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ABSTRACT

We have calculated the mechanical properties of brittle composites with spring-network (SN) model. The composites that we studied involve the transformation toughening effects and the accompanying micro-cracking. Our simulation results are consistent with experiments of $MoSi_2$ toughened with ZrO_2. We found that the continuum mechanics predictions on linear dependence in e^T and f about the transformation toughening are correct except some differences in the critical transformation stress dependence. By monitoring the stress changes due to the transformation and micro-cracking we are able to separate, for the first time, the contributions from these two competing effects. We found that the fracture toughness due to micro-cracks accompanying the transformation is a decreasing function of f and may be negative for large f. We also found that the fracture toughness of the composites increases as the modulus, interfacial cohesion, and uniformity of particle distributions increases.

INTRODUCTION

Potential high temperature structural materials like intermetallics and ceramics suffer room temperature brittleness and have low fracture toughness [1, 2]. This low temperature brittleness prevents the wide-spread use of these materials. One way to improve the materials fracture toughness is to make use of composite toughening, notably by transformation toughening, crack deflection and whisker toughening. The calculation of the fracture toughness of composites has been a very difficult task [3]. In this report, we used a spring-network (SN) model [4, 5] to simulate the mechanical response of the brittle composites under applied stress.

We used a "spring-network model" (SN) model [4, 5] to include the effects of microstructure, second-phase particle toughening and micro-cracking phenomena. We treat the system as a two-dimensional triangular lattice of nodes, each of which is connected to its six nearest neighbors by elastic springs. The microstructure is generated with a Potts model. Transformation of a second-phase particle is modeled by irreversibly increasing the equilibrium spring length when the mean stress on the second-phase particle exceeds a prescribed critical stress, σ_c. Cracking of a bond is modeled by irreversibly turn off the interaction between the nodes that was connected originally when the corresponding breaking length is reached. We assigned a 1% increase in the length of the springs (e^T=0.01) connected to second phase particles upon transformation. The breaking length for the bulk bonds is $r^b{}_m$=1.005 and for the grain boundaries and second phase particles, $r^b{}_{gb}=r^b{}_p=r^b{}_{pm}$=1.003. We note that this choice of breaking strains ensures that there should be a mixture of trans- and intergranular fracture in polycrystals [6]. Finally, we choose a mean stress criterion for transformations with critical stresses σ_{cj} of 0.0005, 0.0010, and 0.0015 and other values (for j equal to 1, 2 and 3, respectively) to test the effect of σ_c on the enhancement of the fracture toughness. Also the second phase particles are randomly distributed in the matrix with a particle size of 1 node (labeled as Sj in the figures, where j representing composites with a transformation stress of σ_{cj}) or cluster of 7 nodes (labeled as Lj in the figures) to study the microstructural effects on the fracture toughness.

RESULTS AND COMPARISONS WITH EXPERIMENTS AND THEORIES

In this section, we describe our method of calculating fracture toughness and present results on the increase of fracture toughness, ΔK_{1c}, as a function of the dilatational strain, e^T, volume fraction of particles, f, and the critical transformation stress, σ_c. We then compared these results with available experimental measurements in $MoSi_2/ZrO_2$ (ZTM) [7], Al2O3/ZrO2 (ZTA) [8] and the continuum theory [1, 8, 9]. Unless specifically stated, all the parameters are associated with composites mixed with small particles having e^T=0.01, σ_c=0.0005, f=0.25, k_m=k_p=1.0, $r^b{}_{gb}$=1.03, and $r^b{}_m$=1.05 (i.e. composite S1). The simulation was performed on a 2-dimensional triangular 50x50 nodes with periodic boundary condition in x-direction and free boundaru condition in y-direction. The composites with finite volume fraction of small and large second-phase particles do not fracture along grain boundaries as compared to the pure polycrystals. The change of the fracture path is accompanied by an increase in fracture toughness, ΔK, as the transforming second-phase particles expand and shield the crack by exerting compressive stresses on the original crack.

