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Effects of particle size and matrix grain size and volume fraction of particles on the toughening of ceramic composite by thermal residual stress

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Abstract

Toughening by thermal residual stresses is one of the effective toughening mechanisms in ceramic composites when the coefficient of thermal expansion of the dispersed particles is larger than that of the matrix grains. Based on this toughening mechanism, exploration of factors affecting the toughening of ceramic composite by thermal residual stress is the focus of the investigation in the present study. Results are verified for the TiB_2/SiC ceramic composite. It is shown that particle size and matrix grain size and volume fraction of particles and the particle size ratio is closely related to the toughening increment. It suggests that the investigation will lead practically to actual applications of the findings in the composition design and the microstructural design of the particulate reinforced ceramic composites.

Keywords: B. Composites; C. Toughness and toughening; Thermal residual stress

1. Instruction

The intrinsic brittleness is a key problem in the widespread application of the ceramic materials. In order to reduce the brittleness and to increase the strength and the toughness, varieties of toughening mechanisms [1–6], such as the crack deflection, crack bridging, crack branching, crack bowing, crack pinning, microcracking, thermal residual stress toughening, transformation toughening and the synergism toughening, etc. have been proposed in the past decades. Furthermore, methods of the ceramic material design such as the interfacial stress design have been adopted to help solve the problem [7]. As a result, the mechanical properties and performances and the reliability of the composite ceramic materials have been significantly improved. However, the problem remains unscanned and the scope of the current researches still remains to be widened and deepened to achieve viable results in the selection and design of reinforcing phases and toughening mechanisms.

When the coefficient of thermal expansion of the dispersed phase is larger than that of the matrix material, thermal residual stress resulted from the mismatch between the coefficients of thermal expansion (CTEs) of the ceramic matrix and particulates will become the dominant toughening mechanism in ceramic materials. At first, Wei and Becher [8] considered the thermal residual stress which is induced by CTE mismatch as a major cause for the crack deflection although they have made no quantitative estimate of the increased toughness. Virkar et al. and Evans et al. [9–11] explained the toughening mechanism in ZrO_2–Zr composite material and cemented carbides with the periodic thermal residual stress model. Then, Taya et al. [12] studied the toughening mechanism of the particulate reinforced TiB_2/SiC ceramic material. They pointed out that the toughening effect of thermal residual stress is approximately three times higher than that of crack deflection. Results predicted from the analytical model based on the compressive thermal stress in the matrix in particulate

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reinforced ceramic matrix composite are in better agreement with the experiments.

On the other hand, a maximum volume fraction of the dispersed phase exists just because of the thermal residual stress [13,14]. When the content exceeds the maximum value, the material will fracture without any external load resulted from the overload in the tensile thermal stress and then mechanical properties of the materials will undoubtedly decrease. According to this, two models have been built for the determination of the maximum volume fraction of the dispersed phase in ceramic composites when the coefficient of thermal expansion of the dispersed phase is less than that of the matrix. These models are used well in the development of the Al_2O_3/TiB_2 [13] and Al_2O_3/SiC/(W, Ti)C [14] ceramics. Therefore, the determination of the maximum volume fraction of the dispersed phase is one of the significant parts in the composition design of the composite ceramic materials.

In the present study, effects of particle size and matrix grain size and volume fraction of particles and the particle size ratio on the toughening of ceramic composite are analyzed in detail with an example of TiB_2/SiC ceramic material based on the toughening mechanism of thermal residual stress.

