Effect of thermal cycling on whisker-reinforced dental resin composites

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The mechanical properties of dental resin composites need to be improved in order to extend their use to high stress-bearing applications such as crown and bridge restorations. Recent studies used single crystal ceramic whiskers to reinforce dental composites. The aim of this study was to investigate the effects of thermal cycling on whisker-reinforced composites. It was hypothesized that the whisker composites would not show a reduction in mechanical properties or the breakdown of whisker–resin interface after thermal cycling. Silicon carbide whiskers were mixed with silica particles, thermally fused, then silanized and incorporated into resin to make flexural specimens. The filler mass fraction ranged from 0% to 70%. The specimens were thermal cycled in 5 °C and 60 °C water baths, and then fractured in three-point bending to measure strength. Nano-indentation was used to measure modulus and hardness. No significant loss in composite strength, modulus and hardness was found after 10^5 thermal cycles (family confidence coefficient = 0.95; Tukey's multiple comparison test). The strength of whisker composite increased with filler level up to 60%, then plateaued when filler level was further increased to 70%; the modulus and hardness increased monotonically with filler level. The strength and modulus of whisker composite at 70% filler level were significantly higher than the non-whisker controls both before and after thermal cycling. SEM revealed no separation at the whisker–matrix interfaces, and observed resin remnants on the pulled-out whiskers, indicating strong whisker–resin bonding even after 10^5 thermal cycles. In conclusion, novel dental resin composites containing silica-fused whiskers possessed superior strength and modulus compared to non-whisker composites both before and after thermal cycling. The whisker–resin bonding appeared to be resistant to thermal cycling in water, so that no loss in composite strength or stiffness occurred after prolonged thermal cycling.
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1. Introduction

Dental resin composites are generally composed of silanized fillers in an acrylic monomer matrix that is subsequently polymerized to form a solid restoration [1–3]. The filler size and shape, volume fraction, composition of the resin matrix, filler–matrix interfacial bonding, and cure conditions all influence the mechanical properties of the composites [1–9]. The fillers that reinforce the composites are usually composed of particulate silicate glasses [1–3]. Silanization of the filler particles [1, 8, 10] improves filler–matrix bonding and increases the strength, modulus and wear resistance of the composites

[11]. Mixing filler particles of different sizes to achieve a bimodal distribution generally enhances composite properties [2, 12, 13]. Heat-curing or post-cure heat treatment of resin composites increases the degree of polymerization conversion and slightly improves the composite strength [14–18]. Short fibers [19] and networked fibers [20] also can be used to reinforce dental resin composites, which resulted in modest increases in composite strength. More recently, ceramic whiskers were used to reinforce dental resin composites [21]. Silica particles were fused onto the whiskers to facilitate silanization, minimize whisker entanglement,

and enhance whisker retention in the matrix by roughening the whisker surfaces [22]. Whisker composites demonstrated flexural strength and fracture toughness nearly two times those of currently available dental composites [23].

In the oral cavity, restorations and teeth are subjected to thermal stresses. For example, the temperature of ice water is close to 0 °C, and that of hot soup or tea can surpass 60 °C. The difference in the thermal expansion coefficient between the restoration and the tooth structure in the presence of normal thermal cycling that occurs in the mouth may cause restoration–tooth debonding and microleakage [24–29]. In the case of resin composite restorations, there can also be internal thermal stresses due to the wide difference in the thermal expansion coefficients between the resin matrix and the fillers, causing cyclic stresses inside the composite during thermal cycling. The cyclic thermal stresses, together with the presence of water and other fluids, may degrade the filler–matrix interfaces and also lead to stress corrosion of the fillers [30, 31]. While microleakage of composite-tooth interfaces induced by thermal cycling has been fairly well studied [24–29], few investigations have studied the effects of thermal cycling on the degradation of the composite itself [30–32]. Montes-G and Draughn [30] observed that thermal cycling caused surface degradation and a decrease in abrasion resistance for dental resin composites. Mair and Vowles [31] reported that thermal cycling caused a slight decrease in the fracture toughness of dental resin composites, while Tanaka et al. [32] showed a loss in flexural strength and flexural modulus and found that the extent of loss was dependent on the composite composition. However, no study has been performed on the effects of thermal cycling on dental resin composites containing silica-fused whiskers.

The aim of the present study, therefore, was to investigate the effects of thermal cycling on the mechanical properties of dental resin composites containing silica-fused whiskers at various filler levels. Two commercial indirect resin composites without whiskers also were tested in the same manner as comparative controls. Silica particles were fused onto the whiskers to enhance whisker silanization and bonding to the resin matrix. It was hypothesized that the silica-fused whisker composite would not show a reduction in mechanical properties after thermal cycling or a breakdown of the whisker–resin interfaces, and therefore would posses significantly higher strength than the controls both before and after thermal cycling.

