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Fracture Toughness of Nanohybrid and Hybrid Composites Stored Wet and Dry up to 60 Days

Sookhakiyan M[a] DDS; Tavana S[a] DDS; Azarnia Y[a] DDS; Bagheri R[b*] DDS, PhD

[a]Shiraz Dental School, Shiraz University of Medical Sciences, Shiraz, Iran
[b]Dental Materials Department and Biomaterials Research Centre, Shiraz Dental School, Shiraz University of Medical Sciences, Shiraz, Iran

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Corresponding Author:
Rafat Bagheri
Dental Materials Department and Biomaterials Research Centre, Shiraz Dental School, Shiraz University of Medical Sciences, Shiraz, Iran
Email: bagherir@unimelb.edu.au
Tel: +98-71-36286251
Fax: +98-71-36285276

Abstract

Statement of Problem: Patients' demand for tooth-colored restoratives in the posterior region is increasing. Clinicians use universal nanohybrid resin composites for both anterior and posterior regions. There are few published reports comparing fracture toughness of nonohybrids and that of hybrid composite stored wet and dry.

Objectives: To investigate the fracture toughness of three nanohybrids compared to that of a hybrid resin composite stored dry or wet up to 60 days, using four-point bending test.

Materials and Methods: Four resin composites were used: three nanohybrids; Filtek Supreme (3M), Ice (SDI), TPH3 (Dentsply) and one hybrid Filtek P60 (3M). For each material, 40 rectangular notched beam specimens were prepared with dimensions of 30 mm × 5mm × 2mm. The specimens were randomly divided into 4 groups (n = 10) and stored at 37ºC either in distilled water or dry for 1 and 60 days. The specimens were placed on the four-point test jig and subjected to force (N) using universal testing machine loaded at a crosshead speed of 0.5mm/min and maximum load at specimen failure was recorded and K_{Ic} was calculated.

Results: Three-way ANOVA showed a significant interaction between all the factors (all $p < .0001$). Except for TPH3, all tested materials showed significantly higher K_{Ic} when stored dry than stored wet ($p < 0.05$). After 1 day of dry storage, Ice showed the highest K_{Ic} (2.04 ± 0.32) followed by Filtek P60 and the lowest was for Filtek Supreme (1.39 ± 0.13). The effect of time on fracture toughness was material dependent.

Conclusions: Wet storage adversely affected the fracture toughness of almost all materials. Keeping the restoration dry in the mouth may increase their fracture toughness. Therefore, using a coating agent on the surface of restoration may protect them from early water uptake and increase their strength during a time period.

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Introduction

Fracture toughness of aesthetic restorative materials has grown significantly since they are used for posterior restoration in the load bearing area. On the other hand, the patients' increasing demand for having tooth-colored restoratives in the posterior region motivated the manufacturers to introduce and market a stronger resin composite. These composites consist of higher filler volume and smaller particle sizes of nanoscale, such as nanohybrids have shown to withstand greater mastication forces than the older composites [1].

A variety of resin composites are available in the market, being categorized into hybrid, microfilled, microhybrid, and nanohybrid with different physical and mechanical properties. Nowadays, nanohybrid composites are extensively used due to their mechanical improvement that makes them suitable for use in anterior and posterior restorations [2-4]. Nanohybrid is a hybrid resin composite with nanofiller in a prepolymerised filler form [5,6]. Nanotechnology, at a scale of 0.1–100 nanometers, is the invention of functional materials and structures by numerous physical and chemical methods [7].

The oral cavity restorations are subjected to complex forces such as bending, shear, fatigue and fracture. Fracture toughness (*K*Ic) is an important mechanical property of dental materials that signifies the ability of a material against crack propagation [4]. In order to measure fracture toughness of resin composites, single edge V-notched beam (SEVNB), ISO 6872 is one of the reliable recommended methods [2]. A recent study [3] evaluated three different fracture toughness methods of seven resin composites and found that the four-point method was a more perceptive method determining the fracture toughness (*K*Ic) value for resin composites with different percent filler contents. The results of this study [3] showed that hybrid resin composite with the highest filler vol% and the largest filler sizes had significantly higher fracture toughness compared to the nanohybrid, microhybrid and microfill composites.

