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Improving fracture toughness of dental nanocomposites by interface engineering and micromechanics

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Abstract

The fracture toughness of dental nanocomposites fabricated by various methods of mixing, silanization, and loadings of nanoparticles had been characterized using fatigue-precracked compact-tension specimens. The fracture mechanisms near the crack tip were characterized using atomic force microscopy (AFM), transmission electron microscopy (TEM), and scanning electron microscopy (SEM). The near-tip fracture processes in the nanocomposites were identified to involve several sequences of fracture events, including: (1) particle bridging, (2) debonding at the poles of particle/matrix interface, and (3) crack deflection around the particles. Analytical and finite-element methods were utilized to model the observed sequences of fracture events to identify the source of fracture toughness in the dental nanocomposites. Theoretical results indicated that silanization and nanoparticle loadings improved the fracture toughness of dental nanocomposites by a factor of 2 to 3 through a combination of enhanced interface toughness by silanization, crack deflection, as well as crack bridging. A further increase in the fracture toughness of the nanocomposites can be achieved by increasing the fracture toughness of the matrix, nano-filled particles, or the interface.

Keywords

dental nanocomposites; fracture toughness; toughening mechanisms; interface engineering

1.0 Introduction

The dental restoratives commonly known as “microfilled” composites resins are usually based on fumed colloidal silica fillers. Such fillers reinforce the matrix while offering high polishability, high optical translucency, and low initial wear rates compared to other composite technologies. However, silica is not inherently radiopaque and therefore does not provide the contrast needed for the diagnosis of marginal leakage and secondary caries. Furthermore, fumed silica is difficult to disperse homogeneously in monomer due to particle chain formation, which increases resin viscosity at even modest filler loading and results in decreased ease of placement and poor functional adaptation. For these reasons, there are considerable recent interests in developing new dental composites reinforced with nano-sized particles with near-zero shrinkage rate during curing and are highly translucent and radiopaque after cured [1–6]. Other desired properties of the nanocomposites include high strength, good fracture toughness, and excellent wear resistance [6].

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While it is apparent that nanosized filler particles can provide substantial improvements in the composite strength and wear resistance, it is not obvious how nano-sized particles can enhance the fracture resistance of dental nanocomposites since fracture toughness usually scales with the $\frac{1}{2}$ power of the characteristic microstructural length scale controlling the fracture process. To be an effective toughening agent, the nanosized particles must increase the process zone size at the onset of critical fracture either through an increase in the fracture strength due to their small size scale or the inducement of one or more new toughening mechanisms by virtue of their size scale or the large surface areas to volume ratio.

The objective of this paper is to give an overview of the fracture mechanisms in selected dental nanocomposites whose fracture toughness is mostly controlled by fracture along the particle/matrix interface. For these nanocomposites, the fracture toughness can be enhanced by silanation to increase the interface toughness and the energy dissipated during the fracture process. The fracture process is examined in details by considering several toughening mechanism including crack deflection, crack trapping, and crack bridging by nanoparticles via microemchanical modeling. The theoretical results are compared with experimental data of fracture toughness to identify possible means to further improve the fracture resistance in dental composites reinforced with nano-sized particle fillers.

2.0 Overview of Fracture Mechanisms in Dental Nanocomposites

The Stöber process was utilized to prepare non-associated colloidal silica particles in the 40 to 120 nm range. The nanoparticles were then silanized by direct addition of γ -methacryloxypropyl trimethoxysilane (MPTMS), and dispersed in a bis-GMA based monomer blend (GTE) to form homogeneous composites of up to 70 wt.% (43% by volume), which are referred to as GTE/Stöber SiO_2 nanocomposites. In addition, nanoparticles ($\approx 0.4 \mu\text{m}$ in size) of Schott glass, which is a mixture of 50% SiO_2 , 30% BaO, 10% Al_2O_3 , and 10% B_2O_3 by weight, were silanized by direct addition of γ -methacryloxypropyl trimethoxysilane (MPTMS), and dispersed in a bis-GMA based monomer blend (GTE) to form homogeneous composites of up to ≈ 79 wt.% (43% by volume). Several different mixing methods were utilized to disperse the Schott glass particles in order to achieve the various loading levels in the GTE/Schott glass nanocomposites. Details of the fabrication techniques and processing conditions are described in the Appendix and in a future publication [7]. Table 1 summarizes the various proceeding methods used to fabricate the nanocomposites.

