JOURNAL OF MATERIALS SCIENCE 22 (1987) 3508–3516

An investigation into the crack propagation behaviour of tooth-coloured particulate-filled dental restorative materials

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Particulate-filled resins, or dental composites, are being increasingly used to restore the load-bearing surfaces of teeth. If these restorations are not to fracture in service, and if improvements are to be made, an understanding of the fracture behaviour of these materials is essential. The fracture parameter used is the stress intensity factor at crack instability (K_{IC}). This has been calculated using the double torsion test in an Instron universal testing machine. The fracture behaviour was studied by varying the amount of filler (7, 15, 26 and 41 vol %); the surface treatment of the filler (coated or uncoated); the environment (air or water); and the crosshead rate (0.05, 0.5, 5 and 50 mm min^{-1}). Fracture was found to occur in either a continuous or stick–slip manner. The stick–slip behaviour was due to blunting of the crack tip, which was controlled by the yield behaviour. If there was no significant blunting, continuous crack growth occurred. A unique fracture criterion was shown to apply, which was that a critical stress of approximately three and a half times the yield stress must be obtained at a critical distance ahead of the crack tip.

1. Introduction

Particulate fillers incorporated into a polymer are used in dentistry to fill cavities in teeth. These restorative materials are capable of giving a good colour match with natural teeth and so, initially, their use was confined to anterior teeth. Amalgam, which has higher tensile and compressive strengths compared to composite restorative materials, is generally used to fill cavities in posterior teeth where appearance is not so important. However, composites are now being increasingly used to restore posterior teeth. If improvements are to be made in these composite restorative materials, and if they are not to fracture in service, a basic understanding of the fracture behaviour of this type of material is required.

A fracture mechanics approach has been used previously on various proprietary dental composites by Lloyd [1], Lloyd and Iannetta [2], and Lloyd and Mitchell [3]. Other, more limited, work has been done by Roberts *et al.* [4] and Koblitz *et al.* [5]. However, there has not been any systematic approach to the effect of filler loading, filler surface treatment, or environment on the fracture behaviour of dental composites. In the wider materials field, a number of authors have investigated the effects of rigid particulate fillers on fracture behaviour [6–12].

The search for a suitable tooth-coloured, direct-filling material is one which has concerned the dental profession for very many years. The first tooth-coloured filling material to be widely used was silicate cement, having been introduced to England in 1871 [13]. Initially, these materials give a very good colour match with the tooth, and the presence of leachable fluoride increases the resistance of the tooth to further decay. However, such restorations tend to discolour after a short time and gradually disintegrate in the oral fluids. Silicate is brittle and tends to fracture upon impact, which makes it unsuitable for use in large restorations, particularly if they are load-bearing. Also, the early acidity of the restoration damages the tooth pulp and many even cause its death.

Room-temperature polymerized methylmethacrylates were introduced to the dental profession in an attempt to overcome the considerable disadvantages of silicate cement. Whilst these polymers give a good colour match and are virtually insoluble in oral fluids, certain inherent characteristics limit their effectiveness as a restorative material. These are: high setting shrinkage; high coefficient of thermal expansion; low strength; and low abrasive resistance. In addition, the exothermic setting reaction and irritant monomer lead to pulpal inflammation.

It was against this background that filled, room-temperature polymerized polymers, or dental composites, were developed for use in dentistry. Early composites used room-temperature polymerized methylmethacrylates as the matrix, but these still proved unsatisfactory. Dental composites are now mostly based on bis-GMA polymer. This thermosetting resin was developed by Bowen [14] and is primarily

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an addition-reaction product of bis-phenol A and glycidyl-methacrylate. Bis-GMA has the advantage, in comparison with methacrylate polymers, of shrinking less during polymerization; producing less heat during polymerization; being harder; and also less toxic to the pulp.

Its principal drawback is its high viscosity, and so lower molecular weight diluents, such as ethylene glycol dimethacrylate, are added. In addition, bis-GMA is not completely colour-stable, and because it is a mixture of high molecular weight optical isomers it cannot be purified by distillation or by crystallization, which results in poor storage stability.

The filler is glass or quartz and has a particle size with a mean range of 10 to 20 μm. The filler is coated with a vinyl silane coupling agent to achieve bonding between the filler and polymer. The inorganic phase forms about 50 vol % of the composite. Setting is usually acheived by using a peroxide–amine free radical system.

Recent developments have seen the introduction of a number of modifications. The bis-GMA polymer has been replaced in some composites by urethane dimethacrylate because of its better colour stability. Composite restorative materials present difficulties in achieving a smooth surface to the restoration. In order to overcome this problem so-called "microfine composites" have been produced in which the filler particle size is reduced to 0.05 μm. Although this results in a smoother surface, microfine composites show greater water uptake, greater thermal expansion, lower compressive strength, and greater creep when compared to the conventional systems. Most recently, composites have been produced in which the filler consists of both the large and the small sizes in an attempt to produce the best of both the conventional and the microfine systems.

A further modification concerns the method of cure. The peroxide–amine system gives a limited time in which to pack the material into the cavity and so "command setting" systems have been introduced. In these the composite does not set until either a blue light or an ultraviolet light is shone on to the restoration. This gives the dental surgeon time to pack and shape the restoration correctly.

