

Grain-size dependence of fracture stress in anisotropic brittle solids

V. D. KRSTIC

Department of Metallurgical Engineering Queen's University, Kingston, Ontario, Canada

The stress concentrations that occur at grain boundaries due to thermal expansion anisotropy and elastic stress concentration are discussed, and the stress intensity factor that results from these stresses is estimated. The procedure for the stress intensity factor calculation is based on the model in which a spherical crystal (grain) is forced into a cavity of equal size possessing annular or radial cracks emanating from the boundary. The stress intensity factor equation thus obtained is extended to include the effect of elastic stress concentration due to the presence of a cavity, and is subsequently used to predict the grain-size dependence of strength in anisotropic brittle ceramics. In assessing the degradation of strength with increasing grain size in non-cubic ceramics, it is shown that, in addition to grain size, the effect of pre-existing crack size must also be considered. Cubic ceramics, on the other hand, are known to exhibit no thermal expansion anisotropy and, based on the present model, their strength is predicted to be governed by the pre-existing flaw size, rather than the grain size.

1. Introduction

It is well known that non-cubic polycrystalline ceramics exhibit a strong grain-size dependence of strength [1, 2], whereas cubic ceramics show only weak grain-size dependence in a wide grain-size range [3]. The strong grain-size dependence of strength in non-cubic polycrystalline ceramics is believed to be associated with thermoelastic stresses developed as a result of thermal expansion anisotropy, which is also known to cause spontaneous microcracking [4, 5]. This microcracking can be suppressed in a single-phase material if the average grain size is kept smaller than some critical value [2, 6]. In polycrystalline materials possessing second-phase particles or inclusions, microcracking can likewise be prevented if the second-phase or inclusion size does not exceed a critical value [7, 8]. Even approximate predictions of the grain- or particle-size dependence of spontaneous cracking and the material's capability of supporting stress is of great significance if strong and thermal shock-resistant material is to be designed.

The spontaneous cracking in two-phase systems possessing second-phase particles of different thermal expansion, and in a single-phase anisotropic ceramic, have been analysed and modelled by a number of authors [8-12]. All these models concerned the role of internal stresses and the grain-size effects on microcrack initiation without the presence of external stresses. However, in many practical situations the microstructural features responsible for spontaneous cracking, which are sub-critical in the absence of external stress, become critical when subjected to applied stress. This will normally lead to undesirable premature failure of the structural component. Furthermore, based on the premise that the inherent flaw size is equivalent to the grain size, the grain-size

exponent from the strength against grain-size plot should always be $-1/2$. However, the experimental data in a wide variety of polycrystalline ceramics have shown that the grain-size exponent is not $-1/2$ [1, 13] and that the crack size is not limited by the grain size [1, 14, 15].

The aim of the present paper is to further examine the mechanism of crack extension in the presence of boundary stresses arising from anisotropic thermal contraction, and also to make some quantitative predictions regarding the extent that these stresses should be expected to influence the fracture-stress-grain-size relationship in such solids.

2. Condition for spontaneous cracking in two-phase systems

For a two-phase system containing a single, isolated spherical particle of radius R , and with thermal expansion coefficient smaller than that of matrix ($\alpha_p < \alpha_m$), the total stress intensity factor at the tip of an annular flaw emanating from the particle-matrix interface was found to consist of two terms. The first term is due to a radial component of the stress concentration outside the spherical boundary, and is given [12] by the expression

$$K_t^r = \frac{2PR^{1/2}}{\pi^{1/2}} [1 + (s/R)]^{1/2} \times \left[1 - \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \right] \quad (1)$$

where P is the thermoelastic stress and s is the annular flaw size. The second term is due to the tangential component of thermoelastic stress concentration and may be expressed [12] in the form

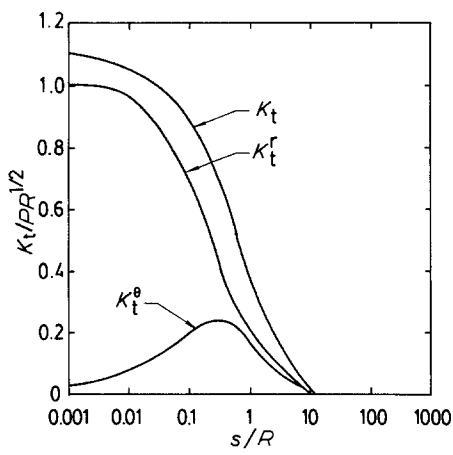


Figure 1 Stress-intensity-factor solution for annular crack emanating from a spherical inclusion.