Small disc-shaped compact-tension, DC(T), specimens of 6 mm in width (measured from the load-line to the edge of the specimen) and 3 mm in thickness with a crack length of about 2.6 – 3.2 mm were tested at ambient temperature to determine the fracture toughness as a function of the loading levels (weight percent) of nanoparticle fillers. Six specimens were tested at each of the various loading levels. Fracture toughness tests were performed in a Sintech 20/G screw-driven testing machine under a displacement rate of 0.025 mm/s until specimen fracture. The maximum load at fracture was utilized to compute the critical stress intensity factor, K_{IC} , at fracture using the ASTM E399 procedure [8]. Fracture mechanisms were determined by performing TEM and AFM on partially fractured specimens.

The TEM micrograph in Figure 1(a) shows the microstructure of the bis-GMA/TEGDMA/Bis-EMA(GTE) composites with silanized Stöber-silica nanoparticles filler. As shown in Figure 1(a), the Stöber-silica particles, which are about 30 – 40 nm, are well dispersed with uniform particle spacing in the nanocomposites. The crack path in the GTE composite with silanized Stöber-silica nanoparticles tended to follow the matrix/particle interface or resided within the matrix. It was sometimes difficult to distinguish the interface crack path from that in the matrix since the nanoparticles were very close together. Figure 1(b) shows two microcracks of a crack length in the range of 300 – 600 nm and a few microcracks in the range of 15 – 90 nm. Some

of the smaller microcracks appear to form at the matrix/particle interface and go around the particles along the interface, and then link with another interface crack in the contiguous nanoparticles. Fracture of the SiO₂ particles was not observed.

Figure 2(a) and (b) show the AFM images of the path of a monotonically loaded crack in the GTE/Schott glass nanocomposite during fracture toughness testing. For both cases, the micrograph on the left (labeled height) shows the topography of the microstructure, while the one on the right (labeled phase) shows the hardness variation in the microstructure. At the low magnification micrographs in Figure 2(a), the crack was seen to propagate in the GTE matrix, intersect the particles, and grow around the particles by following the matrix/particle interface. The higher magnification micrographs in Figure 2(b) show that the crack advances along the particle/matrix interface, but some of the nanoparticles appear to be either cracked or clumped together. In many instances, interface debonding is evident in particles located ahead of the crack tip, as shown in Figure 3(a). Fractography revealed that the fracture surfaces exhibited a lumpy appearance, which is typically associated with fracture along the matrix/particle interface, and the absence of particle fracture, Figure 3(b). Taken together, the results in Figures 1–3 indicate that interface fracture is the prevalent fracture mechanism in these two dental nanocomposites.

3.0 Finite-Element Analysis of Interface Crack Deflection Around Particles

It is well known that crack deflection by particles can lower the local crack-tip stress intensity factor and enhance fracture resistance. There are several crack-tip shielding models [8–10] in the literature that treat crack deflection by particles to a tortuous path from the mode I crack orientation, the crack-tip shielding effect and consequently the fracture toughness enhancement increase with increasing values of the crack-deflection angle θ which measures the tilt or the twist angles of the deflected crack path, as shown in Figure 4(a). In comparison, the deflected crack paths observed in the two dental composites often initiated from the poles of the particles, followed the interface, and propagated around the particles, as shown schematically in Figure 4(b). The local stress intensity factors for the latter type of crack deflection are not known and need to be investigated in order to develop a deeper level of understanding of the origin of fracture resistance in the nanocomposites.

Finite-element method was utilized to analyze the growth of an interface crack around an elastic particle embedded within an elastic/plastic matrix. Figure 5 shows the finite-element mesh of the composite unit cell used in the stress intensity factor associated with the interface crack growth calculation. The unit cell of dimensions $L \times L$, which contains 50% particle and 50% matrix by area, is subjected to principal stresses σ_1 and σ_2 applied along the y-axis and x-axis, respectively. Two boundary layers of elements of uniform size were specified along the interface between the matrix and the particle for avoiding any artifact that may result from nonuniform element size. A small interface crack of length a was placed between the two boundary layers, as shown in Figure 6, by releasing the appropriate nodes at the apex of the circular particle. For a given set of the applied stresses, the stress field around the interface crack was computed. A critical normal stress criterion was applied to an element located at a distance of $0.002L$ ahead of the tip of the interface crack. The interface crack was extended by releasing the nodes in the near-tip elements where the normal stress exceeded the specified critical value of 100 MPa.