2. Experimental details

2.1. Material and specimen preparation

The resin used in this study was prepared from bisphenol A and a compound of glycidyl methacrylate and glycidyl acrylate and is a clear, viscous, light-coloured resin (Nupol resin, Freeman Chemicals Ltd., Liverpool, UK). Methylmethacrylate and ethylene glycol dimethacrylate monomers were used to dilute the resin and facilitate mixing. The mix was cured using lauroyl peroxide and NN-dimethyl-*p*-toluidine. The components were combined in various proportions in order to determine the most satisfactory handling and working properties. The best result was obtained by combining 10 g of resin with 1 g of ethylene glycol dimethacrylate, 1 g of methylmethacrylate, 0.03 g of lauroyl peroxide and 0.01 g of NN-dimethyl-*p*-toluidine.

In the first stage of preparation the resin, ethylene glycol dimethacrylate, and NN-dimethyl-*p*-toluidine were mixed together and stored at 55° C to allow any entrapped air to escape. This portion was then allowed to cool to 40° C. The methylmethacrylate was combined with the lauroyl peroxide and then added to the first portion. The set resin is referred to as "the dental resin" throughout the rest of the work.

The filler was a barium-containing glass [15]. The filler was provided with and without a silane coating which was γ-methacryloxypropyl-trimethoxysilane. The particles were irregular in shape, having an average diameter of 5 μm, with a range of 1 to 20 μm. The filler was added to the resin along with the ethylene glycol dimethacrylate and the NN-dimethyl-*p*-toluidine. The filler was incorporated in amounts of 15, 28, 45 and 62 wt % (7, 15, 26 and 42 vol %). A standard ashing test was carried out on samples of some specimens to check the consistency of the filler content, and showed that the variation in filler content was within 10% for each filler fraction.

All the moulds were constructed from hard brass and could be taken apart, so that no force was required to remove the set material. All the moulds were treated with PTFE spray to facilitate removal of the set materials. The moulds were preheated to 40° C. Once the moulds had been filled a polyvinylchloride sealing strip was placed over the top mould and a metal plate clamped down on to this. A PVC sealing strip was also used between the bottom of the mould and the base plate.

The materials were cured for 24 h at 40° C. To prevent sedimentation of the filler particles during the initial curing, the moulds were rotated through 180° every 30 sec for the first 5 min. At the end of this time polymerization had started and the moulds were then rotated every 5 min for the next 30 min, after which rotation was considered unnecessary. The surfaces of all specimens were smoothed, prior to testing, using Grade 400 waterproof silicon carbide paper as a rotating disc under a water spray. All specimens were stored in either air or distilled water for one week prior to testing.

2.2. Stress intensity factor

The double torsion test was used to measure the stress intensity factor [16]. The specimens used for this were 75 mm × 30 mm × 4 mm. There was a shallow groove 0.5 m deep cut along the centre of the lower surface and an edge notch at one end. The notch was cut initially using a fretsaw, and a crack was then produced at the depth of the cut using a stainless steel single-edge razor blade.

The double torsion specimens were tested in an Instron universal testing machine, Model 1195 (manufactured by Instron Ltd, High Wycombe, England) under compressive loading using a constant crosshead speed. The stress intensity factor was calculated using the following equation [11]:

$$K_{\mathrm{IC}} = PW_{\mathrm{m}}\left(\frac{1+\nu}{Wt^3t_{\mathrm{n}}k}\right)^{1/2} \qquad (1)$$

where K_{IC} = stress intensity factor at crack instability,

W_m = moment arm, ν = Poisson's ratio, W = plate width, t = plate thickness, t_n = plate thickness in the plane of the crack, k = geometric factor which depends on the ratio $(W/2)/t$ and has a value of 2.78 for the testpiece used in this work, and P = applied load.

2.3. Yield stress

Cylindrical specimens, 12 mm long × 6 mm diameter, were used to calculate the yield stress. They were deformed in uniaxial compression between lubricated plates using an Instron universal testing machine. The yield stress was calculated from the applied load P, the initial cross-sectional area A, and the strain e:

$$\sigma_y = \frac{P}{A}(1 - e) \qquad (2)$$

Under some test conditions there was no clearly defined yield point, and in these circumstances a 0.1% offset proof stress was calculated.

2.4. Young's modulus

This was determined using a three-point bend test performed in an Instron universal testing machine. The tests were carried out on one half of the double torsion specimens. Young's modulus was calculated from the initial slope of the load–displacement curve and the specimen dimensions. The displacement was determined from the movement of the crosshead, taking into account the machine softness. The Young's modulus was calculated using the following equation:

$$\frac{P}{y} = \frac{4Ebh^3}{L^3} \qquad (3)$$

where E = Young's modulus, b = breadth of specimen, h = height of specimen, L = span of specimen, P = applied load and y = displacement.

2.5. Test conditions

The stress intensity factor, the yield stress and Young's modulus were determined for every possible combination of the test conditions listed below:

Environment	Air or water at 23 ± 1° C
Filler surface treatment	Coated or uncoated
Crosshead rate	0.05, 0.5, 5, 50 mm min^{-1}
Filler loading	7, 15, 26, 41 vol %

3. Results and discussion

3.1. Double torsion test

Under most conditions the crack propagated in a stick–slip mode with the crack progressing through the test piece in a series of jumps. This type of behaviour occurred for crosshead rates of 0.05, 0.5 and 5 mm min^{-1}. Thus two mean values of the stress intensity could be obtained – one for crack initiation, K_{ICi} and one for crack arrest, K_{ICa}. As the crosshead rate increased so the value for crack initiation reduced, until at 50 mm min^{-1} the crack propagated in a continuous manner (see Fig. 1).