2. Relationship between the toughening effect and volume fraction and particle size

2.1. Theoretical basis

According to Taya et al. [12], the thermal residual stress field in a TiB_2/SiC composite consists of two regions: the tensile stress region in the particulates and their vicinity in the matrix, and the compressive stress region in the bulk of the matrix, as shown schematically in Fig. 1. Consider a semi-infinite crack surrounded by a particulate reinforced ceramic matrix composite with a thermal residual stress distribution as shown in Fig. 1. For the purpose of fracture mechanics analysis, this residual compressive stress can be approximated by an averaged uniform compressive stress, acting normal to the crack plane ahead of a semi-infinite crack. The semi-infinite crack is assumed to advance from one concentrated tensile region to its nearest counterpart. Since the existence of the periodic thermal residual stress field in the particulate reinforced ceramic composite, the fracture toughness K_{IC} of the material is expressed as [11,12]:

$$K_{IC} = K_{I0} + 2q\sqrt{\frac{2D}{\pi}} \tag{1}$$

where K_{I0} is the critical stress intensity factor of the matrix, q is the local thermal residual compressive stress, D is the length of the compressive stress zone which in this case is the average particulate spacing. If the dispersed phase of the same grain size is distributed uniformly in the matrix, then $D = \lambda - d$, where the average distance between the particles of the dispersed phase is λ.

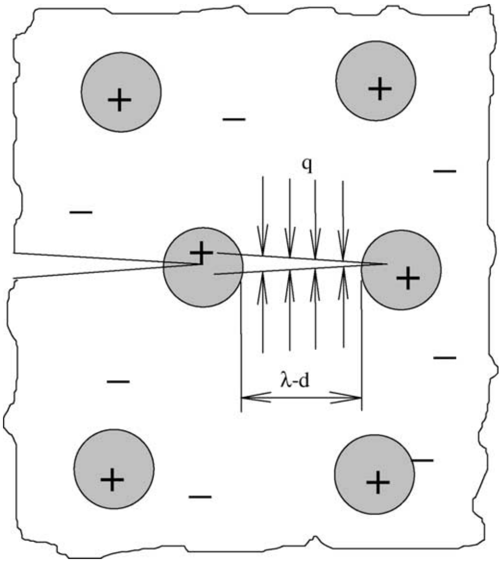

Fig. 1. Analytical model of the toughening mechanism by thermal residual stress [12].

Eq. (1) is based on a stress intensity factor solution by Tada et al. [15] for a semi-infinite two-dimensional crack with a compressive stress zone of intensity q and length D. The compressive thermal residual stress in the matrix is generated when the CTE of the particulate exceeds that of the matrix. Indeed this is the case with the TiB_2-particulate/SiC-matrix [16–18] and TiC-particulate/SiC-matrix composites [17]. Both Cutler and Virkar and Chermant and Osterstock experimentally observed the $\sqrt{D}$ dependence of K_{IC} [10,19]. It was also found [10] that q is a strong function of the volume fraction of the second phase and larger values of q resulted in larger K_{IC}.

The existence of the local stress, q, for TiB_2/SiC composite example, decreases the stress intensity factor, ΔK_I. From Eq. (1) this decrease is

$$\Delta K_I = K_{IC} - K_{I0} = 2q\sqrt{\frac{2(\lambda - d)}{\pi}} \tag{2}$$

where [12]

$$\lambda = \frac{1.085d}{V_P^{1/2}} \tag{3}$$

$$q = \langle\sigma_m\rangle = \frac{2E_m V_P \beta \alpha_1^*}{A} \tag{4}$$

$$A = (1 - V_P)(\beta + 2)(1 + \nu_m) + 3\beta V_P(1 - \nu_m) \tag{5}$$

where $\langle\sigma_m\rangle$ is the average stress field in the matrix, V_P means the volume fraction of the dispersed phase, α_1^* indicates the strain inside the particulate resulted from the difference in the thermal expansion, $\alpha_1^* = \int_{T_P}^{T_R}(\alpha_P - \alpha_m)\,dT$, where T_R means the room temperature, T_P is the temperature where the plastic deformation of the matrix can be neglected. $\beta = (1 + \nu_{\mathrm{m}}/1 - 2\nu_{\mathrm{P}})(E_{\mathrm{P}}/E_{\mathrm{m}})$, E,ν expresses the elastic modulus and the Poisson's ratio, respectively, while the subscript P and m indicates the dispersed phase and the matrix, respectively.

