# Role of residual stress field interaction in strengthening of particulate-reinforced composites

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A quantitative analysis was conducted on the effect of residual thermoelastic stress concentrations on the strength of particle-reinforced brittle matrix systems. The analysis is derived from the stress intensity factor for a periodic array of coplanar cracks emanating from the matrix-particle interface. It is shown that the major drop in strength occurs at smaller volume fractions of second phase where the residual stress field interaction effects are minimal. The effect of volume fraction on strength becomes important at larger volume fractions (normally above 10–15%). The theory is compared with experimental measurements of strength for glass and alumina matrix composites as a function of the particle volume fraction, its size, and thermal mismatch  $\Delta \alpha$ .

#### 1. Introduction

The cracking that occurs around second-phase particles due to thermal expansion mismatch between the matrix and particulate phase has received much experimental and theoretical attention over the last two decades [1-4]. Whenever a multiphase material experiences processing at elevated temperatures, the differences in the thermal expansion and the elastic constants of the constituent phases result in residual stresses upon cooling to room temperature [5]. These residual thermoelastic micromechanical stresses have always been of interest from both the strengthening and the strength-reducing perspectives. Experiments have shown that strengthening normally occurs when  $\Delta \alpha$  (=  $\alpha_m - \alpha_p$ ,  $\alpha_m$  and  $\alpha_p$  being the thermal expansion coefficients of the matrix and particle, respectively) approaches zero, or exhibits small negative values [6-8]. Weakening, on the other hand, invariably occurs in the systems of positive  $\Delta \alpha$  and the extent of weakening was found to be directly related to the particle size and the level of  $\Delta \alpha$  [9, 10]. Based on an energy balance approach, Wang and Stevens [11] have shown that there is a critical volume fraction of zirconia addition in alumina which leads to crack linkup or coalescence. Similarly, Miyata et al. [9] have shown a consistent reduction of the strength of glassmatrix composites with increase of  $\Delta \alpha$ . No reduction of strength was observed only in systems with  $\Delta \alpha = 0$ . The strong effect of  $\Delta \alpha$  on strength and the existence of a critical volume fraction at which a sharp reduction in strength occurs suggest that the residual stress field interaction between adjacent particles may play an important role.

The aim of the current paper is to develop a quantitative analysis of the problem, based on crack-tip stress field interaction between the neighbouring cracks emanating from the particle-matrix interface. For the present analysis a model system is chosen which consists of spherical particles uniformly distributed in a brittle matrix of higher thermal expansion.

## 2. Stress field interaction effects

Before discussing the crack-tip stress field interaction effects in a brittle matrix containing a large number of particles under residual stress, it is considered appropriate to first examine the case of a brittle matrix containing a single isolated particle. Also, it will be assumed that the particle is of a spherical shape with an annular flaw of length *s* emanating from the particle-matrix interface (Fig. 1b). When an external stress  $\sigma_a$  is applied at infinity, the strength of such a solid can be calculated from the expression [12]

$$\sigma_{\rm s} = \frac{1}{\phi_{\rm c}} \left( \frac{\pi \gamma E}{D[1 + (s/R)](1 - v^2)} \right)^{1/2} - AP \frac{\phi_{\rm t}}{\phi_{\rm c}}$$
(1)

where  $\gamma$  is the fracture energy of the matrix, *E* is Young's modulus,  $\nu$  is the Poisson's ratio, *D* is the particle diameter (*D* = 2*R*), *A* is a constant, *P* is the residual thermoelastic stress which can be calculated from the well-known Selsing equation, and  $\phi_e$  and  $\phi_t$ are given by

$$\phi_{c} = \left(1 - \frac{1}{\left[1 + (s/R)\right]^{2}}\right)^{1/2} \\ \times \left(1 + \frac{1}{4\left[1 + (s/R)\right]^{2}} + \frac{3}{4\left[1 + (s/R)\right]^{4}} \\ \times \left\{1 + \frac{1}{3}\left[\left(1 + \frac{s}{R}\right)^{2} - 1\right]\right\}\right)$$





*Figure 1* (a) Coplanar row of cracks perpendicular to the applied stress; (b) coplanar row of spherical particles with annular cracks emanating from the particle-matrix interface.

$$\phi_{t} = 1 - \left(1 - \frac{1}{\left[1 + (s/R)\right]^{2}}\right)^{1/2} + \frac{1}{2\left[1 + (s/R)\right]^{3/2}} \left(1 - \frac{1}{\left[1 + (s/R)\right]^{2}}\right)^{1/2}$$

