigma_o)\right)^m \qquad (2)$$

where m was the Weibull modulus, which determines the slope of the distribution function and characterizes the spread of the failure strength data with respect to σ; σ_o was the characteristic strength, namely the stress level at which 63% of the specimens failed; and σ_u was the theoretical failure stress at which the failure probability approached zero, known as the threshold stress (MPa). The correlation coefficient, *r*, was calculated with a linear regression analysis from the logarithmic transformations and *r*-values above 0.9 were considered statistically acceptable.[27]

Results

The one-way ANOVA indicated that the fiber direction of the FRC substrate had a significant effect on the bond strength values ($p < 0.001$). Group A (where the fibers were placed perpendicular to the surface) showed the highest bond strength values (44.8 ± 3.4 MPa), Group B demonstrated lower bond strength values (37.2 ± 7.4 MPa), and Group C (where the fibers were located transverse to the load) produced the lowest bond strength values (24.8 ± 6.3 MPa) (Figs 3 and 4). The Weibull parameters are summarized in Table 2. The characteristic strength values were as follows: Group A (46.5 MPa, 12.1), Group B (40.6 MPa, 4.6), and Group C (27.6 MPa, 3.5).

The image analysis study performed on the substrate surfaces prior to bonding revealed that the percentage of polymer matrix and fibers on the surface did not differ between the groups [Group A 67.3% (7.4), Group B 67.8% (2.2), Group C 66.4% (4.1)].

A visual examination of the fracture surfaces combined with SEM micrographs (Figs 5–7) showed qualitatively different results for each group after shear bond testing. For Group A, where the fibers were oriented perpendicular to the specimen surface, the fracture occurred within fiber ends in the matrix part (Fig 5a). The polymer matrix in Group A was partly cracked out from the surface, with the fiber ends exposed at the surface (Fig 6); however, the fracture did not penetrate deep into the FRC substrate. Fracture analysis of Group B (fibers longitudinal to the load) showed fiber breakage along the FRC surface (Fig 5b), and the fracture did not penetrate into the surface but continued along the surface, tearing the fibers from the surface. Group C (fibers transverse to load) showed fractures where the fibers were pulled out from the edge part of adhered material (Figs 5c and 7).

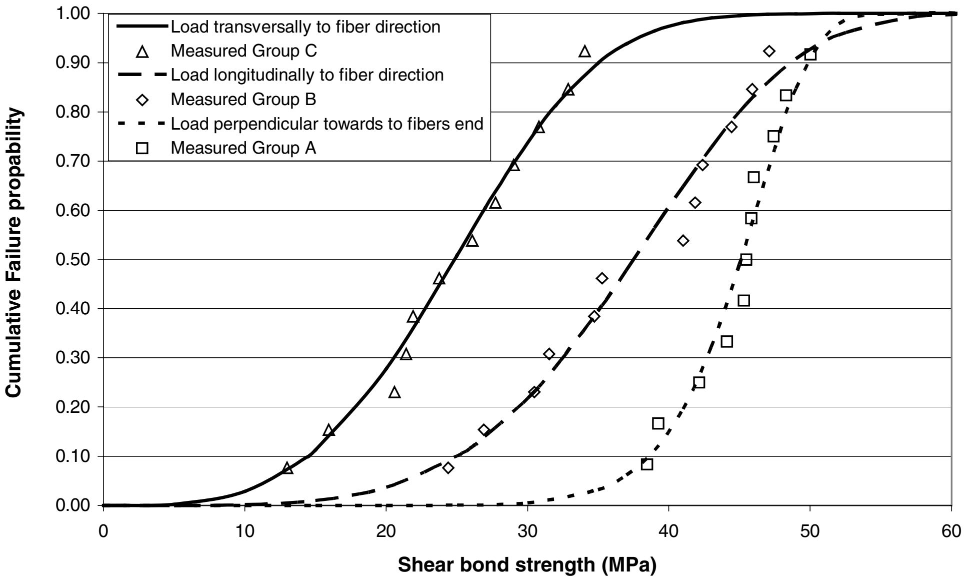


Figure 3. The cumulative failure probability of FRC to particulate filler composite loaded with the fibers placed in different directions.