In addition, water sorption by resin composites has been shown to have a degradation effect on the fracture toughness of the materials [8]. Dental restorative materials are exposed to saliva and beverages in the oral environment that could adversely affect their strength due to aging [9,10]. It has been shown that storing resin composites in dry condition increased their load bearing capacity in comparison to storing them in the wet condition [11]. Others also showed a significant decrease in fracture toughness or resin composites following water uptake [12,13]. Curtis *et al.* [13] in their study investigated the influence of water uptake and mechanical properties of a nanofilled compared with a conventional resin composite, reporting a decrease in fracture toughness following 3-12 months of water immersion. They revealed that strength degradation occurred at different rates between material types. The size and morphology of nanoparticles were important factors in water uptake and mechanical properties of the tested materials [13].

There are a few studies comparing the fracture toughness of glass fiber-reinforced resin composites stored dry and wet with conventional resin composites [14-17], but there are no published reports on the comparison of fracture toughness of nanohybrid and hybrid composites stored dry and wet. Therefore, this study aimed to compare the effect of water uptake on the fracture toughness of three nanohybrid resin composites to that of a hybrid composite up to 60 days of immersion. The null hypothesis is that storing resin composites in dry or wet condition does not affect their fracture toughness after one or 60 days.

Materials and Methods

In this study, four different resin composites (Table 1) were selected to be used in the four-point fracture toughness test. A four-point bending mould consists of a stainless steel detachable set up and prefabricated notch placed in the center of the beam (1 mm long and 0.1 mm wide) to enable the fabrication of tests specimens. A total of 40 rectangular notched beam specimens were constructed with dimensions of 30 mm × 5mm × 2mm for each material. The resin composite was placed into the mould using a flat double-ended stainless steel instrument and compressed, then covered by a clear plastic strip and a stainless steel metal plate was used to level out the material and extrude the excess. The stainless steel plate was then removed and the specimen was light cured along its length in 20 sec intervals using a SDI Radii plus LED light curing unit, (SDI, Bays water, Victoria, Australia) with a wavelength of 440-480 nm/output of 1500 mW/cm^2. The mould was then dismantled and the specimen was turned over to cure the bottom surface again using the same curing regimen. The excess (flash) was clipped from the edges and the sides were polished using fine 1200 grit

Table 1: Detail information of the materials used

Material	Filler type /content	Matrix content	Batch No	Manufacturer	Classification
Filtek Supreme XT	78.5 w% Zirconia/ silica clusters (0.6-1.4 µm)	Bis-GMA, UDM, TEGDMA, Bis-EMA	N599645	3M /ESPE, St Paul, MN, USA	Nanohybrid
Filtek P60	61 v % Zirconia/silica (0.01-3.5) µm	Bis-GMA, UDMA, Bis-EMA	N511095	3M /ESPE, St Paul, MN, USA	Hybrid
Ice	77 w% inorganic filler SAS, AS (40nm-1.5 µm)	UDMA/ Bis-EMA/ TEGDMA	131194T	SDI, Vic, Australia	Nanohybrid
TPH3	76 w% Barium glass (0.8), Nano silicon dioxide particles (10-20nm)	Bis-GMA, Bis-EMAand TEGDMA	1109241	Dentsply	Nanohybrid

UDMA: urethane dimethacrylate, Bis-GMA: bisphenol A glycidyl dimethacrylate, TEGDMA: triethyleneglycol dimethacrylate, Bis-EMA: ethoxylated bisphenol-A dimethacrylate, SAS= Strontium alumino silicate, AS= amorphous silica

silicon carbide paper (Tufbak waterproof sanding sheets; Scour Pads, Melbourne, Victo- ria, Australia).

The specimens were then divided into four groups of 10 and stored either dry or wet (in distilled water) for one or 60 days in a vented container at 37ºC. The specimens were tested after each time interval in universal testing machine (Zwick/Roll Z020, Zwick GmbH & Co, Germany) using a four-point bend test jig, loaded at a crosshead speed of 0.5mm/min, and the maximum load at specimen failure was recorded.

Before testing, a new razor blade was used under hand pressure to create a sharp crack in the notch. Crack length was measured using a stereomicroscope (BS-3060C, Beijing, China) at 60x. The width and height of each specimen was measured using a digital calliper with accuracy of 0.1mm (mini Electronic Caliper, Qingdao Preco Imp. & Exp. Co., Ltd., Zhejiang, China). The K_{Ic} ($MPa.m^{0.5}$) was calculated using the following equation:

$$K_{Ic} = \frac{L_{max}}{w\sqrt{h}} \cdot \frac{l_0 - l_1}{h} \cdot \frac{3 \cdot r_M \sqrt{d/h}}{2.(1-d/h)^{3/2}}$$

$$r_M = 1.9887\text{-}1.326\frac{d}{h} \cdot \frac{[3.49-0.68\, d/h + 1.35(d/h)^2]\, d/h(1-d/h)}{(1+d/h)^2}$$

Where: L_{max} = fracture load,
l_0: outer span; l_1: inner span,
w = specimen width, α = a/w,
d = notch depth, h = specimen height

Statistical analysis

Data were analyzed using SPSS version 21.0 (SPSS Inc., Chicago, IL, USA). Three-way ANOVA was used to find if any interaction is found between materials, times and conditions. One-way ANOVA and Tukey's post-hoc test were conducted for subgroup analysis comparing each factor individually. $p < 0.05$ was considered as statistically significant.