In essence, Equation 1 predicts the condition under which a penny-shaped crack will extend under the combined action of applied stress,  $\sigma_a$ , and a residual thermoelastic stress, *P*. It should be pointed out, however, that in developing Equation 1 it was assumed that both radial,  $\sigma_r$ , and tangential,  $\sigma_{\theta}$ , thermoelastic stresses act in the direction to open the annular (penny) crack. If, on the other hand, only radial stress is assumed to act on the crack,  $\phi_t$  in Equation 1 reduces to

$$\phi'_{\iota} = 1 - \left(1 - \frac{1}{\left[1 + (s/R)\right]^2}\right)^{1/2}$$
 (2)

An important conclusion that can be inferred from Equation 2 is that, besides the influence of residual stress, there is a strong effect of particle size and annular flaw size on strength. Furthermore, Equation 1 shows that at low volume fractions of second-phase particles, where the crack-tip stress field interaction effects are minimal, the strength of the composite is governed by the level of residual thermoelastic stress, the particle size and the inherent or annular flaw size.

Let us now extend our analysis from a single particle to a multiple-particle system as shown in Fig. 1b. The ratio of the failure stress for the infinite row of particles, perpendicular to the applied stress,  $\sigma_F$ (Fig. 1b), to the failure stress for a single pore,  $\sigma_c$ , may be obtained from the equation [13, 14]

$$\sigma_{\mathbf{M}} = \sigma_{\mathbf{c}} \left(\frac{\pi c}{2b}\right)^{1/2} \left(\tan\frac{\pi c}{2b}\right)^{-1/2}$$
(3)

where c is the total crack length and 2b the distance between the centres of adjacent particles. In the pre-

sent analysis, it will be assumed that each particle possesses an annular flaw of length, *s*, extending from the particle-matrix interface. These annular flaws are taken to be an integral part of a total crack length. In order to incorporate the annular flaw size and the interparticle separation into the equation for strength, the following relationship must be established:

$$c = R + s = R\left(1 + \frac{s}{R}\right) \tag{4}$$

$$2b = 2R + 2s + d = 2R\left(1 + \frac{s}{R} + \frac{d}{2R}\right)$$
 (5)

$$d = \frac{2D(1-V)}{3V} = \frac{4R(1-V)}{3V}$$
(6)

where V is the volume fraction of the second phase. On substituting Equations 4, 5 and 6 in Equation 3, the strength reduction ratio becomes

$$\frac{\sigma_{\rm M}}{\sigma_{\rm c}} = \left(\frac{\pi [1 + (s/R)]}{2\{1 + (s/R) + [2(1 - V)/3V]\}}\right)^{1/2} \times \left(\tan \frac{\pi [1 + (s/R)]}{2\{1 + (s/R) + [2(1 - V)/3V]\}}\right)^{-1/2}$$
(7)

The ratio  $\sigma_M/\sigma_c$  is plotted as a function of particle volume fraction in Fig. 2. It is seen that the ratio  $\sigma_M/\sigma_c$  decreases very slowly with particle volume fraction for small s/R ratios (s/R = 1 to 0.001). For s/R ratios larger than approximately 5–10, the strength of the solid containing a large number of interacting cracks drops very quickly, even at very small volume fractions of less than 10%.

The equation for the strength of a solid containing annular cracks at distance d (Fig. 1b) can be developed by combining Equations 1 and 7. Thus, substituting Equation 1 in Equation 7 and replacing  $\sigma_c$  with  $\sigma_s$  and  $\sigma_M$  with  $\sigma_F$ , this gives

$$\sigma_{\rm F} = \left[\frac{1}{\phi_{\rm c}} \left(\frac{\pi\gamma E}{D[1 + (s/R)](1 - v)^2}\right)^{1/2} - AP\frac{\phi_{\rm t}}{\phi_{\rm c}}\right] \\ \times \left(\frac{\pi[1 + (s/R)]}{2\{1 + (s/R) + [2(1 - V)/3V]\}}\right)^{1/2} \\ \times \left(\tan\frac{\pi[1 + (s/R)]}{2\{1 + (s/R) + [2(1 - V)/3V]\}}\right)^{-1/2}$$
(8)



Figure 2 Predicted variation of strength (using Equation 7) with particle volume fraction and s/R ratio.



