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STRENGTH, ABRASIVE WEAR, AND DURABILITY PROPERTIES OF DENTAL COMPOSITES CONTAINING A FLUORINATED DIMETHACRYLATE REACTIVE DILUENT

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Key Words: Dental Composites, Fluorinated Triethyleneglycol Dimethacrylate, Mechanical Properties, Scanning Electron Microscopy

ABSTRACT

The effects of the fluorinated triethyleneglycol dimethacrylate (F-TEGDMA) on the mechanical properties of composites made with this monomer, under both dry and aged conditions, was investigated. Photo-polymerizable formulations of 50/50, 30/70 and 10/90 mol:mol% F-TEGDMA/BisEMA (F5E5, F3E7 and F1E9), and 50/50 mol/mol% TEGDMA/BisEMA (T5E5) and TEGDMA/BisGMA (T5G5) were mixed with 76 wt% of silanated BaSiO₂ filler to form the experimental composite formulations. The experimental T5E5 and T5G5 formulations and a commercial composite HXR (Herculite XR, Kerr), having a similar formulation to the T5G5 group, served as the controls. The compressive strength (CS), diametral tensile strength (DTS), flexural strength (FS) and abrasive wear rate (WR) were determined and compared. Scanning electron microscopy (SEM) techniques were used to evaluate the fracture surfaces. ANOVA and Tukey-Kramer tests were used to determine between group differences.

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The study shows that replacing TEGDMA with F-TEGDMA does not significantly alter the CS, FS and WR but increases DTS of the composite materials. Conditioning in a simulated oral fluid of 75% alcohol for a 3 month period did not significantly affect the DTS of the materials. However, the alcohol conditioning did significantly affect the FS of all materials except material F1E9. This experimental formulation retained its original FS. This work suggests that small amounts of the fluorinated diluent may help resist chemical degradation in composites resin formulations.

INTRODUCTION

Dental composite resins, originally developed in the early 1960's, have been applied commercially to the dental profession for more than thirty years. The most commonly used monomers in composite resins include bisphenol A glycol dimethacrylate (BisGMA), ethoxylated bisphenol A dimethacrylate (BisEMA), and triethyleneglycol dimethacrylate (TEGDMA). BisGMA is still used as the main dimethacrylate oligomer and TEGDMA the main reactive diluent in dental composite materials formulations in the marketplace. These BisGMA resin based composites possess many advantages, including good mechanical properties, satisfactory esthetics, and good adhesion to acid-etched enamel surfaces. However, their clinical use has been somewhat limited to areas of low stress and easy access [1]. High water sorption, slow chemical degradation, along with high polymerization shrinkage and incomplete curing, can be detrimental to many physical and mechanical properties [2].

Interaction with the many substances in the oral environment may be a factor contributing to the long term durability of dental composites. It has been shown that BisGMA based polymers are highly susceptible to chemical softening. Its surface layer can be softened by chemicals with a solubility parameter ranging from $1.82-2.97 \times 10^4 (\text{J/m}^3)^{1/2}$ [3]. This solubility parameter range covers a wide variety of chemicals. It was assumed that the *in-vivo* degradation of non-stress-bearing composite restorations was appreciably influenced by the oral chemical environment. The observed surface damage was attributed to softening and possible removal of portions of the polymer matrix by certain chemicals [4]. The degradation of composites *in-vivo* has been simulated *in vitro* by the storing of composites in solvents, such as ethanol, which appear in FDA guidelines as an appropriate food-simulating liquid [3]. Some mechanical properties

of various composites have been reported to decrease after long-term storage in solutions and the aging effect appears to be dependent upon composition, as well as the testing mode [5, 6].

To enhance the service performance of dental composites, better chemical and water resistant monomers need to be introduced to the resin phase of the composites. One experimental approach has been to synthesize various fluorinated analogues of BisGMA to increase the water resistance of the resin matrix. Several studies have compared the mechanical properties of 2,2-bis[(p-2'-hydroxy-3'-methacryloxypropoxy) phenyl] hexafluoropropane (BisGMA-F) based composites to conventional BisGMA resins. The BisGMA-F based composite resins were found to retain a higher percentage of their initial properties than the conventional BisGMA composites when exposed to either water or thermal fatigue [7-9]. Recent studies in our laboratory, involving the use of dimethacrylates derived from 1,3-bis(hexafluoro-2-hydroxy-2-propyl) benzene, also suggest this to be a fruitful path for study [10]. With recent chemical processing advances [20], the use of fluorine modified TEGDMA (F-TEGDMA) has become another potential approach to enhance the clinical performances of dental composites [11]. Our previous studies have found that neat resin formulations made of F-TEGDMA/BisEMA show significantly lower water sorption than similar resin compositions formulated with TEGDMA [12]. As a continuation of this research, we have evaluated the effect of the F-TEGDMA on the mechanical properties and degradation resistance of select composite resin formulations.