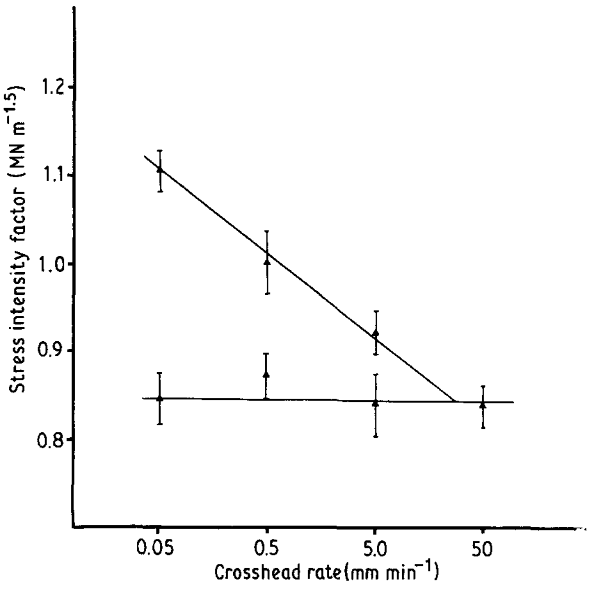

Figure 1 Double torsion test. Effect of crosshead rate on the stress intensity factor. Dental resin + 26% coated filler tested in air at 23 ± 1° C.

The crack arrest values were independent of crosshead rate and were similar to the values of the stress intensity factor for continuous propagation. Similar behaviour was found irrespective of filler loading, whether coated or uncoated fillers were used, and whether the test was carried out in air or water. This effect of crosshead rate upon the mode of crack propagation has been reported in particulate-filled epoxy resin by Spanoudakis and Young [6] and by Moloney *et al.* [8]. Similar behaviour has also been reported in unfilled epoxy resins by Yamini and Young [17] and by Kinlock and Williams [18].

The stress intensity factor increased as the filler content increased (see Fig. 2). This is similar to behaviour reported by Spanoudakis and Young [6] and by Moloney *et al.* [8]. However, they also reported that at higher filler concentrations the mode of crack propagation changed from a stick–slip to a continuous mode. In the present study the extent of crack jumping, i.e. the difference between the crack initiation and crack arrest values, increased until a filler content of 26% was reached, after which the difference between these two values started to decrease. It is possible that further increases in the filler content would produce continuous crack propagation. Unfortunately it was not possible to increase the filler content beyond 41 vol % without incorporating an unacceptable amount of air.

The absence of coating on the filler produced an increase in the stress intensity factor for crack initiation, whilst the arrest values remained largely unaffected (see Fig. 2). The effect of filler–matrix adhesion on crack behaviour is not entirely clear. Brown [12] and Broutman and Sahu [10] reported a decrease in fracture energy with improved adhesion. Moloney *et al.* [8] reported a decrease in the stress intensity factor for crack initiation with improved adhesion. Spanoudakis and Young [7], on the other hand, showed an increase in the stress intensity factor with improved adhesion. It is, however, clear that any change in the stress intensity factor produced by altering the filler–

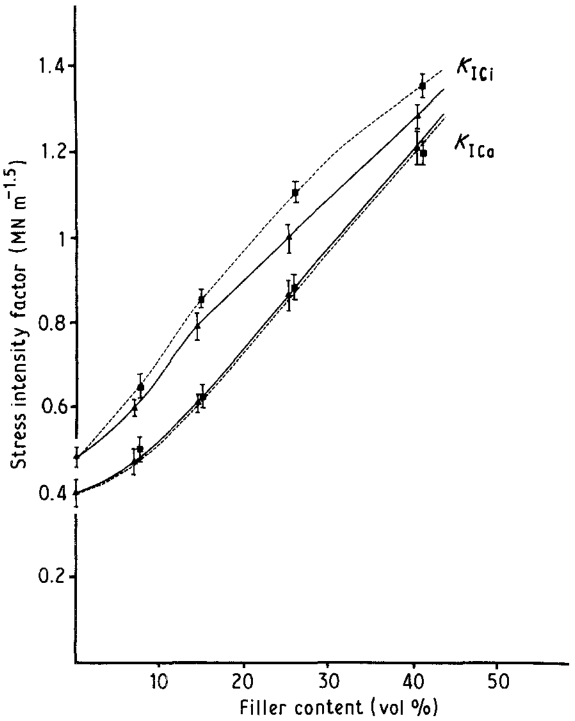

Figure 2 Double torsion test. Effect of filler content on the stress intensity factor. Crosshead rate 0.5 mm min^{-1}, tested in air at 23 ± 1° C: (▲) coated filler, (■) uncoated filler.

matrix interface is small in comparison to the large increase produced by the presence of the filler.

The nature of the filler–matrix interface in affecting the stress intensity factor is quite complicated. For example, the effects of the filler will depend upon the nature of the matrix. Manson and Sperling [19] indicate that for a brittle matrix a rigid filler will increase the fracture toughness, especially if the filler–matrix adhesion is poor. On the other hand, in an otherwise ductile matrix the filler will decrease the fracture toughness as it will inhibit plastic deformation of the matrix, and this will be more effective if there is filler–matrix adhesion.