Figure 3 Change of strength with particle volume fraction for a random array of particles having thermal expansion coefficient smaller than that of the matrix. All values for strength were calculated using A = 0.1, particles size of 300 µm, and varying residual stress from  $P \rightarrow 0$  to P = 196 MPa. The temperature change was assumed to be  $\Delta T = 600$  °C.

Fig. 3 illustrates the change of fracture strength ( $\sigma_F$ ) with particle volume fraction for a given s/R. As expected, there is a weak dependence of particle volume fraction on strength for small s/R (s/R < 0.1), and a much stronger effect at large s/R. A sharp drop of strength at low volume fractions and at high s/R ratios suggests that the residual stress generated by the thermal mismatch and/or the s/R ratio plays a much more important role in controlling the strength of a composite than the crack-tip stress field interaction of the neighbouring cracks.

So far our discussion has been concentrated on the effect of the particle volume fraction and s/R ratio on strength, assuming no change of particle radius. However, Equation 8 shows a strong dependence of strength on particle size (Fig. 4). It is clear that the particle size, along with s/R ratio, plays the dominant role in controlling the strength response of a composite, particularly at lower particle volume fractions.

### 3. Discussion

It is now well accepted that second-phase particles in a brittle matrix of different thermal and elastic properties can serve as fracture origins. The residual thermal stress fields developed on cooling from the fabrication temperature are expected to mutually interact, leading



In calculating the strength of the composite in Fig. 5, it was assumed that there is a change of s/Rratio from 0.04, for a glass matrix of equal thermal expansion with that of the glass beads, to 25 for a glass matrix having a thermal expansion coefficient 5.7 times larger than that of the glass beads [9]. The difference in thermal expansion coefficients between the matrix and particles may influence the strength behaviour of the composite in two ways; through the stress-field interaction effects, and through the generation of radial cracks on cooling from the fabrication temperature. In the first case, the crack-tip stress fields of the adjacent particles interact to magnify the stress intensity factor so that the applied stress required to cause fracture is reduced. In the second case, the cracks are formed either prior to the external stress application or during stressing. Here, the cracks are not visible prior to stressing and can be detected only after fracture. The ultimate result of this interaction is crack linking and a drastic reduction in strength of the composite, Microcracking and crack linking are well known phenomena in two-phase systems [14, 15].



Figure 4 Predicted variation of strength (using Equation 1) with particle size for a given s/R ratio.



Figure 5 Comparison between predicted (using Equation 8) and measured strength for glass matrices with different thermal expansion containing spherical glass particles (beads) of 300  $\mu$ m size [9].

In order to understand better the mechanism of crack extension and consequently the fracture behaviour, it is necessary to find out whether the cracks are formed prior to the application of external load or during loading. Clearly, if the residual stresses are high, soon after the external load is applied the annular crack-tip stress field will reach a critical value and crack extension will occur. The extent of crack propagation after initiation can be estimated using a crackopening displacement concept [12]:

$$s = \frac{\pi (ER \,\Delta \alpha \Delta T)^2}{(1 - v^2)^2 \,K_{\rm lc}^2} - R \tag{9}$$

where s is the annular/radial flaw size and  $K_{\rm le}$  is the fracture toughness of the matrix material. The important result of Equation 9 is that there is a strong dependence of arrested crack length, s, on thermoelastic strain,  $\Delta \alpha \Delta T$ . Assuming no contribution from the kinetic energy of the crack, Equation 9 may be used to predict the change of strength with change of  $\Delta \alpha$  by combining Equations 8 and 9.

Fig. 6 shows the comparison between measured and predicted strength (from Equations 8 and 9) as a function of  $\Delta \alpha$  for a glass matrix containing 300  $\mu$ m size glass beads. It is clear from Fig. 6 that good agreement between predicted and measured values for strength is achieved at larger  $\Delta \alpha$ , whereas at small  $\Delta \alpha$ Equation 8 gives somewhat underestimated values for strength. It may also be noticed that Equation 8 gives consistently lower values for strength than those actually measured. This behaviour may suggest that the extent of crack propagation after initiation is controlled not only by the crack opening but also by microstructural features such as porosity and the particle-matrix interface. A quantitative analysis using Equations 8 and 9 shows that, for example, for a volume fraction of glass beads of 0.1 and  $\Delta \alpha = 4 \times 10^{-6}\,^{\circ}\mathrm{C}^{-1},$  the mean interparticle spacing is



Figure 6 Variation of strength as a function of thermal expansion mismatch  $\Delta \alpha$ , for a glass matrix containing spherical glass particles of 300 µm size and V = 0.1; ( $\bigcirc$ ) experimental, (——) theoretical (Equation 8).