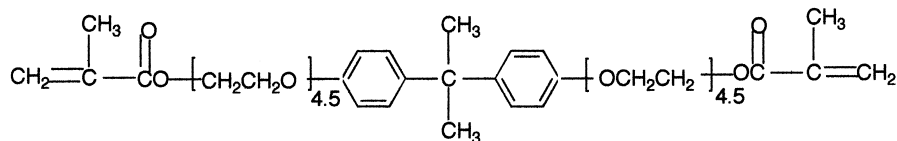
The purpose of this study was to investigate the effect of the fluorinated triethylene glycol dimethacrylate (F-TEGDMA) on the mechanical properties of composites in both dry and aged conditions. An ethanol/water (75/25 vol/vol) oral simulation liquid was used to store samples for up to 3 months. Further, scanning electronic microscopy (SEM) was used to analyze the fracture surfaces of the materials.

EXPERIMENTAL

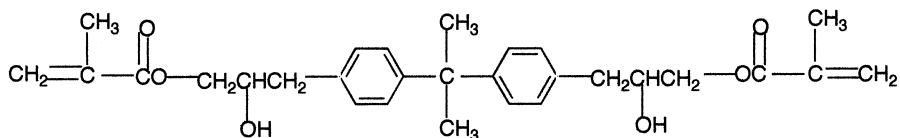
Materials

The resin matrix phase of the experimental dental composites investigated in this study consisted of various oligomer/diluent comonomer mixtures. Figure 1 summarizes the name, source, structure, and molecular weight (MW) of

Monomers / Oligomers

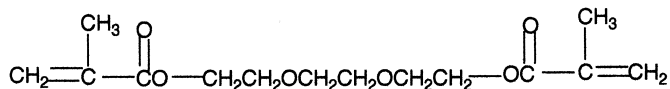


BisEMA (MW = 732.29), Esschem, Inc. (Essington, PA)



BisGMA (MW = 512.59), Cook Composites and Polymers (Port Washington, WI)

Diluents



TEGDMA (MW = 286.33), Aldrich Chemical Co. (Milwaukee, WI)



FTEGDMA (MW = 430.25), Exflour Research Corp. (Austin, TX)

Figure 1. Structures of the monomers or oligomers used in the study.

the oligomers and diluents used in the comonomer mixtures. The abbreviation code and brief descriptions of the materials used in the study are listed in Table 1. Since the novel F-TEGDMA was not miscible to any great extent with BisGMA, no F-TEGDMA/BisGMA mixtures could be evaluated. Commonly used T5G5 and T5E5 composition and a similarly formulated commercial product HXR (Herculite XR, Kerr) were used as controls.

The initiator camphorquinone (0.5 wt%) and accelerator 2-diethylaminoethyl methacrylate (1 wt%) were added to the comonomer solutions to allow for photo-polymerization. Filled composite resins were then prepared by mixing 76 wt% of silanated BaSiO₂ (average size 0.6 μm, supplied by Kerr Co.) with each of the photo-polymerizing comonomers to form composite pastes. The various pastes were formed into samples of various dimensions, visible light-

TABLE 1. Materials Used in the Study

Code	Material Description
HXR	Herculite (Kerr Dental Corporation, USA), 78 % (wt %) filler
T5G5	TEGDMA : BisGMA = 50 : 50 (mol %), 76 % (wt %) filler
T5E5	TEGDMA : BisEMA = 50 : 50 (mol %), 76 % (wt %) filler
F5E5	FTEGDMA : BisEMA = 50 : 50 (mol %), 76 % (wt %) filler
F3E7	FTEGDMA : BisEMA = 30 : 70 (mol %), 76 % (wt %) filler
F1E9	FTEGDMA : BisEMA = 10 : 90 (mol %), 76 % (wt %) filler

cured under nitrogen, and the selected mechanical and physical property evaluations were performed.

Mechanical Strength and Chemical Durability

Cylindrical specimens were prepared in glass molds, formed from tubing, with the dimensions of 4 mm in diameter by 8 mm in length and 4 mm in diameter by 2 mm in thickness for compressive strength (CS) and diametral tensile strength (DTS) tests, respectively. The ends of the specimens for CS tests were ground flat and parallel on a small lathe. The specimens for the flexural strength (FS) test were prepared using a split rectangular Teflon mold with a dimension of 2 mm in width by 2 mm in depth by 25 mm in length. The specimens were removed from the glass tubing and the mold after curing and divided into two groups. One group was tested directly and the other was conditioned in a 75% alcohol aqueous solution for three months.

