Factors Involved in Mechanical Fatigue Degradation of Dental Resin Composites

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ABSTRACT

The design of clinical trials allows for limited insights into the fatigue processes occurring in resin composites and the factors involved therein. In vitro studies, in contrast, can fundamentally narrow study interests to focus on particular degradation mechanisms and, to date, represent the major contributors to the state of knowledge on the subject. These studies show that microstructural features are important in determining strength and fracture toughness, whereas fatigue resistance is mainly related to the susceptibility of the matrix and the filler/matrix interface to mechanical and chemical degradation. In this review, we focus on fracture mechanisms occurring during fatigue, on the methods used to assess them, and on additional phenomena involved in the degradation of initial mechanical properties of resin composites.

KEY WORDS: composite resins, cyclic loading, fracture toughness, viscoelasticity, hydrolysis, crack propagation.

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INTRODUCTION

Clinical evidence on the performance of resin composite restorations points to fracture as one of the 2 main factors leading to clinical failure. Unfortunately, clinical efforts engaged in the long-term evaluation of resin composite restorations contribute rather superficially to our understanding of their fracture behavior. Because complete observation periods are usually unavailable, partial observations often lead to premature interpretations, due to the dynamic behavior of survival curves (Bayne, 2012). Adding to the confusion, fractures in clinical trials are usually treated statistically along with other failure criteria, resulting in fracture distribution patterns that are not readily clear. Differences in the mechanical behavior between classes of resin composites are therefore difficult to evaluate by only looking at clinical data. In a recent meta-analysis, no differences in bulk fracture rates could be observed among microfilled, macrofilled, and hybrid composites in studies evaluating class II restorations for up to 10 years (Heintze and Rousson, 2012). In function, fatigue mechanisms occurring in different composites can take longer than that to result in a perceptible difference in mechanical performance (Da Rosa Rodolpho *et al.*, 2011). Interestingly, much of the clinical data points to the importance of studying the fatigue of resin composites in bending (Brunthaler *et al.*, 2003; Pallesen and Qvist, 2003; Van Neuiwenhuysen *et al.*, 2003; Opdam *et al.*, 2004, 2007).

For *in vitro* test methods to achieve clinical relevance (Della Bona and Watts, 2013), the assessment of composite degradation due to fatigue may emerge as an important clinical predictor (Ferracane, 2013). The purpose of this review is to better understand this complex phenomenon, by reviewing the work during the past 5 years on the mechanical properties of dental composites and their potential degradation in relevant environments.

INFLUENCE OF MICROSTRUCTURE ON MECHANICAL PROPERTIES

Elastic Modulus and Strength

The use of model composites is the ideal experimental strategy for systematic testing of the effects of alterations in filler or monomer formulation on the mechanical responses of composites. Studies involving commercial products may yield results that have a greater relevance for the clinician—for example, when composite classes are compared—but are not reliable for distinguishing the effects of compositional variations. As a rule, studies of commercial and experimental materials agree that the elastic modulus (Braem *et al.*, 1986; Sabbagh *et al.*, 2002; Lohbauer *et al.*, 2006; Masouras *et al.*, 2008b;

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Gonçalves *et al.*, 2010; Leprince *et al.*, 2010; El-Safty *et al.*, 2012a) and the flexural strength (Kim *et al.*, 1994, 2002) of resin composites increase with filler content. This relationship was found to be exponential for the former (Braem *et al.*, 1986; Gonçalves *et al.*, 2010; Kim *et al.*, 1994, 2002) and linear for the latter (Kim *et al.*, 1994).