significantly larger than the crack length, s, while at higher  $\Delta \alpha$  the length of an arrested crack is larger than the interparticle spacing. This means that the probability of crack arrest by a neighbouring particle (or its interface) is smaller at lower  $\Delta \alpha$  than at larger  $\Delta \alpha$ . which appears to be consistent with the results presented in Fig. 6. Experimental results on glass matrix-glass bead composites, used to test the present theory, have shown that severe cracking occurred in all composites that contained glass particles of smaller thermal expansion [9]. However, in composites with a relatively small thermal mismatch  $(\Delta \alpha = 1.8)$  $\times$  10<sup>-6</sup>  $^{\circ}\mathrm{C}^{-1}$  ), the microcracks were observed only between closely spaced particles while all other randomly distributed particles with larger interparticle spacings did not contain cracks after cooling. Also, coalescence of microcracks was readily observed in composites of higher volume fraction of particles and in composites of smaller volume fractions, but only between the closely spaced particles.

These observations are in agreement with the theoretical prediction made in the present paper and serve as a confirmation that the residual thermoelastic stresses are of a short range and their interaction can influence the crack propagation behaviour only in composites with a large number of particles (high particle volume fractions) or in highly segregated composites. Perhaps the most important implication of the present theoretical analysis and the experimental results cited is that the magnitude of the residual stresses plays the dominant role in controlling the strength response of the composite, rather than the crack-tip stress field interaction. In other words, in systems with high thermal expansion mismatch, the critical particle size for crack extension\* will be small and cracking will occur on cooling from the fabrication temperature, prior to external stress application. In systems with lower thermal expansion mismatch, the critical particle size for crack extension will normally be large and microcracking will occur only during external stress application. In both cases, partial or nearly complete relaxation of residual stresses will occur. The resultant body will be free of residual stresses but will contain a large number of cracks. Clearly, the mechanical response of such solids will depend entirely on the number and size of these cracks.

To further test the theory, Fig. 7 shows the change of strength of alumina matrix-partially stabilized zirconia composite with zirconia volume fraction at different zirconia particle sizes. As with the glass matrix-glass particle composite discussed above, the effect of particle size on strength of the aluminazirconia composite is relatively much stronger than the effect of zirconia particle volume fraction on strength. For example, an increase of zirconia particle size from 1.25 to 5.4  $\mu$ m caused a drop in strength from ~ 550 to ~ 250 MPa, whereas an increase in zirconia volume fraction from ~ 2 to 20% caused a drop in strength of only 100 MPa. In calculating the strength variation with zirconia volume fraction in Fig. 7, a constant value for s/R was assumed, i.e.

<sup>\*</sup> Often called the critical particle size for spontaneous cracking.



Figure 7 Comparison of measured and calculated strength in Al<sub>2</sub>O<sub>3</sub>-ZrO<sub>2</sub> composites containing two different zirconia particle sizes. Experimental values (taken from Claussen [1]): ( $\bigcirc$ ) D = 1.25, ( $\triangle$ ) D = 2.84. Theoretical values: ( $\oplus$ ) D = 1.25, ( $\triangle$ ) D = 2.84. (D in  $\mu$ m).

s/R = 15. The residual stress responsible for fracture was assumed to be the thermoelastic stress generated due to the presence of a thermal mismatch of magnitude P = 468 MPa. The transformation stress was assumed to be of lower magnitude and thus to exhibit a minor effect on the strength of the composite.

## 4. Conclusions

The analysis of the interactions between closely spaced cracks emanating from the particle-matrix interface has established the conditions for crack linking prior to catastrophic fracture. Due to the shortrange order interaction effect of the two coplanar cracks, crack linking occurs at large particle volume fractions (small interparticle spacings). At small volume fractions where the interaction effects are minimal, the dominant factor affecting the strength response of a composite is the difference in thermal expansion coefficients between the matrix and particulate phase,  $\Delta \alpha$ . Crack extension may occur during external stress application due to the combined action of applied stress and residual thermal stress. At large  $\Delta \alpha$ , however, radial/annular crack extension may occur prior to external load application and the strength response of the solid will be controlled entirely by the large pre-existing (arrested) cracks.

It is shown that the strength of two-phase particulate composites is governed by the thermal mismatch ( $\Delta \alpha$ ), the particle size, *D*, the ratio of flaw to particle size, *s/R*, and the particle volume fraction.

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