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Fracture toughness of composite materials reinforced by debondable particulates

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Abstract

In this paper, cleavage cracking across a regular array of debondable particles was discussed through an energy analysis. It was found that the toughening effect of the particles is dominated by a parameter χ. Increasing the particle volume fraction or the particle size has beneficial effects to the overall fracture toughness.

Keywords: Fracture; Toughness; Particulate reinforced composites

1. Introduction

Over decades, it was repeatedly reported that in many particulate reinforced polymer and metal based composite materials debonding along the filler–matrix interface could occur before or at the early stage of the plastic deformation, which had considerable influence on the fracture, viscoelastic, and thermal properties [1–5]. At high temperature, the microvoiding triggered by the relatively weakly bonded particles, e.g. rubber particles in high impact polystyrene (HIPS) [6], could promote the plastic deformation at the crack tip. At low temperature, in intrinsically brittle or quasi-brittle matrix the plastic deformation became difficult while the particles could serve as obstacles to the cleavage crack advance. In engineering practice this transition in fracture mode from ductile to brittle must be predicted accurately. There are two basic mechanisms related to the toughening effect of the particles in brittle matrix. The first one is the geometrically necessary increase of the roughness of the fracture surface. The second one is the crack trapping effect. The retardation of the crack propagation promotes the crack-tip plastic relaxation beneficial to lowering the ductile-to-brittle transition temperature.

Rose suggested a model for brittle materials reinforced by well-bonded particles [7]

$$\widetilde{K} = \sqrt{(1 - 2R/L) + (2R/L)\widetilde{K}_{\mathrm{p}}^2} \tag{1}$$

where R is the particle radius, L is the center-to-center distance of the particles, $\widetilde{K} = K_{\mathrm{C}}/K_{\mathrm{m}}$, and $\widetilde{K}_{\mathrm{p}} = K_{\mathrm{p}}/K_{\mathrm{m}}$, with K_{C}, K_{p} and K_{m} being the fracture toughness of the composite material, the particle, and the matrix, respectively. This problem was further investigated by Bower and Ortiz through the simulation of the profile evolution of the cleavage front penetrating between tough

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obstacles [8,9]. In a crack arrest experiment it was demonstrated that the numerical results fit with the experimental data quite well if R/L was in the range of 0 to about 0.3 [10,11]. In this model it was assumed that the crack front could cut through the particles gradually and the peak resistance was reached when the front advanced to the centers of the particles.

For relatively weakly bonded particles, however, the failure mode is the fracture-type separation along the particle–matrix interface after a certain "preparatory" plastic shear deformation as depicted in Fig. 1a, which results in quite different behavior of the cleavage front. Furthermore, for cleavage cracking in heterogeneous materials size effect was often found to be important since the problem was not scalable with fixed particle size and volume fraction [12]. This crack length dependence was attributed to the effect of the second derivative of strain energy, $\partial^2 U/\partial a^2$, which has been studied intensively in the R-curve analysis. In the models introduced above where the crack front behavior was described by a single parameter K_C the size effect could not be captured.

In order to obtain the optimum properties and cost-performance balance, very often the filler content needs to be maximized. Numerical simulation of the chronology of cleavage front advance in composites with high particle volume fraction is very difficult. In the following discussion, we will show that K_C can be estimated through a relatively simple energy method, in which the calculation of the complicated front profile is avoided and the size effect can be predicted.

2. Toughening effect of weakly bonded particles

When a cleavage crack propagating in a brittle matrix encounters an array of relatively weakly bonded particles, part of the crack front will be trapped locally along the particle boundaries and the rest of the front will penetrate between the particles resulting in the relatively high driving