According to Eq. (2), if $q < 0$, i.e., $\Delta K_{\mathrm{I}} < 0$, which means that the stress intensity factor is decreased by ΔK_{I} as a result of the action of the thermal residual stress. However, the decrease in the stress intensity factor by ΔK_{I} is equivalent to the increase in the crack growth resistance by the same amount ΔK_{I}, which will lead to the toughening of the material. On the contrary, the fracture toughness will be decreased. Let the increase in the fracture toughness be $\Delta K(\Delta K > 0)$, then the toughening condition will be:

$$\Delta K_{\mathrm{IC}} \geq \Delta K \tag{6}$$

i.e.,

$$-2q\sqrt{\frac{2(\lambda - d)}{\pi}} \geq \Delta K \tag{7}$$

Taking Eqs. (3)–(5) into account, and let

$$B = 4E_{\mathrm{m}}\beta\alpha_1^*\left(\frac{2d}{\pi}\right)^{1/2},$$

$$F = -(\beta + 2)(1 + \nu_{\mathrm{m}}) + 3\beta(1 - \nu_{\mathrm{m}}),$$

$$C = (\beta + 2)(1 + \nu_{\mathrm{m}}) \tag{8}$$

Then, Eq. (7) can be expressed as an unequality of V_{P} by means of the two order polynomial expansion, i.e.,

$$\left(\frac{0.593B^2}{(\Delta K)^2} + F^2\right)V_{\mathrm{P}}^2 + \left(2FC - \frac{0.814B^2}{(\Delta K)^2}\right)V_{\mathrm{P}} + C^2 + \frac{1.085B^2}{8(\Delta K)^2} \geq 0 \tag{9}$$

If the following condition is met,

$$\left(2FC - \frac{0.814B^2}{(\Delta K)^2}\right)^2 - 4\left(\frac{0.593B^2}{(\Delta K)^2} + F^2\right) \times \left(C^2 + \frac{1.085B^2}{8(\Delta K)^2}\right) \geq 0 \tag{10}$$

then Eq. (9) will have the solution

$$V_{\mathrm{P}} \leq V_{\mathrm{P1}}, \quad V_{\mathrm{P}} \geq V_{\mathrm{P2}} \quad (V_{\mathrm{P1}} < V_{\mathrm{P2}}) \tag{11}$$

where

$$V_{\mathrm{P1,2}} = \frac{((0.814B^2/(\Delta K)^2) - 2FC) \pm [(2FC - (0.814B^2/(\Delta K)^2))^2 - 4((0.593B^2/(\Delta K)^2) + F^2) \times (C^2 + (1.085B^2/8(\Delta K)^2))]^{1/2}}{2((0.593B^2/(\Delta K)^2) + F^2)} \tag{12}$$

After detailed calculation, it is found that one of the solutions V_{P2} is always larger than 1.0. So, solution V_{P2} can be omitted since the case is not in accordance with the fact that the volume fraction can never be larger than 100%. For the ceramic composite, reasonably let

$$V_{\mathrm{Pmax}} = V_{\mathrm{P1}} \tag{13}$$

where V_{Pmax} is the maximum volume fraction of the dispersed particles.

It must be pointed out that the above deduction is actually an ideal case since the effect of the porosity is not considered. As a matter of fact, the measured density is usually very close to its theoretical value for the ceramic composites fabricated by the hot pressing technique or hot iso-static pressing technique. The porosity of the material is so small, usually less than 2% [12], that it is a reasonable approximation.

If the porosity V_{V} is taken into account, Eq. (5) will become [12]:

$$A = (1 - V_{\mathrm{V}}) \times \left[\left(1 - \frac{V_{\mathrm{P}}}{1 - V_{\mathrm{V}}}\right)(\beta + 2)(1 + \nu_{\mathrm{m}}) + \frac{3\beta V_{\mathrm{P}}(1 - \nu_{\mathrm{m}})}{1 - V_{\mathrm{V}}}\right] \tag{14}$$

The solution of the maximum volume fraction V_{Pmax} can be kept unchanged in form if the parameter C in Eq. (8) is changed into $C = (1 - V_{\mathrm{V}})(\beta + 2)(1 + \nu_{\mathrm{m}})$.

