CONDITIONS FOR TOUGHENING OF PARTICULATE BRITTLE COMPOSITES

V. D. Krstic and A. K. Khaund

Department of Metallurgy and Materials Science
McMaster University
Hamilton, Ontario, Canada

ABSTRACT

The effect of interfacial strength and stress concentration on toughening of particulate brittle composites are discussed. When thermal and elastic stresses are eliminated and satisfactory bonding is formed between the glass matrix and metal particles, increases in toughness over 60 times that of the glass was achieved.

KEYWORDS

Brittle composites, fracture toughness, elastic mismatch, thermal mismatch, interfacial bonding.

INTRODUCTION

The need for toughness improvement of brittle ceramic matrices has led to extensive theoretical and experimental studies of crack-particle interaction as a possible means for toughening of particulate composites. Since Forwood and Forty (1965) showed the metallic Ag particles in NaCl single crystals decelerated propagating cracks, various combinations of ductile metal particles and brittle matrices have been formulated in an attempt to increase the toughness of these composites. For example, spherical metal particles have been mechanically dispersed in ceramic matrix, i.e. Mo in Al_2O_3 (McHugh and others, 1966; Rankin and others, 1971), W in MgO (Rossi, 1969), Fe, Co and Ni in MgO (Hing and Groves, 1972), Ni in glass (Stett and Fulrath, 1968), W in glass (Nivas and Fulrath, 1970) and Zr in ZrO_2 (Virkar and Johnson, 1977). All these investigations indicated that the metal particles have substantially increased either the fracture strength (McHugh and others, 1966; Hing and Groves, 1972; Stett and Fulrath, 1968; Nivas and Fulrath, 1970) or increased the thermal shock (Rossi, 1969) resistance of the ceramic matrices. However, the fracture toughness increases are small (Rankin and others, 1971; Virkar

and Johnson, 1977) even though high toughness metal particles were incorporated. The common characteristics of all these studies are that metal particles have not been utilized.
This paper discusses the importance and the role of internal stresses and interfacial properties in toughening brittle matrices as well as the conditions for effective crack-particle interaction and utilization of inherent toughness of second phase dispersion.

CONDITIONS FOR TOUGHENING

If an infinite isotropic body containing a spherical particle of radius (a) with elastic constants different from that of the matrix, is subjected to a uniform applied stress (σa), a stress concentration is developed around the particle and is usually expressed in terms of radial (σrr) and tangential ($\sigma\theta\theta$) stress components. According to Goodier (1933), the radial component of the elastic stress concentration is:

$$\sigma_{rr} = \sigma_a \left\{ 2A\left(\frac{a}{r}\right)^3 - \frac{2\nu m}{1-2\nu m} C\left(\frac{a}{r}\right)^3 + 12B\left(\frac{a}{r}\right)^5 \right.$$

$$\left. + \left(-\frac{2(5-\nu m)}{1-2\nu m} C\left(\frac{a}{r}\right)^3 + 36B\left(\frac{a}{r}\right)^5 \right) \cos 2\theta \right\} + \frac{\sigma_a}{2} (1 + \cos 2\theta) \qquad (1)$$

and the tangential component;

$$\sigma_{\theta\theta} = \sigma_a \left\{ -A\left(\frac{a}{r}\right)^3 - \frac{2\nu m}{1-2\nu m} C\left(\frac{a}{r}\right)^3 - 3B\left(\frac{a}{r}\right)^5 \right.$$

$$\left. + \left(C\left(\frac{a}{r}\right)^3 - 21B\left(\frac{a}{r}\right)^5 \right) \cos 2\theta \right\} + \frac{\sigma_a}{2} (1 - \cos 2\theta) \qquad (2)$$

where

$$A = \frac{1}{4} \frac{Gm - Gi}{(7-5\nu m)Gm+(8-10\nu m)Gi} \times \frac{(1-2\nu i)(6-5\nu m)2Gm+(3+19\nu i-20\nu m\nu i)Gi}{(1-2\nu i)2Gm+(1+\nu i)Gi}$$

$$+ \frac{1}{2} \frac{\left\{ (1-\nu m)\left(\frac{1+\nu i}{1+\nu m}-\nu i\right)Gi - (1-2\nu i)Gm \right\}}{(1-2\nu i)2Gm + (1+\nu i)Gi}$$

$$B = \frac{1}{4} \frac{Gm - Gi}{(7-5\nu m)Gm + (8-10\nu m)Gi} \qquad C = \frac{1}{4} \frac{5(1-2\nu m)(Gm-Gi)}{(7-5\nu m)Gm + (8-10\nu m)Gi}$$