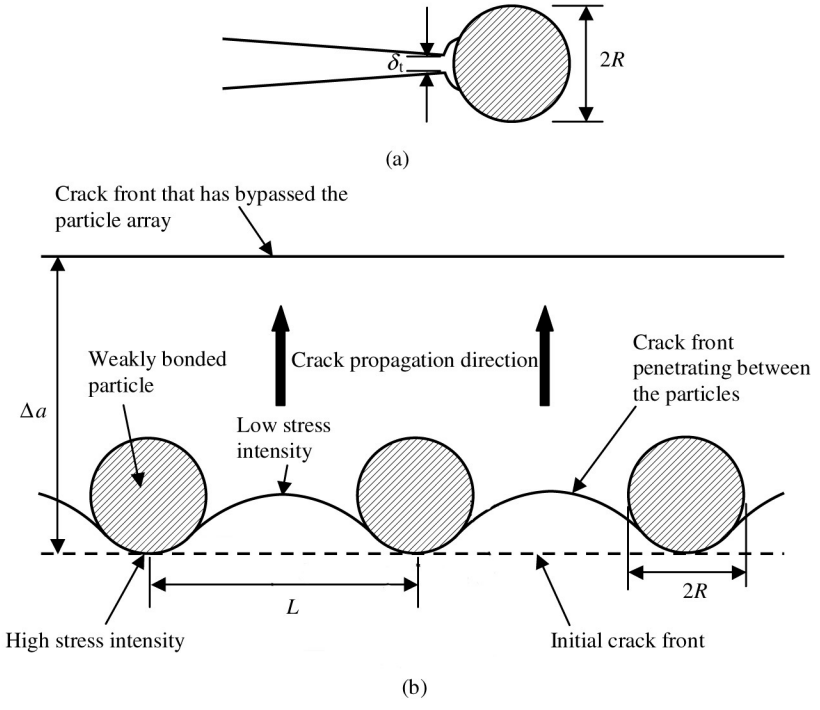

Fig. 1. (a) Fracture-type separation of the weakly bonded particle from the matrix following the "preparatory" plastic shear distance δ_t; (b) the cleavage front bypassing a regular array of particles.

force at the concave part, as depicted in Fig. 1b. When the critical condition is reached, debonding will occur along the particle–matrix interface and the crack front will propagate across the particle array. As shown in Fig. 1a, usually the particles are separated through mixed mode fracture triggered by shear deformation. The shear strain can be stated as

$$\gamma_p = \frac{\delta_t}{2R} = \frac{K_C^2}{2\pi YER} \tag{2}$$

where δ_t is the shear distance, which is taken as the crack tip opening displacement (CTOD), K_C is the critical stress intensity factor for the crack front to overcome the resistance of the particles, E is the Young's modulus, and Y is the yield strength. Through Eq. (2), if we can estimate the range of γ_p the fracture toughness K_C can be obtained. However, direct experimental observation of the separation of the particles from the matrix is difficult and the useful data are rare.

To calculate K_C consider the propagation of a cleavage crack in the displacement controlled test depicted in Fig. 2. Initially the crack tips are trapped by the regular arrays of debondable particles at points "A_1" and "A_2". Since the presence or not of other particles should not have influence on the front behavior, we assume that the material is homogeneous elsewhere. The stress intensity factor caused by the crack opening force per unit thickness, P, is [13]

$$K_I = \frac{P}{\sqrt{\pi a_0}} \tag{3}$$

where a_0 is the initial half length of the crack. Through Eq. (3), the energy release rate can be obtained as

$$G_I = \frac{(1-\nu^2)P^2}{\pi E \tilde{a}_0 L} \tag{4}$$

where ν is the Poisson's ratio and $\tilde{a}_0 = a_0/L$. Since $G = -(1/B)\partial U/\partial a$, we have

$$\frac{U}{B} = \frac{(1-\nu^2)P^2}{\pi E} \ln \tilde{a}_0 \tag{5}$$

where U is the strain energy, B is the sample thickness, and a is the crack length. Note that $U/B = P\delta/2$, with δ being the crack opening displacement. Through Eq. (5), it can be seen that by increasing the crack opening displacement quasi-statically, the wedging force P rises linearly