Testing of specimens was performed on a Universal Testing Machine (INSTRON, Model 4202) with a cross-head speed of 0.5 mm/min for all CS, DTS and FS tests. The FS test was performed with the help of a four-point bending assembly, with a span of 20 mm between supports and 6.7 mm between two loading points. A sample size of six was used for each of the tests. Formulas 1, 2, and 3 were used to calculate the various strength values.

Calculation formulas used are shown in Formulas 1, 2, and 3.

$$CS = P/pr^2 \quad (1)$$

$$DTS = 2P/pDT \quad (2)$$

$$FS = Pl/WT^2 \quad (3)$$

where P = the load at fracture, r = the radius of the specimen, D = diameter of specimen, l = the distance between two supports, W = the width of the specimen, and T = the thickness of the specimen.

Abrasive Wear Resistance

The cylindrical specimens were prepared in glass tubing molds, with the dimensions of 3.5 mm in diameter by 5 mm in length. The specimens were conditioned in distilled water at 37°C for one week prior to testing. The wear resistance (WR) of the materials was determined using a pin-on-disc-type apparatus. The apparatus was designed to produce continuous sliding contact between the materials studied and a substrate disk (30- μm diamond abrasive disk, Beuhler Ltd., Lake Bluff, IL 60044), as shown in Figure 2. The sample and substrate surface was continuously flushed with tap water at a constant rate of 0.25 gallons per minute. Details of the apparatus were described previously by Seghi, *et al.* [13]. This apparatus has been used to study the abrasion of enamel against various ceramic surfaces and the abrasive wear rate of dentin [14]. The specimen cylinder was placed in the holder, and the loading and drive arm was activated. The specimen was then held in contact with the substrate surface with a 1.5 N load and traced a circular orbiting pattern around the diamond disk. The length of the sample was recorded at 2 second intervals for the entire test period with an LVDT to an accuracy of 0.0005 mm. The time of the experiment was controlled by an electronic timer, and the number of cycles was counted digitally. The substrate disk was cleaned after each trial. The wear rate of human enamel was also determined and used for comparison. The change in length of the samples was plotted against the number of revolutions and a linear regression analysis performed. The slope of the regression line was used to determine the wear rate ($\mu\text{m}/\text{rev}$) for each sample. Ten specimens were averaged to obtain a mean wear rate for each composite material.

Statistical Analysis

One-way analysis of variance (ANOVA) and Tukey-Kramer post-hoc multiple comparison test was used to determine significant differences between material groups at the 0.05 alpha level.

Scanning Electronic Microscopy (SEM)

The fracture surface of one specimen for each material from the FS tests were observed with a scanning electron microscope (Philips XL-30 CP, Philips Electronics N.V., Eindhoven, The Netherlands). The specimens were mounted on SEM stubs and then sputter-coated with gold in a vacuum. A high vacuum (5.5×10^{-5} mbar) was used for dehydration of the specimens before SEM analysis.

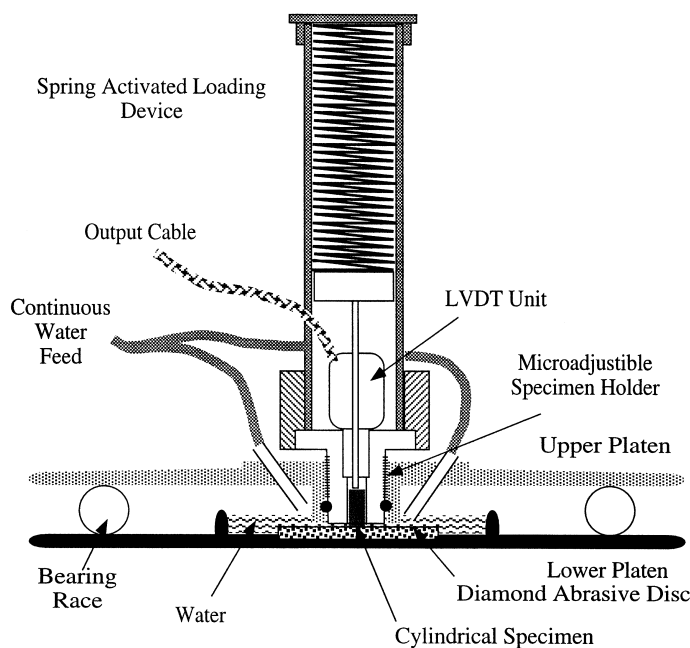


Figure 2. Schematic diagram of two-body abrasive wear apparatus.

