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The effect of aging methods on the fracture toughness and physical stability of an Oxirane/Acrylate,Ormocer, and Bis-GMA based resin composites

Hamad Algamaiah^{1,2}, Robert Danso³, Jeffrey Banas¹, Steve R. Armstrong¹, Kyumin Whang³, H. Ralph Rawls³, Erica C. Teixeira¹

¹Operative Dentistry Department, University of Iowa College of Dentistry, Iowa city, IA, 5222

²Department of Restorative Dental Science, King Saud University College of Dentistry, Riyadh, Saudi Arabia, 11545

³Department of Comprehensive Dentistry, The University of Texas Health Science Center at San Antonio School of Dentistry, San Antonio, TX, 78229

Abstract

Purpose: To determine the effect of aging methods on the fracture toughness of a conventional Bis-GMA based resin composite (Filtek Supreme), an ormocer-based resin composite (Admira), and an experimental hydrophobic Oxirane/Acrylate interpenetrating network resin System (OASys)-based composite.

Methods: A 25 × 5 × 2.8 mm stainless-steel mold with 2.5 mm single edge center notch, following ASTM standards [E399–90], was used to fabricate 135 specimens (n=15) of the composite materials and randomly distributed into groups. For the baseline group, specimens were fabricated and then tested after 24-hours storage in water. For the biofilm challenge, specimens were randomly placed in a six well tissue culture plate and kept at 37°C with bacterial growth media (Brain Heart Infusion (BHI); *Streptococcus mutans*) changed daily for 15 days. For the water storage challenge, specimens were kept in 5 ml of deionized distilled autoclaved water for 30 days at 37°C. μ CT evaluation by scanning the specimens was performed before and after the proposed challenge. Fracture toughness (K_{IC}) testing was carried out following the challenges.

Results: μ CT surface area and volume analyses showed no significant changes regardless of the materials tested or the challenge. Filtek and Admira fracture toughness was significantly lower after the biofilm and water storage challenges. OASys mean fracture toughness values after water

Correspondent author: Erica C. Teixeira, Operative Dentistry Department, University of Iowa College of Dentistry, S-243 Dental Science Building, The University of Iowa, Iowa City, Iowa 52242, erica-teixeira@uiowa.edu, +1 319 335 7270.

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aging were significantly higher than that of baseline. Toughness values for OASys composites after biofilm aging were not statistically different when compared to either water or baseline values.

Conclusion: The fracture toughness of BisGMA and ormocer-based dental resin composites significantly decreased under water and bacterial biofilm assault. However, such degradation in fracture toughness was not visible in OASys-based composites.

Clinical significance: Current commercial dental composites are affected by the oral environment, which might contribute to the long-term performance of these materials.

INTRODUCTION

Recent resin-based dental composite materials have improved in regard to their physical and mechanical properties becoming both patients' and clinicians' first choice for restorative dentistry. As such, over 800 million dental composite restorations were placed in 2015. However, a meta-analysis shows that by 2025, 32 million of these restorations will be replaced or repaired due to fracture [1]. Therefore, the mechanical and physical properties of direct restorative filling materials require further improvement and new materials should be evaluated before widespread clinical use.

In particular, it has been shown that dental resin composite suffers from degradation when in contact with the oral environment [2]. Water absorption into the composite can contribute to leaching of unreacted monomer and chemicals, and degradation can lead to leaching of resin byproducts [3–5]. These phenomena can lead to defective margins and loss of retention [6, 7]. Beyond concerns with clinical durability, leached materials affects biocompatibility [8, 9]. *In-vitro* studies have used water for different amount of time to promote a challenge environment for the specimens.

To exasperate the situation, *in-vitro* studies have shown that resin monomers that have leached into the oral cavity can alter the expression of virulence factors and promote the growth of cariogenic bacteria [10]. Additionally, biofilms form more preferably on resin composites as compared to other restorative materials [11, 12].