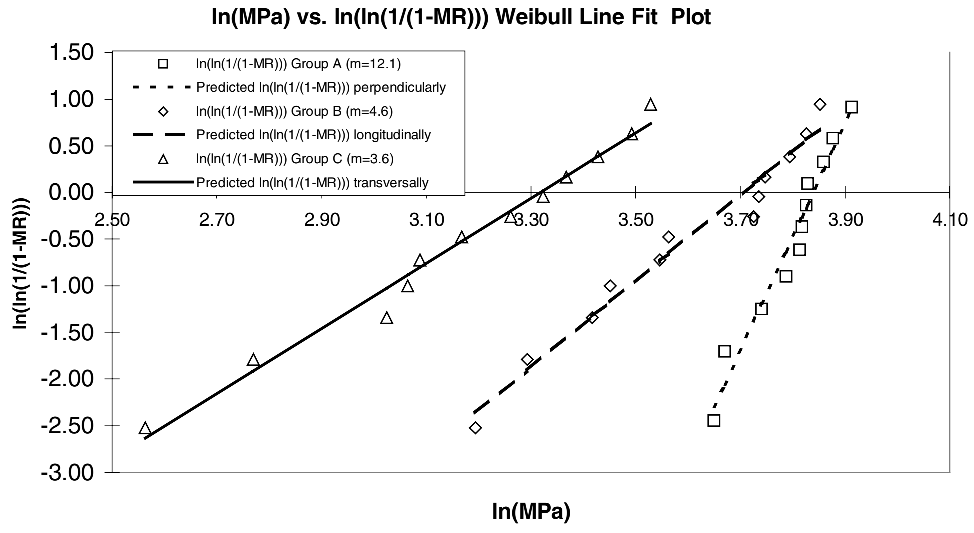


Figure 4. Weibull graph showing the slope of the distribution and characterizes the spread of the failure data.

Discussion

This study was designed to evaluate the anisotropic nature of FRC substrates with regard to their bonding using a shear bond test setup. A notable feature of the studies evaluating shear bond strength tests is the observation that failure mode is predominantly cohesive within the substrate and attributed to the nature of the stresses generated within the adherence zone.[17,23] FEA models indicate that the shear bond strength configuration develops tensile surface stresses within the substrate, close to the adherend-substrate interface edge, nearest to the applied shear load.[23] In the case of isotropic materials, the reason for cohesive bulk fractures was attributed to the unfavorable stress distribution, and therefore governed by the resistance of the base to the surface tensile stresses.[23] This might have great importance, especially when the base material is not isotropic but rather has different tensile strength properties depending on the orientations of the fibers and the interfacial adhesion between the fiber and the polymer matrix.[28,29]

The results of this study highlighted significant differences in bond strength values between PFC and FRC depending on the fiber orientation of the substrate. These results parallel the findings of previous studies concerning the anisotropic nature of tooth structure.[10,20,25] Enamel, with its prismatic, rod-like apatitic morphology, showed the highest bond strength, when loaded perpendicular to the enamel prisms. Similarly, glass fibers having a perpendicular orientation to the applied load resulted in the highest bond strength values; however, when fibers were oriented along the substrate surface, the bond strength values decreased. There was also a significant difference between fibers that were oriented longitudinally or transversally along the surface to the load direction. Longitudinally oriented fibers produced higher bond values, which could be caused by the

Table 2. Weibull Parameters and the Mean Shear Bond Strength and Standard Deviations According to Fiber Orientation

Parameter	*Group A Perpendicular*	*Group B Longitudinal*	*Group C Transversal*
Weibull modulus = m	12.1	4.6	3.5
Characteristic strength = σ_o (MPa)	46.5	40.6	27.6
r	0.96	0.98	0.98
Stress for 10% failure probability	38.7	25.0	14.5
Mean shear bond strength (MPa)	44.8	37.2	24.8
Standard deviation	3.4	7.4	6.3

Figure 5. Fracture surfaces. (*a*) Group A: original magnification 25×, (*b*) Group B: original magnification 35×, (*c*) Group C: original magnification 25×.

fibers' ability to carry the load along the direction of the applied load and share stresses among greater number of fibers at the margin from where the high stress concentration exists and debonding starts. Transversally oriented fibers could act as a crack stopper but the high stresses are not shared evenly among all fibers, causing each fiber interface to break sequentially, one fiber to another, causing fiber breakage along the whole adhered stub. The results of the present study are in agreement with a previous study by Scheirs,[28] which suggests that the fillers in the adhesive area can reduce the static adhesive strength if the filler aspect ratio is increased, especially if these fillers were transversally oriented.

Another important factor is the difference in the elastic moduli causing changes in the stress distribution. Previously it was suggested that the elastic modulus and compliance of the substrate have effect on bond strength values. This is particularly important with the use of an anisotropic material. It is well known that anisotropic materials have at least five or six independent elastic moduli depending on the packing symmetry of the system,[29] which may also cause dramatic changes in the elastic modulus of the substrate and may produce different bond strength results depending on the fiber orientation.