RESULTS AND DISCUSSION

Strength and Abrasive Wear of Experimental Composites

The means and standard deviations of the flexural strength, diametral tensile strength compressive strength values in MPa and the wear rates in $\mu\text{m}/\text{rev}$. of the specimens under dry conditions are shown in Table 2. The results of the ANOVA test indicate that the polymer formulation significantly affects ($p < 0.01$) the resulting FS and DTS, but does not significantly affect the CS and WR of the composite materials. The results of the post-hoc test are indicated with superscripts. The values with the same superscript letters were not found to be significantly different at the 0.05 alpha level. Material F1E9 resulted in the lowest mean FS value while material F5E5 resulted in the highest mean DTS values. The materials formulated with the fluoro-substituted reactive diluent resulted in generally lower flexural strengths and higher diametral tensile strengths. Both the flexural and diametral tensile test are designed to estimate the tensile strength of a material. Both tests, however assume that the materials fracture in a substantially brittle manner.

TABLE 2. Mechanical Strength and Abrasive Wear of Composite Resins

Code	Flexural Strength ^A (MPa)	Diametral Tensile Strength ^B (MPa)	Compressive Strength ^B (MPa)	Wear Rate ^{C†} ($\mu\text{m}/\text{rev}$)
HXR	105.64 (8.28) ^{a,b}	50.58 (8.71) ^c	309.45 (27.26)	0.624 (0.067)
T5G5	114.05 (13.61) ^a	42.29 (3.18) ^c	362.84 (29.84)	0.615 (0.064)
T5E5	101.28 (7.59) ^{a,b}	64.37 (5.11) ^d	384.24 (22.06)	0.701 (0.077)
F5E5	101.13 (8.93) ^{a,b}	74.93 (3.81) ^e	307.04 (22.43)	0.643 (0.058)
F3E7	97.62 (10.19) ^{a,b}	65.32 (6.29) ^{d,e}	351.60 (25.42)	0.702 (0.070)
F1E9	92.69 (14.77) ^b	63.00 (6.54) ^d	316.82 (29.97)	0.645 (0.068)

Entries are mean values with standard deviations in parentheses. For each test, the strength values with the same superscript letters were not significantly different ($p > 0.005$).

† wear rate for control enamel is 0.217 (0.069) $\mu\text{m}/\text{rev}$

A n=5

B n=6

C n=10

The DTS test is designed to replace the direct tension test since the measured tensile strengths are often too low and produce big variations for brittle materials [19]. The DTS test however is significantly influenced by the modulus and flow characteristics of the material and is not considered appropriate for significantly elastic materials [17]. We speculate that the apparent increase in the DTS values of the F-TEGDMA groups are a direct result of the increased elasticity and/or plasticity of the polymer phase. This test may not be appropriate when comparing the different monomer formulations used in this experiment.

The FS test is designed to consider a combination of several stresses at the same time. This test is a simultaneous collective measurement of tensile (at the lower surface of the specimen beam), compressive (at the upper surface) and shear (in the direction which is parallel to the load) strengths [16]. Prosser, *et al.* [17] thought that the most appropriate measure of the strength was considered to be a flexural test because a material could only fail by the separation of the planes of atoms (i.e., tensile failure), or by the slipping of the planes of atoms (i.e., shear failure). Under the appropriate conditions, the mode of failure is primarily tensile stress applied at the sample surface. While this test suffers from being sensitive to surface flaw distributions, it is not as greatly affected by modulus changes and is more appropriate in evaluating these materials. Only the F1E9 material was significantly lower than the commercial HXR material. However, the latter difference in strength was less than 10% and is well within the range of many products used for clinical application.

Since the CS and WR values did not show significant between group differences, it was decided that these tests may not be sensitive to polymer differ-

ences and therefore not good candidates for the durability test. Many dental materials have a tensile strength that is markedly lower than the compressive strength. These materials fail by crack propagation that is favored by tensile rather than compressive loading. Degradation of material can cause flaws both on the surface and inside the material. Flaws do not play a significant role when the material is subjected to an external compressive force. The compressive stress that develops in the material tends to close the flaws or cracks, and thus stress distribution is more uniformly distributed [15]. In the case of abrasive wear, our test is a very aggressive wear test that measures the abrasive wear rate of the bulk structure and not just the surface. The alcohol degradation process has been shown to be confined to the outer surface of the specimen [3] which would not affect the wear rate of the bulk structure as tested with our device.

Chemical Durability

The mean FS and DTS values and standard deviations of the composite materials stored under both dry and alcohol conditions, are summarized in Table 3. The data in Table 3 suggests that the storage of the materials in alcohol reduced the mean DTS but the reduction was not significant. The flexural strength (FS) data, however, resulted in a significant reduction in the strength of most materials stored in the alcohol solutions. Only material F1E9 did not result in a significantly lower mean FS value. While all the other groups resulted in a significant degradation in FS, the F-TEGDMA containing formulations resulted in generally less degradation than the TEGDMA containing formulations, suggesting that the reactive diluent, F-TEGDMA, provides some type of degradation resistance to the materials.