Increasing filler packing to enhance elastic modulus and strength works up to a certain filler level, beyond which there is little additional benefit (Pick *et al.*, 2011; Hahnel *et al.*, 2012) or even a slight decrease in strength (Ikejima *et al.*, 2003). In model composites, an increase in filler size has been shown to increase the elastic modulus (Masouras *et al.*, 2008a), but limited effects on mechanical properties for nanometer-sized (between 7 and 40 nm) particle-based model compositions have been observed (Karabela and Sideridou, 2011). Filler size seems to play a minor role in flexural strength of commercial composites, with nanofilled, nanohybrid, and microhybrid composites performing similarly (Beun *et al.*, 2007; Leprince *et al.*, 2010; Ferracane, 2011; Ilie *et al.*, 2013). Likewise, filler shape has shown little effect on mechanical properties in commercial composites (Beun *et al.*, 2007). However, model composites with irregularly shaped fillers were shown to have increased elastic modulus in comparison with those with spherical shape, in both unimodal and multimodal compositions (Masouras *et al.*, 2008a), likely due to the enhanced interactions of the particles during loading.

Invariably, interpretations of strength measurements in resin composites need to consider influences of test design and specimen geometry (Rodrigues *et al.*, 2008a; Pick *et al.*, 2010). Strength tests also magnify the role of surface flaws in crack initiation. In a four-point-bending configuration, for example, a mean roughness R_a of 2.1 μm has been shown to be the threshold size beyond which initial flaws created by surface treatments would be large enough to have a significant effect on strength values (Lohbauer *et al.*, 2008).

Fracture Toughness

Toughening mechanisms in composites may be attributed to the matrix, fillers, and their interfaces, but interactions between the crack front and second-phase particles predominate. The probability of a particle being hit by a crack in a favorable orientation for energy dissipation increases with an increase in filler packing. This effect is implied by the results of Wang *et al.* (2008), who showed that the decreased energy release rate is maximized when the crack encounters a particle at its center. At offset trajectories, the stress induced by the higher modulus of the particle can still “attract” the oncoming crack if they are close enough to each other; the crack is then deflected and momentarily loses energy. The reported positive correlation between fracture toughness and filler content reflects this mechanism (Ferracane *et al.*, 1987; Ferracane and Condon, 1990; Kim *et al.*, 1994).

Regardless of filler shape, the matrix/filler interface provides a considerable contribution to the overall toughness of the composite. Modeling the interfacial toughness to be equal to 2 to 3x the toughness of the matrix has shown a good correlation to the overall toughness of the composite obtained experimentally (Chan *et al.*, 2007). The fracture toughness is also affected by

residual tensile stresses built up around particles (Feng *et al.*, 2010) due to polymerization shrinkage. In this case, the interfacial stress amplifies the stress intensity as the crack tip reaches the particle, ultimately reducing the overall composite toughness (Sun *et al.*, 2006).

The effect of filler volume on toughening is limited, and in one study no improvement was shown for filler contents exceeding 55 vol% (Kim *et al.*, 2002). In a comparison of 49 commercial resin composites, the threshold for maximum fracture toughness due to filler loading was found to be 57 vol%, with a plateau until 65 vol% and a slight decrease thereafter (Ilie *et al.*, 2012). Microhybrid,Ormocer-based and packable composites were ranked equally in the same study, while only the tested nanofilled and flowable composites ranked worse than microhybrids. Although the sizes of fillers appear to be a factor in commercial composites (Watanabe *et al.*, 2008), recent studies in model composites do not support this relationship (Hosseinalipour *et al.*, 2009; Elbishari *et al.*, 2012).

FATIGUE OF RESIN COMPOSITES

Mechanical degradation of materials below a critical failure stress is commonly termed “fatigue” and involves the growth of subcritical defects at subcritical loads. Evaluation of fatigue behavior can be performed either through crack-growth rate measurements or by phenomenological approaches with lifetime data analysis. While the former is fracture-mechanics-based (see Fig. 1) and follows the development of stress intensity and crack velocity during crack extension, the latter is based on indirect evaluation of residual strength after fatigue-loading (Baran *et al.*, 2001). Experiments can either be done on specimens with initial notches or pre-cracks, or can be based on the inherent natural flaw population.