2. Materials and methods
2.1. Filler powder and resin preparation
Ceramic silicon carbide whiskers (SiC_w T-Grade, Advanced Refractory Technologies, Inc., Buffalo, NY) had diameters ranging from about 0.1 μm to 3 μm with a mean of approximately 0.9 μm, and lengths ranging from about 2 μm to 100 μm with a mean of approximately 14 μm. Many of these whiskers were slightly bent, and the diameters of whiskers appeared to vary slightly along their lengths. The whiskers were mixed with silica

having a nominal particle size of 0.04 μm (Aerosil OX50, Degussa Corp., Ridgefield, NJ) in a 2 : 1 mass ratio by dispersing both with stirring in ethyl alcohol until all the solvent was removed and a dry powder was obtained [22, 23]. To fuse silica onto the whiskers, the dried mixture was heated in air for 30 min at 800 °C [23]. The heated powder was silanized by mixing it with mass fractions of 2% n-propylamine (Aldrich, Milwaukee, WI) and 4% 3-methacryloxypropyltrimethoxysilane (MPTMS) (Aldrich, Milwaukee, WI) in cyclohexane by means of a rotary evaporator under moderate vacuum in a 90 °C water bath until dry.

2.2. Specimen fabrication
The silanized silica-fused whiskers were manually mixed by spatulation with a resin monomer consisting of mass fractions of 48.965% of an oligomeric urethane derivative of Bis-GMA (NCO/Bis-GMA, Caulk/Dentsply, Milford, DE), 48.965% triethylene glycol dimethacrylate (TEGDMA) (Esstech, Essington, PA), 0.070% 4-methoxylphenol (MEHQ) (Aldrich, Milwaukee, WI), and 2.000% benzoyl peroxide (BPO) (Aldrich, Milwaukee, WI). Five batches of whisker-resin paste were prepared at one of the following five whisker/(whisker + resin monomer) mass fractions: 0%, 20%, 40%, 60%, and 70%. The purpose of the 0% (unfilled resin) was to see if there was degradation in the matrix due to cyclic stress and/or hydrolysis. The different filler levels were used because they increased the whisker–resin interfaces in the composite, and would reflect differences in the composite properties if there was stress-induced interface degradation or water-induced degradation of the silane coupler. The paste of each filler level was placed into steel molds of 2 mm × 2 mm × 25 mm dimensions and heat-cured in an oven (Model 48, Fisher Scientific, Pittsburg, PA) at 140 °C for 30 min at room atmospheric pressure to make specimens [33]. The temperature was measured by an analog thermometer (Kessler, Westbury, NY) installed inside the oven. Twenty-four specimens were thus made at each of the five filler levels. All the composite whisker specimens were cured in this oven at atmospheric pressure.

In addition, specimens of two commercial non-whisker composites were fabricated as comparative controls. The paste of an indirect composite (Concept©, Ivoclar North America, Amherst, NY) was placed into the same molds and cured in the Concept© heat integrated processor at 120 °C for 10 min under a pressure of 0.6 MPa, according to the manufacturer's instructions. Concept© consists of a mass fraction of 76% of silicate fillers in a urethanedimethacrylate resin (Technical Data Sheet, Ivoclar North America). Concept© is denoted in this paper as "control c". The paste of a second indirect composite (Artglass©, Heraeus Kulzer GmbH, Wehrheim, Germany) was placed into the molds and cured in a Dentacolor XS© photo-curing unit (Heraeus Kulzer GmbH) for 90 s on each side of the specimen. According to the manufacturer, Artglass© contains a mass fraction of 70% barium glass in a resin with tetra- and hexa-functional groups in addition to

conventional bi-functional methacrylates. Artglass® is denoted in this article as "control a". Twenty-four specimens were made for each control.

2.3. Thermal cycling

Thermal cycling was performed using a computer-controled two-temperature cycler (American Dental Association Health Foundation, Paffenbarger Research Center) with two baths of distilled water at temperatures of 5 °C and 60 °C, respectively. One cycle consisted of 15 s immersion in each water bath and a travel time of 8 s, yielding a total time of 38 s. The two temperatures were chosen to approximate the minimum and maximum temperatures found in the oral cavity. The dwell time of 15 s was chosen based on the justification of Wendt et al. [28]. The water baths were constantly stirred with two stirrers (Arrow Engineering Co., Hillside, NJ). The variation in the temperature of each water bath was within 1 °C of the set temperature.

All the cured specimens were immersed in distilled water at 37 °C for 24 h. The specimens were then divided into four groups. Each group contained six specimens of each of the seven materials: the whisker composites at five different filler levels, control a, and control c.

For the four groups of specimens, group one was subjected to a flexural test and a nano-indentation test without thermal cycling (0 cycle). Groups two and three were thermal cycled for 10^4 and 10^5 cycles, respectively, and then subjected to the flexural and indentation tests. It took approximately 105.6 h to complete 10^4 cycles, and 1056 h to complete 10^5 cycles. Group four was not cycled but was immersed in distilled water for the equivalent time taken by 10^5 cycles, i.e., 528 h in the 5 °C bath and 528 h in the 60 °C bath, and then subjected to the flexural and indentation tests. The purpose of group four with immersion but no thermal cycling was to differentiate whether any degradation seen was caused by hydrolytic breakdown of the resin or silane coupler, or was caused by fatigue from cyclic thermal stresses.

2.4. Mechanical testing

A standard three-point flexural test [34] with a span of 10 mm was used to fracture the specimens at a crosshead speed of 1 mm/min on a computer-controled Universal Testing Machine (model 5500R, Instron Corp., MA) [23]. The flexural strength values of the four groups of specimens were measured. The halves of specimens from the flexural test were used for nano-indentation measurement of elastic modulus and hardness [35–38], in order to examine whether the surface layer of the specimens was degraded due to thermal cycling in water.