$$K_t^\theta = \frac{PR^{1/2}}{\pi^{1/2}} \frac{1}{[1 + (s/R)]^{3/2}} \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \quad (2)$$

The total stress intensity factor is obtained by the addition of Equations 1 and 2:

$$K_t = K_t^r + K_t^\theta = \frac{2PR^{1/2}}{\pi^{1/2}} \left\{ [1 + (s/R)]^{1/2} \left[1 - \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \right] + \frac{1}{2[1 + (s/R)]^{3/2}} \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \right\} \quad (3)$$

Fig. 1 illustrates the change of normalized stress intensity factor as a function of s/R . Close examination of Fig. 1 and Equation 3 leads to several important conclusions. First, it is immediately evident that the stress intensity factor due to the tangential component of stress concentration (K_t^θ) is much smaller than the stress intensity factor due to the radial component (K_t^r). This means that if only K_t^θ is used to define the critical condition for spontaneous cracking, without taking into account the other component of stress intensity factor (K_t^r), the total stress intensity factor will appear to be so small that in most practical situations the crack would hardly be initiated and spontaneous cracking would never be observed. For example, in the case of polycrystalline alumina possessing fracture toughness $K_{IC} \sim 4 \text{ MPa m}^{1/2}$, with a maximum grain size $D = 100 \mu\text{m}$ and $s/R = 0.1$, a residual stress of $P = 2000 \text{ MPa}$ would be required in order to initiate crack extension. Even if single-crystal values for fracture toughness are used ($K_{IC} \sim 2 \text{ MPa m}^{1/2}$), residual stresses in excess of $P = 1380 \text{ MPa}$ are required to cause crack extension. However, the maximum thermoelastic stress that may develop in polycrystalline alumina as a result of thermal expansion anisotropy is found to be of the order of 200 to 300 MPa [5, 13]. At these stresses, a critical grain size required to initiate crack extension would be around 3 mm, which is several orders of magnitude larger than the observed critical grain size for spontaneous cracking in a polycrystalline alumina, namely $100 \mu\text{m}$ [5]. Thus, it is clear that the stress intensity factor due to the tangential component of stress concentration

alone is not capable of explaining the phenomenon either on physical grounds or quantitatively. It is therefore considered necessary to take into account the radial component of the stress concentration (σ_r) which acts along the grain boundary, but also in the direction to open the crack. For such a crack-particle geometry, it is self-evident that the half crack length (C) consists of a cavity radius (R) and the length of an annular flaw, namely $(R + s)$. The major benefit of this approach is that it gives a strong particle-size dependence of spontaneous cracking and it also facilitates the inclusion of the entire residual stress (P) into the equation for the grain-size dependence of strength.

3. Condition for crack extension in anisotropic solids

Although Equation 3 has been developed for the case of a spherical inclusion dispersed in a brittle matrix of higher thermal expansion [12], it can also be used to interpret the data on the spontaneous cracking of a single-phase material with thermal expansion anisotropy [13]. Statistically, in any anisotropic polycrystalline solid a certain number of grains will be oriented with respect to their neighbouring grains, such that the axis of minimum thermal contraction of the central grain is parallel to the axis of maximum thermal contraction of the surrounding grains (Fig. 2a). This condition leads to the generation of mostly compressive stresses within the central grain (A), while the neighbouring grains will be subjected mostly to tensile stresses. Under such circumstances, an annular flaw is likely to be formed by the linking of small radial cracks on individual neighbouring grains. Clearly, due to random orientation, some of the surrounding grains may be under compression while the central grain is locally under tension. This grain orientation problem was analysed by Davidge [16], who showed that the maximum misorientation and therefore the maximum anisotropic stress causing crack initiation does not depend only on the maximum difference in

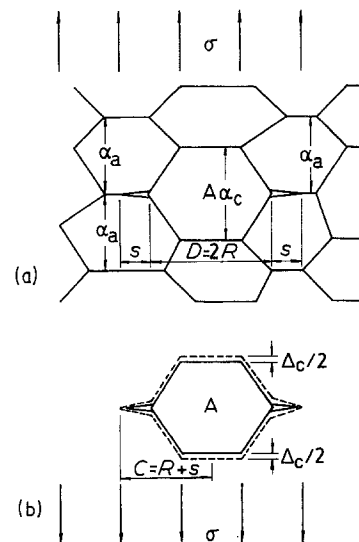


Figure 2 Polycrystalline aggregate containing annular flaw. (a) Hexagonal grain under compression; (b) opening of the crack due to thermal expansion. α_a and α_c are the coefficients of thermal expansion along the a and c axes, respectively.

thermal expansion anisotropy but also on the grain orientation.