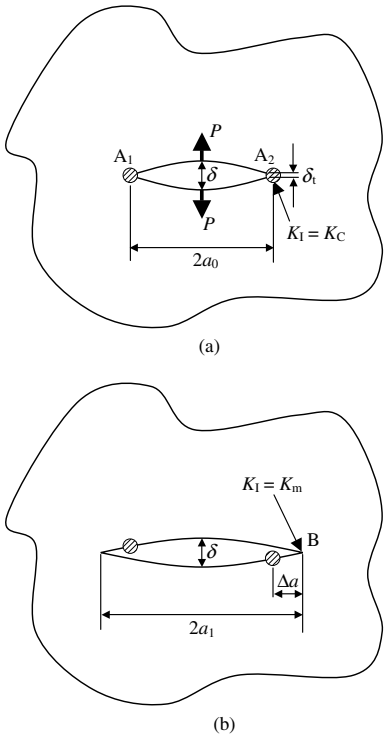

Fig. 2. Cleavage cracking across the regular arrays of particles at points "A_1" and "A_2": (a) before the crack jump; (b) after the crack jump.

$$P = \frac{\pi E \delta}{2(1-\nu^2)\ln \tilde{a}_0} \tag{6}$$

Note that the above discussion is valid only when $\tilde{a}_0 \gg 1$. It will be shown shortly that in this model the range of the crack length under consideration is $\tilde{a}_0 > 10$.

Once the crack tip opening displacement is increased to δ_t, the particle–matrix interface fails and the crack will propagate unstably by a distance Δa, until the stress intensity decreases to the critical value to arrest the propagating crack, K_m. According to the experimental observations of dynamic crack behavior, K_m is about the same as the resistance of a stationary crack [14]. Just before the crack jump, through Eqs. (3) and (6), the stress intensity factor at the crack tip is

$$K_C = \frac{P}{\sqrt{\pi a_0}} = \sqrt{\frac{\pi E^2}{4(1-\nu^2)^2 \tilde{a}_0 L}} \frac{\delta}{\ln \tilde{a}_0} \tag{7}$$

Since the crack jump is dynamic, we assume that the change of the crack opening displacement δ is negligible. When the crack front is stopped at point "B" in the matrix,

$$K_m = \frac{P}{\sqrt{\pi a_1}} = \sqrt{\frac{\pi E^2}{4(1-\nu^2)^2 \tilde{a}_1 L}} \frac{\delta}{\ln \tilde{a}_1} \tag{8}$$

where $a_1 = a_0 + \Delta a$ and $\tilde{a}_1 = a_1/L$. Thus, we have

$$\widetilde{K} = \sqrt{1 + \frac{\Delta \tilde{a}}{\tilde{a}_0}} \frac{\ln \tilde{a}_1}{\ln \tilde{a}_0} \tag{9}$$

where $\widetilde{K} = K_C/K_m$ and $\Delta \tilde{a} = \Delta a/L$.

Substitution of Eqs. (4) and (6) into (5) gives

$$\frac{U}{B} = \frac{\pi E \delta^2}{4(1-\nu^2)\ln \tilde{a}} = G_I L \cdot \tilde{a} \ln \tilde{a} \tag{10}$$

where $\tilde{a} = a/L$ is the normalized crack length. The strain energy change ΔU associated with the crack jump can be stated as

$$\begin{aligned} \Delta U &= U_2 - U_1 \\ &= G_C L \cdot \tilde{a}_0 \ln \tilde{a}_0 - G_m L \cdot \tilde{a}_1 \ln \tilde{a}_1 \end{aligned} \tag{11}$$

where U_1 and U_2 are the values of the strain energy before and after the crack jump, respectively, which were calculated through Eq. (10); G_C and G_m are the critical energy release rates of the composite material and the matrix, respectively.