Thus, Bis-GMA-alternative monomer compositions have been introduced to improve durability. One alternative is the ormocer-based dental composite material (Organic-Modified ceramic), which consists of inorganic-organic co-polymers. Ormocer-based composite materials were proposed to have less polymerization shrinkage than conventional Bis-GMA-based composites due to its larger three dimensionally cross-linked ceramic polysiloxane monomer [13]. Cavalcante et al. [14] also showed that pure ormocer composites (no added acrylate-based monomers) better maintained surface integrity *in vitro* after seven days of water storage and had the least hardness change after seven days of ethanol storage as compared to a partial ormocer composite and a bis-GMA-based resin composite. However, a recent systematic review has shown that ormocer composite materials have a higher global failure rate and poorer long-term clinical behavior than conventional resin composites [15].

More recently, an experimental resin material based on an Oxirane/Acrylate interpenetrating network resin **System** (OASys) was reported to have increased hydrophobicity and decreased polymerization shrinkage stress. Both of these properties were due to the inherent properties of oxirane and its ring-opening polymerization mechanism [16]. The addition of a fluorinated acrylate to an OASys-based composite further increased contact angles to great than 115° [17].

Fracture toughness and flexural strength have been proposed as mechanical properties that have some correlation with the clinical performance of composite resin restorations. Illie et al (2012) showed that the fracture toughness of sixty-nine direct tooth-colored materials measured after 24 hours in distilled water (baseline) varied according to each material [18]. In addition, Lin and Drummond [19] observed that a 50/50 mixture of ethanol and water was the most detrimental aging media to a nano-filled, hybrid, and micro-filled dental composites after cyclic loading. However, no study has evaluated the effect of a biofilm challenge on the fracture toughness of dental composite materials.

Thus, the objective of this work is to determine the effect of water and bacterial (*Streptococcus mutans*) biofilm challenge on Bis-GMA-based (Filtek Supreme Ultra), ormocer-based (Admira Fusion) and OASys-based dental composite degradation and fracture toughness (K_{IC}).

MATERIALS AND METHODS

Samples preparation

A stainless-steel mold of $25 \times 5 \times 2.8$ mm with a 2.5 mm single edge center notch, following ASTM standards (E399–90 [20]) was used to fabricate specimens using three different dental composite materials (n=45) [Table 1]. Specimens were light cured with a polywave curing light (Valo light-curing system, Ultradent Products Inc., USA) on both sides to deliver a 20 J/cm² radiant exposure. The specimens were polished with aluminum sandpaper 600–1200 grit-size (Buehler, Illinois, USA) for 10 seconds on each side, then carefully examined to ensure that no surface defects were present.

Specimens were randomly and equally assigned (n=15) to nine groups after 24 hours in water [Figure 1] as follows: G1: Bis-GMA-based dental composite (Filtek) baseline (control); G2: Ormocer-based dental composite (Admira) baseline (control); G3: Experimental oxirane/acrylate-based dental composite (OASys) baseline (control); G4: Filtek with 15-day biofilm challenge; G5: Admira with 15-day biofilm challenge; G6: OASys with 15-day biofilm challenge; G7: Filtek with 30-day water storage; G8: Admira with 30-day water storage; G9: OASys with 30-day water storage.

Biofilm challenge

Streptococcus mutans (ATCC, 215175, American Type Culture Collection) was revived from frozen stocks on Trypticase Soy Agar (TSA) with 5% sheep's blood for 24 hours. Colonies from the blood agar plates were inoculated into Brain Heart Infusion (BHI) broth using a sterile Q-tip. Specimens were sterilized and placed in six wells tissue culture plates (Falcon 353046, Corning Inc.) and kept at 37° C. BHI medium was then added and

supplemented with 0.5% sucrose for 24 hours to promote the establishment of a biofilm on the composite sticks. The (BHI) growth medium was changed every day for the 15 days [21].

Water challenge

Specimens were randomly placed in tissue culture plates (Falcon 353046, Corning Inc.) with 5 ml deionized distilled autoclaved water and kept at 37°C. The autoclaved water was changed once every week for 30 days.

MicroCT evaluation

After 24 hours in water, specimens (n=3) from each group were randomly selected for baseline surface area and volumetric μ CT evaluation (Xradia Versa 3D XRM-520, Zeiss, Germany). Specimens were then submitted to the proposed challenges then scans were repeated. The x-ray settings were as follows: voltage (kV): 90, power (watts): 8, rotation: 360, projections: 1601, exposure time (seconds): 1.25, objective: 0.4x, reconstructed pixel size: 25 microns, beam hardening correction, reconstruction binning: 1. These settings were based on composite density. The constructed 3D models of the pre/post challenge were analyzed using Object Research System (ORS) software (ORS, Canada) for surface area and volumetric analyses. The software reconstructs the 3D model using a specific region of interest (ROI) for each composite material based on the material density.