Gm, νm and Gi, νi are the rigidity modulus and Poisson's ratio for matrix and the second phase particle respectively, and r is the distance from the centre of the particle (see Fig. 1)
Figure 2 illustrates two dimensional variation of radial (σrr) and tangential ($\sigma\theta\theta$) component of the elastic stress concentration around a spherical particle possessing elastic properties higher than those of the matrix. It has been shown (Khaund and others, 1977; Tirosh and Tetelman, 1976) that when a crack propagates through an array of second phase particles with different elastic properties, it responds to the sign and magnitude of the induced stresses. On approaching spherical particle ($G_i > G_m$) equatorially the crack experiences a decrease of ($\theta=90°$) stress intensity factor since tangential ($\sigma\theta\theta$) component of the elastic stress concentration is smaller than the applied stress (σa).
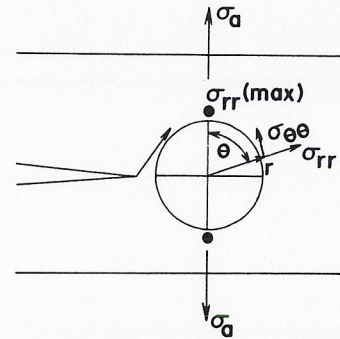
Fig. 1. Crack path in the near vicinity of the Ni particles in glass matrix ($\theta=0°$ is the loading axis).
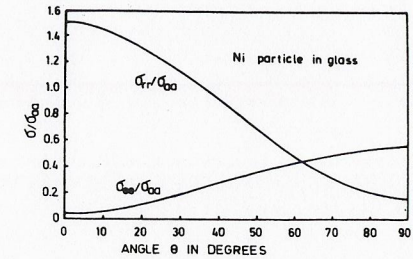

Fig. 2. The relative change of radial (σrr) and tangential ($\sigma\theta\theta$) component of elastic stress concentration generated around an Ni particle in a glass matrix ($\theta=0°$ represents the loading axis).

For the composites in which the particle possesses sufficiently higher elastic properties than those of the matrix, the relative change of stress intensity factor K_I (elastic) is given by Khaund and others (1977):

$$\frac{K_I(\text{elastic})}{K_I} = 1 - 0.78 A\left(\frac{a}{t}\right)^3 - \frac{1.56\nu m}{1-2\nu m} C\left(\frac{a}{t}\right)^3 - 1.64 B\left(\frac{a}{t}\right)^3 \qquad (3)$$

where t is the distance between the spherical inclusion and the crack front. However, more practical case would be the crack approaching particle off the equatorial plane (i.e. along $\theta=90°$ in Fig. 1). In such a case, the radial component (σrr) of the elastic stress concentration at the poles of the particles ($\theta=0°$) will tend to deflect the propagating cracks toward the position of maximum stress concentration (see Figs. 1 and 2). Provided that other stress concentrations would not interfere, this crack-particle interaction will result in a crack avoiding the particle completely. From the view point of toughening of brittle matrices with spherical metal particles this is the most undesirable combination and the main limitation for significant toughness improvement.
The next combination of matrix and second phase is the case of elastically soft particle dispersed in an elastically hard matrix ($G_m > G_i$). For such composite, constants A, B and C in Eq. (3) will have opposite sign and the stress intensity factor K_I (elastic) for approaching crack (along $\theta=90°$) will be increased and the crack will be attracted towards the particle (Fig. 3).
The phenomenon of crack avoiding the particles in hot pressed glass-Ni composite ($\alpha_i = \alpha_m$ where α_i, α_m being the thermal expansion coefficient of Ni and glass respectively) is shown in Fig. 4 (Khaund, 1978). Figure 4 proves that utilization of inherent toughness of Ni particles were not achieved even when satisfactory interfacial bonding existed between inclusion and matrix due to presence of elastic stress concentration. On the other hand, in hot pressed glass-partly oxidized aluminum particles ($\alpha_i = \alpha_m$) substantial deformation of Al particles was found (see Fig. 5) as in this composite $G_i = G_m$ and no elastic stress

concentration is present to deflect the crack front or to break the interface (Khaund, 1978).
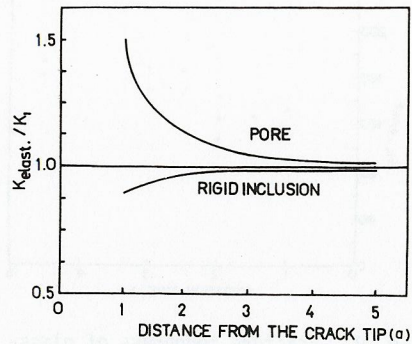

