

Important Dental Fiber-Reinforced Composite Molding Compound Breakthroughs

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COLUMN ARTICLE

Fiber-reinforced composite (FRC) molding compounds with micromechanic requirements for fiber lengths longer than critical length (L_c) are now the most important breakthrough in Dentistry since the amalgam. The L_c is a measure of the minimum perfectly aligned fiber length dimension needed before maximum fiber stress transfer starts to occur within the cured resin [1-3]. Mechanical properties tested on flexural strength, yield strength, modulus, resilience, work of fracture (WOF), critical strain energy release (S_{ic}), critical stress intensity factor (K_{Ic}) and Izod impact toughness for FRCs using pure quartz fibers have shown large statistically significant increases over the dental particulate-filled composites (PFCs) as 3M Corp. Z100® and Kerr Corp. Herculite® [4-10] and a PFC with microfibers that cannot satisfy L_c as Alert® from Jeneric Pentron [5-8,10]. In addition, FRCs have shown large statistically significant increases for all mechanical properties tested except modulus when compared to a widely used amalgam alloy Tytin® from Kerr Corp. [9,10]. Further, FRCs have shown other greatly improved properties for wear less than enamel [10,11], significantly increased condensing packability force with significant larger interproximal contacts [10,12] and ability to incorporate the antimicrobial triclosan without PFC sticky glueyness [10]. Industrially FRCs are accepted as high-performance molding compounds that can pack with control to form into intricate geometric cavities and used extensively in the electrical and automotive industries so that FRC

development for Dentistry can proceed on firm dedicated principles.

Common problems with poor service longevity for dental PFCs when compared to amalgams [13-21] accentuate the importance for dental FRC molding compound use as an amalgam alternative [10]. Evidence-based randomized controlled clinical trials over 5 to 7 years have determined that the current PFCs used in dentistry fail at a rate 2 to 3 times greater than the amalgam [16,17,19,20]. Both the PFCs and amalgams generally fail due to secondary caries at the margins where PFC secondary caries failure rates have been shown to be 3.5 times greater than amalgams [17]. Recent accurate mechanical tests show that PFCs have an extremely low modulus compared to the amalgam modulus that can be compared much better to the modulus for enamel [9,10]. Subsequent lower dental PFC modulus filling material that deflects much greater should be more susceptible to increased interlaminar shear stress debonding at a higher modulus tooth adhesive interface that helps to account for more occlusal marginal leakage with related secondary decay [9,10]. On the other hand, FRC molding compounds can have much higher moduli that are closer to the modulus of amalgam [9,10]. Larger occlusal fillings and restorations with more than three surfaces then accentuate PFC failures [16,17]. Subsequent larger fillings with more margin exposure to occlusal loading would increase the probability that debonding occurs at a margin [9,10]. Also, the ADA has recommended that dental PFCs not be used in "stress-bearing" areas periodically since 1994 [22-24]. Al-

though accurate mechanical tests comparing dental PFCs to amalgams show superior PFC properties for strengths and fracture toughness [9], moisture adsorption greatly reduces dental PFC strength [25] that could be accelerated by low modulus PFC strain-related microcracking to determine the eventual fracture failure in larger PFC fillings [9,10].

PFCs have many other problems that can account for increased secondary caries compared to amalgams. PFCs wear at a much greater rate than amalgams with much deeper related marginal ditching [26] that would tend to collect more bacteria in a pool next to margins on the occlusal surface [10,26]. Also, PFC wear rates increase with wider cavity sizes correlated with reduced “sheltering” by the enamel margins [27]. In addition, PFCs were shown to require 7 - 8 times more repairs than amalgams [16]. Dental PFCs are extremely technique sensitive [28,29] while the amalgam is far easier to fill a cavity preparation than the PFC [30-36]. Dental PFCs require about double the time to finish as a similar amalgam [31]. The dental PFC is a tacky paste as a difficult material to pack due to low viscosity or consistency where matrix resins further have a tendency to adhere to packing instruments resulting in noticeable voids [37]. Subsequent sticky tack and low consistency in dental PFCs are then known to produce problems related to class II fillings with voids in the proximal box [9,10,38-43], overhangs difficult to remove [9,10,30,36] and inadequate interproximal contacts [30,32,34-36,39,44-55] with food impaction [34,56-59]. Also, poor interproximal contacts are associated with a higher caries risk [34]. Consequential plaque is found 3.2X more frequently on interproximal margins with dental PFCs than amalgam [60] and interproximal secondary decay has been detected 5.4 times more frequently on the gingival margin in dental PFCs than amalgams [15]. As a related concern for voids in the dental PFC proximal box, the ADA Council on Dental Materials expressed alarm early in 1980 standard requirements that radiopacity be measurable for the detection of voids on x-rays [40]. Conversely, fibers greatly increase polymer consistency so that FRCs pack with positive controlled pressure into complex mold cavities to prevent void formation such as in the proximal box [9,10,12]. Further, FRCs have shown large significant statistically improved reductions in voids over dental PFC polymerization shrinkage test samples ($p < 0.00001$), in-

creased interproximal contact areas over both high-viscosity Tytin® FC amalgam and Z100® PFC ($p < 0.0001$), and much higher packing forces than both high-viscosity Tytin® FC amalgam and Z100® PFC ($p < 0.001$) [10,12].