Fracture toughness

Fracture toughness testing was carried out using a universal test machine (Zwick Roell Group, Germany) with a load cell capacity of 500 N. Single edge notch (SENB) three-point bending was performed with a 20 mm span at a cross-head speed of 0.5 mm/min. The width, thickness, and the notch length of each specimen were carefully measured and verified using a digital caliper.

The mode I critical stress intensity factor (K_{IC}) was calculated using the following equation:

$$K_{IC} = \frac{3PL}{2BW^{3/2}} \left\{ 1.93 \left(\frac{a}{W} \right)^{1/2} - 3.07 \left(\frac{a}{W} \right)^{3/2} + 14.53 \left(\frac{a}{W} \right)^{5/2} - 25.11 \left(\frac{a}{W} \right)^{7/2} + 25.8 \left(\frac{a}{W} \right)^{9/2} \right\}$$

Where K_{IC} is mode I crack opening, P is load at fracture (N), L is the span length in mm, W is the specimen width in mm, B is the specimen thickness in mm, and a is the notch length in mm [20].

Hydrophobicity (Contact Angle Measurements)

Contact angles were determined with a VCA1000 video contact angle instrument (AST Products, Billerica, MA). Three images of ~0.05 mL drops (one on the center, two on opposite edges) of deionized water were captured by the camera 15 seconds after placement of water droplets on the specimens and the angles determined.

Statistical analysis

Two-Way ANOVA was carried out to evaluate the effect of the material type and the challenge on the fracture toughness (K_{Ic}). A statistical significant interaction between material and challenge was observed ($p = 0.007$). A one-way ANOVA was then used and followed by Bonferroni *post-hoc* test. One-way Repeated Measure ANOVA was used to evaluate the surface area and volume before and after aging. Levene's test of Equality confirmed homogeneity for the surface area and volume measurements. All tests were at 5% level of significance.

RESULTS

Fracture Toughness results

Filtek Supreme composites had significantly higher baseline K_{Ic} ($1.37 \pm 0.29 \text{ MPa m}^{1/2}$) than Admira and OASys composites ($0.88 \pm 0.52 \text{ MPa m}^{1/2}$ and $0.575 \pm 0.28 \text{ MPa m}^{1/2}$, respectively, $p < 0.05$) [Table 2]. The same pattern was observed after the biofilm challenge. Filtek and Admira K_{Ic} significantly decreased following biofilm and water challenges. On the contrary, OASys K_{Ic} significantly increased following water storage ($0.795 \text{ MPa m}^{1/2}$) as compared to the baseline measurement ($0.575 \text{ MPa m}^{1/2}$, $p < 0.05$), while no significant difference in K_{Ic} was found between the OASys biofilm group and either baseline or water groups.

Surface area and Volume changes

OASys composites had significant lower volumes than Admira and Filtek composites [Tables 3,4]. However, no significant change was observed in the surface area and volume after aging regardless of the material ($p > 0.05$).

Hydrophobicity

An average and standard deviation of 108.2(9.2) was observed for OASys-based composites, showing a high degree of hydrophobicity. Lower average values were observed for Filtek [79.9 (11.9)] and Admira groups [59.39 (12.6)].

DISCUSSION

Degradation is a major factor in the shortened longevity of dental composite restoratives [2, 22]. Recent advancements in dental materials technology to overcome this challenge includes the development of ormocer-based composites and an experimental oxirane/acrylate interpenetrating polymer resin (OASys)-based composites [16, 17]. Fracture toughness (K_{Ic}) has been shown to be a good predictor of a clinical performance for direct restorative materials that are used in stress-bearing areas [1, 2].