TABLE 3. Chemical Durability of Composite Resin Strength Properties

Code	Flexural Strength (MPa)		Diametral Tensile Strength (MPa)	
	Dry*	Alcohol*†	Dry**	Alcohol**†
HXR	105.64 (8.28) ^{a,b}	16.27 (3.23) ^a	50.58 (8.71) ^a	43.54 (7.12) ^{a,b}
T5G5	114.05 (13.61) ^a	21.17 (3.27) ^{a,b}	42.29 (3.18) ^a	39.35 (5.43) ^a
T5E5	101.28 (7.59) ^{a,b}	31.11 (3.91) ^c	64.37 (5.11) ^b	55.35 (9.50) ^{b,c}
F5E5	101.13 (8.93) ^{a,b}	42.12 (2.04)	74.93 (3.81) ^c	71.60 (11.52) ^d
F3E7	97.62 (10.19) ^{a,b}	28.60 (1.53) ^{b,c}	65.32 (6.29) ^{b,c}	63.69 (4.28) ^{c,d}
F1E9	92.69 (14.77) ^b	95.18 (10.24)	63.00 (6.54) ^b	65.73 (6.03) ^{c,d}

Entries are mean values with standard deviations in parentheses. For each test, the strength values with the same superscript letters were not significantly different ($p > 0.005$).

†Materials were conditioned in 75% alcohol for three months.

*n=5

**n=6

Mechanical properties are closely related to structural features such as regularity of polymer structure, double bond conversion (degree of polymerization), molecular weight and inter- or intermolecular forces. In this study, the fluorinated triethylene glycol dimethacrylate (F-TEGDMA) monomer was used as a diluent to replace the regular TEGDMA and to combine with the BisEMA, with three molar ratios. As shown in Figure 1, most of the protons on the ethylene glycol were replaced by the fluorines in the F-TEGDMA, compared to the TEGDMA. These fluorines may play a key role in determining the changes in properties. Under dry conditions, there were no significant differences observed among the materials in the FS test and there were only a little differences observed among the materials in the DTS test. After being conditioned in alcohol for three months, significant decrease in FS were observed for all the materials except the F1E9, which contained less F-TEGDMA but more BisEMA. The exact mechanism of degradation resistance is unclear. Possible explanations may be related to the degree of polymerization, the solubility parameter of the matrix polymer and the filler/matrix interface. According to the thermodynamic solution theory [18], a maximum softening effect is expected when the value of the magnitude of the solubility parameter of a liquid is equal to that of the composite matrix polymer. Another issue for concern is associated with the particles used in this study, which were silanated by a non-fluorine containing silane coupling agent. More F-TEGDMA in the polymer matrix may lead to higher chemical resistance of the matrix, but this may also alter filler/matrix bonding.

SEM Analysis

Figures 3-5 are SEM photomicrographs (2723 X) of the fracture surfaces of select materials shown in Tables 2 and 3, i. e., SEM photomicrographs of materials F1E9, T5G5 and HXR, respectively. Figures 3A, 4A, and 5A represent the fracture surfaces of the specimens stored under dry conditions, while Figures 3B, 4B, and 5B show the fracture surfaces of the specimens stored in alcohol/water for three months. Figure 3A and 4A appear very similar in surface appearance. These materials have identical solid filler phases with different polymer matrices. The roughened surfaces in these photos and the particles on the fractured surface do not appear as cleanly defined and the appearance of plastic deformation within the polymer phase is evidenced by the small tag-like processes extending out of the surface.

Figure 5A appears to have a slightly different surface pattern than the two experimental materials shown in 3A and 4A. The commercial product is similar in composition and filler content to material T5G5 (Figure 4A) with the excep-