Fracture Mechanics Approach

Mechanisms of fatigue crack growth in resin composites have been shown to be governed by composition, microstructure, and susceptibility to degradation. Large particles have been shown to increase the crack resistance threshold of composites, while cracks in composites containing pre-polymerized resin fillers grow at lower stress intensity threshold (ΔK_{th}) values (Kawakami *et al.*, 2007; Takeshige *et al.*, 2007). A microhybrid (Filtek Z250, 3M ESPE, Minneapolis, MN, USA) has been shown to endure higher levels of cyclic stress before the onset of stable crack initiation when compared with a nanofilled composite with smaller particle size (Filtek Supreme Plus, 3M ESPE) (Shah *et al.*, 2009a). A change in crack behavior is readily seen by water-induced degradation. Takeshige *et al.* (2007) showed that the resistance to crack initiation in resin composites increases during the first few days in water, but ΔK_{th} decreases considerably during subsequent months. Crack resistance deterioration was more related to the degradation of the interface than to matrix hydrolysis, but the latter was linked to crack acceleration once a crack had initiated (decrease in n -value). Severe degradation of the bond between matrix and nanoparticle clusters in Filtek Supreme Plus was the cause of the erratic behavior observed over regime II in the study of Shah *et al.* (2009a). For Filtek Z250, interface degradation was less severe,

Figure 1. The subcritical crack behavior during cyclic loading can be characterized according to the parameters defined by linear elastic fracture mechanics. A plot of the applied stress intensity factor range (ΔK) against the crack growth rate (da/dN) yields the whole range of stable crack behavior (a). The subcritical crack growth interval is bordered at the onset of crack extension by the stress intensity range threshold (ΔK_{th}) and by the critical stress intensity factor (K_{IC}), at which the crack becomes unstable. Close to these 2 points, the crack advances rapidly, but in between (i.e., regime II), da/dN has a nearly linear relationship with ΔK , and crack behavior is described by the Paris Law: $da/dN = C(\Delta K)^n$, where $\Delta K = K_{max} - K_{min}$ and C , a material constant. The Paris exponent n represents the slope of the curve and determines the sensitivity to fatigue degradation. In some brittle materials, a single-value K_c (or critical energy release rate, G_c) is sufficient to describe the onset of unstable crack extension. In others, more complex microstructures act in ways to alleviate the stress concentration at the crack tip so that higher loads are required to reach the critical stress level for crack growth. Consequently, the value of K increases with crack extension for the time these mechanisms are in play. A crack resistance curve (R-curve) then forms and K stabilizes in a plateau value (K_p). This behavior is made visible by when K is plotted as a function of crack extension Δa for a monotonically loaded pre-cracked specimen. In (b) the behavior of 3 different materials is shown. Material A shows no R-curve behavior (i.e., K does not increase as the crack extends), in contrast to the other 2. Materials B and C show R-curve behavior, with material B having lower K_p in comparison with material C. Material behaviors that show considerable plasticity and/or fracture processes that envelop the crack tip beyond the length scale of the zone controlled by K are better described by the elastic plastic fracture mechanics (EPFM) approach, with the J-integral method. In this method, K parameters are replaced by J parameters describing the stress field. From the J-integral values, analogous values for K can be calculated through the applied energy release rate according to the relation: $J = G_I = K_I^2/E$, where E is the elastic modulus.

and crack behavior in regime II followed the Paris Law; the derived low n -value (5.2 m/cycle) was suggested to reflect some ductile material response on the microscale. Soappman *et al.* (2007) found a similar value for n (5.3 m/cycle) for another microhybrid composite (Vit-I-escence, Ultradent, Logan, UT, USA).

R-curve Behavior: LEFM vs. EPFM (J-integral)

R-curve behavior (see Fig. 1b) has been recently described for dental resin composites. In 2 complementary studies, as-processed, hydrated, and post-cured samples of the microhybrid Filtek Z250 and the nanofilled Filtek Supreme Plus were evaluated by Shah *et al.* (2009b,c). In the as-processed state, both composites showed similar fracture patterns, with interparticle crack propagation mainly through the resin matrix. However, the microhybrid showed a steeper fracture resistance curve and a higher plateau value for K (K_p) than did the nanofilled composite, possibly due to the slightly higher filler content and larger particles of the former, along with the higher constraint imposed on the matrix phase (Shah *et al.*, 2009b). With full hydration, a significant decrease in K_p was observed only for Filtek Supreme, whose fracture pattern evolved to include interfacial fracture and cluster debonding from the matrix (Shah

et al., 2009c). Enhancing the matrix properties through post-cure treatment did not increase the plateau values of K for either composite, but rather resulted in shorter and steeper R-curves. In all conditions, crack deflection was observed over the whole range of crack extension, while crack bridging was identified as the main mechanism for the increase in crack resistance.