The results of this study revealed that the type of resin composite material had a significant effect on the fracture toughness K_{Ic} ($p < 0.05$). At baseline, Filtek Supreme composite had a significant higher K_{Ic} than Admira and OASys composites. Other studies have demonstrated similar results in regards to Filtek and Admira [18, 23–25]. The mechanical properties, including fracture toughness, of ormocer based composites has been reported to be

comparable to the conventional hybrid and nanohybrid when stored in water for 24h [15, 18]. However, differences in the amount and type of filler in the composite matrix can be significant contributors to the differences in K_{Ic} of these materials [26, 27]. Filtek has a higher filler content compared to that of Admira and OASys. In addition, it could be hypothesized that the nanoclusters present in the Filtek Supreme might contribute to crack deflection [28]. On the other hand, Admira has larger glass fillers that might elicit less resistance to crack propagation [29]. Although OASys composite material had voids, most likely due to it being an experimental composite without a delivery device, its fracture toughness was similar to Admira.

Following water storage, the K_{Ic} of both Filtek and Admira composite materials significantly decreased. Some studies have reported no change in K_{Ic} values after water storage [25] while others have shown significant reductions [30, 31], [4]. Dental composites tend to slowly absorb water, some of which accumulates between the inorganic fillers and matrix. This process could take up to 2 months until equilibration [31, 32]. The accumulated water molecules separate the chains of the polymer network resulting in unpredictable polymer expansion and chemical degradation [33], which can affect the mechanical properties of composites.

OASys composites had a significant increase in K_{Ic} after being aged in water. Ilie et al. (2009)[34] has stated that silorane based resin composites (a monomer that contains siloxane and oxirane) showed good mechanical properties compared to Bis-GMA- based resin composite and was mechanically stable following 5000 cycles of thermocycling and four weeks of water, artificial saliva, or alcohol storage [34]. It is possible that the hydrophobic surface of the OASys composite as demonstrated by the contact angle measurement prevented water inhibition into the composite and the slower oxirane polymerization continued, showing higher fracture toughness.

An array of aging methods can be used to evaluate mechanical and physical properties of dental materials. Studies have shown that water and artificial saliva had similar effect on the mechanical properties of resin composites after cyclic fatigue [19, 22, 34]. The main degradation mechanism of composite materials aged in water and/or artificial water is possibly due to hydrolytic degradation of the filler-matrix interface. When using water as the environmental challenge, hydrolytic degradation is the only anticipated mechanism.[3, 22] In our study, both 15 days of biofilm challenge and 30 days of water challenge reduced the fracture toughness of Filtek and Admira resin composites but did not reduce the fracture toughness of OASys composites. This could be related to the high degree of hydrophobicity of the experimental composite.

Although the biofilm challenge has high technical demands compared to water storage, it includes a potential enzymatic activity known to be responsible for degradation of dental composite materials[35, 36]. *Streptococcus mutans* biofilm is known to be associated with dental plaque and dental caries [37, 38]. Lactic acid and enzymes, such as esterase, that are produced by bacteria, such as *Streptococcus mutans*, can hamper the resin composite via hydrolysis and chemical breakdown of ester bond in resin matrix and affect its surface roughness and physical stability [11, 12]. In this study, for Filtek and Admira, the effect of

biofilm aging was similar to that of water storage aging with a significant reduction of K_{IC} . This is in agreement with Zhou *et al's* [39] report that *Streptococcus mutans* biofilm aging produced a significant flexural strength reduction in resin composites. Interestingly, there was no such reduction in fracture strength for OASys composites after bacterial biofilm aging, further lending evidence that the hydrophobic surface may have prevented or reduced degradation by preventing water and enzyme imbibition.

Several studies have confirmed the accuracy and validity of using μ CT to evaluate the volumetric change of resin composite materials [40–44]. The small increase in volume of the specimens were mostly attributed to hygroscopic expansion caused by water sorption. Water molecules are slowly absorbed between the inorganic particles and organic matrix [3]. This accumulated water might interact with the monomer through hydrogen bond, allowing for further accumulation of water molecules inside the polymer. Moreover, resin composites that contained Bis-GMA/TEGDMA co-polymers have up to 3.3% increase in volume due to hygroscopic expansion [45]. Boaro et al.[46] has shown that the same Filtek Supreme resin composite has a high sorption rate compared to seven other resin composite materials and silorane based resin composite (a monomer that contains siloxane and oxirane) had a low sorption rate. The surface area and volume change results from this work, however, did not show any significant increases for any of the materials.