The energy dissipated in the crack jump process is

$$W = G_m L \cdot \Delta \tilde{a} + \chi_0 (2\pi R^2)/L \tag{12}$$

where $\chi_0 = \chi_{eff} - G_m$, with χ_{eff} being the effective work of separation of the particle–matrix interface. Here we only study the case where $\chi_0 \geqslant 0$. The first term at RHS of Eq. (12) reflects the work to separate the primary fracture surfaces in the matrix; and the second term reflects the contribution of the debonding of the particles from the matrix. Since ΔU and W should be equal to each other, we have

$$\widetilde{K}^2 = \frac{(\Delta \tilde{a} + \tilde{a}_0 \chi) \cdot \ln(\tilde{a}_0 + \Delta \tilde{a})}{\tilde{a}_0 \ln \tilde{a}_0 \cdot [\ln(\tilde{a}_0 + \Delta \tilde{a}) - \ln \tilde{a}_0]} \tag{13}$$

where $\chi = 2\pi(\chi_0/G_m)(R/a_0)(R/L)$. The combination of Eqs. (9) and (13) gives Δa and K_C. Since the only independent variables in the two equations are the parameter χ and the initial crack length a_0, K_C must be function of them.

3. Discussion

Fig. 3 shows that the overall fracture toughness, which is taken as the critical stress intensity factor to overcome the regular array of the particles, K_C, rises rapidly with χ and decreases slightly with increasing a_0. When χ is 0, $\widetilde{K}$ tends to 1, i.e. the toughening effect of the particles vanishes, as it should. In the range of $a_0/L > 10$ where the assumption of the regular particle array along the crack front is reasonable, the numerical solution can be regressed as

$$\begin{aligned} \widetilde{K} &= \frac{K_C}{K_m} \\ &= 1 + 1.04 \cdot \chi^{0.49} \cdot (0.27\bar{a}^2 - 0.43\bar{a} + 2.10) \end{aligned} \tag{14}$$

where $\bar{a} = \tilde{a}_0/100$. Through Eq. (2), if the shear strength of the particle–matrix interface is estimated as $k = \mu\gamma_p$, with μ being the shear modulus, for the first order approximation the effective work of separation associated with the debonding can be calculated as $\chi_0 = (2R)k = \mu K_C^2/\pi YE$. This relationship indicates that $\widetilde{K} \propto \chi^{1/2}$, which is quite compatible with Eq. (14).

If we assume that the crack advance is uniform along the front, the fracture toughness can be

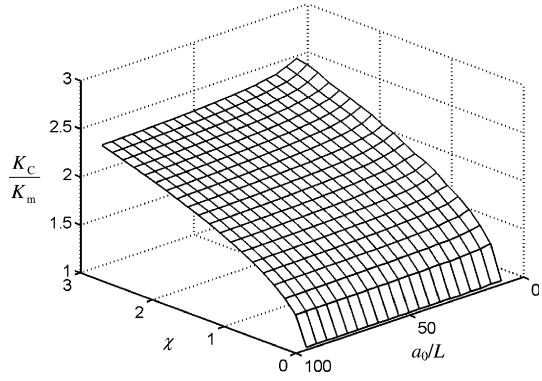

Fig. 3. The effects of χ and a_0 on the fracture toughness K_C.

simply estimated through the area average of the fracture work

$$\frac{1-\nu^2}{E}K_{\mathrm{C}}^{*2}=\frac{\chi_0(2\pi R^2)/L+G_{\mathrm{m}}\Delta a}{\Delta a} \tag{15}$$

It can be seen that K_{C}^{*} is considerably lower than K_{C} when χ is larger than 0.1. The difference between them is caused by the crack trapping effect.

If the crack length is comparable to the characteristic microstructure length, e.g. when $a_0/L<10$, the assumption of the front behavior depicted in Fig. 1b is not acceptable [8,9], thus the above discussion is no longer valid. In the range of $a_0/L>10$, according to Eq. (14), the fracture toughness of the composite material reinforced by debondable particles is not a material constant. The toughening effect is more pronounced for shorter cracks, which is consistent with the fact that the cleavage crack is not scalable when the crack length varies while the particle size is constant. At the front of a longer crack, the particle size appears to be "smaller", leading to the lower resistance to cleavage cracking. The factor of the crack length also comes in by affecting the rate of change of the energy release rate, which is reflected by both of the factor $\chi^{0.49}$ and the term in the bracket at RHS of Eq. (14). This effect is quite similar to the crack length dependence in the R-curve analysis, where the second derivative of the strain energy plays an important role in determining the critical condition of the unstable crack advance.