The numerical scheme was utilized to simulate the growth of interface crack growth for three values ($\lambda = 0, 0.5$, and 1) of the biaxial stress ratio, σ_2/σ_1 , where σ_1 is along the y-axis and σ_2 is along the x-axis. Figure 6 summarizes the development of the crack-tip plastic zone as the interface crack extends in length under uniaxial tension, $\sigma_2/\sigma_1 = 0$. At small interface crack length, the near-tip stresses are elastic and contain high normal stress components. As the

interface crack increases in length and extends around the circular particle, the crack opening displacement increases and an asymmetric elastic-plastic zone develops at the crack tip. The near-tip distributions of the von Misses stresses are as shown in Figures 6a, b, and c for a/L of 0.1, 0.3, and 0.5, respectively.

The mode I and II stress intensity factors of the interface crack were computed as a function of the crack length. Figure 7(a) shows the results of K_I and K_{II} , normalized by $\sigma_1(\pi a)^{1/2}$ as a function of the circular length, a , of the interface crack normalized by the length L of the unit cell. The results indicate that the mode I stress intensity factor, K_I , increases with increasing crack length for small values of a/L , but then decrease with increasing a/L when $\theta = \tan^{-1}(a/L)$ exceeds 1.8° . In contrast, the mode II stress intensity factor, K_{II} , decreases rapidly with increasing a/L . Figure 7(b) presents the results of equivalent K , defined in terms of K_I and K_{II} , increase with a/L and then decreases with increasing a/L when $a/L > 0.031$. For $a/L > 0.25$, K_{eq} decreases to $0.6 - 0.67$ of the $\sigma_1(\pi a)^{1/2}$ value and remains relatively constant for $a/L > 0.25$. The result indicates that the driving force for crack growth decreases with increasing crack length when an interface crack grows around a particle from the pole position to the lateral position. For $a/L > 0.25$, the toughening ratio is relatively constant with a value of $1.49 - 1.67$. In comparison, the toughening ratios for tilt- and twist-type crack deflection are about 1.3 to 1.6, respectively. Thus, the toughening ratios by tilt- and twist-types crack deflection are similar to those achieved by shielding of an interface crack advancing from the pole position and around the particle/matrix interface when the interface toughness and matrix toughness are identical ($K_{in} = K_m$).

4.0 Modeling of Toughening Mechanisms and Fracture Toughness

The fracture process of crack deflection and interface cracking were modeled to investigate their role in the fracture toughness of the nanocomposites with various levels of particle fillers. In addition, toughening mechanisms such as crack-trapping and particle bridging have been modeled to explore their potential applications to improving the fracture toughness of nanocomposites. A summary of the various toughening models is described in the sub-sections below.

4.1. Crack Deflection and Interface Cracking

Crack deflection is a shielding mechanism that improves the fracture resistance lowering the local stress intensity factors at a crack tip. For spherical particles with tilt-induced deflection, the fracture toughness of the composite is given by [9–11]

$$K_C = [1 + \alpha_{tl}\alpha_{ds}V_f]^{1/2} K_m \quad (1)$$

where K_C and K_m are the critical stress intensity factors (fracture toughness) of the composite and matrix, respectively; V_f is the volume fraction of the particles and $\alpha_{tl} = 0.87$ is a constant related to toughening by pure tilt-induced crack deflection. The parameter α_{ds} represents the toughening increase due to crack deflection by particles with a distributed spacing and $\alpha_{ds} = 1.6$ for distributed spherical particles. For a deflected crack advancing on the particle/matrix interface, the fracture toughness composite, which arises from contributions of the matrix and the interface, is given by

$$K_C = \left[1 + \alpha_{tl}\alpha_{ds}V_f \left(\frac{K_{in}}{K_m} \right)^2 \right]^{1/2} K_m \quad (2)$$

where K_{in} is the interface toughness. The parameter α_{ds} is replaced by α_{pr} with $\alpha_{pr} = 1.49 - 1.67$ for an interface crack advancing from the pole and around the particles. For simplicity, $\alpha_{pr} \approx \alpha_{ds} \approx 1.6$.