Conclusion

Within the limitations of this study, both biofilm aging and water storage significantly reduced the fracture toughness of the commercially available dental composite materials - Filtek Supreme Ultra and Admira Fusion. However, biofilm aging did not affect the fracture toughness of the OASys dental composites and water storage actually increased the fracture toughness. All tested dental composite materials showed no significant surface area or volume changes after the environmental challenges as measured using μ CT. The use of a hydrophobic OASys-based dental composite shows promise for increasing restoration longevity while further studies are warranted.

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Figure 1:
Flowchart of the experimental study design

Table 1:

Materials used in this study, manufacturer, composition, and LOT numbers

Material	Manufacturer	Matrix	Fillers	Fillers by weight/volume %wt/%vol	Lot Number
Admira Fusion (A)	VOCO, Cuxhaven, Germany	Ormocer resin multi-functional urethane thioetheroligo(meth)acrylate alkoxysilanes	SiO ₂ , Ba-Al-B-Si-glass	77%/54%	1651310 1628174 1734497 1623194 1633336
Filtek Supreme Ultra (F)	3M ESPE, St. Paul, MN, USA	BisGMA, BisEMA, UDMA, TEGDMA	ZrO ₂ /SiO ₂ cluster SiO ₂ nanofiller	78.5%/63%	N820171 N853944 N870984
OASys (OA)	UTHSCSA	- 25% DPHA - 25% Epalloy 8370 -20% UDMA -30% PFOEA	- 0.7 µm barium glass filler - acrylate silanated	70%/NI	

Abbreviations: NI= no information available; Bis-GMA = bisphenol A-glycidyl methacrylate; BisEMA = ethoxylated bisphenol-A dimethacrylate; UDMA= urethane dimethacrylate; TEGDMA= triethylene glycol dimethacrylate; DPHA= dipentaerythritol hexa/pentaacrylate; PFOEA = perfluorooctylethyl acrylate

Table 2:

Mean (SD) of the fracture toughness K_{Ic} results (MPa m^{1/2}). (Different uppercase letters in columns and lowercase letter in rows represent statistical significant differences)

n=15	Baseline (control) mean(SD) (MPa m ^{1/2})	Biofilm (15 days) mean(SD)	Water (30 days) mean(SD)
OASys	0.575(.28) Ba	0.69(.16) Bab	0.795(.08) Ab
Filtek	1.37(.29) Aa	0.97(.18) Ab	0.95(.33) Ab
Admira	0.88(.52) Ba	0.50(.24) Bb	0.56(.22) Bb

Table 3:

Volumes of composites before and after aging (Different uppercase letters in columns and lowercase letter in rows represent statistical significant differences)

	Water		Biofilm	
Materials n=3	Mean(std)($\times 10^{11} \mu\text{m}^3$) Before	Mean(std) ($\times 10^{11} \mu\text{m}^3$) After	Mean(std) ($\times 10^{11} \mu\text{m}^3$) Before	Mean(std)($\times 10^{11} \mu\text{m}^3$) After
OASys	3.08(0.06)Aa	3.09(0.07)Aa	3.146(0.01)Aa	3.15(0.01)Aa
Filtek	3.19(0.05)Bb	3.23(0.05)Bb	3.23(0.03)Bb	3.26(0.03)Bb
Admira	3.15(0.03)Bb	3.17(0.03)Bb	3.26(0.06)Bb	3.27(0.06)Bb

Table 4:

Surface areas of composites before and after aging (Different uppercase letters in columns and lowercase letter in rows represent statistical significant differences)

	Water		Biofilm	
Materials n=3	Mean(std) ($\times 10^8 \mu\text{m}^2$) Before	Mean(std) ($\times 10^8 \mu\text{m}^2$) After	Mean(std) ($\times 10^8 \mu\text{m}^2$) Before	Mean(std) ($\times 10^8 \mu\text{m}^2$) After
OASys	5.08(0.06)Aa	5.1(0.08)Aa	4.83(0.04)Aa	4.84(0.06)Aa
Filtek	4.42(0.03)Bb	4.57(0.09)Bb	4.46(0.04)Bb	4.45(0.01)Bb
Admira	4.41(0.05)Bb	4.46(0.07)Bb	4.41(0.1)Bb	4.48(0.12)Bb