A nano-indentation system (Nano Instruments, Knoxville, TN) with a diamond Berkovich indenter, which is a three-sided pyramid with the same depth-to-projected area ratio as the Vickers indenter [38], was used to produce the indentations. The indentation loads and the corresponding displacements were recorded continuously throughout a loading–unloading cycle, enabling the measurement of the elastic modulus of the indented specimen. The calculation of hardness and elastic modulus was made according to a method

described previously [35]. The method involves the extrapolation of a tangent to the top of the unloading curve to determine the depth (a combination of elastic and plastic displacement) over which the indenter tip is in contact with the specimen at the maximum load, P_{max}. This depth, and the knowledge of the indenter geometry, gives the contact area, A; hardness H then follows directly from [35, 39]:

$$H = P_{max}/A \qquad (1)$$

The slope of the unloading curve also provides a measure of the contact stiffness, which can be used with the contact area to determine the elastic modulus. The modulus obtained, sometimes referred to as the indentation modulus, E, is related to the Young's modulus E_Y [35, 39] by

$$E = E_Y/(1 - v^2) \qquad (2)$$

where v is the Poisson's ratio. E_Y can be obtained via Equation (2) for materials with known v. For materials with v of approximately 0.25 [39], $E_Y = 0.94 E$. The present study measured indentation modulus E, without trying to calculate E_Y.

Twenty-four indentations were made for each material with four indentations in each of six specimens. This process produced a total of 672 indentations for the four groups of specimens. P_{max} of 1 N was used to yield an indentation contact area of about 600 μm² to 3000 μm², depending on the hardness of material. This method ensures that the measured hardness and modulus approximate those of the composite bulk, rather than the resin phase or filler particles.

Selected specimen fracture surfaces were sputter-coated with gold and examined with a scanning electron microscope (SEM, model JSM-5300, JEOL, Inc., Peabody, MA). Two-way ANOVA was performed to detect significance ($\alpha = 0.05$) and interaction of filler level and treatment conditions. One-way ANOVA and Tukey's multiple comparison test were used at a family confidence coefficient of 0.95 to compare the groups of composites. Student's t test was used to compare the properties of materials after 10^5 thermal cycles to those immersed for the equivalent time taken by 10^5 cycles.

3. Results

Fig. 1 plots flexural strength as a function of filler level for composites without thermal cycling, after 10^4 and 10^5 thermal cycles, and after immersion for the equivalent time taken by 10^5 cycles. The solid symbols are for the whisker composite at various filler levels, and the open symbols represent the two control composites (a for Artglass® and c for Concept®). Each datum is the mean with the error bar showing one standard deviation (SD), $n = 6$. One standard deviation is used throughout the manuscript as the standard uncertainty of the mean. Two-way ANOVA on four levels of treatment (0 cycle, 10^4 cycles, 10^5 cycles, and immersion only) and seven levels of material (whisker composite at five filler levels, controls a and c) showed that there was no statistically significant interaction between treatment and material ($p = 0.23$). In all four treatments, the strength of the
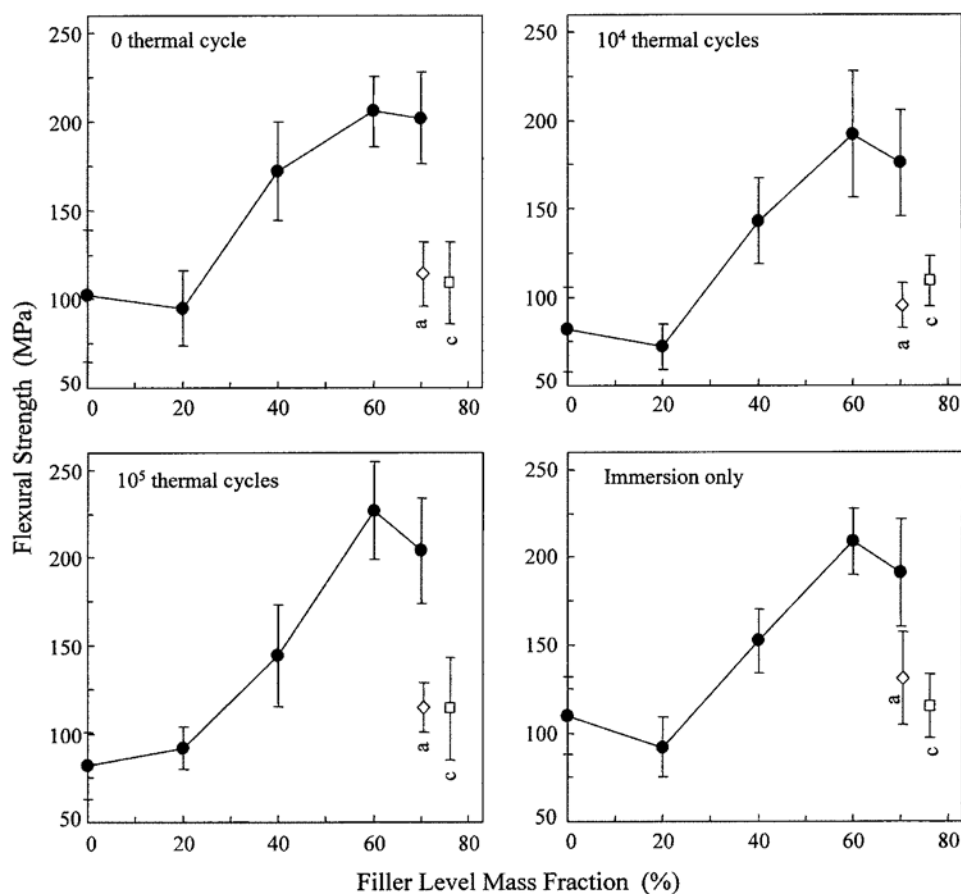
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Figure 1 Flexural strength vs. filler level for composites without thermal cycling (0 cycle), after 10^4 and 10^5 thermal cycles, and after immersion for the equivalent time taken by 10^5 cycles. The solid symbols are for the whisker composite at various filler levels, and the open symbols represent the two control composites (a: Artglass$^{®}$; c: Concept$^{®}$). Each datum is the mean with the error bar showing one standard deviation (SD), $n = 6$.