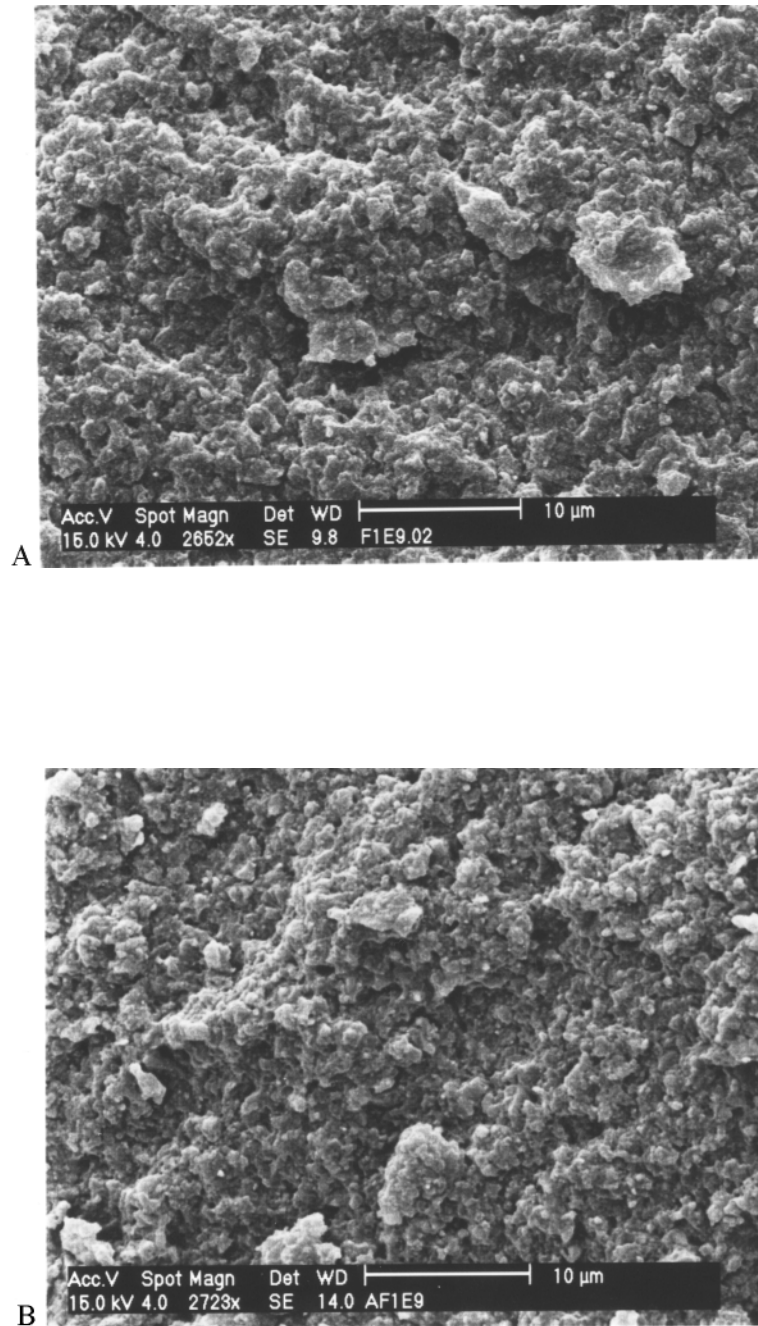


Figure 3. The fracture surfaces of the F1E9 material formed by a four-point being flexure test. The A (top) SEM photomicrograph is under the dry condition, while the B (bottom) SEM photomicrograph is for the alcohol/water conditioned sample.

