The effect of intragranular microstress in Al_2O_3–SiC nanocomposites*

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A theoretical model is established to investigate the intragranular particle residual stress in Al_2O_3–SiC nanocomposites. Using this model, we calculate the average compressive stress on the Al_2O_3 grain boundary (GB) and the average tensile stress within Al_2O_3 grains caused by SiC nanoparticles. The normal compressive stress strengthens the GB, and the average tensile stress weakens the grains. The model gives a reasonable interpretation of the strength changes of Al_2O_3–SiC nanocomposites with the number of SiC particles.

Keywords: nanocomposite, grain boundary strengthening, internal stress, weakening

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1. Introduction

Nanocomposite materials have attracted much attention due to their unique electronic, thermal, optical, magnetic, and mechanical properties, etc.[1−22] Ceramic nanocomposites show a much higher strength and/or toughness than matrices themselves.[7−20] The nanocomposites are divided into three types: intra-type, inter-type, and nano/nanotype.[7] The intragranular and intergranular nanocomposites indicate that the nanoscale particles are dispersed mainly within the matrix grains and at the grain boundaries (GBs) of the matrix, respectively, while the nano/nano-type composites are composed of the dispersoids and the matrix grains, which are both of nanometer size. Among ceramic nanocomposite systems, Al_2O_3–SiC has been researched most extensively.[8−25] The intragranular second phase SiC particles can significantly strengthen the nanocomposites. The fracture strength increases with the increase in SiC content until a maximum value is reached at about 5 vol% of SiC particles, and the strength then begins to decrease as the SiC content increases further.[20−22] Meanwhile, the fracture mode of the nanocomposites evolves gradually from intergranular to transgranular mode.[20,25]

Various mechanisms have been proposed to explain the behaviour.[8−25] The strength improvement was ascribed to the reduction in processing defect size, matrix grain refinement or subgrain formation, particle-bridging, and dislocations or the dislocation network and the residual surface compressive stresses induced during the machining process.[8−18] However, none of these mechanisms has been widely accepted.[21] Besides the above mechanisms, several researchers have claimed that the compressive stresses on GBs induced by the mismatch between the coefficients of thermal expansion (CTEs) of the matrix and particles could elucidate the strengthening mechanism in Al_2O_3–SiC nanocomposites.[19−21] Furthermore, qualitative or quantitative analyses of the normal compressive stresses on GBs have been tried by several research groups, modifying the inconsistent and, maybe, plausible formula calculus.[20,21]

On the other hand, weakening mechanisms at higher particle contents have also been put forward.[23,24] It is proposed that the average internal stress existing within the grain may weaken the strength of the matrix.[23] The particle agglomerates can produce crack-like voids which result in strength-degrading.[24] The weakening by the internal stress or particle agglomerates has been analysed only concep-

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tually.

As is well known, many factors influence the measured fracture strength, and the corresponding mechanisms may be interweaved with each other. It is very difficult, sometimes even impossible, to separate out the contribution from each factor involved. Theoretical estimation of the possible intensity of the effect of a particular factor on the strength is, however, significant for the interpretation of a mechanism and the design of a better structure. As for the Al_2O_3–SiC nanocomposite, the intragranular particle and the mismatch between the CTEs of the matrix and particles are two features raising much discussion,[19–21] and their relation to the SiC content dependence of fracture behaviour may be an ingredient in the understanding of strengthening in the Al_2O_3–SiC nanocomposite.

In the present study, we present a theoretical model to estimate the internal stress caused by the mismatch between the CTEs of the matrix and the intragranular particles in Al_2O_3–SiC nanocomposites. We try to elucidate whether the normal compressive stresses on GBs and the average tensile stresses within grains will make a critical difference in strengthening and weakening Al_2O_3–SiC nanocomposites, respectively. The normal compressive stresses on GBs are calculated, using standard tensor manipulation in Section 2, and the average tensile stresses within grains are computed in Section 3; their relevance to the fracture behaviour is then discussed.