According to the definition of χ, the larger the effective work of separation of the particle–matrix interface, the higher the fracture toughness. Since $\widetilde{K}\propto\chi^{1/2}$, K_{C} should be proportional to $\chi_0^{1/2}$, or, equivalently, $G_{\mathrm{C}}\propto\chi_0$, i.e. the additional fracture work associated with the crack trapping effect is proportional to the effective particle strength. Note that when χ_0 is larger than a certain critical value such that the particle can bridge across the crack flanks even after the crack front bypasses them, the particles can no longer be considered as weakly bonded. Under this condition the problem must be analyzed through the detailed simulation of the evolution of the crack front profile [8–11].

As shown in Fig. 3, if the crack length is much larger than L, the crack length dependence becomes negligible. By substituting the definition of χ into Eq. (14) and taking a_0 as $100L$, the numerical result of the toughening effect for long cracks can be simplified as

$$\widetilde{K}_0=1+0.5\cdot\left(\frac{\chi_0}{G_{\mathrm{m}}}\right)^{0.5}\frac{R}{L} \tag{16}$$

where $\widetilde{K}_0$ is the value of $K_{\mathrm{C}}/K_{\mathrm{m}}$ for long cracks. It can be seen that for long cracks the fracture toughness is linear to the size/spacing ratio of the particles, R/L. For spherical particles, the particle volume fraction can be calculated as $c=4\pi R^3/L^3$. Consequently, $\widetilde{K}_0\propto c^{1/3}$, which, as shown in Fig. 4, fit with the experimental data quite well. The experimental data in Fig. 4 were obtained in a notched Izod fracture test of the polypropylene reinforced by the debondable calcium carbonate particles [5]. The value of $K_{\mathrm{C}}/K_{\mathrm{m}}$ was taken as the square root of the normalized fracture work. For the best fit of the data, the value of χ_0/G_{m} was taken as 1.05 and, quite acceptable.

On the other hand, for relatively short cracks, according to Eq. (13), $\widetilde{K}\propto c^{1/6}$, indicating that the beneficial effect of adding more particles is more significant when the crack length is small. According to Eq. (14) and the definition of χ, with constant a_0 and L, the overall toughness rises with the particle size R, which should be attributed to the fact that with larger particle size the total separation work of the particles becomes larger, which promotes the toughening effect.

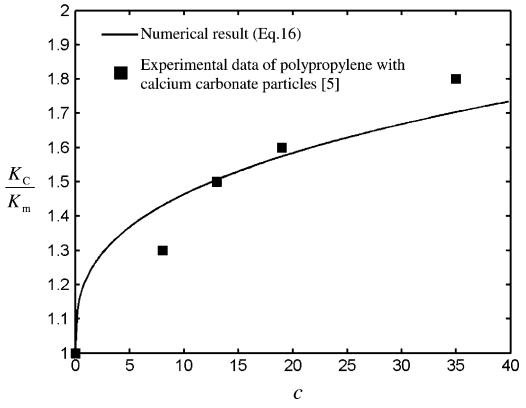

Fig. 4. The fracture toughness, K_{C}, as function of the particle volume fraction, c.

4. Conclusions

In this paper, the toughening effect of debondable particles in brittle matrix was discussed through an energy method. Both of the increasing of the fracture surface roughness and the crack trapping effect were accounted for and the following conclusions were drawn:

(1) The crack trapping effect of the relatively weakly bonded particles is significant, and is dominated by a single parameter χ collecting together the effects of the strength of the particle–matrix interface, the particle size, and the particle volume fraction.
(2) Due to the non-self-similar nature of the crack advance the value of K_C decreases with increasing crack length. However, for long cracks this size effect is negligible.
(3) For relatively short cracks, the overall fracture toughness is proportional to $c^{1/6}$ with c being the particle volume fraction. For cracks much longer than L, $\widetilde{K} \propto c^{1/3}$.
(4) Increasing particles size has a beneficial effect on K_C. With the same particle content, the toughening effect of coarse particles is more profound than that of fine particles.

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