whisker composite increased with filler level up to 60%, then plateaued when the filler level was further increased to 70%. One-way ANOVA and Tukey's multiple comparison test showed that, for each treatment, the strength values of the whisker composite at 60% and 70% filler levels were significantly higher than those of controls a and c (family confidence coefficient $= 0.95$). The whisker composite at 60% and 70% filler levels had strengths of (228 ± 28) MPa and (204 ± 30) MPa after 10^5 thermal cycles, respectively, similar to (209 ± 19) MPa and (191 ± 31) MPa after immersion for the equivalent time taken by 10^5 cycles ($p > 0.1$; Student's t).

The results on elastic modulus are plotted in Fig. 2 as a function of filler level for composites without thermal cycling, after 10^4 and 10^5 thermal cycles, and after immersion for the equivalent time taken by 10^5 cycles. The solid symbols are for the whisker composite at various filler levels, while the open symbols represent the two control composites. Unlike the flexural strength which plateaued after a filler level of 60%, the elastic modulus increased monotonically with filler level up to 70%. In each group, the whisker composite at 70% filler level had a significantly higher modulus than the controls (Tukey's multiple comparison test; family confidence coefficient $= 0.95$). Compared to the composites in the 0 cycle group, thermal cycling for 10^4 and 10^5 cycles caused no significant loss in elastic modulus of the composites ($p > 0.1$; Student's t).

Fig. 3 plots the hardness value versus filler level for the

whisker composites, together with the two controls in open symbols. Similar to the elastic modulus in Fig. 2, the hardness value of the whisker composites increased monotonically with filler level up to 70%. In each of the four groups, the whisker composite at 70% filler level possessed a hardness value similar to that of control c ($p > 0.1$; Student's t), but significantly higher than that of control a ($p < 0.05$; Student's t). For each composite, thermal cycling for 10^4 and 10^5 cycles did not cause a significant loss in hardness compared to the 0 thermal cycle group ($p > 0.1$; Student's t).

Representative SEM micrographs of fracture surfaces of specimens after 10^5 thermal cycles are shown in Fig. 4A for control c (Concept$^{®}$), and Fig. 4B for control a (Artglass$^{®}$). These fracture surfaces were relatively flat compared to those of the whisker composites in Fig. 5.