Fig. 3. Relative change of stress intensity factor (from Eq. 3) in front of circular particle as a function of distance from the centre of the particle.
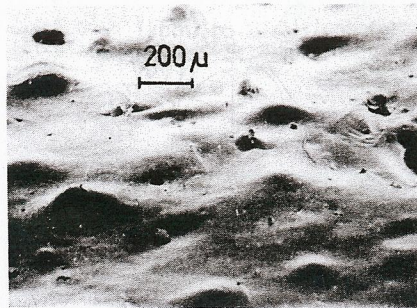

Fig. 4. Scanning electron micrograph showing the fracture surface of the glass-Ni composite. (Ni particles oxidized at 700^{0}C for 2.5 h ; 7 vol.%).

A similar argument can be applied to the interaction of a crack-tip stress field with thermal stress developed as a result of thermal expansion mismatch. When a crack approaches thermally mismatched spherical particle, Khaund and others (1977) obtained the solution for the stress intensity factor in the form:

$$K_{thermal} = K_I + 0.94 \, P \, \frac{a^3}{t^{5/2}} \qquad (4)$$

where

$$P = \Delta T (\alpha m - \alpha p) \left(\frac{1 + \nu m}{2Em} + \frac{1 - 2\nu p}{Ep} \right)^{-1} \qquad (5)$$

and K_I is the stress intensity factor due to applied stress, a is the particle radius, $\Delta \alpha$ the difference in thermal expansion of the matrix and the second phase, ΔT the temperature gradient and t is the distance between the crack tip and the centre of the particle.
The sign of the constant P in Eqs. (4) and (5) depends on the sign of $\Delta \alpha$. When $\alpha_m < \alpha_p$ the stress P is negative and local stress intensity factor in Eq. (4) will decrease, causing deceleration (providing there is satisfactory bonding between the matrix and the second phase). Relative change of stress intensity factor for these two different cases are illustrated in Fig. 6. The tensile component of thermal stress (σrr) will superimpose to reinforce the (σxx) component of the crack front; since crack propagation in a real composite material proceeds in a non-planar fashion, this sudden reinforcement of (σxx) component will cause tilting of principal axis of crack driving stress (σyy) and so would force the crack to avoid the particle. This behaviour of the crack has been confirmed by Davidge and Green (1968) in glass-thoria composite in which glass crack did bypass high modulus high thermal expansion thoria spheres. Under this condition ($\alpha_m < \alpha_p$) inherent toughness of the particles will not be utilized.

When $\alpha_m > \alpha_p$, the stress P is positive (from Eq. (4)), the local stress intensity factor $^mK_{thermal}$ would increase by the action of ($\sigma\theta\theta$) and the crack would be attracted towards the particle. This case of crack-particle interaction can, in principle, lead to utilization of inherent toughness of second phase.
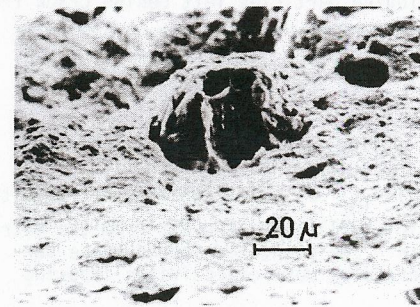

Fig. 5. Tilted fracture surface showing local necking of Al particle. (After Khaund,1978)
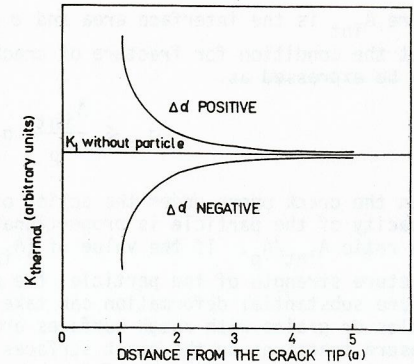
Fig. 6. Relative change of local stress intensity factor for the crack approaching spherical particle with thermal stress field.