4.2. Crack Trapping and Bridging

The trapping and bowing of a crack front around an array of spherical, ductile particles in a brittle matrix have been analyzed by Bower and Ortiz. Their analysis indicated that crack trapping and bowing can provide a slight increase in the fracture resistance of the composite. A larger increase in the composite fracture resistance can be attained if the crack front bows around the ductile particles, advances into the brittle phase, and leaves intact particles in the crack wake bridging the crack surfaces. The onset of particle pinning and crack bridging occurs when the fracture toughness of the ductile particles exceeds a critical value, which is about three times of the fracture toughness of the brittle phase [12]. The analysis of Bower and Ortiz [12] was subsequently modified by Chan and Davidson [13] to cover the entire range of volume fraction of ductile phase from 0 to 1 and to include the effect of plastic constraint on fracture toughness. According to the modified analysis, the fracture toughness of a composite that exhibits a combination of crack trapping, bowing, and bridging process is given by [13]

$$K_c = K_m \left[1 + \sqrt{1 - V_f} \left(\left[\frac{K_p}{K_m} \right]^2 - 1 \right) \right]^{1/2} \quad (3)$$

where K_c , K_m , and K_p are the critical stress intensity factors (fracture toughness) for the composite, the matrix phase, as the particles, respectively. In the presence of plastic constraint, the fracture toughness of a composite reinforced with toughening particles is reduced according to the expression given by [13]

$$K_c = K_m \left[1 + \sqrt{1 - V_f} \left(\left[\frac{K_p}{K_m} \right]^2 \exp \left\{ -\frac{8q}{3} \left[\frac{V_f}{1 - V_f} \right] \right\} - 1 \right) \right]^{1/2} \quad (4)$$

$V_f < V_{crit}$, but [12]

$$K_c = K_p \left[1 + \sqrt{1 - V_{crit}} \left(\left[\frac{K_p}{K_m} \right]^2 \exp \left\{ -\frac{8q}{3} \left[\frac{V_{crit}}{1 - V_{crit}} \right] \right\} - 1 \right) \right]^{1/2} \quad (5)$$

for composites with $V_f \geq V_{crit}$, where q is a constant with a typical value of 1 and V_{crit} is the critical volume fraction of particles at which the particles are in contact with each others and becomes the continuous phase.

4.3. Rule-of-Mixtures

The fracture toughness expression based on the rule of mixtures has been derived earlier by Chan and Davidson [13] on the basis of elastic strain energy rates dissipated by the constituent phases in the fracture process zone at the onset of the critical fracture event. According to this analysis,

$$K_c = K_m \left[V_f + (1 - V_f) \left(\frac{K_p}{K_m} \right)^2 \right]^{1/2} \quad (6)$$

where K_p the fracture toughness of the toughening particles, while K_m is the fracture toughness of the matrix or the toughened phase.

5.0 Model Applications to Dental Nanocomposites

The material input to the fracture toughness models include the fracture toughness values of the constituent phases, and the volume fractions of the particle filler in the composites. Although the theoretical fracture toughness models are based on volume fractions, there is a need to convert volume percents to weight percents through the particle and matrix densities because the dental composites are fabricated and the corresponding fracture toughness data are generally reported in terms of the filler loading levels or weight percents. The K_{Ic} value of the matrix phase ($K_{Ic} = 0.22 \text{ Mpa } \sqrt{\text{m}}$) was measured experimentally using compact-tension specimens fabricated from the matrix material using the composite processing technique but without particle fillers. The measured value of $K_{Ic} = 0.22 \text{ Mpa } \sqrt{\text{m}}$ is in agreement with literature data [14]. The fracture toughness of the Stöber SiO_2 nanoparticles were taken to be $1 \text{ Mpa } \sqrt{\text{m}}$, which is slightly higher than that of fused silica ($0.8 \text{ Mpa } \sqrt{\text{m}}$) [15] while that of the Schott glass was estimated based on the weighted average of the fracture toughness of the constituent particle ($K_{Ic} = 0.8 \text{ Mpa } \sqrt{\text{m}}$ for SiO_2 [15] and B_2O_3 ; $K_{Ic} = 1.2 \text{ Mpa } \sqrt{\text{m}}$ for Al_2O_3 [16]; $K_{Ic} = 1.3 \text{ Mpa } \sqrt{\text{m}}$ for BaO [17]). Since the dental composites was processed and reported in the dental literature on the basis of weight percents or loading levels, the volume fractions of the particles were computed on the basis of the weight fractions and the density of the matrix (1.2 g/cm^3) [18] and those of the nanoparticles ($2.46, 3.80, 1.75$, and 5.72 g/cm^3 for B_2O_3 , Al_2O_3 , SiO_2 , and BaO , respectively) [18]. The only unknown was the interface toughness (K_{Ii}) and it was varied from 1 to 3 times of that of the matrix toughness, while using $\alpha_{II} = 0.87$, $\alpha_{ds} = 1.6$, and $\alpha_{pr} = 1.6$ for interface cracking around the particles.