Fig. 5 shows SEM micrographs of the fracture surfaces of the whisker composites, after 10^5 thermal cycles, at filler levels of (A) 0% (unfilled resin), (B) 20%, (C) 40%, (D) 60%, and (E) 70%. The magnification was the same for (A)–(E), and (F) is a higher magnification of (E). The fracture surfaces of specimens without cycling and with only immersion were similar to those after 10^5 cycles. The fracture surface of the unfilled resin (Fig. 5(A)) was flat with brittle fracture lines. However, the fracture surfaces of the whisker composites were noticeably rougher, with relatively large fracture steps (long arrows in Fig. 5(B)–5(E)) and whisker pullout (short arrows). These features constituted a high surface area, consuming energy by creating new surfaces. The whiskers
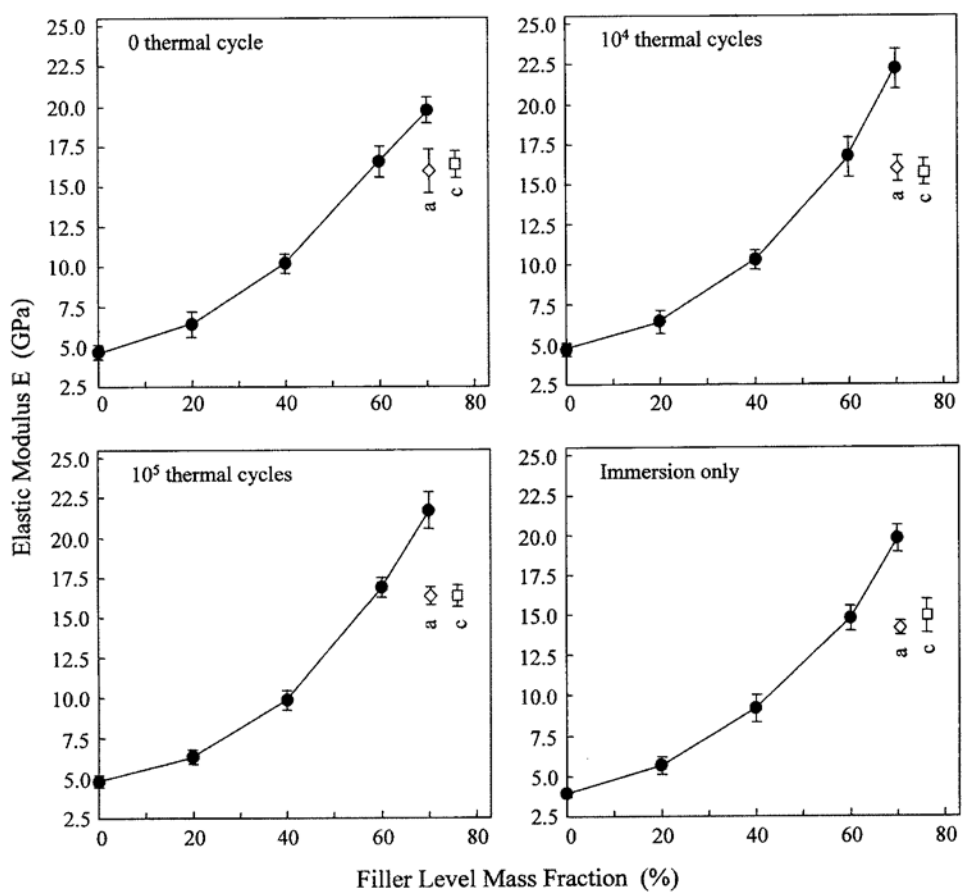
e 2 Elastic modulus measured by nano-indentation as a function of filler level for composites without thermal cycling, after 10^4 and 10^5 thermal s, and after immersion for the equivalent time taken by 10^5 cycles. Solid symbols are for the whisker composites, open symbols are for controls a lass[®]) and c (Concept[®]).
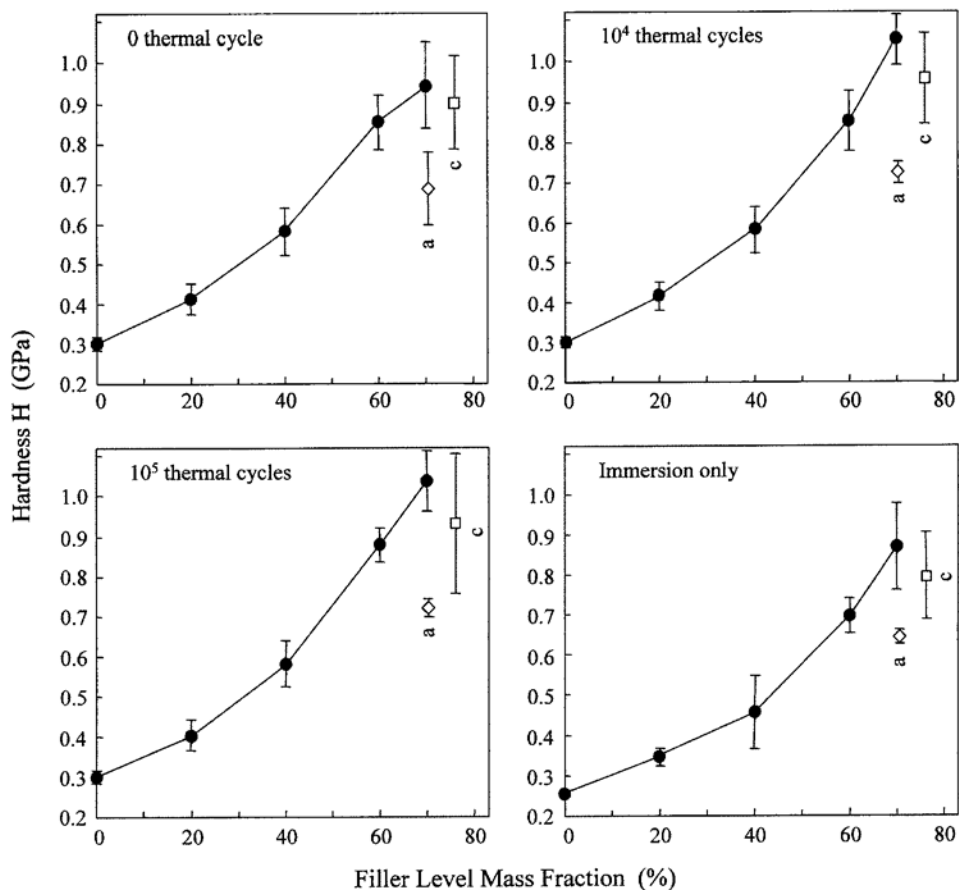