Results

Three-way ANOVA showed a significant interaction among all three factors of material, time, and wet/dry condition (all $p < 0.001$). Results of one-way ANOVA and pairwise comparisons are shown in Table 2 and graphically in Figure 1 All materials had higher fracture toughness when kept dried after either 1 or

Table 2: Mean ($MPa.m^{0.5}$) and standard deviation of K_{Ic} after one and 60 days of wet and dry storage for all resin composites (n=10)

Material	One Day Wet	One Day Dry	60 Days Wet	60 Days Dry
Filtek Supreme	^{Ba}1.28 ± 0.11	^{Bb}1.39 ± 0.13	^{Bab}1.34 ± 0.33	^{Bc}1.54 ± 0.48
Filtek P60	^{Aa}1.52 ± 0.13	^{Ab}1.81 ± 0.17	^{Ab}1.78 ± 0.40	^{Ac}1.93 ± 0.25
Ice	^{Aa}1.62 ± 0.15	^{Ab}2.04 ± 0.32	^{Cc}1.09 ± 0.29	^{Ba}1.63 ± 0.34
TPH3	^{Aa}1.7 ± 0.13	^{Bb}1.43 ± 0.17	^{Bb}1.39 ± 0.69	^{Cc}1.06 ± 0.33

Different upper case letters show significant differences between materials in a column
Different upper case letters show significant differences between conditions in a row

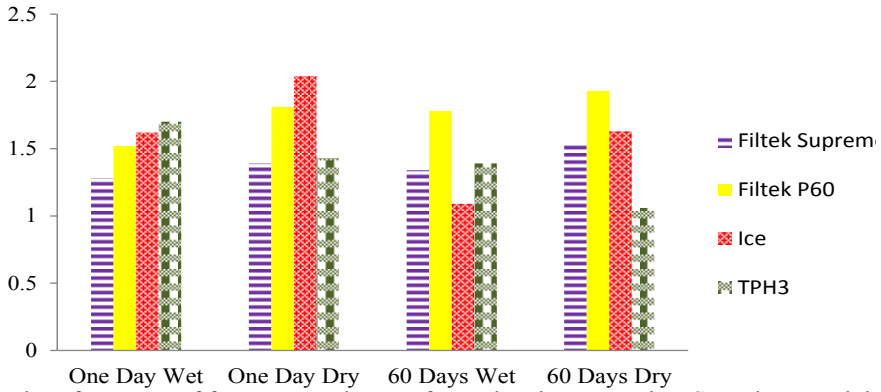

Figure 1: Graphical performance of fracture toughness of tested resin composites Stored wet and dry at one or 6o days

60 days of storage with an exception of TPH3 that showed a higher resistance when stored in distilled water. The effect of time on fracture toughness was material dependent. Filtek Supreme and Filtek P60 showed a significant increase after 60 days of storage in both wet and dry condition while Ice and TPH3 showed a significant decrease in the same condition ($p < 0.05$). Among the materials, in dry condition, Ice showed significantly higher fracture toughness (2.04 ± 0.32) than Filtek P60 (1.81 ± 0.17) followed by TPH3 (1.43 ± 0.17) and Filtek Supreme having the lowest K_{Ic} value (1.39 ± 0.13). A similar trend was observed at wet condition. However, after 6o days of storage in water, Ice had the most decrease in fracture toughness value compared to all other materials (1.09 ± 0.29).

Discussion

Comparison of the fracture toughness of three nanohybrids with that of hybrid composite revealed comparable fracture toughness in different times and conditions. Filtek P60 showed significantly greater K_{Ic} values than Filtek Supreme but slightly lower than Ice and TPH3. However, after 60 days of water storage, Filtek P60 showed significantly higher fracture toughness than all other nanohybrids. That is, the K_{Ic} values of two nanohybrids (Ice and TPH3) decreased significantly after 60 days of immersion in water while Filtek P60 and Filtek Supreme showed a significant increase in toughness. This result indicated that the midterm water storage adversely affected the fracture toughness of the nanohybrids compared to the hybrid composite. This effect may be related to less water uptake of hybrid than the others. Curtis *et al.* [13] in their study of water uptake and strength characteristics of a nanofilled composite found an increased water uptake and degradation of the filler/matrix interface for nanofilled. They speculated that it could be due to the larger surface area to volume ratio of the fillers in nanofilled compared with those of the microhybrid composite. They also concluded that the presence of nanoparticles and clusters in the nanofilled led to diverse mechanical and physical properties of this material [13].