The J-integral method was used by de Souza *et al.* (2011) to study the fracture of small hybrid double-cantilever beams due to the reduced linear-elastic stress field in relation to the fracture process zone. The authors measured the R-curve of a nanofilled composite (Filtek Z350, equivalent to Filtek Supreme Plus) and a nanohybrid composite (Concept Advanced, VigoDent, São Paulo, Brazil) composed of irregular fillers with a small size distribution. The observed behavior of the nanofill differed from that described by Shah *et al.* (2009b), although comparable values of K_{IC} for crack initiation were found. In contrast, de Souza *et al.* (2011) found frequent fracture of the nanofiller clusters, and little evidence of crack bridging in the nanofilled system. The crack resistance curve of the nanofill increased for small crack extensions (50-100 μm) up to 2 times the energy values for crack initiation. Crack extension during the curve-rising interval was significantly longer for the nanohybrid composite (up to 1.6 mm) toward a plateau value 3 times that of the initial value. In addition, large-scale crack bridging was observed at

the crack tip and crack wake, sometimes leading to crack blunting. Single and multiple microcrack formation strongly contributed to raising the crack resistance in the nanohybrid material.

Fatigue Degradation and Phenomenological Lifetime Prediction

Contrary to initial, static, *quasi*-static, or dynamic loading, cyclic loading simulates the clinical mastication on dental materials more precisely. The number of cycles to failure at different stress levels provides a good estimation of the fatigue resistance and serves well for material comparisons. Fig. 2 shows a typical S-N-curve for a microfill (Heliomolar) and a microhybrid composite (Filtek Z250). An obvious difference in fatigue resistance is seen at high stress levels, but this difference is narrowed as the stress amplitude decreases. For the same composites, a decrease in stress to failure of 35% to 75% has also been reported with the staircase approach (Lohbauer *et al.*, 2003b; Abe *et al.*, 2005). This highlights the degradation effects of mechanical stress relative to the amount of the resin phase in resin composites. After challenges of 2,000 cycles at loads from 20 to 100 N, a nanofilled composite (Filtek Supreme) showed better fatigue resistance in comparison with 2 microhybrid composites (Filtek Z250 and Z100) and a nanohybrid (Grandio, Voco, Cuxhaven, Germany) (Curtis *et al.*, 2009), also pointing to an important influence of the microstructure on the fatigue resistance of resin composites. In restored teeth subjected to chewing simulation, the degradation of properties in the bulk of the composite, rather than solely surface microcracking, was shown to govern fatigue performance (Pieniak and Niewczas, 2012).

Fracture toughness, in turn, has been shown to be minimally affected by cyclic loading when specimens are un-aged or aged in air alone (Ravindranath *et al.*, 2007; Lin and Drummond, 2010), while cyclic loading in water and/or solvents leads to a significant decrease in fracture toughness. The effect of mechanical fatigue is highlighted by the progressive deterioration of K_{Ic} with the number of loading cycles, because the matrix phase is especially vulnerable to repetitive strain challenges. Consequently, higher degrees of conversion (DC) in the resin matrix result in greater fatigue resistance, as has been shown for heat-processed composites (Drummond *et al.*, 2009; Lin and Drummond, 2010).

Few studies have correlated fatigue data to clinical outcomes, but Garcia-Godoy *et al.* (2012) reported that the clinical fracture rates at 6 years for a nanohybrid (Grandio, Voco) and a microhybrid (Tetric Ceram, Ivoclar-Vivadent) composite were similar, despite the fact that the nanohybrid had superior fatigue resistance *in vitro*. They did, however, observe accelerated marginal deterioration for the less fatigue-resistant material.