2.2. Results and discussions

According to the above theoretical analyses, corresponding calculations are carried out with an example of $\mathrm{TiB_2}$/SiC ceramic composite where SiC is the matrix and $\mathrm{TiB_2}$ acts as the dispersed phase. In order to make the comparison, results are shown in Fig. 2 with the data of the material properties coming from the literature [12] which are shown in Table 1 with λ = 11.1 μm, $\alpha_1^* = -5.15 \times 10^{-3}$. It seems that the maximum volume fraction of the dispersed $\mathrm{TiB_2}$ is closely related with the increase in the fracture toughness and the particle size. The larger the increase in the fracture toughness is, the higher the content of the dispersed phase is. But the larger particle size of the dispersed phase will result in the lower maximum volume fraction with the same toughening increment. But when the particle size is larger than a certain value that is concerned with the toughening increment, the maximum volume fraction will be approximately a constant.In Taya et al. [12] research, the measured particle size of $\mathrm{TiB_2}$ is 3.06 μm and the volume content is 16%, the corresponding toughening increment is 1.56 MPam$^{1/2}$; while according to the prediction, the maximum volume fraction of $\mathrm{TiB_2}$ is about 34%. Thus, it suggests that the adopted volume content of $\mathrm{TiB_2}$ in Taya et al. experiment is suitable as it falls in the scope of the predicted maximum volume faction (Fig. 3).

When the particle size of $\mathrm{TiB_2}$ is increased from 2 to 10 μm, the predicted maximum increment of the fracture toughness is also enhanced from 1.6 to 3.6 MPam$^{1/2}$ (Fig. 3).

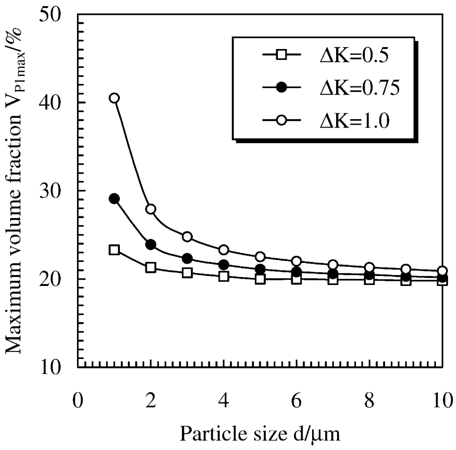

Fig. 2. Maximum volume fraction of TiB_2 in TiB_2/SiC ceramic composite.

It denotes, on the one hand, that a maximum value of the toughening effect of the thermal residual stress exists as well as the maximum content of the dispersed phase. On the other hand, the increase in the particle size, to some extent, is beneficial for the improvement of the fracture toughness of the material. However, when the toughening increment is relatively small, the particle size has little effect on the content of the dispersed phase. At the same time, it has to be noticed that the larger particle size will result in the decrease of the strength of the ceramic material. In fact, the particle size must be smaller than its maximum value. Otherwise, spontaneous microcracking will happen in the material.

As a matter of fact, the calculation of the maximum volume fraction of the dispersed particles is in close relationship with the particle size. Hence, not only the volume content but the particle size can be determined according to the result of the theoretical calculation, which will become the basis for the selection and determination of the microstructural parameters and then for the microstructural and compositional design of the ceramic composite.

3. Relationship between the toughening effect and the matrix grain size

It is well known that the particle size and the content of the dispersed phase are the two significant factors affecting the thermal residual stress in ceramic matrix composite. However, effects of the matrix grain size on the thermal residual stress and then on the fracture toughness have seldom been reported which should actually not be neglected. Since thermal residual stress will eventually be distributed at each interfaces or grain boundaries, it results in the interface stress in the particulate reinforced ceramic composites. If the processing technology is well controlled to decrease the matrix grain size, the quantity of the interfaces will be increased correspondingly and the thermal residual stress distributed at each interfaces will not exceed the maximum value. Moreover, with the full utilization of these interface stresses, the extending cracks can be inhibited, deflected or pinned and, thus, this process will lead to the toughening of the material.