A distinction must also be made between fracture energy and the stress intensity factor. They are related by the equation

$$2\gamma = G_{IC} = \frac{K_{IC}^2}{E} \quad (4)$$

where γ is the fracture energy, G_{Ic} is the strain energy release rate, and E is Young's modulus. Many workers, including Lange and Radford [20], Broutman and Sahu [10] and Young and Beaumont [11] have shown that the fracture energy reaches a maximum at about 30 vol % filler content. Further increases in filler content cause the fracture energy to fall. On the other hand, the stress intensity factor continues to increase as the filler content increases. This difference is because the modulus increases as the filler fraction increases, but at a faster rate than the stress intensity factor. So, although the fracture energy reaches a maximum, the stress intensity factor continues to increase. This shows that great care must always be taken when comparing different sets of results.

The presence of water increased both the values of K_{ICi} and K_{ICa} where K_{ICi} refers to the stress intensity factor at crack initiation and K_{ICa} to the stress intensity factor at crack arrest. The main effect is on K_{ICi}, with K_{ICa} showing only a small change. Beaumont and Young [21] demonstrated an increase in the stress intensity factor when they tested their silica-particle filled epoxy resin composite in distilled water. The water can be expected to influence the interfacial bonding between the matrix and the filler, as well as plasticizing the matrix.

3.2. Young's modulus

As would be expected, as the filler content increased so did the values for Young's modulus. The principal effect of the filler arises from the partial occupancy of volume by rigid and immobile masses, and this effect is independent of the nature of the interface between the filler and the matrix.

The presence of a coupling produced an increase in the value of Young's modulus, but the increase was small when compared to the overall increase produced by the presence of the filler. The presence of water reduced the value for Young's modulus.

3.3. Yield stress

The yield stress increased as the filler content and the crosshead rate increased. The presence of water reduced the yield stress, as did the absence of a coating on the surface of the filler. As the filler content increased it became progressively more difficult to define a yield point. The stress–strain curves for the dental resin, dental resin + 7% filler, and dental resin + 15% filler all showed a drop in stress after yield. The curves for filler contents of 26 and 41% did not show such a drop. Even for those three groups which did show a drop in stress after yield, the yield point was still difficult to identify at the crosshead rate of 0.05 mm min^{-1}. Under these conditions a 0.1% offset proof stress was used.

The relationship between yield stress and the stress intensity factor for crack initiation is illustrated in Fig. 3. It can be seen that for each composite, i.e. for each combination of dental resin and filler, there exists a master curve indicating a unique relationship between the two properties. As the filler loading increased this curve was displaced to higher values, and for each filler loading as the yield stress fell the stress intensity factor for crack initiation increased.

The values for the yield stress were obtained in uniaxial compression as very little yield occurred in tension. Ideally, the same strain rates should be used for calculating the stress intensity factor and the yield stress. In this work the same crosshead rates have been used. Obviously this results in different strain rates, but it is envisaged that the error caused by this approximation is small.

3.4. Critical distance

The stress intensity factor for crack initiation is clearly related to the yield behaviour. The yield stress represents a diffuse yielding which surrounds the crack tip and can be expected to blunt it. It would therefore be useful to consider the stress distribution around the

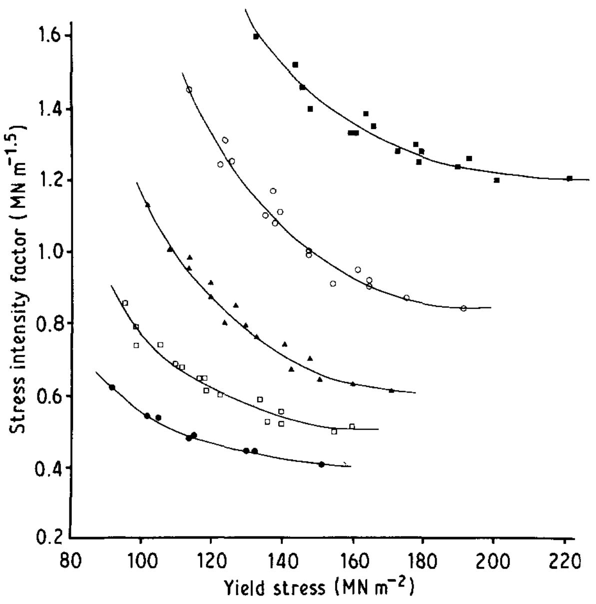

Figure 3 Relationship between the stress intensity factor for crack initiation and yield stress. Coated and uncoated filler tested in air and distilled water at 23 ± 1° C and at four crosshead rates. (●) Dental resin alone, (□) 7% filler content, (▲) 15% filler content, (○) 26% filler content, (■) 41% filler content.

blunt tip. Williams [22] has shown that for a crack with tip radius ϱ and length a, then the stress σ_{yy} normal to the axis of the crack at a small distance r ahead of the tip is given by the equation

$$\sigma_{yy} = \frac{\sigma_0 a^{1/2}}{(2r)^{1/2}} \frac{[1 + (\varrho/r)]}{[1 + (\varrho/2r)]^{1.5}} \tag{5}$$

where σ_0 is the applied stress.