The local stress, σ_L, for composites with second phase particles is linearly related to the local stress intensity, K_L. The same relation exists between the far-field stress , σ_∞ and stress intensity, K_∞. The reduction of the stress (or ΔK) over the block of nodes can be monitored as a function of the number of particles transformed as the strain (or stress) is applied. The increase of the fracture toughness is described by

$$\Delta K_{1c} = K_\infty - K_L = \alpha(\sigma_\infty - \sigma_L) = \alpha(-\Delta\sigma_{xx}) \qquad (1),$$

where α is a constant that depends on the simulation cell. This correlation of the stress change and the ΔK enable us to quantify the change of the fracture toughness. It has been shown by continuum fracture mechanics [1, 8, 9] that the increase of the fracture toughness, ΔK, due to the transformation is proportional to the modulus of the materials, E, the dilatational strain, e^T, the volume fraction of the transforming particles, f, and is inversely proportional to the critical stress of the transformation, σ_c. The formula is represented by

$$\Delta K = \alpha_1 E e^T f(\sigma_c)^{-1} / (1-\nu) \qquad (2),$$

where ν is the Poisson's ratio, α_1 is a constant value.

The first way to change the fracture toughness is to change the dilatational strain, e^T, of the particles. In the simulation, we held all parameters the same except the value of e^T to study the dependence of ΔK_{1c} on e^T. As shown in Fig. 1, ΔK is linear in e^T for values from -0.0025 to +0.025. Unfortunately, there has no independent experimental studies on the linear dependence of e^T existed yet. Therefore, direct comparisons with experiments are not possible at this moment. But this linear dependence is consistent with the continuum results as expressed in eq. (2) [1]. We found that for $e^T > 0$ the composite is tougher than the single phase material so it is good for transformation strain to be large and positive if massive micro-cracking (shattering of the sample) can be avoided [10]. On the other hand, if e^T is negative the composite is less tough than the matrix material. This reduction in fracture toughness has been shown in $LnBO_3$ (where Ln represents Lanthanide) and $MgO.SiO_2$ weakened composites [10].

The second quantity in eq. (2) that can be easily changed is the volume fraction of the transformable particles, f. Assuming all the parameters the same except f, we calculated the ΔK as a function of the volume fraction of particles. The volume fraction dependence of ΔK has been shown previously by Chen et al [4, 5], and we only summarize the results here. It was found that the increase in the fracture toughness associated with the transformation toughening is linear in the

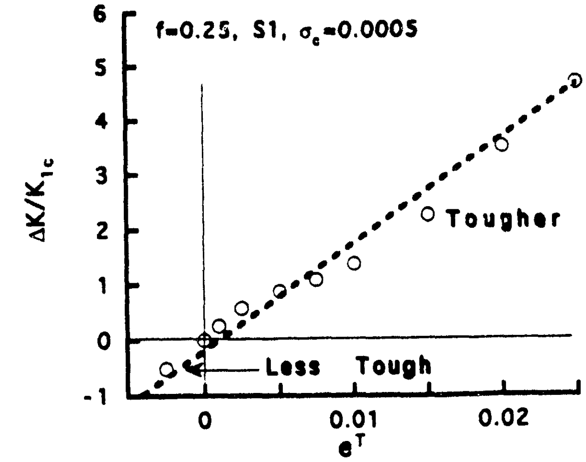


Fig. 1: (a) The linear dependence of ΔK vs. dilatational transformation strain, e^T.

volume fraction, f, for small and large particles (Fig. 2a). We noted that the increases of the composite toughness with large particles or the particles which clusters are smaller than the composites with small particles (assuming the same σ_C). There are about 10 to 15% variations in the calculated values of ΔK. The measurement of ΔK indicates a linear dependence of f of ZrO_2 in ZTM [7] shown in Fig. 2b is a direct confirmation of the simulation results. This calculated linear dependence of ΔK as a function of f is also consistent with the theory of continuum mechanics as represented by eq. (2).