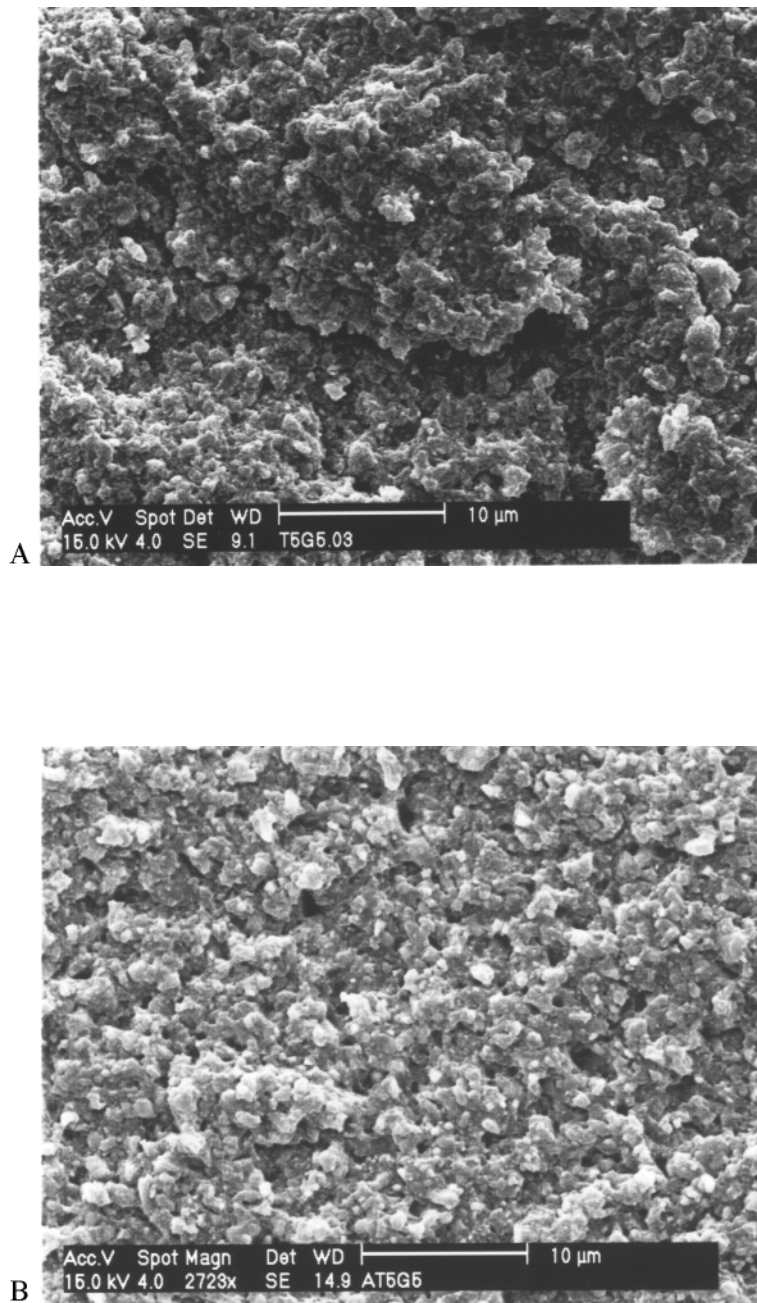


Figure 4. The fracture surfaces of the T5G5 material formed by a four-point bending flexure test. The A (top) SEM photomicrograph is under the dry condition, while the B (bottom) SEM photomicrograph is for the alcohol/water conditioned sample.

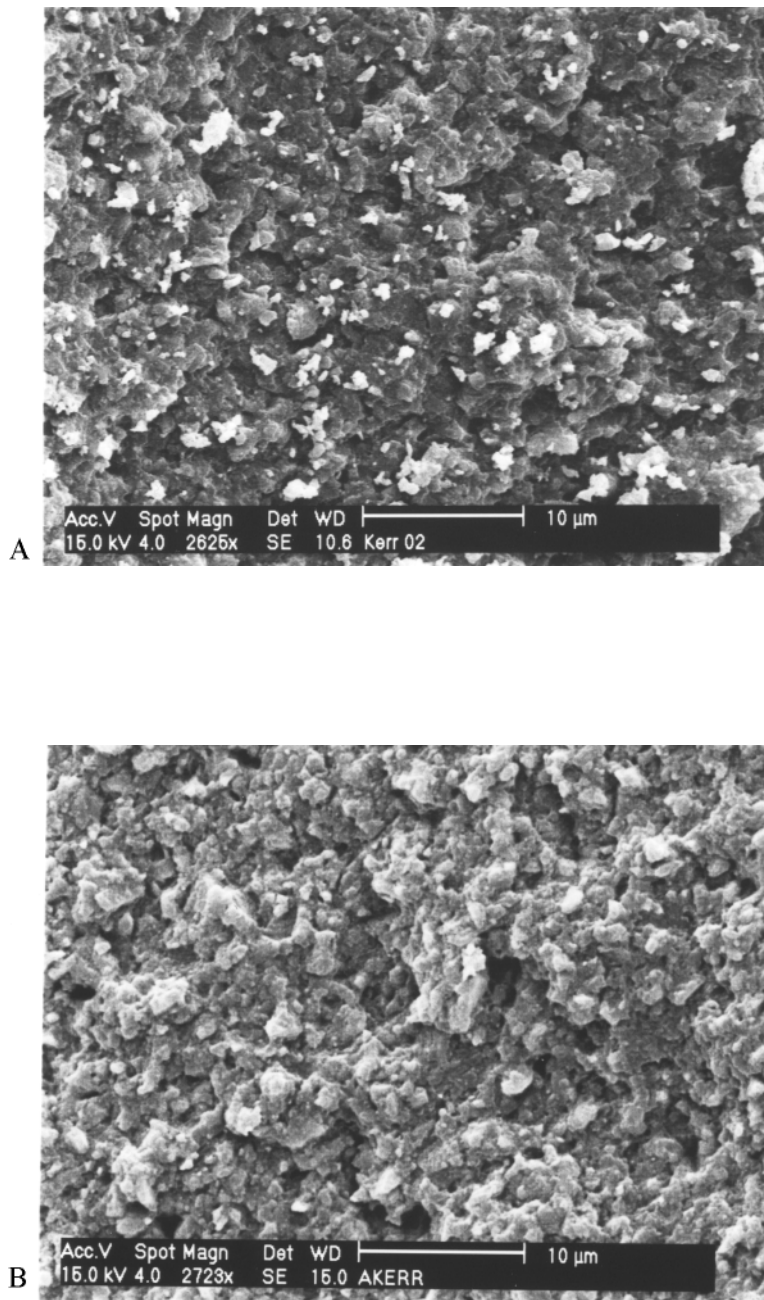


Figure 5. The fracture surfaces of the commercial HXR material formed by a four-point bending flexure test. The A (top) SEM photomicrograph is under the dry condition, while the B (bottom) SEM photomicrograph is for the alcohol/water conditioned sample.

tion that an additional small percentage of colloidal silica particles are also added. These particles tend to agglomerate and are likely to be the white particles/clusters seen in Figure 5A.