In general, almost all resin composites performed with higher K_{Ic} values when stored dry in both time intervals of one or 60 days. This result is in agreement with those of other recent studies [11,14-17]. Farmani *et al.* [11], under hertzian indentation test, showed that nanohybrid composites had comparable or even higher load bearing capacity than that of hybrid composite. Specimens stored in distilled water showed significantly lower load bearing capacity in comparison with dry condition. Bijelic-Donova *et al.* [16] compared fracture toughness and some other mechanical properties of short fiber-reinforced composite (everX Posterior, eXP) with that of microfilled, nanofilled and bulk-fill composites. The specimens were stored dry for 7 days or in water for 30 days. They showed that water storage weakened the fracture toughness of all materials and this reduction of K_{Ic} was material dependent. Higher fracture toughness of the resin composites reveals the fact that immersion in water probably facilitates and accelerates the crack propagation by water diffusion in the interface between the resin matrix and the filler, leads to softening the polymer matrix by swelling the network and reducing the frictional forces between the polymer chains [18,19].

On the other hand, Hoshika *et al.* [18] evaluated the degree of conversion and mechanical properties of Bis-GMA / TEGDMA / HEMA (60:30:10) containing 0–15 mass% QAMs (quaternary ammonium-methacrylates) after 3 days of dry or water storage. They reported higher frac-

ture toughness for wet specimens ($p < 0.05$). The author [18] speculated that the significant increase in fracture toughness seen after water immersion for three days might have been due to slight decreases in the elastic modulus of the resins after water plasticization. This may have increased the ability of the matrix in absorbing water to resist crack propagation. Indrani *et al*. [19] assessed the effect of aging in water on the K_{Ic}, elastic modulus and fracture energy. The results indicated an increase in the fracture toughness and fracture energy as well as a decrease in the elastic modulus. The reported that water sorption also occurred mainly during the first 2 weeks. They interpreted variations in the mechanical properties to be due to plasticization of the resin matrix by water, which seems to lower the yield stress and increase in the size of the plastic zone ahead of the crack, thus leading to an increase in K_{Ic} and fracture energy [19].

Among all the tested resin composites, TPH3 behaved differently in terms of the effect of the condition, i.e. specimens stored in water had higher K_{Ic} values (1.7 and 1.39) than that of those stored dry (1.39 and 1.06). Polymeric matrix composition of TPH3 may justify the dissimilar results for this material compared to others. Although the exact percentages of oligomers in each material is not released by the manufacturer, it seems that TPH3 consisted of less amount of TEGDMA in which more water is absorbed compared to Bis-EMA and Bis-GMA; therefore, less degradation of the oligomer occurs in water [20,21,22]. It has been found that physical and mechanical properties of resin composites are affected by different monomer structures of the materials. Gonçalves *et al.* [23] in their study determined the influence of variety of concentration of Bis-GMA/ TEGDMA/Bis-EMA on the flexural strength of resin composites. The result revealed that Bis-EMA mixtures presented a statistically lower flextural strength followed by TEGDMA + Bis-EMA mixtures, and then by TEGDMA mixtures [23].

In general, the results of this study showed that water storage for up to 60 days reduced the fracture toughness of almost all the tested resin composites in comparison to dry specimens. One of the limitations of this study was the dry and wet storage of the specimens in a short time. Further long-term studies of more than a year are needed to find out if aging and wet storage in the fracture toughness of resin composites affects the fracture toughness of resin composites more objectively.

Conclusions

Within the limitations of this study, the following conclusions are drawn. Hybrid composite (Filtek P60) showed higher fracture toughness than one of the nanohybrids (Filtek Supreme) and less than the other two nanohybrids after one day of storage in distilled water but higher toughness than all the three nanohybrids after 60 days of aging. In general, dry specimens had greater K_{Ic} values than wet specimens after one or 6o days with an exception of TPH3. The results revealed that keeping the restoration dry in the mouth may increase their fracture toughness. Therefore, using a resin-coating agent on the surface of the restoration may protect them against early water uptake and increase their strength during a time period.

Conflict of Interest: None declared.

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