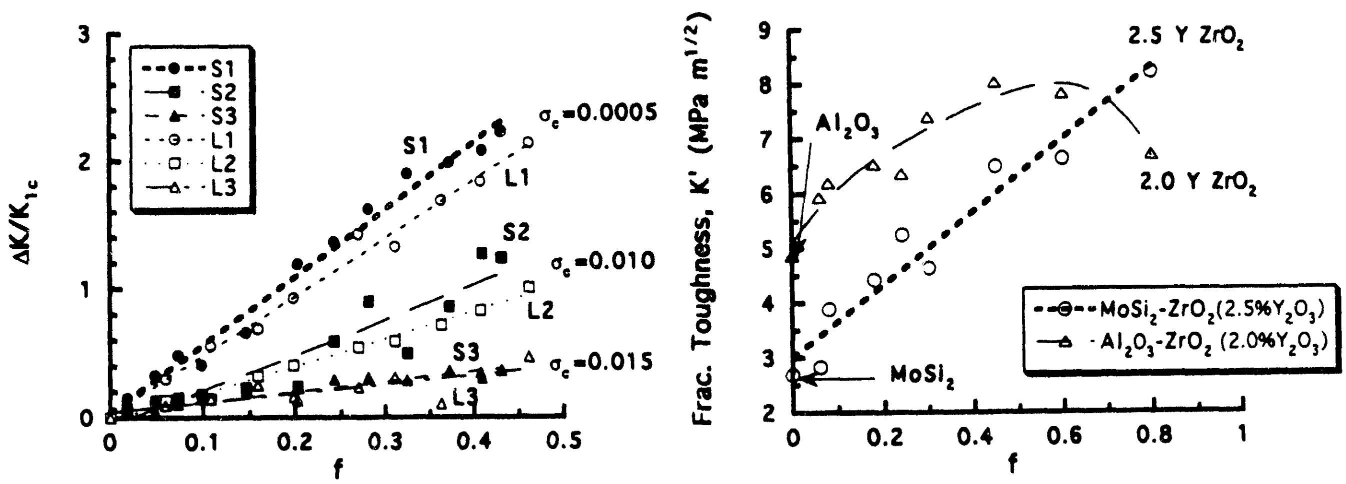


Fig. 2: The linear dependence of ΔK vs. f for (a) small and large particles. (b) The experimental results of ΔK as a function of f for $MoSi_2/ZrO_2$ (ZTM) and Al_2O_3/ZrO_2 (ZTA).

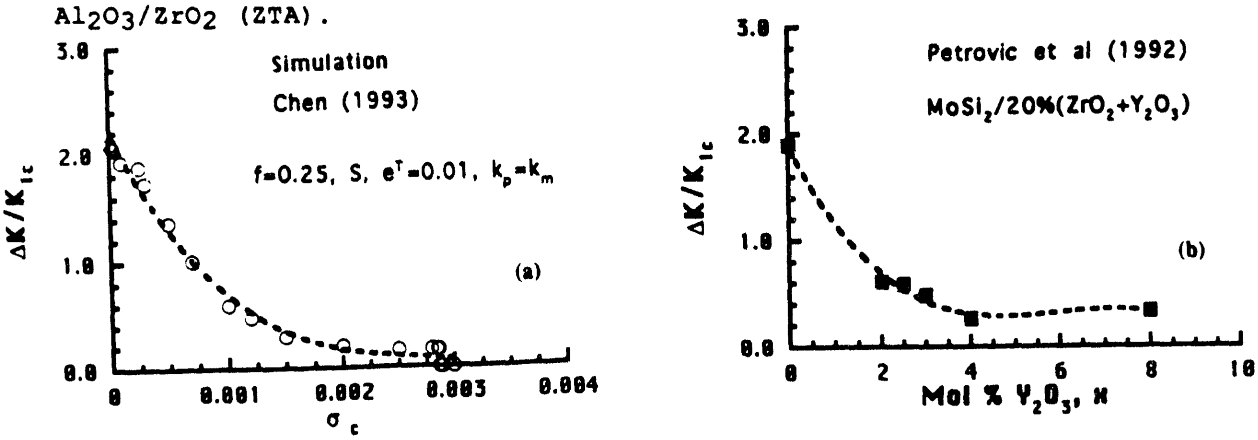


Fig. 3: (a) The ΔK dependence on the critical transformation stress, σ_C. (b) The experimental ΔK of the ZTM as a function of x Mole % of Y_2O_3 stabilizer in ZrO_2 particles. These two curves of simulation and ZTM (both fitted to a 3rd order polynomial) are strikingly similar.

The third way in changing the composite fracture toughness is to modify the critical transformation stress of the particles, σ_c. The change in σ_c can be achieved by the addition of stabilizers like (CaO, MgO, or Y_2O_3) in transforming ZrO_2 particles. The alloying of CaO, MgO, or Y_2O_3 (roughly from 0% to 10 molecular %) in ZrO_2 stabilizes the tetragonal and cubic ZrO_2 and thus increases the critical transformation stress even though an explicit equation relating σ_c to the content, x, of the stabilizer is still lacking [1, 2]. We found the simulation results (Fig. 3a) are in good agreement with experiments of the $MoSi_2/ZrO_2$(+ x% of Y_2O_3) composites (Fig.3b)[7]. The similarity between the measurements and the simulation results are striking. A direct comparison with continuum theory (eq. (2)) reveals some new features. If we exclude the portion that does not transform because σ_c is too high, we found that for different stress levels the ΔK vs. σ_c curve can be broken into two regions that have their own power law dependence:

(a) for large σ_c (> 0.0003), $\Delta K = \beta_1 \sigma_c^{-1.3}$,
(b) for small σ_c (< 0.0003), $\Delta K = \beta_2 \sigma_c^{-0.05} = \sim\beta_2$ (3)

where β_1 and β_2 are constants. The uncertainties in the values of n are about 15%. For high σ_c, the simulation results indicate a less toughening than the results predicted by eq. (2) which means that the continuum is overestimating the transformation toughening effect by treating every transformation event as an independent event.