2. Strengthening mechanism

The residual stresses on GBs are derived from the mismatch of the CTEs between the particles and the matrix. A hydrostatic stress at the surface of the particle developed as a result of the thermoelastic difference is given by[26]

$$p = (\alpha_{\rm p} - \alpha_{\rm m})(T_{\rm p} - T_{\rm R})\left(\frac{1+v_{\rm m}}{2E_{\rm m}} + \frac{1-2v_{\rm p}}{E_{\rm p}}\right)^{-1}, \quad (1)$$

where α, E, v are the CTE, Young's modulus, and Poisson's ratio, respectively; the subscripts m and p denote the matrix and the particle, respectively; and $T_{\rm R}$ and $T_{\rm p}$ are the room temperature (~ 20 °C) and the composite processing temperature, respectively.

The hydrostatic pressure will cause a radial stress $\sigma_{\rm r}$ and a tangential stress $\sigma_{\rm t}$ in the matrix:[27]

$$\sigma_{\rm r} = -p\left(\frac{r}{R}\right)^3, \quad (2)$$

$$\sigma_{\rm t} = \frac{1}{2}p\left(\frac{r}{R}\right)^3, \quad (3)$$

where r is the radius of the second phase particle and R is the distance from the centre of the particle.

Equations (2) and (3) are deduced in the spherical coordinate system. In order to calculate the stress conveniently (see below), the stress can be expressed in the Cartesian coordinates system. This can be realized using standard stress tensor transformation law.[28] If a particle is located at point (X, Y, Z), the stress that is generated at point (x, y, z) outside the particle should be expressed by a tensor with six components, three principal components (σ_x, σ_y, and σ_z), and three shear components (σ_{xy}, σ_{xz}, and σ_{yz}). Relevant to our analyses below are the three principal components, and the one that is in the z direction can be written as

$$\sigma_z = \frac{pr^3}{2}\frac{(x-X)^2 + (y-Y)^2 - 2(z-Z)^2}{((x-X)^2 + (y-Y)^2 + (z-Z)^2)^{5/2}}. \quad (4)$$

The increasing strength of the Al_2O_3–SiC nanocomposites may be explained, to some extent, by the normal stress experienced by the GB, which we try to estimate in the present work. For simplicity, we assume that the intragranular SiC particles of the second phase and the Al_2O_3 grains of the matrix are both spherical, and their mean sizes are d and D, respectively. The intragranular SiC particles are distributed randomly within the Al_2O_3 grains and the particles do not interfere with each other. From these hypotheses, we can find that the normal stress at any point on the GB is equal. This microstructure model has also been adopted by Lü *et al.*[21]

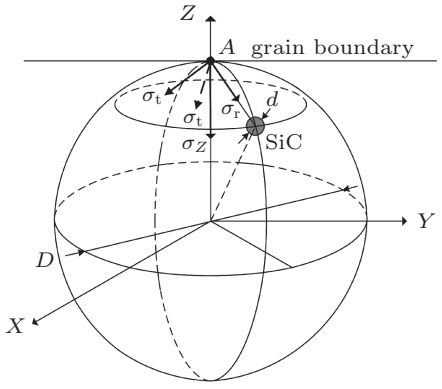


Fig. 1. Schematic diagram showing an intragranular SiC particle within an Al_2O_3 grain and the stress exerted on the GB.

We locate the centre of a spherical Al_2O_3 grain at the coordinate origin and choose to analyse the principal stress exerted at a point $A(0, 0, D/2)$ on the GB

by an SiC particle at point (x, y, z) (Fig. 1). Since point A is also on the Z axis, the stress in the normal direction at this point on the GB is

$$\sigma_z = \frac{pd^3}{16}\frac{x^2+y^2-2(z-D/2)^2}{(x^2+y^2+(z-D/2)^2)^{5/2}}. \quad (5)$$

For the uniformly randomly distributed SiC particles, the probability of finding an SiC particle at any position is the same within the spherical grain, except in the outer shell with a thickness of (d/2), since the SiC particle is intragranular. So the normal stress at A due to one SiC particle should be obtained by the volume-averaged σ_z as

$$\sigma_Z = \frac{\iiint_{\Omega}\sigma_z \mathrm{d}V}{V} = \frac{\iiint_{\Omega}\frac{pd^3}{16}\frac{x^2+y^2-2(z-D/2)^2}{(x^2+y^2+(z-D/2)^2)^{5/2}}\mathrm{d}x\mathrm{d}y\mathrm{d}z}{V}, \quad (6)$$

where V is the volume of the Al_2O_3 grain, and $V = \pi D^3/6$ and Ω is the interior of the Al_2O_3 grain, except the outer shell.