Table 1
Material properties of SiC and TiB_2 [12]

Property	SiC	TiB_2
Young's modulus (GPa)	410	531
Poisson's ratio	0.19	0.28
Average size (μm)	Grain size 3	3.06
Volume fraction (%)	84	16
CTE ($\times 10^{-6}$ per °C)	4.02	4.6
Melting point (°C)	2540	2790

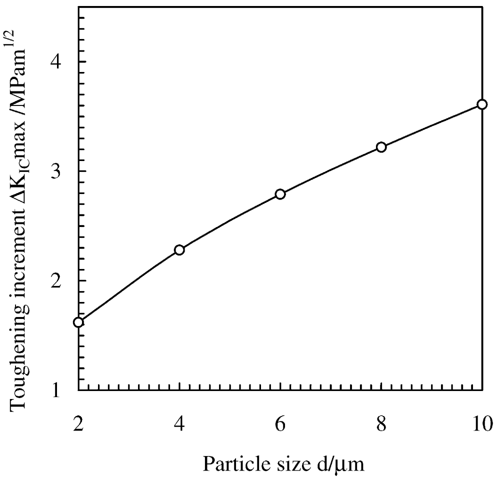

Fig. 3. Maximum toughening increment for different particle sizes (ΔK = 2 MPam$^{1/2}$).

When the grain size of matrix is taken into account, ΔK_I will be modified into:

$$\Delta K_I^* = \Delta K_I f(r,d) \tag{15}$$

where [20] $f(r,d)$ is

$$f(r,d) = 1 + \left(\frac{h}{r/d + e}\right)^n \tag{16}$$

where r and d is the grain size of the matrix and the dispersed phase, respectively; h, e and n are the constants related with the particulate characteristic of both matrix and dispersed phase. Therefore, during the deducing process of the volume content of the dispersed phase, if ΔK in Eq. (12) is substituted with $\Delta K^* = \Delta K f(r,d)$, the expression for the description of the maximum volume faction of the dispersed phase will be achieved when the grain size of the matrix is considered.

Variations of the maximum volume fraction with the sizes of both the matrix grains and the dispersed particles of TiB_2/SiC ceramic composite are illustrated in Figs. 4 and 5 where h = 0.55, e = 0.15, n = 0.7, [20,21]. It is noted that the grain size of the matrix SiC has little effect on the maximum volume fraction of the dispersed

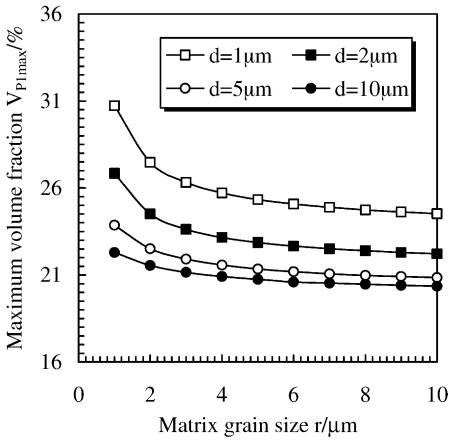

Fig. 4. Variation of the maximum volume fraction with the sizes of the matrix grain and the dispersed particle (ΔK = 0.5 MPam$^{1/2}$).

phase TiB_2 when the toughening increment is small. With the increase in the toughening increment, the effect is becoming more obvious. Generally, the maximum volume fraction of the dispersed phase decreases with the increase in the grain size of the matrix. It must be noticed, however, that the physical meaning of the parameters *h*, *e* and *n* still remains ambiguous which needs further investigation.