The theoretical results were compared against experimental data to elucidate the effects of interface and reinforced particles on the fracture toughness of GTE/ SiO_2 and GTE/Schott glass nanocomposites. Figure 8 shows a comparison of the measured and computed fracture toughness values of GTE/ SiO_2 nanocomposites as a function of wt.% Stöber silica particles. The toughening mechanisms considered in the theoretical modeling included crack deflection, crack trapping and bridging, and the rule-of-mixture. The rule-of-mixture tends to overpredict the fracture toughness of the nanocomposites, while the crack-trapping and bridging model and the crack deflection model are in reasonable with the experimental data. The agreement with the crack trapping and bridging model may be fortuitous since particle fracture was not observed. Three different levels of the interface toughness are utilized in the crack deflection model with $K_{Ii}/K_{Im} = 1, 2$, and 3 . For $K_{Ii}/K_{Im} = 1$, fracture toughness enhancement is entirely due to the shielding of the crack tip by a tortuous crack path. The toughness enhancement due to crack-tip shielding is relatively small (about 60%) and cannot explain the fracture toughness levels observed in the nanocomposites. For $K_{Ii}/K_{Im} > 1$, the interface toughness and crack-tip shielding both contribute to the fracture toughness of the nanocomposite. A ratio of K_{Ii}/K_{Im} of 2 to 3 gives the best agreement with the measured fracture toughness values, suggesting that the interface fracture toughness of the GTE/ SiO_2 nanocomposites is about two to three times higher than that of the GTE matrix.

Similarly, Figure 9 shows the observed fracture toughness for the GTE/Schott glass nanocomposites compared against model calculation based on crack deflection (Eq. 2) and interface fracture with three different ratios of interface toughness to matrix (GTE) toughness. At $K_{Ii}/K_{Im} = 1$, the toughness of the interface is identical to that of the matrix and only a small toughness enhancement can be achieved by crack deflection at the particle/matrix interface. Crack deflection enhances the fracture toughness in the nanocomposite when $K_{Ii}/K_{Im} = 2$ or 3 . A comparison of the computed and measure fracture toughness again suggests that the enhanced composite fracture toughness might originate from a two- or three-fold increase in the interface toughness. The interface toughness, estimated to be 0.44 to $0.66 \text{ Mpa } \sqrt{\text{m}}$, is

bounded by the fracture toughness ($0.22 \text{ Mpa} \sqrt{\text{m}}$) of the matrix and the fracture toughness ($0.66 \text{ Mpa} \sqrt{\text{m}}$) of the particles.

To understand the absence of particle fracture in the GTE/Schott glass nanocomposites, fracture toughness enhancement due to crack-tip trapping and bridging mechanisms is compared against those predicted by the rule-of-mixtures and interface fracture in Figure 9. The results indicate toughness enhancements that can be achieved from interface fracture is slightly lower than the other toughening mechanisms. The lower fracture toughness by interface fracture explains the absence of particle fracture in the nanocomposites. Furthermore, a further increase in the interface toughness by improved processing techniques would not increase the composite fracture toughness, but would change the fracture mechanism to crack-tip bridging and particle fracture, unless the toughness of the particles is also increased.

6.0 Discussion

The FEM K solutions indicate that an interface crack initiated at the pole of a spherical particle and advancing around a circular particle experiences an increasing K_I value with increasing crack length for small crack lengths ($a/L < 0.03$). Because of the rising K_I , the growth of the apex crack is expected to be unstable initially but becomes stable subsequently after the K_I and K_{II} values both decrease with increasing crack length for $a/L > 0.03$. The FEM K_I and K_{II} solutions are reminiscent of the analytical solutions obtained by Sendecky [19] for debonding of rigid cylindrical inclusions in a composite.