e 3 Hardness vs filler level for composites without thermal cycling, after 10^4 and 10^5 thermal cycles, and after immersion for the equivalent taken by 10^5 cycles. Solid symbols are for the whisker composites, open symbols are for controls a (Artglass[®]) and c (Concept[®]).
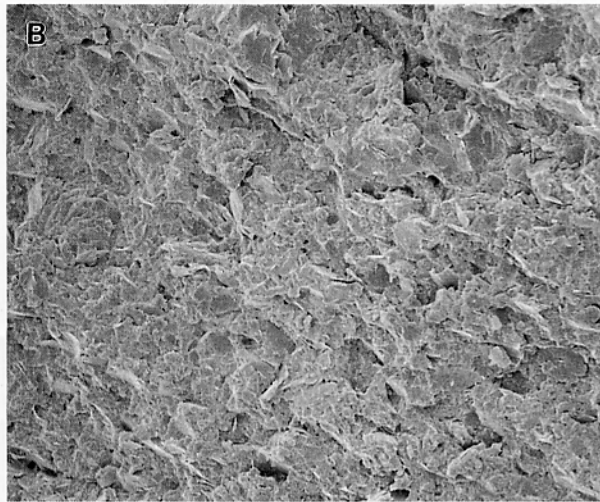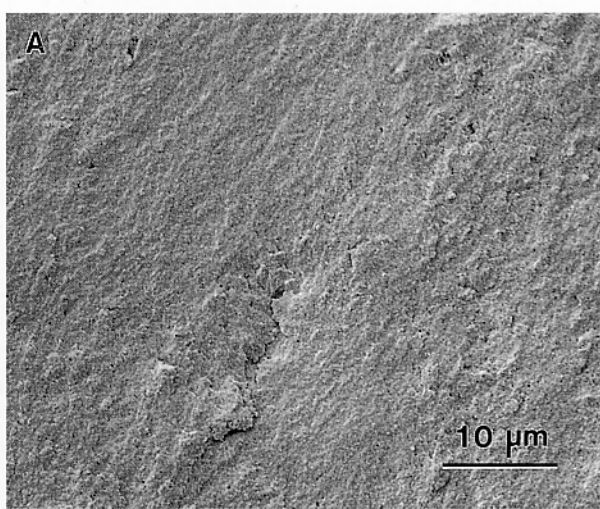
Figure 4 SEM micrographs of fracture surfaces of specimens after 10^5 thermal cycles in water baths of 5 °C and 60 °C for (A) control c (Concept™), and (B) control a (Artglass™).

were not broken and appeared well bonded with the matrix. Examination at magnifications up to 30,000 revealed no separation between the whiskers and the matrix for composites after 10^5 thermal cycles, similar to those with 0 cycles and with water immersion only. In Fig. 5(F), where W denotes pulled-out whiskers and M denotes the resin matrix, the small arrows point to remnants of matrix resin on the surfaces of the pulled-out whisker, indicating whisker–matrix resin bonding even after 10^5 thermal cycles.

4. Discussion

No significant loss in strength, elastic modulus or hardness was measured after 10^5 thermal cycles between 5 °C and 60 °C water baths. The results of the unfilled resin suggested no degradation in the matrix of the composites. The results from the immersion group without thermal cycling showed no degradation due to hydrolytic breakdown of the resin matrix. No differences were also found between immersed and cycled groups, indicating that adding fatigue cycling to the immersion also did not lead to degradation. Since there was no noticeable degradation of the whisker–resin interface, the

increase in filler level and corresponding increase in interfacial area did not result in more degradation after thermal cycling. Simply assuming 50 thermal cycles per day experienced in the oral cavity, it would take about 5.5 years to complete 10^5 cycles. The *in vivo* situation is certainly more complicated with the composite restoration in prolonged contact with oral saliva and food bolus exposed to masticatory stresses. The following are factors that may cause strength loss to a composite due to thermal cycling in water: (1) failure of the filler–matrix interfacial bonding due to water attack; (2) microcracking in composite due to fatigue from cyclic residual stresses; and (3) hydrolysis of the fillers and/or the matrix. These factors are discussed below:

1. SEM observations revealed that the whiskers were still well bonded to the resin matrix after 10^5 thermal cycles, and resin remnants were bonded on the pulled-out whiskers. A previous study showed that the addition of *n*-propylamine enhanced silanization of 3-methacryloxy-propyltrimethoxysilane to silica (SiO_2) surfaces, and cyclohexane yielded a more water-resistant silica–silane bond [10]. In the present study, the fusion of silica onto the silicon carbide whiskers, and the use of *n*-propylamine with silane in cyclohexane for the silanization of the silica-fused whiskers may have contributed to the strong whisker–silane bonding.