4. Relationship between the toughening effect and the particle size ratio

It is well known that the smaller the size ratio between the dispersed particles and the matrix grains is, the more uniformly the dispersed particulates will distribute. Conversely, it is more difficult to be well distributed. Jin et al. [22] have ever studied the size effect of the reinforcement phase in composite ceramic materials. They pointed out that the smaller size ratio between the dispersed particles and the matrix grains will result in the higher strength of the material. Comprehensive effects of other factors must be considered, however, during the practical fabrication of the ceramic materials. Not only the strengthening effects but also the toughening effects have to be taken into account in order to fulfill the aim of the material design with good comprehensive mechanical properties.

Let $l = d/r$ where l is the size ratio between the dispersed particles and the matrix grains, then Eq. (16) becomes:

$$f(l) = 1 + \left(\frac{h}{1/l + e}\right)^n \tag{17}$$

In the same way, if ΔK in Eq. (12) is substituted with $\Delta K^* = \Delta K f(l)$, the equation of the maximum volume fraction will be achieved with the consideration of the particle size ratio.

Results are shown in Fig. 6 with the example of TiB_2/SiC ceramic material. It seems that with the increase in the toughening increment effect of the particle size ratio on the maximum volume fraction is becoming more obvious. The larger the particle size of the dispersed phase is, the larger particle size ratio will accordingly lower the required maximum volume content of the dispersed phase with the same toughening increment. The case is noticeably different from the point of view of the strengthening. Moreover, according to the analysis given above, the larger particle size of the dispersed phase will undoubtedly lead to the higher toughening increment which is the conclusion drawn from the viewpoint of the toughening of the ceramic materials. Therefore, both toughening and strengthening are needed to be considered comprehensively in the selection and determination of the volume content of the dispersed phase, the sizes of both matrix grain and dispersed particle, and the size ratio between the dispersed particle and the matrix grain, etc. Only in this way can the requirement of the ceramic material design be fulfilled.

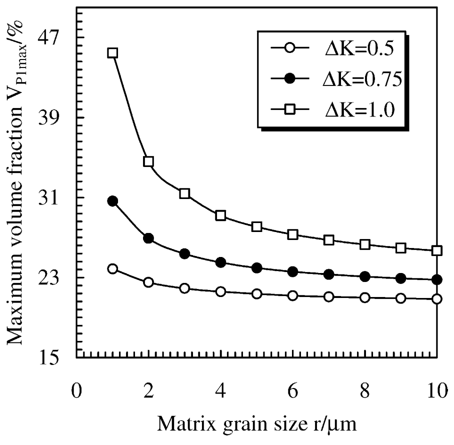

Fig. 5. Variation of the maximum volume fraction with the matrix grain size under different toughening increments (d = 5 μm).

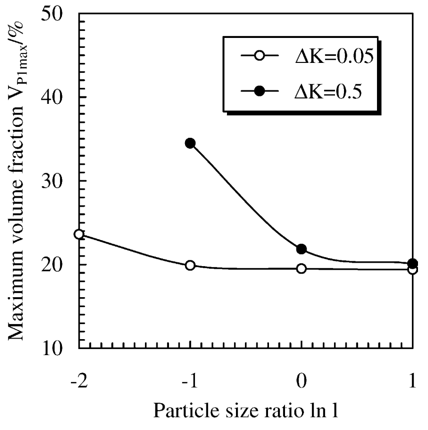

Fig. 6. Variation of the maximum volume fraction with the particle size ratio.

5. Conclusions

Based on the toughening mechanism of thermal residual stress, relationships between the toughening effects and the volume fraction of the dispersed phase, the sizes of both the matrix grain and the dispersed particle, and the size ratio between the dispersed particle and the matrix grain in particulate reinforced ceramic composite have been analyzed and discussed theoretically. From the point of view of the strengthening of the ceramic material, the smaller particle size of the dispersed phase and the smaller particle size ratio will be the better. But from the viewpoint of toughening, the larger particle size of the dispersed phase and the larger particle size ratio will result in the higher fracture toughness. Hence, both toughening and strengthening effects need to be considered in the selection and determination of the material parameters in order to get good comprehensive properties for the particulate reinforced ceramic composites.

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