The interface crack path around a spherical particle gives essentially the amount of crack-tip shielding as those that can be achieved by tilt- or twist-induced crack deflection. In all three cases, the toughening ratio ranges from 1.3 – 1.6 or 30% to 60% improvement in the composite fracture toughness. This level of fracture toughness enhancement is considerably less than the 2 – 3 times enhancement observed in the dental nanocomposites, implying the presence of an additional toughening mechanism. Since crack deflection and interface crack growth are the observed fracture mechanisms, the finding suggests that the fracture toughness enhancement originates from increases in the interface toughness resulting from the use of coupling agents silanization during the composite fabrication process. Silanization is beneficial for interface toughness because it helps to establish Si-based bonds between the matrix and the particles [20,21]. Since fracture of both the dental nanocomposites is dominated by interface fracture, it appears that the interface bonds are not sufficiently strong to induce particle fracture. The nano-sized particles appear to enhance fracture toughness of the nanocomposites in two ways, which are (1) the large surface to volume ratio that aids to improve interface bonding and consequently the interface toughness, and (2) the high strength of the nano-sized particles that helps to prevent particle fracture during interface cracking. With nano-sized particles, the interface toughness can be increased to higher levels without the risk of causing particle fracture and thus allows a higher fracture toughness value to be attained in a nanocomposite. Since the computed fracture toughness values based on these toughening are comparable, a further increase of the interface toughness may cause fracture of the bridging particles, resulting in a change of fracture mechanism but not necessarily an increase in the fracture toughness unless the fracture toughness of the particles is also increased.

7.0 Conclusions

The conclusions reached in this study are as follows:

1. The dominant fracture mechanism in GTE/SiO₂ and GTE/Schott glass nanocomposites is crack deflection and crack growth along the matrix/particle interface.

2. Interface crack growth around a particle lower the near-tip effective stress intensity and increase the fracture toughness by about 30 to 60% of the matrix toughness.
3. The fracture toughness of nanocomposites with silanized nanoparticles are two to three times higher than that of the matrix toughness.
4. Nano-sized particles can improve the fracture toughness of dental composites by enhancing the interface bonding between the particle and matrix through a higher surface area to volume ratio and a high particle strength.
5. Further increase in the interface toughness of GTE/SiO₂ and GTE/Schott glass nanocomposites might cause a change of the dominant fracture mechanism from interface fracture to particle fracture.

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APPENDIX

Composite resins were formulated with a 3-component monomer solution containing 37.5% (w/w) bis-GMA, 37.5% (w/w) bis-EMA, and 25% (w/w) TEGDMA. A liquid photoinitiator system comprised of camphorquinone and dimethylaminoethyl methacrylate (0.4 g:1.0 g) was added to the monomer solution at a total level of 3% (w/w) prior to formulation with fillers. Three different barium-modified glass fillers with an average particle size of 0.4 μm were selected for the study: a 9.4% silanated filler (Product #8235-UF0,4-sil9,4, Schott Glass, GmbH), a 5.6% silanated filler (Product #8235-UF0,4-sil5,6, Schott Glass, GmbH), and a 50% (w/w) dry mixture of the two for a combined silane level of 7.5%. Fillers were admixed into each respective resin batch at a level of up to 79% (w/w) based on the total weight of monomer, including photoinitiator. Mixing was performed on 15 g batches using a SpeedMixer (Model DAC150FV, FlackTek, Inc., Landrum, SC). Mixing was performed for a total of 3 minutes at increasingly high speed in the range of 3000 – 3500 rpm with vacuum applied during the final minute. Disc-shaped compact-tension, DC(T), specimens 6 mm in width (W) measured from the load-line to the edge of the specimen and 3 mm in thickness ($B = W/2$) were prepared by packing the composite resins into a stainless steel mold after applying a perfluorinated release agent (Krytox, DuPont) to the metal surfaces. The stainless steel mold contained two screws to create the loading holes and a razor blade to create a sharp notch. The loading holes were 1.5 mm (W/4) in diameter and the nominal notch length was about 2.5 mm. The specimens were cured for 1 minute using a handheld dental curing lamp (Optilux 400, Demetron Research Corp.), prior to release from the mold, and then post-cured in a halogen light box (CureLite Plus, Jeneric/Pentron, Inc.) for 10 additional minutes. After released, the notch length of the DC(T) specimens was further extended to 2.6 – 3.2 mm by inducing a small crack at the tip of the notch via a razor blade. The sum of the notch length and crack length was taken to be the total crack length. Fatigue precracking was attempted but the composites were too brittle to obtain a fatigue precracking without fracturing the specimen.