SEM fracture analysis revealed that adhesion between the FRC and the PFC was strong enough in Groups B and C (where the fibers were placed along the surface) to pull the fibers away from the surface causing a cohesional fracture pattern; however, SEM analysis also confirmed the different fracture mechanism, demonstrating in the transversally oriented FRC that the fibers were typically pulled away from whole area of adherend stub (Fig 5c), whereas in the longitudinally

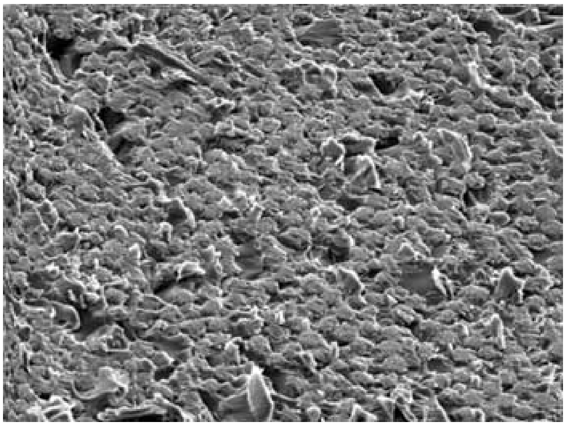


Figure 6. Fracture surface for Group A. At the fracture surface it can be seen that fiber ends are surrounded with fractured polymer matrix. Original magnification 300×.

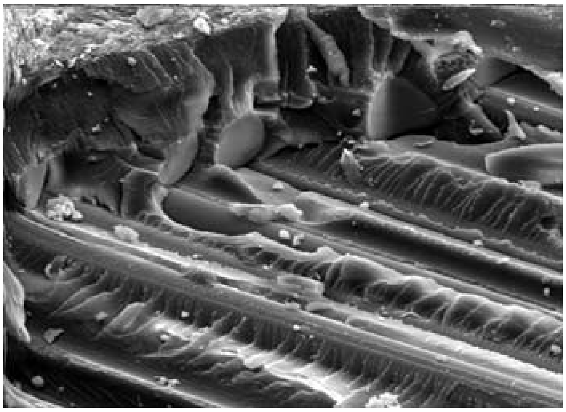


Figure 7. Fracture surface for Group C. Typical fracture mechanism is that outer layer of fibers are pulled away from the FRC surface. Original magnification 1000×.

oriented FRC, the fibers usually broke along the whole surface (Fig 5b). This difference may be due to the transversally oriented fibers' inadequate capability to carry and share the applied load evenly, which causes stress concentration and crack propagation. The crack is propagated sequentially to the next glass fiber, and when the fibers were oriented longitudinal to the load, the fibers were more likely to be pulled out and be cut. When one fiber becomes broken, other fibers carried the applied load. Contrary to the observations of the studies with ceramics and composites highlighting deep cohesive fractures of the substrate,[23] the cohesive fracture only stayed at the surface of the substrate and did not penetrate inside. Consequently, the load was carried by the fiber regions of the outer surface of the substrate. The interface between the FRC and the PFC may provide the most likely propagation pathway because of the stress concentrations from flaws or irregularities. A crack propagates along the fiber's surface; however, when fiber breakage occurs, the crack cannot penetrate into the substrate surface, but the crack continues along another fiber surface.

Group A, with perpendicular fibers, showed the highest bond strength, even though the fracture pattern could be categorized as an adhesive fracture. While the fracture line existed between the substrate and adhered material, a closer examination highlighted that the crack was propagated in the substrate by pulling pieces of the polymer matrix away from the fibers (Fig 5a). This suggests good adhesion of the adhered composite resin to the polymer matrix of the FRC, which has also been shown previously,[13,14] and the perpendicular fiber orientation may resemble the crack propagation of particulate composites, which can increase the toughness by transferring the stress to the strong particulate fillers that are well coupled to the resin matrix.

The Weibull characteristic strengths indicated similar behavior to the mean bond strength values; however, when Weibull modulus values are compared, Group A shows clearly higher values (12.1) compared with Groups B (4.6) and C (3.5), indicating a more reliable bond and less scattered bond values for Group A (Figs 3 and 4). The stress at the 10% failure probability was clearly higher for Group A (38.7 MPa), compared with Groups B (25.0 MPa) and C (14.5 MPa).

Conclusions

Within the limitations of this study, highest shear bond strength values were obtained when the fibers were oriented perpendicular to the bonding surface.

Clinically, this study might suggest how the PFC needs to be bonded to FRC to optimize the attachment of a veneering resin composite to FRC framework. During manufacturing of FRC crowns or fillings, in an optimal framework design the fiber should be placed perpendicular to the PFC veneering composite surface. Such an optimized design could simulate enamel structure, with its prismatic, rod-like apatite toward the surface. The risk of delamination of the PFM from the FRC could be diminished in this way. Technically the fabrication of such an optimized design needs further development. Furthermore, it should be emphasized that when the shear bond strength values with FRC materials are measured and reported, the anisotropic behavior of the FRC should be taken into consideration to avoid the false interpretation of the resultant bond strength data.