CORRELATION OF MECHANICAL FATIGUE DEGRADATION WITH INTRINSIC MATERIAL RESPONSE

Viscoelastic Material Response

The polymeric matrix of dental composites displays viscoelasticity, meaning that its response to stress is not totally elastic, but

Figure 2. S-N-curve for a microfiller composite (Heliomolar, Ivoclar Vivadent, Schaan, Liechtenstein) and a microhybrid composite (Filtek Z250, 3M ESPE). One hundred cyclic loads at stress amplitudes between 100% and 40% of the initial fracture strength were performed until the specimen failed. The *n*-value obtained by the regression line correlates to the susceptibility to degradation as expressed by the Paris Law. The fatigue resistance can be distinguished between low-cycle fatigue (LCF) and high-cycle fatigue (HCF). Because this type of experiment is extremely time consuming and accompanied by a high scatter in data, a simplified test procedure (staircase approach) has been established to assess the fatigue degradation at a set number of cycles, e.g., 10,000 cycles.

contains a non-linear element due to the dissipation of energy by some permanent deformation. The extent of this effect is based upon the time scale of observation, with viscoelastic materials behaving mostly linearly when stressed at a rapid rate, but showing viscoelastic deformation when stressed at a slow rate. It is well-known, and was reinforced by a recent article (Masouras *et al.*, 2008b), that dental composites display greater elastic behavior, and a higher modulus of elasticity, when filled to higher levels. This is logical, since the viscous effect occurs within the resin matrix, and matrix volume is reduced at higher filler loads. The viscoelastic deformation, or damping, within the polymer matrix may provide toughening due to energy dissipation, thus requiring a crack to be further loaded at a higher stress to continue to advance.

For a viscoelastic material, stress relaxation experiments reveal a time-dependent reduction in stress under constant strain. Creep tests, in contrast, monitor the change in strain under constant load. Both tests provide evidence for an internal re-organization of the polymeric material, which may involve polymer chain scission, oxidation or hydrolysis reactions, chain sliding, molecular re-organizations, or other events that cause the polymer to deform and relax the internal stresses. A recent study on the viscoelastic nature of dental composites followed the development of elastic modulus and the loss of damping during the initial stages of polymerization, showing the rate of these 2 events to be a function of filler content and monomer composition, as well as polymerization rate (Kim *et al.*, 2010). The viscoelastic behavior may also reflect changes in the polymer structure during aging, which may be related to the long-term stability of the restoration. A study on cyclic fatigue of resin composites suggested that viscoelasticity substantially

Figure 3. SEM micrographs showing typical macroscopic fracture patterns on fast-fractured (a) and on cyclic-fatigued (b) bending bars (nanohybrid resin composite: GrandioSO, Voco). The tensile surface is at the bottom. The higher energy involved in fast fracture led to rough surfaces and evidence of multiple crack planes (a). Typical fracture patterns for brittle failure can be distinguished as: fracture origin (1), mist and hackle region (2), and compression curl (3). The underlying, energy-dissipating mechanisms are crack deflection and crack branching. In contrast, the slower crack growth due to cyclic fatigue produced a smooth and unremarkable appearance. Less fracture energy was involved, resulting in the lack of brittle surface patterns. Even the obvious fracture origin (subsurface pore, arrow) did not produce clear markings on the fracture plane, though the compression curl is evident at the top of the bar.

influenced mechanical degradation due to the repetitive slow loading of the materials (Lohbauer *et al.*, 2003b). Recent studies have shown good correlations between static and dynamic creep of composites (Kaleem *et al.*, 2012a), and between bulk creep and that determined by nanoindentation (El-Safty *et al.*, 2012b). Studies have shown creep to be greater in composites with higher resin volume, especially for those with non bis-GMA resins (El-Safty *et al.*, 2012c). The latter is explained by the rigidity of the bis-GMA molecule and the greater resistance that imparts to the polymer network as compared with a more flexible base monomer system. Filler shape and size may also influence creep, with composites made with spherical fillers showing less resistance to creep than those with a similar size but irregular shape, due to the reduced activation energy needed for inter-particle sliding (Kaleem *et al.*, 2012b). Comparably, hybrid composites with a broader filler distribution have shown an improved resistance to slow crack growth (Ornaghi *et al.*, 2012).