If fracture occurs when a critical stress σ_c, is attained at a distance c ahead of the crack tip, then $\sigma_{yy} = \sigma_c$ and $r = c$, and Equation 5 can be rewritten as

$$\frac{\sigma_0(\pi a)^{1/2}}{\sigma_c(2\pi c)^{1/2}} = \frac{[1 + (\varrho/2c)]^{1.5}}{1 + (\varrho/c)} \tag{6}$$

The term $\sigma_c(2\pi c)^{1/2}$ can be considered as the critical stress intensity factor K_{Ic} for a sharp crack, and $\sigma_0(\pi a)^{1/2}$ as the stress intensity factor K_{Ib} for a blunt crack. Thus Equation 6 becomes

$$\frac{K_{\text{Ib}}}{K_{\text{Ic}}} = \frac{[1 + (\varrho/2c)]^{1.5}}{1 + (\varrho/c)} \tag{7}$$

K_{Ib} represents the value of the stress intensity factor for crack initiation when the crack propagates in a stick–slip manner (K_{ICi}). K_{Ic} represents the value of K when the crack propagates in a continuous manner, and is similar to the crack arrest values (K_{ICa}). For stable, continuous crack growth K_{Ib} and K_{Ic} have the same value, so $K_{\text{Ib}}/K_{\text{Ic}} = 1$.

It is very difficult to measure ϱ directly. Kinloch and Williams [18] produced a series of epoxy resin samples containing blunt cracks which had been formed by drilling holes of known diameter at the crack tips. They then obtained values of K_{Ib} (stress intensity factor for crack initiation) for the blunted cracks. They showed that the relationship given by Equation 7 could be used for these materials.

Kinloch and Williams [18] were also able to demonstrate that the crack tip radius ϱ is proportional to the crack opening displacement δ. Thus

$$\varrho = k\delta \tag{8}$$

where k is the proportionality constant. In the work described above, Kinloch and Williams took the value of k to be unity. This approximation was successful, with both experimental and theoretical results being in excellent agreement. Thus Equation 7 can be rewritten

$$\frac{K_{\text{Ib}}}{K_{\text{Ic}}} = \frac{K_{\text{ICi}}}{K_{\text{ICa}}} = \frac{[1 + (\delta/2c)]^{1.5}}{1 + (\delta/c)} \tag{9}$$

Values for the crack opening displacement can be calculated using the standard relationship for a Dugdale plastic zone:

$$\delta = \frac{K_{\text{IC}}^2}{\sigma_y E} \tag{10}$$

Fig. 4 shows the values of $K_{\text{ICi}}/K_{\text{ICa}}$ measured as a

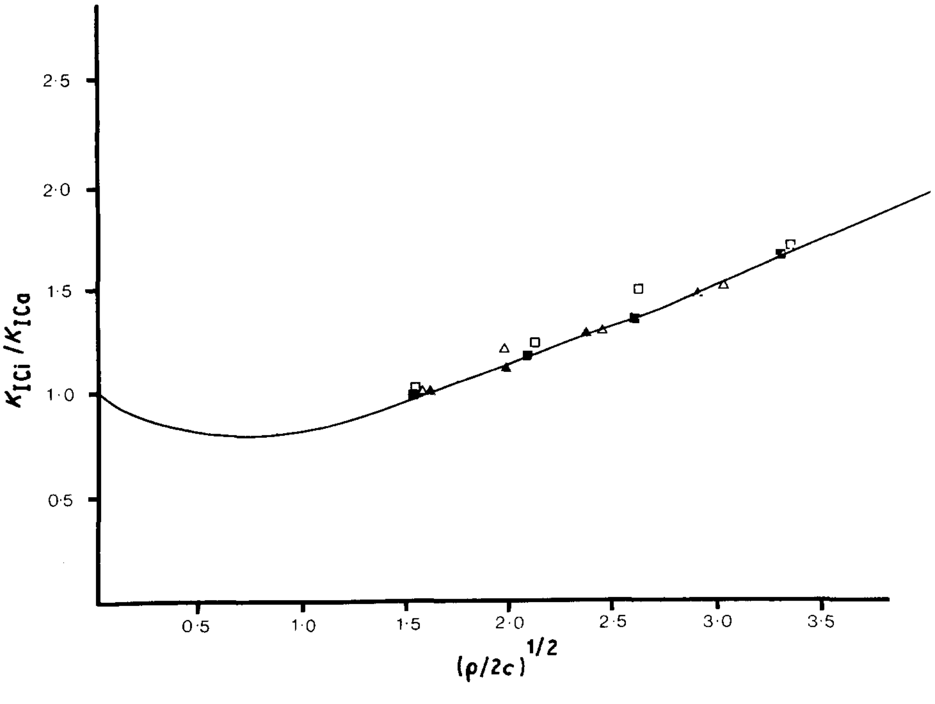

Figure 4 Relationship between $K_{\text{ICi}}/K_{\text{ICa}}$ and $(\varrho/2c)^{0.5}$ for 7% filler content: (▲) coated filler in air, (△) uncoated filler in air, (■) coated filler in water, (□) uncoated filler in water.