Because the integral field Ω is spherical, it is more convenient to make the integration in the corresponding spherical coordinates system. Then equation(6) becomes

$$\sigma_Z = \frac{\frac{pd^3}{16}\int_0^{2\pi}\int_0^{\pi}\int_0^{(D-d)/2}\frac{r^2\sin^2\theta-2(r\cos\theta-D/2)^2}{[r^2\sin^2\theta+(r\cos\theta-D/2)^2]^{5/2}}r^2\sin\theta\mathrm{d}r\mathrm{d}\theta\mathrm{d}\phi}{V}. \quad (7)$$

Note that the upper integral limit of $\mathrm{d}r$ is $(D-d)/2$.

The average number of dispersion particles in one Al_2O_3 matrix grain, n, can be obtained according to Eq. (8) as

$$n = f\frac{V}{V_\mathrm{p}}, \quad (8)$$

where V_p is the volume of one SiC particle, $V_\mathrm{p} = \pi d^3/6$, and f is the volume fraction of intragranular SiC particles.

Thus the net normal stress σ at any point on the GB is n times the σ_Z value, namely

$$\sigma = \frac{3pf}{8\pi}\int_0^{2\pi}\int_0^{\pi}\int_0^{(D-d)/2} \times \frac{r^2\sin^2\theta-2(r\cos\theta-D/2)^2}{[r^2\sin^2\theta+(r\cos\theta-D/2)^2]^{5/2}} \times r^2\sin\theta\mathrm{d}r\mathrm{d}\theta\mathrm{d}\phi. \quad (9)$$

Stress is a second-order tensor and its transformation should comply with the tensor transformation rules.[28] Our treatment of the residual stress (Eq. (4) and equations thereafter) ensures a pertinent analysis of the model. The formula calculus, which did not regarded stress as a tensor of second order,[20,21] is highly questionable.

Figure 2 shows the compressive stress on the GB, calculated from the model. The following data of the Al_2O_3–SiC nanocomposite given by Lü *et at.*[21] are used: $\Delta\alpha = \alpha_\mathrm{p} - \alpha_\mathrm{m} = -4.1\times10^{-6}$ K^{-1}, $\Delta T = 1100$ K, $\nu_\mathrm{m} = 0.25$, $\nu_\mathrm{p} = 0.17$, $E_\mathrm{m} = 389$ GPa, $E_\mathrm{p} = 450$ GPa, $d = 100$ nm, and $f = 0$, 1, 2.5, 5, 7.5, and 10 vol%, $D = 2.4$, 3.5, 2.6, 2.1, 1.9, and 1.8 μm. The compressive stress increases almost linearly with the increase in the volume fraction of the intragranular SiC particles. The increment in the strength reported in Ref. [21] is also shown in Fig. 2. $\sigma_\mathrm{f}f$ is the measured fracture strength when the content of SiC is f vol%. When the volume fraction of SiC is less than 5%, the compressive stress on the GB that is calculated is less than the corresponding increment of the strength, ($\sigma_\mathrm{f}f$–$\sigma_\mathrm{f}0$). The values of the ratio of σ to ($\sigma_\mathrm{f}f$–$\sigma_\mathrm{f}0$) are 41%, 47%, and 60% when the values of f are 1, 2.5, 5 vol%, respectively. For the Al_2O_3–SiC nanocomposites, the fracture mode is typically intergranular when the SiC content is less than or equal to 4 vol% and the mode becomes a mixture mode of intergranular mode and transgranular mode when the SiC content is at about 5 vol%.[20] The compressive stresses can tighten matrix GBs and protect weak ones. The comparison between the increment of the strength and the compressive stress on the GB indicates that the normal compressive stress on the GB may be one of the major factors in strengthening the nanocomposites at least when the SiC content is ≤ 0.05. Strengthening mechanisms such as particle-bridging and surface residual stress may also play roles in strengthening the nanocomposites.[14,17]