Hydrolysis and Long-term Relaxation

Dental composites absorb water predominantly *via* the polymer matrix and along the interface between the fillers and matrix, with the rate being accelerated under cyclic fatigue conditions. The applied tensile stress opens cracks and expands the material volume, enabling water to enter the affected region. The repetitive pumping effect contributes to further hydrolysis (Truong *et al.*, 1990). The water also may serve to relax internal stresses within the polymer, most likely by serving as a plasticizer that separates the polymer chains and reduces the rigidity of the network (Ferracane, 2006). During prolonged exposure, water and other solvents may produce chemical degradation of the

polymer through attack on ester linkages, breaking bonds in the polymer chain and reducing rigidity (Ferracane, 2006).

Water sorption is generally increased with resin content and monomer hydrophilicity (Sideridou *et al.*, 2011). The effect of diffusion-controlled water sorption may be the first to cause a softening of the matrix at the surface, with time needed for water to reach the interior (de Moraes *et al.*, 2008). A study on the effect of water sorption on fracture strength of a microhybrid composite showed a reduction in strength of 30% within the first 3 months (Lohbauer *et al.*, 2003a). In contrast, the authors did not observe a change in cyclic fatigue resistance within this period, indicating that the degradation due to water sorption was limited, or that the specimen was not totally saturated. In other, longer term, experiments in which full saturation of the entire polymer network with water or solvents occurred, the softening of the network, in conjunction with other long-term degradative effects, was shown to cause a reduction in the failure resistance of dental composites (Ravindranath *et al.*, 2007), as evidenced by the polymer network becoming less rigid and more yielding to external loads (Pastila *et al.*, 2007). While some plasticization of the network may help to provide an enhanced plastic zone ahead of crack tips for energy dissipation, the reduced properties of the compromised network facilitate the propagation of cracks at lower energies, negating any beneficial effects.

Perhaps of greater significance than the effects of water and other chemicals in the oral environment on the polymer matrix are the effects of salivary and microbial enzymes. Enzymes, such as esterases, are capable of attacking the methacrylate molecules and cleaving the ester linkages, thus reducing the structural integrity of the cross-linked polymer network (Finer and Santerre, 2004). This effect is enhanced for certain monomers, but has also been shown to be enhanced in composites with less filler concentration (Finer and Santerre, 2007). Recent studies have shown chemical alteration of the surfaces of composites exposed to conditioned media containing enzymatic activity (Gregson *et al.*, 2009), which may, at a certain level, contribute to surface degradation and flaw initiation in the long term.

Matrix/Filler Interface Degradation

Recent studies of composites with equal amounts of nano-sized fillers have shown that as filler size decreases, the extent of water and ethanol sorption is significantly increased, likely reflecting the increased surface area resulting from the smaller fillers (Karabela and Sideridou, 2011). While these filler surfaces are treated with a silane coupling agent to make them hydrophobic, the coating is not uniform or perfect, and the greater hydrophilic surface area results in enhanced pathways for water and solvent uptake. The mechanical properties, $\tan \delta$, T_g , and storage modulus, were most optimum when a higher proportion of silane was coated onto the fillers (Sideridou and Karabela, 2009). Other studies have shown a significant amount of surface degradation of flowable resin composite when exposed to acidic and alcoholic solutions *in vitro* over 14 days (Han *et al.*, 2008). The authors concluded that surface degradation was greatest for the composites with the least amount of filler due to an incomplete DC of the polymer matrix and non-optimized matrix/filler coupling.