2. The residual stresses in the composite are caused by the thermal expansion mismatch between the fillers and the matrix: $\sigma_r = E \cdot \Delta\alpha \cdot \Delta T$, where σ_r is residual stress, E is elastic modulus, and $\Delta\alpha$ is thermal expansion difference between filler and matrix. $\Delta T = T_g - T$ where T_g is the glass transition temperature of the matrix and T is the temperature of consideration (5 °C or 60 °C in this study) which will be described below [40, 41]. For the whisker composites (for a simple estimation, neglect the complication from the presence of silica particles), the thermal expansion coefficient α of silicon carbide is approximately $4.5 \times 10^{-6}/°C$ [41], and α of resin is about $93 \times 10^{-6}/°C$ [42]. Therefore, when the composite cooled down after curing, the whiskers were under compression, but the resin near the whisker surface was in axial and circumferential tension. The compressive stresses on the whiskers may have been beneficial in retaining the whiskers in the matrix. However, the tensile stresses in the matrix, coupled with cyclic fatigue, could induce microcracking [40, 41, 43]. For an estimate of the tensile stress in the matrix near the whisker–matrix interfaces (the composites were heat-cured at 140 °C in the present study), take the resin glass transition temperature T_g of approximately 100 °C [44], then at 5 °C, $\Delta T = (100 °C–5 °C)$, $\Delta\alpha = 88.5 \times 10^{-6}/°C$, and $E = 5$ GPa for the matrix [45], hence $\sigma_r = 42$ MPa. Similarly, at 60 °C, σ_r is equal to 18 MPa, and both are tensile stresses. In the present study, no microcracks in the resin near the whisker–resin interfaces were observed with SEM at magnifications up to 30,000, nor was significant strength loss measured due to thermal cycling. While this appears to suggest that cyclic fatigue between tensile stresses of 42 MPa and 18 MPa for 10^5 cycles was not enough to break down the resin, actual cyclic fatigue tests of the unfilled resin specimens are required to examine this.
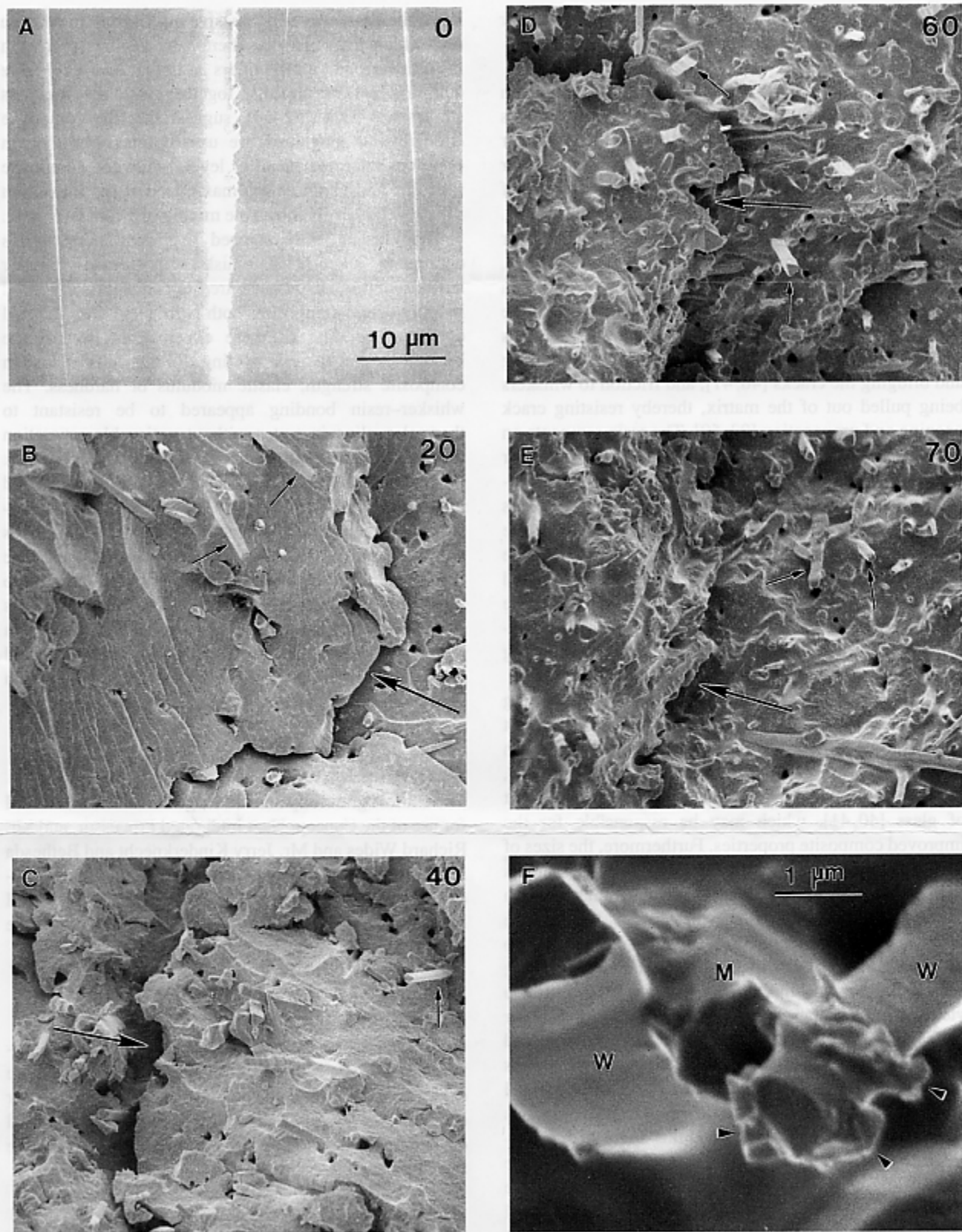
Figure 5 Fracture surfaces of whisker composites, after 10^5 thermal cycles, at filler levels of (A) 0%, (B) 20%, (C) 40%, (D) 60%, and (E) 70%. The magnification was the same for (A)–(E), but (F) is a higher magnification of (E). The fracture surface of unfilled resin was flat with brittle fracture lines; those of the whisker composites were rougher, with steps (long arrows in B–E) and whisker pullout (short arrows). The whiskers appeared well bonded with the matrix. In (F), W denotes whiskers, M denotes matrix, and the small arrows point to remnants of matrix on the pulled-out whisker, indicating strong whisker–resin bonding even after 10^5 thermal cycles.