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References

1. Creugers NH, De Kanter RJ, van't Hof MA: Long-term survival data from a clinical trial on resin-bonded bridges. J Dent 1997;25:239-242
2. Botelho M: Resin-bonded prostheses: the current state of development. Quintessence Int 1999;30:525-534
3. Magne P, Perakis N, Belser UC, et al: Stress distribution of inlay-anchored adhesive fixed partial dentures: a finite element analysis of the influence of restorative materials and abutment preparation design. J Prosthet Dent 2002;87:516-527
4. Vallittu PK: Flexural properties of acrylic resin polymers reinforced with unidirectional and woven glass fibers. J Prosthet Dent 1999;81:318-326
5. Vallittu PK, Sevelius C: Resin-bonded, glass fiber-reinforced composite fixed partial dentures: a clinical study. J Prosthet Dent 2000;84:413-418

6. Ahlstrand WM, Finger WJ: Direct and indirect fiber-reinforced fixed partial dentures: case reports. Quintessence Int 2002;33:359-365
7. Freilich MA, Karmaker AC, Burstone CJ, et al: Development and clinical applications of a light-polymerized fiber-reinforced composite. J Prosthet Dent 1998;80:311-318
8. Tezvergil A, Lassila LV, Vallittu PK: Strength of adhesive-bonded fiber-reinforced composites to enamel and dentin substrates. J Adhes Dent 2003;5:301-311
9. Watanabe LG, Marshall GW Jr, Marshall SJ: Dentin shear strength: effects of tubule orientation and intratooth location. Dent Mater 1996;12:109-115
10. Spears IR, van Noort R, Crompton RH, et al: The effects of enamel anisotropy on the distribution of stress in a tooth. J Dent Res 1993;72:1526-1531
11. Dyer SR, Lassila LV, Jokinen M, Vallittu PK: Effect of fiber position and orientation on fracture load of fiber-reinforced composite. Dent Mater 2004;20:947-955
12. Tezvergil A, Lassila LV, Vallittu PK: The effect of fiber orientation on the thermal expansion coefficients of fiber-reinforced composites. Dent Mater 2003;19:471-477
13. Lastumaki TM, Kallio TT, Vallittu PK: The bond strength of light-curing composite resin to finally polymerized and aged glass fiber-reinforced composite substrate. Biomaterials 2002;23:4533-4539
14. Kallio TT, Lastumaki TM, Vallittu PK: Bonding of restorative and veneering composite resin to some polymeric composites. Dent Mater 2001;17:80-86
15. Gohring TN, Schmidlin PR, Lutz F: Two-year clinical and SEM evaluation glass-fiber reinforced inlay fixed partial dentures. Am J Dent 2002;15:35-40
16. DeHoff PH, Anusavice KJ, Wang Z: Three-dimensional finite element analysis of the shear bond test. Dent Mater 1995;11:126-131
17. Versluis A, Tantbirojn D, Douglas WH: Why do shear bond tests pull out dentin? J Dent Res 1997;76:1298-1307
18. Pecora N, Yaman P, Dennison J, et al: Comparison of shear bond strength relative to two testing devices. J Prosthet Dent 2002;88:511-515
19. Uno S, Asmussen E: Selected variables in bonding to dentin. Scand J Dent Res 1992;100:130-132
20. McDonough WG, Antonucci JM, He J, et al: A microshear test to measure bond strengths of dentin-polymer interfaces. Biomaterials 2002;23:3603-3608
21. Yanagawa T, Finger WJ: Relationship between degree of polymerization of resin composite and bond strength to Gluma-treated dentin. Am J Dent 1994;7:157-160
22. Phrukkanon S, Burrow MF, Tyas MJ: Effect of cross-sectional surface area on bond strengths between resin and dentin. Dent Mater 1998;14:120-128
23. Della Bona A, van Noort R: Shear vs. tensile bond strength of resin composite bonded to ceramic. J Dent Res 1995;74:1591-1596
24. van Noort R, Noroozi S, Howard IC, et al: A critique of bond strength measurements. J Dent 1989;17:61-67
25. Shimada Y, Tagami J: Effects of regional enamel and prism orientation on resin bonding. Oper Dent 2003;28:20-27
26. McCabe JF, Carrick TE: A statistical approach to the mechanical testing of dental materials. Dent Mater 1986;2:139-142
27. Robin C, Scherrer SS, Wiskott HW, et al: Weibull parameters of composite resin bond strengths to porcelain and noble alloy using the Rocatec system. Dent Mater 2002;18:389-395
28. Scheirs J: Compositional and Failure Analysis of Polymers: A Practical Approach. Chichester, UK, Wiley, 2000, pp 600-619
29. Nielsen LE: Mechanical Properties of Polymers and Composites. New York, NY, Dekker, 1974, pp 454-475