Fractographic Observations

Clinical and laboratory-created fracture surface analyses provide indirect evidence for the degradation mechanisms occurring in dental composites over time. The features found in fractured composite surfaces are generally similar to those found on ceramics (Fig. 3). Fractures created by fast fracture (Fig. 3a) show a rougher and highly branched composite surface compared with those produced in fatigue failure (Fig. 3b), due to the greater stored elastic energy present at the higher strength levels achieved in fast fracture tests (Quinn and Quinn, 2010). The roughness of the fracture surface (and thus the involved energy) can be seen in Fig. 4. Typically, fracture surfaces of dental composites created from *in vitro* tests of fracture toughness show crack propagation predominantly through the matrix resin phase, with occasional excursions along the matrix/filler interface (Ferracane *et al.*, 1987). However, at higher magnifications, Figs. 5b and 5d show a residual resin layer on the particle surface, implying that interfacial failure occurs only rarely, unless advanced degradation processes are operational. For microfill composites, which contain large pre-polymerized resin particles that are well cured, there is poorer adhesion between the newly formed resin matrix and the particles, and the crack often propagates along this interface (Ferracane and Condon, 1992). This same crack propagation mode is evident in nanohybrid composites that also contain pre-polymerized resin fillers for the purpose of reducing polymerization shrinkage, though fractures of the pre-polymerized particles can be seen as well (Takahashi *et al.*, 2011). Nanofill composites, which are composed entirely of individual particles smaller than 100 nm, typically show fracture through the resin matrix, though the small size of these particles, and the presence of clusters of nanoparticles, makes it difficult to determine if matrix/filler interfacial failure has occurred (Rodrigues *et al.*, 2008b) (Figs. 5a and 5c).

The fractography of dental composites shows that the toughening mechanisms active in dental composites include crack deflection or branching (Fig. 4a), crack pinning between fillers, and crack bridging (Shah *et al.*, 2009a,b). Thus, composites with higher concentrations of fillers show enhanced toughness, though

Figure 4. SEM micrographs showing fracture surface features of a nanohybrid resin composite (GrandioSO, Voco) after fast fracture at a constant stress rate of 50 MPa/s (a), slow fracture at a constant stress rate of 0.005 MPa/s (b), and after cyclic fatigue of 10,000 cycles at 0.5 Hz (c). While the single-loaded fracture surfaces (a, b) show what appears to be exposed particles, the repetitive loading (c) did produce a clearer interparticle fracture path. Further analysis (see Fig. 5) showed that the exposed particle surfaces actually remained covered with a residual resin matrix layer, providing evidence that, in all cases, the crack path was predominantly through the resin matrix. The fast fracture (a) further showed indication for crack branching, involving multiple crack planes (arrows) in the failure event. This feature is missing on the slow-fractured (b) as well as the fatigued surface (c).

Figure 5. Typical fracture morphologies of a nanofilled composite (Filtek Supreme, 3M ESPE) (a,c) and a nanohybrid composite (Miris II, Coltene) (b,d) under different magnifications in the SEM. (a) The smaller fillers in comparison with those in (b). Under high magnification, both materials present cohesive failure in the resin matrix (c,d), which cannot be concluded from (a) and (b). A residual resin layer remains on the particle surfaces (c,d), implying that the crack front did not touch the matrix/filler interface. Since hoop stresses typically develop around bonded filler particles during photopolymerization of a resin composite, a progressing crack generally deflects away from the filler/matrix interface by the energy associated with the residual stress layer around the particles.

certain mechanisms, such as crack pinning, are reduced or eliminated when the particle size becomes too small and when there is insufficient space between particles for energy-dissipating crack-bowing events to be active. The critical crack length in resin composites has been reported to be between 35 and 100 μm , due to chemical inhomogeneities in the resin matrix rather than to particle agglomeration (Quinn and Quinn, 2010).

CONCLUDING REMARKS

The initial mechanical state of resin composites is unstable under conditions of mechanical stress and environmental corrosion. The extent and nature of these changes are unclear, but may not be appropriately characterized by measures of their initial properties. Current evidence supports the leading role of the organic phase and the matrix/filler interface in the degradation of strength and toughness. Important advances in the mechanical stability of resin composites may be achieved by directing the focus of research toward the development of resin polymer matrices less susceptible to chemical and stress-related degradation. This review points to the importance of considering the behavior of resin composites under fatigue, and potentially involving experiments with clinically relevant geometries and loading conditions, as an important factor to be considered during materials selection.

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