3. Hydrolysis of the whiskers was not a concern as silicon carbide whiskers and fibers are chemically very stable and able to survive hostile environments even at high temperatures [41]. This is in contrast to the observed hydrolysis of some types of glass fillers [46, 47], and the observed strength loss of some dental resin composites in

thermal cycling [10, 32]. The matrix resin did not seem to have significantly degraded in thermal cycling, manifested by the measured strength of the unfilled resin. However, 10^5 thermal cycles in the present study was equivalent to only 1056 h (6.3 weeks) of immersion in water. Longer-term water aging studies up to two years

are underway to examine the behavior of the whisker composites and the unfilled resin.

Reinforcement with silica particle-fused silicon carbide whiskers resulted in substantial improvements in composite mechanical properties both before and after thermal cycling. The flexural strength of the whisker composite at 70% filler level was nearly twice those of the control indirect composites with similar filler levels. The elastic modulus of whisker composite at 70% filler level was larger than those of the control composites, similar to that of human dentin [39], but still smaller than that of enamel [39]. Based on SEM observations of the fracture surfaces (Fig. 5), the reinforcement mechanisms of whisker composite appeared to be whiskers deflecting and bridging the cracks [40, 41], and friction to whiskers being pulled out of the matrix, thereby resisting crack opening and propagation [23, 50]. The resin remnants on the pulled-out whiskers suggest the difficulty in whisker pullout, contributing substantially to the fracture resistance of the composite. These toughening mechanisms resulted in relatively large steps and rough fracture surfaces for the whisker composites, in contrast to the relatively flat fracture surfaces of the unfilled resin and the control composites filled with glass particles. These features constituted a high surface area, further contributing to the composite toughness by consuming energy in creating new surfaces [40, 41].

High strength fillers are less likely to be broken during crack pinning, crack bridging, and frictional pullout [23, 48]. The whiskers possessed a tensile strength an order of magnitude higher than that of glass fibers [49], and fracture toughness of approximately three times that of glass [40, 41], which may be responsible for the improved composite properties. Furthermore, the sizes of whiskers were much finer than those of short glass fibers (i.e., a mean diameter of about $15\,\mu m$, and a mean length of about $100\,\mu m$) used in previous studies to reinforce dental resin composites [2, 19]. Smaller fillers tended to be more uniformly distributed in the matrix, improving composite strength and wear resistance [51], and also yielding a smoother surface after machining [38] or polishing [23]. In addition to filler strength, size and shape, filler level also affected the composite properties [11, 52–55]. Previous studies on the effect of filler level appeared to have controversial results. Some studies showed that the composite modulus increased monotonically with increased filler level [53], and the composite wear depth decreased (or wear resistance increased) monotonically with increasing the filler level [11]. However, other studies showed that the composite strength and toughness first increased with filler level, but then decreased with a further increase in filler level [54]; and the fatigue resistance of composite increased with filler level, reaching a maximum, then decreased with a further increase in filler level [55]. Both phenomena were observed in the present study: the composite strength first increased with filler level up to 60%, then plateaued or slightly decreased, when the filler level was further increased to 70% (Fig. 1); the modulus and hardness, on the other hand, increased monotonically with filler level (Figs 2 and 3). The whisker–resin paste at 60% filler level was relatively easily mixed. At 70% filler level, the paste was still cohesive but starting to become dry, which indicates that there may be barely enough resin to surround all the fillers and bond into a cohesive composite. These results, together with the previous studies [23, 33, 45, 52–55], suggest that the composite strength and toughness are usually maximized at an optimum intermediate filler level, while the composite modulus and hardness are maximized at the maximum filler level that still allows the mixing of a cohesive paste.

The present study showed that dental composites containing silica-fused whiskers possessed superior strength and modulus compared to conventionally filled indirect dental composites both before and after thermal cycling. The data of these experiments support the hypothesis that thermal cycling did not cause a loss in composite strength, elastic modulus or hardness. The whisker–resin bonding appeared to be resistant to thermal cycling in water without noticeable separation or cracking. The flexural test showed that the bulk properties of the composite specimens were not degraded in thermal cycling; the nano-indentation test demonstrated that even the thin surface layer of the specimens was not degraded after 10^5 thermal cycles in water. The whisker composite possessed flexural strength nearly two times those of currently available indirect composites both before and after thermal cycling; this is likely a result of silica-whisker fusion enhancing silanization and filler–resin bonding, as well as the high-strength whiskers bridging and resisting crack propagation.

Disclaimer
Certain commercial materials and equipment are identified to specify the experimental procedure. In no instance does such identification imply recommendation or endorsement by NIST or ADAHF or that the material or equipment identified is necessarily the best available for the purpose.

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