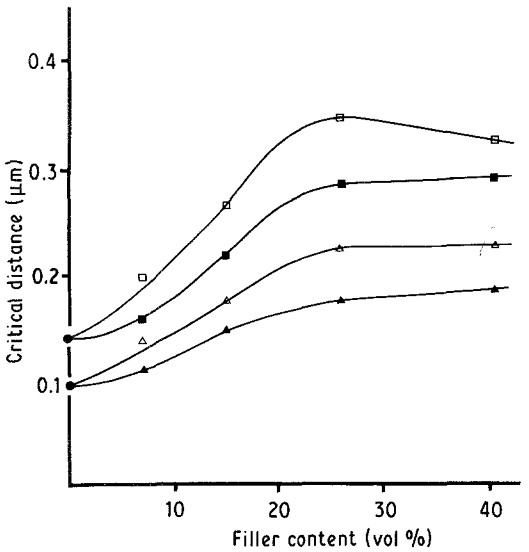

Figure 5 Relationship between critical distance and filler content. The critical distances have been obtained assuming $k = 1$ for 0, 7, 15, 26 and 41% filler content. (▲) Coated filler in air, (△) uncoated filler in air, (■) coated filler in water, (□) uncoated filler in water.

function of $(\varrho/2c)^{1/2}$ for the composite with 7% filler content. The curved line is the relationship between $K_{\mathrm{ICi}}/K_{\mathrm{ICa}}$ and $(\varrho/2c)^{1/2}$ predicted using Equations 7 and 9. The data have been fitted to the theoretical curve by selecting appropriate values of c. Similar curves can be obtained for the dental resin and all its composites. For each composite the critical distance was smallest when a coated filler was used and the test pieces were fractured in air. The critical distance increased when uncoated filler was used and was greatest when the uncoated filler was tested in water. The conditions which produced the greatest critical distances were those conditions which produced the lowest yield stresses. The effect of filler content on the critical distance is shown in Fig. 5. The critical distance increases as the filler content increases until a filler volume content of approximately 26% is reached. Further increases in the filler content fail to produce any marked increase in the critical distance, and it even starts to fall for uncoated filler tested in water. In calculating the critical distances the assumption has been made that the crack tip radius is equal to the crack opening displacement. However, there is no direct evidence that this is true for particulate-filled resins. It is likely that as the amount of filler increases, so the discontinuities at the crack tip also increase, thus changing the value of k in Equation 8. This is discussed in greater detail in Section 3.5.

3.5. Critical stress

Once the value of the critical distance has been determined it is possible to calculate the value of the critical stress using the equation $K_{\mathrm{IC}} = \sigma_{\mathrm{c}}(2\pi c)^{1/2}$. The critical stress is greatest when the test is carried out in air with a coated filler, and least for those tests performed in water using uncoated filler. If the critical stress values are compared to those obtained for the critical distances, the larger the critical distance the smaller the critical stress. The largest values for the stress intensity factor were obtained in water using uncoated filler. These conditions produce the smallest values for the critical stress, but the largest values for the critical distance.

It has been suggested that the relationship between the critical stress and the yield stress is important in deciding whether crack propagation occurs in a stable or unstable manner [22]. Yamini and Young [23] suggest that a stress in the order of three times the yield stress must be reached in the plastic zone at a critical distance ahead of the crack tip for failure to occur in a stable manner. If the critical stress is greater than three times the yield stress then the crack behaves in an unstable manner.

A plot of the ratio of $K_{\mathrm{ICi}}/K_{\mathrm{ICa}}$ against σ_c/σ_y is shown in Fig. 6 for the dental resin, and for dental resin with 7 and 15% filler content. The values of σ_c/σ_y for stable propagation (i.e. $K_{\mathrm{ICi}}/K_{\mathrm{ICa}} = 1$) show a remarkable

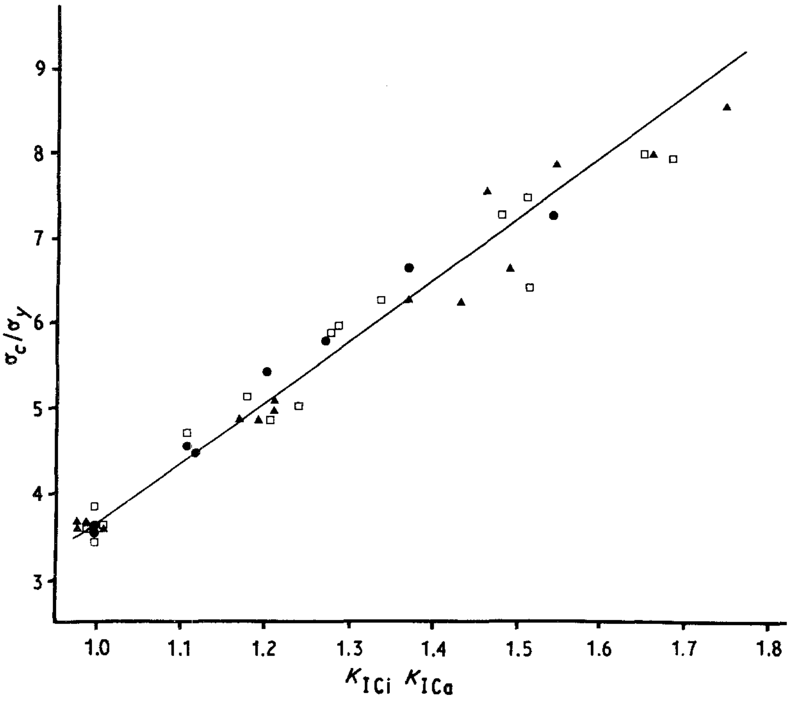

Figure 6 Relationship between σ_c/σ_y and $K_{\mathrm{ICi}}/K_{\mathrm{ICa}}$: (●) dental resin, (□) 7% filler content, (▲) 15% filler content. The results are for coated and uncoated filler tested in air and water at four crosshead rates.