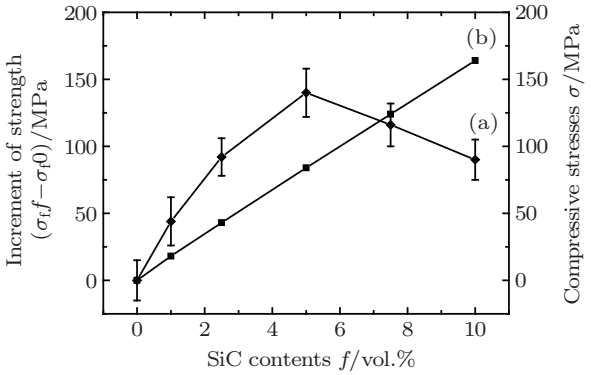


Fig. 2. A comparison between the increment in strength of the composite (a) (i.e. experimental data given in Ref. [21]) and the compressive stress on the calculated GB (b).

3. Weakening mechanism

Tensile residual stress is generated when the CTE of the matrix exceeds that of the particle, which is the case in the Al_2O_3–SiC composite. Internal stresses caused by the mismatch of CTEs have been analysed using micromechanics.[29] The basic concept is described as follows: in the cooling process from processing temperature, T_P, to room temperature, T_R, a misfit strain will be produced. This thermal expansion misfit strain results in an average residual stress σ_m field in the matrix, and the average stress can be calculated by[29]

$$\frac{\sigma_m}{E_m} = \frac{2f\beta\alpha^*}{(1-f)(\beta+2)(1+v_m)+3\beta(1-v_m)}, \quad (10)$$

$$\beta = \left(\frac{1+v_m}{1-2v_p}\right)\left(\frac{E_p}{E_m}\right), \quad (11)$$

$$\alpha^* = \int_{T_p}^{T_R} (\alpha_p - \alpha_m)\delta \, dT, \quad (12)$$

where δ is the isotropic tensor (Kronecher's δ). Figure 3 shows that the average tensile stresses for the values of 1, 2.5, 5, 7.5, and 10 vol.% SiC content are 15, 37, 74, 110, and 147 MPa, respectively. The parameters used for this calculation are the same as those given above in Section 2. The average tensile stress increases almost linearly with the increase in the volume fraction of intragranular SiC particles (Fig. 3). It is also noted that the absolute value of the average tensile stress is slightly lower than that of the compressive stress on the GB (cf. Fig. 2 and Fig. 3).

When the coefficient of the thermal expansion of the matrix is less than that of the particle, the matrix is under compressive stress and the composite is strengthened. This is the case of the SiC (matrix)-TiB_2 (particle) conventional composite (i.e., the non-nanocomposite without an appreciable number of intragranular particles).[29] As for the Al_2O_3–SiC nanocomposite, where $\alpha_p < \alpha_m$, the situation becomes slightly more complicated due to the intragranular feature of the nanocomposite. The monolithic Al_2O_3 ceramic fractures intergranularly. When an SiC particle is introduced, it exerts compressive stress on the GB, acting as a strengthening mechanism, and tensile stress in the matrix grain, acting as a weakening mechanism. As long as the fracture mode remains mainly intergranular, the strengthening mechanism prevails. The weakening mechanism manifests itself when the fracture mode becomes transgranular because of the severe effects of tensile stresses in matrix grains. The two competitive mechanisms reach a balance when both intergranular and transgranular fractures are present and the maximum strength is achieved.[20]