BEYOND EXISTING EXPERIMENTS & THEORIES

The contributions from different mechanisms involving micro-cracks and transformations are monitored by the stress changes before and after the bonds are broken or the particles are transformed. This separation of toughness changes according to separate mechanisms allowed us to monitor the contributions from two separate source of shielding of the main crack due to micro-cracking and transformations.

In Fig. 4a, we show that for $k_p=k_i=k_m=1.0$, the contribution from the transformation (open circle), the micro-crack (open triangle) and the total fracture toughness (solid circle) as a function of the volume fraction of particles, f. The contributions due to the transformations and micro-cracking are linear in f. Therefore, the total fracture toughness, K'_{tot}, is linear in f too. The above results may be only applicable to the linearly related N_b-N_t situations. For the micro-cracking part, we found that the contribution from the micro-cracking is 100% of the fracture toughness at f=0.0 (pure matrix material) and decreases as f increases. This result indicates that the micro-cracking accompanying the transformation is detrimental to the fracture toughness of the composite when compared with the original fracture toughness matrix. For the composites with the largest f (0.45) studied, the micro-cracking actually has a negative contribution to the fracture toughness due to the extensive cracking. This extensive cracking does not shield the crack at all and actually helps the original crack propagate. This negative contribution from the micro-cracking manifests itself in the composite with 45% particles that the total fracture toughness of the composite is lower than the toughness derived from the transformation toughening alone. We further study the effect of the modulus of the transforming particles on the fracture toughness. The modulus of the particle, E_p, is proportional to the particle's force constant, k_p. Therefore, the ratio of the moduli equals the ratio of corresponding force constants, e. g. $E_p/E_m = k_p/k_m = k_p$. As shown in Fig. 4b, the K' dependence on k_p is almost linear except for

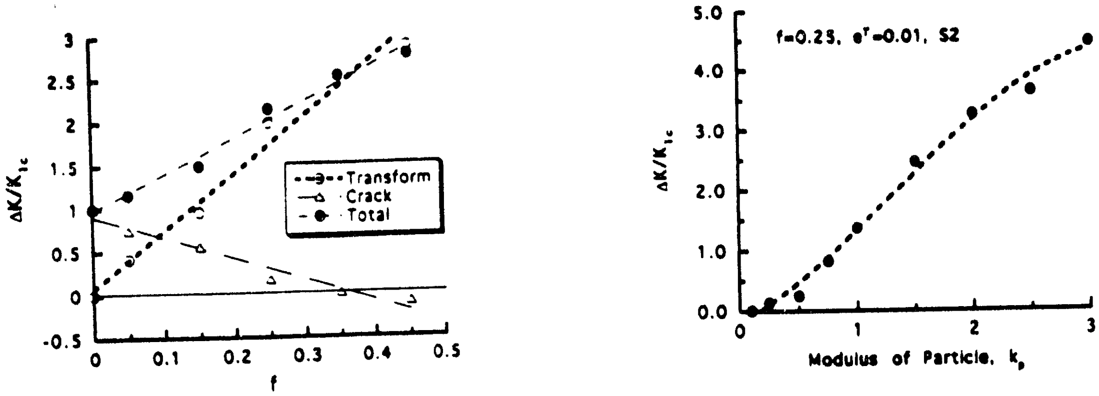


Fig. 4: (a) The K' (transformation, crack, and total) for k_p=1.0 as a function of f. (b) The total fracture toughness K' is plotted as a function of particle force constant, k_p (=E_p/E_m).