The FRC mechanical properties in a Z100® PFC matrix for different fiber lengths from L_c at approximately 0.5 mm for a 9 μm diameter quartz fiber and longer lengths up to 3.0 mm are compared with a Z100® PFC, Alert® PFC with microfibers well below L_c and Tytin® amalgam, Table 1 [9]. For the FRC with fiber lengths of 0.5 mm, at the L_c of 0.5 mm most of the fiber debonds from the polymer matrix that fails before the fiber breaks so that the full strength of the fiber can not be transferred through the composite. Consequently, small reductions in many mechanical properties occurred for the FRC at the 0.5 mm length when compared to the same polymer matrix composed of Z100® PFC. In fact, Alert® with microfibers well below L_c resulted in lower mechanical results for all properties when compared to the Z100® PFC probably due to a large extent from microfiber debonding that creates detrimental defects during the different loading conditions. But, as fiber lengths increase above L_c , increasing the fiber length to fiber diameter or aspect ratio increases strengths and modulus [5,10]. Further, increasing fiber length with aspect ratio above the L_c increases all fracture toughness properties for resilience, WOF, S_{lc} , and K_{lc} [5,10]. Subsequent lower mechanical properties for strength would then increase bulk fracture while lower fracture toughness properties would increase marginal chipping. Also, as fibers with some of the highest moduli known and above L_c bond well along the cavity walls at the occlusal margins interlaminar shear with the tooth and debonding related to secondary caries is expected to diminish greatly.

Fiber Length (mm) 30wt% (28.2 V _v)	Flexural Strength (MPa)	Modulus (GPa)	Yield Strength (MPa)	Resilience (kJ/m ²)	WOF (kJ/m ²)	S _{1c} (kJ/m ²)	K _{1c} (MPa*m ^{1/2})	Strain at Peak Load
0.0 mm (PFC) Z100® 3M	117.6 (0.0012)	19.5 (0.00102)	95.4 (0.01337)	3.03 (0.01882)	4.48 (5.1x10 ⁻⁵)	0.036 (0.16055)	1.71 (0.06584)	0.0079 (0.9740)
0.5 mm (FRC)	113.8 (0.1399)	23.0 (0.0008)	92.8 (0.0372)	2.35 (0.0400)	3.91 (0.0984)	0.075 (0.1794)	1.93 (0.07953)	0.0062 (0.9189)
1.0 mm (FRC)	173.6 (0.00318)	26.2 (0.001875)	126.1 (0.00018)	3.84 (0.00083)	8.7 (0.01879)	0.097 (0.0584)	2.77 (0.00797)	0.0084 (0.2993)
2.0 mm (FRC)	373.9 (5.2x10 ⁻⁵)	34.0 (0.00116)	329.8 (0.00168)	19.7 (0.00287)	28.2 (0.00046)	1.882 (0.0338)	11.01 (0.00579)	0.0121 (0.1290)
3.0 mm (FRC)	374.9 (2.2x10 ⁻⁸)	31.5 (0.01236)	343.5 (0.00014)	23.3 (0.00348)	30.1 (4.2x10 ⁻⁵)	2.4 (0.00296)	12.01 (3.89x10 ⁻⁵)	0.0131 (0.0677)
Alert® (PFC) with microfibers	90.4 (0.7057)	17.6 (0.00019)	62.3 (0.9791)	1.52 (0.0440)	3.23 (0.0077)	0.034 (0.2950)	1.33 (0.7523)	0.0069 (0.7130)
Amalgam Tytin®	86.0	43.6	62.6	0.67	1.40	0.013	0.91	0.0078

Table 1: Averages and T-test (p value) Comparisons Between Composites and Amalgam.

By related FRC strength improvements wear rates are reduced as fibers better support surface loading conditions and in particular as the fiber lengths become longer than the wearing plowing grooves [61,62]. FRCs with high modulus fibers reduce microcracking and water adsorption related to lower strain [63,64] that should further reduce wear. In fact, during a typical University of Alabama at Birmingham 3-body generalized wear simulator test at 400,000 cycles on a flat occlusal tooth sample corresponding to 3 clinical years of service FRC molding compound with fibers above L_c produced wear less than enamel. Accordingly, FRC wear produced a smooth filling material transition with the enamel margin. Conversely, the Alert® PFC with microfibers below L_c wears more than enamel to produce the characteristic material depression with ditch trenches at the enamel margins [10,11]. Profilometer tracings of the wear surfaces for an FRC with quartz fibers above L_c and the Alert® PFC with microfibers that were well below L_c are shown in Figure 1 [10,11]. Although amalgams wear much less than dental PFCs, amalgams still wear more than enamel to create small depressions at the margins [26] and microfills that wear slightly more than amalgams nevertheless fail from low fracture toughness properties resulting in marginal fracture [65,66]. While the polymer matrix is sheltered by nanoparticulate that fit closer together and do not debond by wear the larger particulate of microhybrid PFCs shear under

loading conditions into the polymer matrix and debond to accelerate wear and produce a much deeper marginal ditching trench [27].