THE ROLE OF INTERFACE

So far analysis on the effect of elastic and thermal stress concentrations in particulate composites was based on the assumption that satisfactory bonding between the two phases exists.
To better understand the role of interfacial strength consider a composite made up of metallic second phase dispersion embodied in a brittle matrix of the same or lower modulus and thermal expansion. From foregoing analysis, such a composite provides basic requirements for effective crack-particle interaction and utilization of inherent toughness of second phase, i.e. a propagating crack should not avoid but intersect the particle. If the interface is weak, tri-axial stress state developed at the front of a propagating crack may break the interface in which case pseudo-pores are formed. For systems with strong interface, the matrix crack can reach the interface (provided elastic and thermal mismatch criteria discussed above are met) and circumvent the ductile particle leaving "ligaments" across the crack faces shown in Fig. 5. It is important to note at this point that prerequisite for ligament generation behind the crack front in a well bonded particulate composite is the difference in crack opening displacement between the two phases. If the particulate phase has crack opening displacement higher than that of the matrix material, when a crack reaches particle-matrix interface it will be locally blunted and its segments forced to circumvent the particle, thus bridging the crack along its length. Once the ligaments are left behind the crack front, they impose closure traction on the crack surfaces against opening of crack. The force that each individual particle can supply is associated with interfacial and plastic properties of metallic particle. For example, the force on the particle is:

$$F_p = A_p \, \sigma_f \qquad (6)$$

where A_p is the cross-section and σ_f is the uniaxial fracture strength of the particle. This force is counterbalanced by the force acting across the interface, i.e.

$$F_{int} = A_{int} \; \sigma_{int} \qquad (7)$$

where A_{int} is the interface area and σ_{int} is the interfacial strength. It follows that the condition for fracture of crack-bridging particles left behind crack front may be expressed as:

$$\sigma_f \leqslant \frac{A_{int}}{A_p} \; \sigma_{int} \qquad (8)$$

When the crack opens under the action of an applied stress, the stress bearing capacity of the particle is proportional to the interfacial strength (σ_{int}) and the ratio A_{int}/A_p. If the value of $(A_{int}/A_p) \sigma_{int}$ is sufficiently lower than the fracture strength of the particle, the particle will be pulled-out from the matrix before substantial deformation can take place. The elongated second phase particles or grains with rough surfaces are therefore, more effective in imposing closure tractions on the crack surfaces.
The nature and the strength of the boundary phase formed during fabrication of the composite, is another important factor that governs the toughening effects. For example, when partly oxidized Ni particles are dispersed in a glass matrix possessing thermal expansion coefficient higher than that of nickel, a propagating crack always follows the weak Ni-nickel oxide interface (see Fig. 7a). A different crack path is obtained when partly oxidized Ni particles are dispersed in MgO matrix (Fig. 7b); a propagating crack now tends to follow MgO-nickel oxide interface rather than Ni-nickel oxide interface.
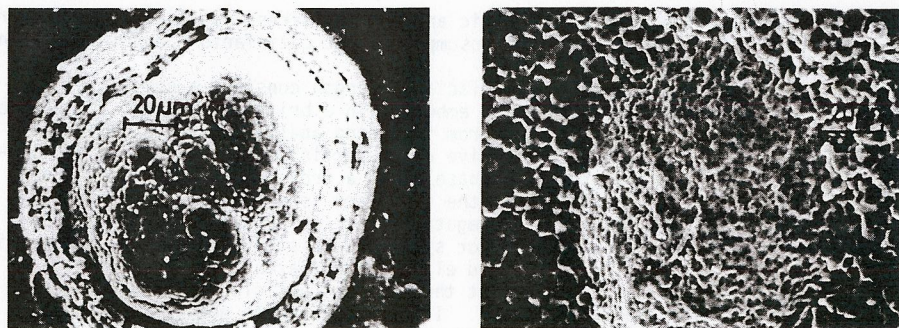

Fig. 7   Crack path in the vicinity of Ni particle (After Krstic, 1979).
(a) Ni in glass   (b) Ni in MgO

The role of the interface in particulate composites can best be understood through its effect on the composite toughness. For example, it has been shown (Khaund, 1978) that in the glass-Ni system, in which thermal expansion coefficients were matched, the increase of oxidation time of Ni particles led to continual increase of fracture toughness up to 30 min of oxidation and thereafter toughness decline (Fig. 8).
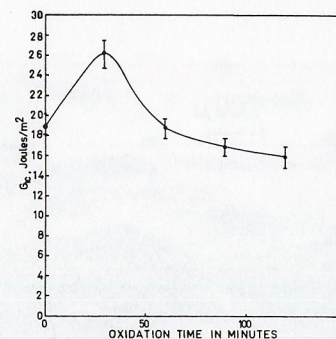
Fig. 8.   Variation of fracture toughness as a function of oxidation time of Ni particles at 750°C. 7 vol. %Ni in glass (After Khaund, 1978).
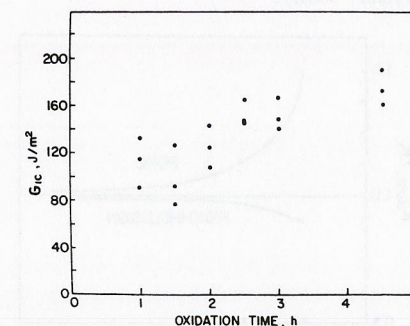
Fig. 9.   Fracture toughness of glass-Al composite as a function of oxidation time of Al particles at 450°C (particle size in all composites was 150 – 106 μm), Volume fraction of Al is 7%.