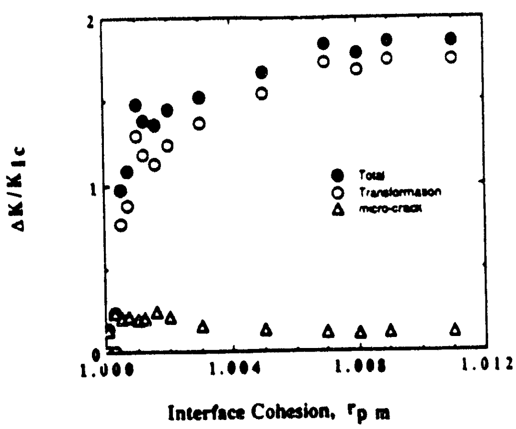
the composite with the largest k_p where the increase in K' starts to saturate. The composite with k_p larger than the matrix has a K' larger than the one with a smaller k_p. This connection of K' with k_p is easy to see by observing that with the same level of e^T, the particle with larger modulus will shrink less after the relaxation. Therefore, the transformed particle puts more compressive stress on the matrix and creates a larger stress reduction or crack shielding. Assuming the rule of mixture applies to the composite modulus, we can rewrite eq. (2) to include explicitly the contribution from the modulus change:

$$\Delta K = \alpha\, Em\, [(1-f) + fE_p/E_m]\, e^T f(\sigma_c)^{-1} / (1-\nu) \quad (4).$$

This formula can be used as a predictive tool to select future tougheners.

In this part, we investigate the influence of the particle-matrix interfacial cohesion by varying the breaking distance of the nodes, r^b_{pm}, on the fracture toughness of the composites with second-phase particles. The results indicate that the stronger the interfacial cohesion, r_{pm}, is the larger the increase of the fracture toughness, ΔK_{1c} (Fig. 5).

Fig. 5 : (a) The ΔK is plotted as a function of the particle-matrix interfacial cohesion, r_{pm}. The contributions due to the transformation are much larger than the contributions derived from micro-cracking.



The results indicate that the transformation toughening is the dominant cause of the increase in ΔK_{1c}, while the micro-cracking only provides less than 10% of the toughening when r_{pm} is relatively large (i.e. r_{pm} > 1.003). For weak interfaces (i.e. when r_{pm} is close to 1.0), none of the particles were transformed because high stresses can not be transfered from the matrix to the particles with cracking of the interfacial bonds and therefore only micro-crackings near the particles are providing the toughening at the level of 0.1 to 0.2 of the original value of K_{1c} (for pure matrix material). The K'_{crack} as a function of N_b (number of broken bonds) has a maximum for intermediate N_b which indicates that micro-cracking contribution to the fracture toughness is maximum for a intermediate particle-matrix cohesion and intermediate micro-cracking situation. Generally speaking, the composite with small r_{pm} has a larger ΔK due to micro-cracking but the total K' is much smaller than the one with large r_{pm}.

It has been shown [4, 5] that with the same critical stress the composites with smaller particles with linear particle to grain size ratio of $R_{p/g}$=0.17 (S1, S2 and S3) is tougher than the composites with large particles with $R_{p/g}$=0.46 (L1, L2 and L3 in Fig. 2a). This result does not take into account of a possible change in the critical transformation stress as a function of the particle size that may existed in the real systems [11, 12]. Therefore, this result indicates clustering of particles (large particles) is detrimental to the fracture resistance of composites.

CONCLUSIONS

The simulation results presented above using Spring-Network (SN) model indicate that the increase in fracture toughness, ΔK is linear in dilatational transformation strain, e^T and volume fraction, f. These results are in good agreement with the available experiments and continuum theory. In particular, we found that $MoSi_2/ZrO_2$ (ZTM) composite is a classic case of transformation toughening dominated material, while Al_2O_3/ZrO_2 (ZTA) involves both transformation and micro-cracking. Detailed analysis of the $\Delta K=\beta\sigma_c^n$ formula shows that at large stresses, the exponent is -1.3 instead of -1.0 predicted by the continuum theory and at small stresses, the exponent is -0.05 which is almost equals 0.0. This calculated flatness of ΔK-σ_c curve at low σ_c is different from the continuum theory (eq. (2)) and the prediction of Evans and Cannon [9]. In addition, we have separated, for the first time, the micro-cracking and the transformation contributions to the fracture toughness by monitoring the changes of stresses induced by the breaking of bonds and the transformed particles. In helping design tougher composites, we found that choosing particles with several characteristics like (1) large dilatational transformation strain (e^T), (2) large volume fraction (f), (3) small transformation stress (σ_c), (4) large particle modulus (E_p), (5) strong particle-matrix cohesion (r_{pm}), and (6) uniform distributions of transformable particles are beneficial in making tougher composites.

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