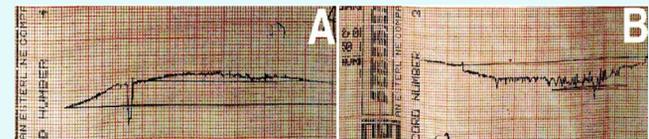


Figure 1: Profilometer wear tracings. (A) FRC with fibers above L_c with less wear than enamel transition with a smooth cleaner surface at the margin (B) PFC Alert® with microfibers below L_c show margins ditched with greater wear than enamel.

Scanning Electron Microscopy (SEM) of the same wear sample surfaces show the discontinuous chopped FRC molding compound with fibers greater than L_c to be vastly smoother and polished even at twice the magnification, Figure 2A, when compared to the rough surface for the PFC Alert® with short microfibers below L_c that lie in random planar fashion, Figure 2B [10,11]. Fibers above L_c will not be found on the wear surface where “sheltering” of the Z100® spherical nanohybrid particulate polymer matrix is accentuated by fibers that align parallel to the occlusal surface

from packing forces. An SEM of the FRC compound at much higher 5000X magnification shows how a high-strength 9 μm diameter quartz fiber wears by thinning until sufficiently skeletal to break up into fine flat plate-like particulate with sizes from much less than 200 nm to about 3 μm that press fairly level back into the Z100[®] PFC polymer matrix, Figure 2C. Subsequent thin flat particulate would not be expected to shear debond from the composite matrix by wear loading but rather break down further into smaller particles. Finer nanoparticulates that debond from the polymer matrix might then tend to even polish the FRC across a dryer surface above the flat sample enamel plane. On the other hand, depressions that exist following PFC wear would pool fluids and bacteria that tend to dissolve the polymer for increased wear and marginal secondary decay. Also, the FRC polymer matrix may experience creep from wear pressure that results in the scission of some polymer molecules thus leaving a free radical on either side of the dissociated bond. Subsequent dangling free radicals may possibly then reinitiate free-radical crosslinking of methacrylate end groups with coupling to quartz particulate and better help explain the ideal smooth wear surface created in Figure 2A.

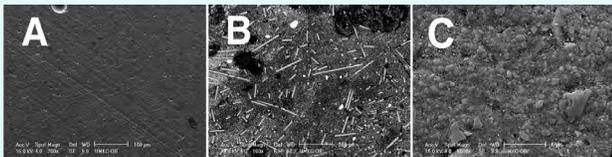


Figure 2: SEMs (A) FRC polished smoothly with extremely low wear surface 200X magnification scale bar 100 μm (B) Rough PFC Alert[®] with debonding microfiber wear surface debris 100X magnification scale bar 200 μm . (C) FRC 5000X magnification reveals how a quartz fiber wears thin into flat plate-like particulate producing the smooth surface in figure 2A, scale bar 5 μm .

PFCs have developed 3.2 times more plaque than amalgam on class II margins [60]. Leachable monomers of dental PFCs [67,68] have been found capable of supporting bacterial growth [67,69]. Further, dental PFCs can support decay under restorations that do not occur below amalgams under identical conditions [67]. However, amalgam has silver antimicrobial properties [70-74]. Similarly, high-viscosity FRC consistency allows incorporation of broad-spectrum

triclosan antimicrobial whereas lower viscosity PFCs are deprived of entire consistency and become gluey when triclosan is added by disrupting the resin and nanoparticulate weak secondary bonds [75-77]. The nonpolar or hydrophobic antimicrobial triclosan is a wetting agent to reduce viscosity during the mixing stage for resin and fiber incorporation, but and on the other hand is a toughening agent for the cured polymer to further increase flexural and adhesive bond strengths [75-77]. The hydrophobic or nonpolar principles for chemistry with triclosan should also reduce material breakdown by repelling polar molecules such as water and acid. An unusual odd alarmist triclosan controversy over bacterial resistance from unwarrantable extreme laboratory conditions that cannot be found in a normal micro-environment has been unjustifiable without any bacterial resistance to triclosan reported in over 40 years resulting in a government report recommendation for triclosan use wherever a health benefit is possible [76,77] as in dentistry. In some similar manner as triclosan disruption of secondary bonding needed for PFC consistency [75-77], incorporation of water-repelling hydrophobic low-viscosity resin that does not form secondary hydrogen bonds is not effective in providing PFCs with adequate consistency [10]. On the other hand, accentuated consistency with high-viscosity FRCs above L_c allows incorporation of more hydrophobic low-viscosity resin with a reduction in leachable monomer to suggest that much better polymer systems can be designed for future dental filling materials [10]. Also, higher FRC packing forces squeeze monomer, resin and particulate away from the molding compound fiber network and cavity margins to seal the adhesive bond with the insoluble high-modulus quartz fibers, nanofibers and particulate [4,9,10]. Subsequent elevated concentrations of insoluble fibers that align parallel to the cavity walls and occlusal plane with particulate along the margins should then better provide an enduring seal as a thin adhesive bond moisture barrier [9,10,63,78].

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