At two hours oxidation at 750°C, the composite toughness dropped to 16 J/m^2 and the crack bypassed most of the particles. The same experiment with glass-Al composite (Krstic, 1979) exhibited entirely different behaviour. Toughness continually rose with oxidation time of Al particles even after 4.5h (Fig. 9).
The increase of toughness with oxidation time in glass-Ni composite is attributed to continual bonding improvement and the associated crack propagation resistance imposed by the interface. Beyond some optimal oxidation time (30 min in this composite) the interfacial strength is sufficient to allow build up of elastic stress concentration and possibly some residual thermal stress which drive the crack in the matrix only (see Fig. 4). This automatically leads to a toughness degradation and prevents utilization of inherent toughness of Ni phase. In the glass-Al system which is free of any residual stresses (as in this work $E_m = E_i$ and $\alpha_i = \alpha_m$) the toughness continually increases with improving bond strength. Figure 10 shows toughness increase with volume fraction of partly oxidized Al particles which were dispersed and hot pressed in a glass matrix with matched thermal expansions.
The toughness value obtained for 2o vol. percent Al is over 60 times that of the glass matrix. Such a large toughness increase is attributed to deformation of Al particles in front of a crack and ligaments fracture behind the crack front.
Typical ductile fracture appearance of the fracture surface of Al particles and dimples extended above the fracture plane (see Fig. 5) of the glass matrix provide the evidence that the bulk of energy dissipated during fracture is confined to fracture of ligaments left behind the crack front.
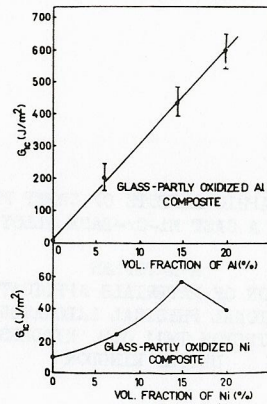
Fig. 10. Fracture toughness
of glass-partly oxidized
Al and Ni composites
as a function of volume
fraction of Al and Ni.
(After Khaund, 1978 and
Krstic, 1979).

SUMMARY AND CONCLUSIONS

Poor particle-matrix bonding and stress concentrations developed as a result of
elastic and thermal mismatch are the main limitations to the utilization of tough-
ness of second phase particles in brittle matrices. Weak interface usually breaks
during cooling from fabrication temperature or fractures ahead of a crack before
the crack front reaches the interface. Sufficiently higher elastic and thermal
properties of particles embodied in a soft matrix (E_m, $\alpha_m < E_p$, α_p) lead to stress
concentrations which drive the crack around the particles. Under these circum-
stances the inherent toughness of the second phase will not be utilized. Signifi-
cant toughness improvement of a brittle matrix can be achieved in well bonded
composites with elastic and thermal properties of dispersed metallic particles
  equal   or lower than that of the matrix.

REFERENCES

Davidge, R.W. and T. J. Green (1968). J. Mat. Sc., 3, 629.
Forwood, C. T. and A. J. Forty (1965). Phil. Mag., 11, 1067.
Goodier, J. N. (1933). J. Appl. Mech., 1,39
Hing, P. and G. W. Groves (1972). J. Mat. Sc., 7,427.
Khaund, A. K., V. D. Krstic and P. S. Nicholson (1977). J. Mat. Sc., 12,2269.
Khaund, A. K. (1978).M. Eng. Thesis, McMaster University.
Krstic, V. D. (1979). Ph.D. Thesis, McMaster University.
Nivas, Y. and R. M. Fulrath (1970). J. Amer. Ceram. Soc., 53,188.
Rankin, D. T., J. J. Stiglich, D. R. Petrak and R. Ruh (1971). J. Amer.
   Ceram. Soc.; 54;277.
Rossi, R. C. (1969). Amer. Ceram. Soc. Bull., 48,736.
Stett, M. A. and R. M. Fulrath (1968). J. Amer. Ceram. Soc., 51,599.
Tirosh, J. and A. S. Tetelman (1976). Int. Journal of Fracture, 12,187.
Virkar, A. V. and D. L. Johnson (1977). J. Amer. Ceram. Soc., 60,514.