(a)

(b)

Figure 1. TEM bis-GMA/TEGDMA/Bis-EMA(GTE) composite with silanized Stöber-Silica nanoparticles filler showing: (a) microstructure, and (b) microcracks on the order of 15 – 90 nm in lengths. Microcracks appear to initiate at and grow along the matrix/particle interface. Loading direction is horizontal.

(a)

(b)

Figure 2.

AFM micrographs of matrix and interface cracks in GTE/Schott glass nanocomposites: (a) low magnification micrograph showing main crack and interface microcracks at particles, and (b) high magnification micrograph showing interface cracks around the particles.

(a)

(b)

Figure 3.

Interface crack propagation in GTE/Schott glass nanocomposites: (a) crack initiated at the pole of a particle and crack propagation around the matrix/particle to link with the main crack; the micrograph labeled height (left) shows the topography and the micrograph labeled phase (right) shows the hardness variations in the microstructure; and (b) fracture surface showing a lumpy appearance due to interface fracture around particles without particle fracture in GTE/Schott glass nanocomposites.

Figure 4. Crack deflection mechanisms: (a) tilt- or twist-type crack deflection by a particle, and (b) crack initiation of the pole of a particle and propagation around a particle to link with the main crack.

Figure 5.
Finite-element mesh of a hard particle embedded in an elastic matrix separated by an interface.

Figure 6.

Figure 6(a). Finite-Element-Method (FEM) modeling of crack propagation along the interface of GTE/Schott glass dental nanocomposite for various normalized circular crack lengths of: (a) $a/L = 0.1$, (b) $a/L = 0.3$, and (c) $a/L = 0.5$. The principal stresses σ_1 and σ_2 are applied along the y- and x-axes, respectively.

Figure 6(b). Finite-Element-Method (FEM) modeling of crack propagation along the interface of GTE/Schott glass dental nanocomposite for a normalized circular crack length of 0.3. The principal stresses σ_1 and σ_2 are applied along the y- and x-axes, respectively.

Figure 6(c). Finite-Element-Method (FEM) modeling of crack propagation along the interface of GTE/Schott glass dental nanocomposite for a normalized circular crack length of 0.5. The principal stresses σ_1 and σ_2 are applied along the y- and x-axes, respectively.

(a)

(b)

Figure 7.

Stress intensity factors normalized by $\sigma_1 \sqrt{\pi a}$ as a function of circular crack length a normalized by unit cell length L for various ratios (λ) of σ_2 to σ_1 : (a) K_I and K_{II} , and (b) equivalent K_{eq} , where $K_{eq} = [K_I^2 + K_{II}^2]^{1/2}$. The principal stress σ_1 is along the y-axis (vertical) and σ_2 is along the x-axis (horizontal).

Figure 8.

Fracture toughness data of GTE/SiO₂ nanocomposites compared against model predictions based on the interface fracture crack deflection model, crack trapping and bridging model, and the rule-of-mixtures.

Figure 9.

Measured fracture toughness values of GTE/Schott glass dental composites compared against model prediction based on crack deflection and interface fracture using three different assumed values for the ratio of interface toughness (K_{in}) to matrix toughness (K_m).

Figure 10.

Measured fracture toughness values of GTE/Schott glass dental composites compared against model predictions for three different fracture models: (1) crack deflection and interface fracture, (2) crack-tip trapping, bridging, and particle fracture, and (3) the rule-of-mixtures.

Table 1

A Summary of the Processing Conditions for GTE/Schott Glass Nanocomposites

Processing Procedure	Particle Loading, wt %	Silane, Wt %	Mixing Method
Method 1	0, 20, 40, 60, 75	9.5	Mortar and Pestle
Method 2	78.5	3	Premixed at FlackTek
Method 3	78.5	3	FlackTek SpeedMixer
Method 4	79	5.6, 7.5, 9.4	FlackTek SpeedMixer
Method 5	77.5	5.6, 7.5, 9.4	FlackTek SpeedMixer
Method 6	73	5.6, 7.5, 9.4	FlackTek SpeedMixer