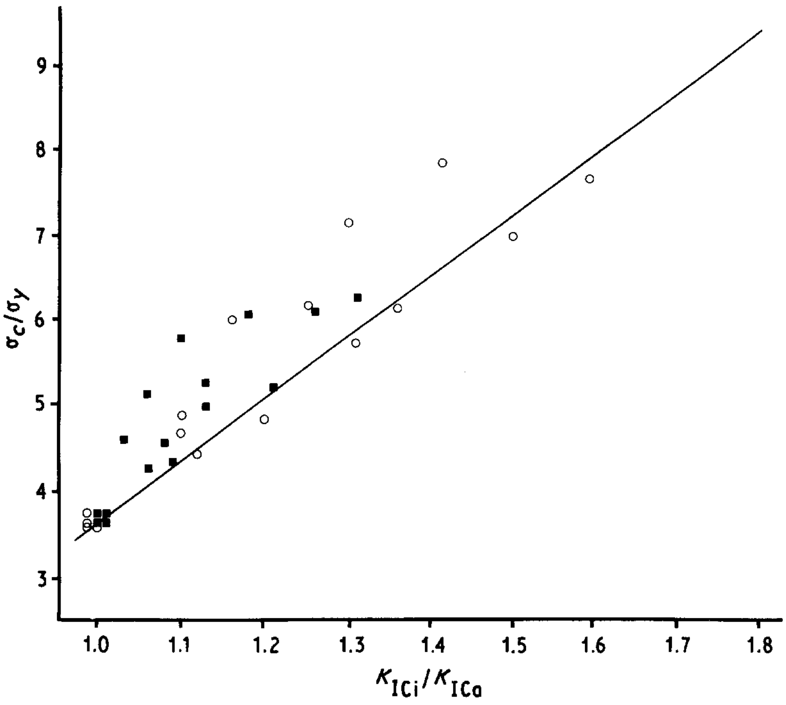

Figure 7 Relationship between σ_c/σ_y and $K_{\rm ICi}/K_{\rm ICa}$: (○) 26% filler content, (■) 41% filler content. The results are for coated and uncoated filler tested in air and water at four crosshead rates.

consistency and give a value of 3.67 ± 0.1 for σ_c/σ_y. This is higher than the figure suggested by Yamini and Young [23], but it still serves to indicate that the relationship between the critical stress and the yield stress is important in controlling the mode of fracture. This relationship affects not only the mode of failure but also the value of the stress intensity factor.

Fig. 6 only gives the points for the dental resin, dental resin + 7% filler, and dental resin + 15% filler. The points for the composites containing 26 and 41% filler content have not been included as they lie well away from the general trend. The values of the critical stress and critical distance have been calculated on the assumption that the crack tip radius is equal to the crack opening displacement. It seems most unlikely that the profile of the crack tip remains unchanged as the filler concentration increases. As the crack tip profile changes, its effective radius will no longer equal the crack opening displacement. Thus, the selection of a suitable value of k in the equation $\varrho = k\delta$ is very important.

The ratio of the critical stress to the yield stress is important in deciding whether stable or unstable crack growth occurs. The evidence suggests, for the composite used in this work, that when this ratio approximates to 3.67 stable crack growth occurs. It is therefore possible to select values of k for the composites containing 26 and 41% filler contents which, in turn, give values for σ_c/σ_y of approximately 3.67 for stable crack growth, i.e. for crack growth at a crosshead rate of 50 mm min^{-1}. The value of k selected for 26% filler content is 1.2 and for 41% it is 1.8.

The effect that this has on the relationship between σ_c/σ_y and $K_{\rm ICi}/K_{\rm ICa}$ is shown in Fig. 7. The values for the composite with 26% filler content fit the master line reasonably well. The results for the composite with 41% filler content still show some scatter, but agreement is much better than the results obtained assuming $k = 1$ for all composites. The effect of altering the value of k on the critical distance is shown in Fig. 8, where it can be seen that the plot of the critical distance against filler content is now reasonably linear, suggesting that as the filler concentration increases so does the critical distance. This is an attractive explanation, but it must be stressed that there is no direct experimental evidence to suggest that the values of k should be 1.2 and 1.8. They have been chosen to give the appropriate value of σ_c/σ_y for stable crack growth.

4. Mechanisms of crack propagation

There are a number of explanations for the increase in the stress intensity factor with increased filler content.

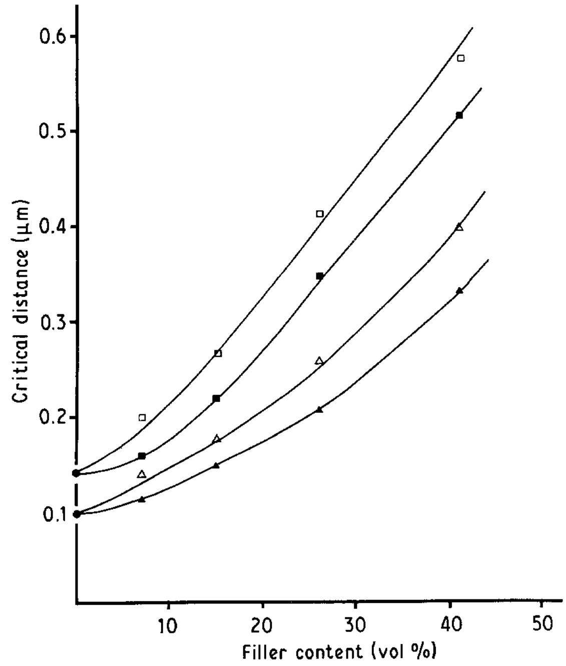

Figure 8 Relationship between critical distance and filler content. The critical distances have been obtained assuming $k = 1$ for 0, 7 and 15% filler content; $k = 1.2$ for 26% filler content; $k = 1.8$ for 41% filler content. (▲) Coated filler in air, (△) uncoated filler in air, (■) coated filler in water, (□) uncoated filler in water.