Figures 4 and 5 show significant differences between the undegraded (dry storage) and chemically degraded (alcohol/water stored) surfaces. The degraded surfaces appear to cleanly expose the surfaces of the filler particles with no resin tags evident. This appearance suggests that the fracture path travels along the filler matrix interface. The lower fracture strengths and different fractured surface topography suggest that some chemical interaction has occurred between the storage medium and the composite material.

Unlike the HXR and T5G5 materials, F1E9 (Figure 5) does not show an obvious difference in appearance of the fracture surface between the dry and alcohol/water stored composite materials. The absence of a mean strength decrease and the similarity in appearance suggest that the chemical effect within this material group was not significant. The exact mechanism of this alcohol resistance is not clear. Previously, we have shown that the F-TEGDMA containing monomers significantly decreased the amount of water sorption. While it is likely that the hydrophobic nature of the polymer contributes to the improved durability, it cannot be solely responsible since the F-TEGDMA formulations were slightly more hydrophobic yet did not result in similar durability. Degree of polymerization, oxygen inhibition, molecular packing, as well as intermolecular bonding all may play a role in the process. Further investigations are needed.

CONCLUSION

Replacing TEGDMA, which is a commonly used diluent in commercial dental composites, with F-TEGDMA does not significantly affect the CS, FS and WR, but increases DTS of the composites in the dry condition. No significant differences were found for the DTS test in each material, after conditioning in alcohol. For FS, all materials studied significantly degraded when stored in alcohol for three months, except the F1E9 material, which retained its original FS.

SEM photomicrographs of F1E9 showed that the fracture surfaces of dry and alcohol treated specimens are similar. The photomicrographs of all the other materials studied showed loose and more exposed glass particles in the alcohol treated composite resins compared to the non-alcohol treated groups.

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REFERENCES

- [1] R. Talib, *J. Nihon. Univ. Sch. Dent.*, 35, 161-70, (1993).
- [2] K. F. Leinfelder, *Adv. Dent. Res.*, 2(1), 115-121, (1988).
- [3] W. Wu and J. E. McKinney, *J. Dent. Res.*, 61(10), 1180-83, (1982).
- [4] W. Wu, E. E. Toth, J. F. Moffa, and J. A. Ellison, *J. Dent. Res.*, 65, 675-680, (1984).
- [5] E. Asmussen, *Scand. J. Dent. Res.*, 92, 257-261, (1984).
- [6] J. L. Ferracane, R. C. Antonio, and R. S. Mathis, *J. Dent. Res.* 66, Abst. No. 1112, (1987).
- [7] M. Kawaguchi, T. Fukushima, and T. Horibe, *Dent. Mater. J.*, 7(2), 174-181, (1988).
- [8] M. Kawaguchi, T. Fukushima, and T. Horibe, *Dent. Mater. J.*, 8(1), 40-45, (1989).
- [9] J. Tanaka, K. Inoue, H. Masamura, K. Matsumura, H. Nakai, and K. Inoue, *Dent. Mater. J.*, 12(1), 1-11, (1993).
- [10] B. M. Culbertson, A. Tiba, and P. E. Cassidy, IADR, Abs. No. 1364, (1995).
- [11] G. Wang, R. R. Seghi, and B. M. Culbertson, IADR, Abs. No. 2239, (1996).
- [12] G. Wang, E. Ezzie, R. R. Seghi, IADR, Abs. No. 494, (1997).
- [13] R. R. Seghi, S. F. Rosenstiel, and P. Bauer, *J. Dent. Res.*, 70(3), 221-225, (1991).
- [14] R. R. Seghi, G. Wang, W. M. Johnston, and B. M. Culbertson, *J. Dent. Res.*, 73, 394, (1994).
- [15] K. J. Anusavice, *Phillips' Science of Dental Materials*, 10th Ed., W. B. Saunders Company, 71-72, (1996).
- [16] R. G. Craig, in *Restorative Dental Materials*, Mosby-Year Book, Inc., (1997).
- [17] H. J. Prosser, D. R. Powis, and A. D. Wilson, *J. Dent. Res.*, 65(2), 146-148, (1986).

- [18] J. H. Hildebrand and R. L. Scott, *Solubility of Non-Electrolytes*, 3rd Ed., Reinhold Publishing Co., New York, NY, 1950, pp. 424-434.
- [19] K. J. Anusavice, in *Phillips' Science of Dental Materials*, W. B. Saunders Company, 1996.
- [20] R. J. Lagow, T. R. Bierschenk, T. J. Juhlke, and H. Kawa, *A New Synthetic Procedure for the Preparation and Manufacture of Perfluoropolyethers in Synthetic Fluorine Chemistry*, G. Olah, R. Chambers, and G. Prakash, Editors, John Wiley and Sons, Inc., 1992.

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