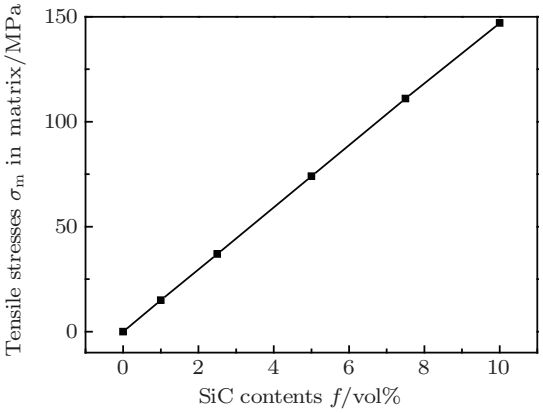


Fig. 3. The average tensile stress within the matrix grain via the volume fraction of SiC particles.

Experimentally, transgranular fracture becomes significant at about 5 vol% and dominant at higher SiC content. The higher the SiC content, the lower the measured strength[21] and the higher the calculated tensile stress (Fig. 3). It is thus interesting to compare the increment in the tensile stress and the decrement in the fracture strength, taking the corresponding values at $f = 5$ vol% as reference. $\sigma_m f$ is the average tensile stress when the content of SiC is f vol%. The increment in the tensile stress ($\sigma_m f$–$\sigma_m 5$) and the decrement in the strength ($\sigma_f 5$–$\sigma_f f$) are shown in Fig. 4. The values of the increment of the tensile stresses ($\sigma_m f$–$\sigma_m 5$) are 0, 36, and 73 MPa for the values of 5, 7.5, and 10 vol% SiC content, respectively, while the values of the decrement in the strength ($\sigma_f 5$–$\sigma_f f$) are 0, 24, and 50 MPa for the values of 5, 7.5,

and 10 vol% SiC content, respectively.[21] The increment in the tensile stress is larger than the decrement in the fracture strength. This should not be considered as unfavourable to the tensile stress weakening mechanism. It should be pointed out that one may not suppose that the macroscopic fracture stress is reduced by an amount equal to the internal stress.[30] In some cases the internal stress may be considerably larger than the macroscopic fracture stress. A comparison between ($\sigma_{\rm f}5$–$\sigma_{\rm f}f$) and ($\sigma_{\rm m}f$–$\sigma_{\rm m}5$) may provide qualitative evidence for the hypothesis that the tensile stresses can be regarded as the main weakening mechanism in the nanocomposites at least when SiC content ≥ 0.06 though other weakening mechanisms such as particle agglomerates may also be an important factor.[23]

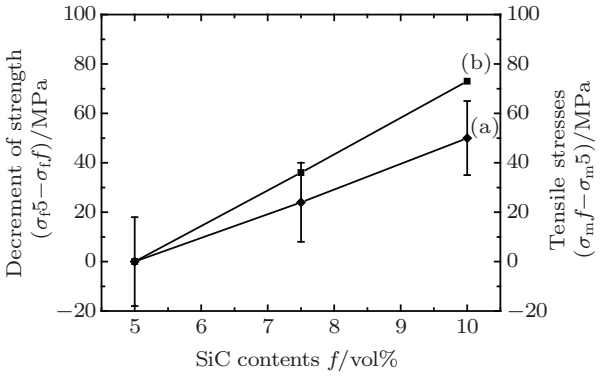


Fig. 4. Comparison between the decrease in the strength (a) and the increase in the average tensile stress within a grain (b). Data for an SiC content of 5 vol% are cited from the references for comparison.

4. Conclusions

The internal stress is calculated to analyse the strengthening and weakening mechanisms in Al_2O_3–SiC nanocomposites. The normal compressive stress on GBs is calculated based on a model. The result shows that the normal compressive stress can tighten matrix GBs and shield weak ones, which can be attributed to the major factors in strengthening the nanocomposites at least when the SiC content is ≤ 0.05. In the meantime, the average tensile stress within the grain is also calculated. The result shows that the tensile stress acts like a weakening effect in the Al_2O_3 matrix, which can be regarded as the main weakening mechanism in nanocomposites with high SiC content.

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