Firstly, the filler causes the crack to deviate from a straight line and so the surface area of the crack increases. Certainly, increased filler content produces rougher surfaces to the crack, but the increase in the surface area is insufficient to account for the rise in the stress intensity factor [20]. Secondly, it is possible that the filler itself may absorb some of the energy by deformation of the particles. This is unlikely to occur with the brittle silica filler used in this study. Thirdly, the increase in stress intensity factor may be due to the filler particles pinning the crack and causing the crack front to bow out between the particles, thus increasing the "line tension" of the crack front. This crack pinning effect was originally advocated by Lange [24], with later modifications introduced by Evans [25] and Green *et al.* [26]. Fourthly, the increase in toughness may arise from an increased plastic deformation of the matrix. It is known that under certain conditions, materials that are thought to be brittle undergo plastic deformation [27]. Lilley [28] considered that in epoxy resins containing filler particles, local plastic deformation around the particles was responsible for the increase in fracture toughness, although this work was done with low filler loadings.

Crack propagation in unfilled epoxy resins is generally explained in terms of localized shear yielding at the crack tip which leads to crack blunting [18, 23]. A clear relationship has been demonstrated between the yield stress and the stress intensity factor for crack initiation, low yield stresses producing high values for the fracture toughness.

In the present study for particulate filler composites, the stress intensity factor for crack initiation is clearly related to the yield behaviour. For each filler loading, those conditions which give low yield stresses give high values for the stress intensity factor. The yield stress represents a diffuse yielding which surrounds the crack tip and blunts it. Thus during loading a plastic zone forms at the crack tip. The absence of a coupling agent, the presence of water, and a slow crosshead rate all make it easier for the matrix to undergo plastic deformation. Propagation then takes place through this area of localized deformation into material which has not undergone deformation. Once the crack has propagated, the rate of release of energy is greater than that required for a stable crack and so the crack rapidly accelerates, resulting in unstable crack growth. The crack comes to rest once this excess energy has been used up. If there is no significant blunting effect, then the crack propagates in a continuous, stable manner.

The increase in the stress intensity factor which occurs with the addition of rigid particles is generally attributed to the effects of crack pinning [6, 8]. Spanoudakis and Young [6] found that if the effects of crack blunting were removed, i.e. if only values of stable crack growth were examined, then the effects of the filler in increasing the stress intensity factor could be explained in terms of crack pinning, with good agreement between theoretical and experimental results.

However, if the effects of blunting were not removed then the agreement between the theoretical and experimental data was not so good. Thus, crack propagation cannot be fully explained only in terms of crack pinning. Equally, the crack behaviour cannot be explained only in terms of crack blunting, because the stress intensity factor increases with increasing filler fraction, even when propagation occurs in a continuous manner. If crack blunting was the only mechanism then an increase in filler loading would have to produce a decrease in the yield stress, and this is clearly not the case with rigid filler particles. However, the close relationship between the yield stress and the stress intensity factor clearly shows that blunting of the crack tip has a part to play in increasing the stress intensity factor, when stick–slip behaviour occurs. The relative contributions of crack pinning and crack blunting will depend upon the test conditions, crosshead rate, and the nature of the filler–matrix interface.

The critical stress and the critical distance are an alternative representation of the plane strain toughness in which the deformation is confined to a very small zone. The plastic deformation represented by the yield stress is a more diffuse yielding which surrounds the plane strain region. It is the deformation arising from this latter region which gives rise to the crack blunting. If there is no significant blunting effect, then the plane strain behaviour predominates and gives rise to stable crack growth. The relationship between the critical stress and the yield stress has been shown to be important, in epoxy resins, in controlling the mode of crack propagation. This relation would seem to hold true for filled particulate composites as well.

5. Conclusions

The fracture toughness, yield stress and Young's modulus were determined for a composite resin with varying filler loadings, with and without a coupling agent, and after storage in air and water. Under most conditions crack growth proceeded in a stick–slip mode. Whereas the K_{ICi} values decreased with crosshead rate, the K_{ICa} values remained constant. At the fastest crosshead rate used ($50\,\mathrm{mm\,min^{-1}}$) continuous crack propagation occurred at the K_{ICa} value. The values of K_{ICi} were higher for uncoated than for coated filler, and for water-stored samples compared with those stored in air. The K_{ICa} values were largely unaffected.

K_{ICi} and yield stress both increased with filler loading up to the maximum value used of 41 vol %. For each filler loading a unique relationship was found to exist between the yield stress and the stress intensity factor for crack initiation for all testing conditions. The behaviour of the crack dependent upon the relationship between the yield stress and the critical stress. If the critical stress had a value greater than approximately three and a half times the yield stress then the crack propagated in a stick–slip mode. Thus it can be seen that the fracture behaviour of dental composites is very similar to the behaviour of other particulate-filled epoxy resins, with the mechanism of toughening being a combination of crack pinning and crack blunting.

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Received 10 September 1986
and accepted 16 January 1987