Although the stress-field interaction of randomly distributed grains in an anisotropic solid is a very complicated problem, a better understanding of the behaviour of such solids under applied stress would be of considerable value. It has already been shown [13] that when an external stress is applied to an anisotropic solid, an additional positive stress intensity factor is generated and the total stress intensity factor is obtained by the addition of the thermoelastic stress intensity factor due to thermal expansion anisotropy, K_t (from Equation 3), and the stress intensity factor due to the applied stress, K_a :

$$K = K_t + K_a \quad (4)$$

Previously [13], this problem was treated by considering that the central grain is forced into a cavity of diameter $D = 2R$ surrounded by the annular flaw of length s (Fig. 2), without taking into consideration the effect of a stress concentration due to the presence of a cavity. In the present analysis, the previous model [13] for microfracture is extended to include the effect of elastic stress concentration due to the applied stress, and to further examine the relative contribution of both the thermal and the elastic stress concentrations. In what follows, it will be assumed that only the tangential component of the stress concentration due to the presence of a spherical cavity is imposed on the crack in the direction to open it. This tensile component of the stress concentration, σ_θ , due to the presence of a cavity [17] is

$$\sigma_\theta = \sigma \left[\frac{4 - 5\nu}{2(7 - 5\nu)} \left(\frac{R}{x} \right)^3 + \frac{9}{2(7 - 5\nu)} \left(\frac{R}{x} \right)^5 + 1 \right] \quad (5)$$

$(x \geq R)$

where σ is the applied stress, R the cavity radius, ν the Poisson's ratio and x the distance from the centre of the cavity.

For an axisymmetric stress distribution problem such as is the case with an annular crack of width s , the stress intensity factor is given [18, 19] by the expression

$$K_a = \frac{2}{(\pi C)^{1/2}} \int_0^C \frac{\sigma(x) dx}{(C^2 - x^2)^{1/2}} \quad (6)$$

where $C = R + s$ and $\sigma(x)$ is the stress distribution. The stress intensity factor due to the applied stress (K_a) can now be obtained by substituting Equation 5 into Equation 6. This procedure yields (from $x = R$ to $x = C$),

$$K_a = 2\sigma \left(\frac{C}{\pi} \right)^{1/2} \left(1 - \frac{R^2}{C^2} \right)^{1/2} \times \left\{ \frac{4 - 5\nu}{2(7 - 5\nu)} \left(\frac{R^2}{C^2} \right) + \frac{9}{2(7 - 5\nu)} \right. \\ \left. \times \left[1 + \frac{1}{3} \left(\frac{C^2}{R^2} - 1 \right) \right] \left(\frac{R^4}{C^4} \right) + 1 \right\} \quad (7)$$

When expressed in terms of s/R , Equation 7 reads (for $\nu = 0.2$)

$$K_a = \frac{2\sigma R^{1/2}}{\pi^{1/2}} \left(1 + \frac{s}{R} \right)^{1/2} \left[1 - \left(1 + \frac{s}{R} \right)^{-2} \right]^{1/2} \\ \times \left(1 + \frac{1}{4[1 + (s/R)]^2} + \frac{3}{4[1 + (s/R)]^4} \right) \\ \times \left\{ 1 + \frac{1}{3} \left[\left(1 + \frac{s}{R} \right)^2 - 1 \right] \right\} \quad (8)$$

The total stress intensity factor obtained as a result of the combined effects of the residual thermal stress due to the presence of thermal expansion anisotropy and the applied stress is (from Equation 4)

$$K_{tot} = \frac{2PR^{1/2}}{\pi^{1/2}} \\ \times \left\{ \left(1 + \frac{s}{R} \right)^{1/2} \left[1 - \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \right] \right. \\ \left. + \frac{1}{2[1 + (s/R)]^{3/2}} \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \right\} \\ + \frac{2\sigma R^{1/2}}{\pi^{1/2}} \left(1 + \frac{s}{R} \right)^{1/2} \left(1 - \frac{1}{[1 + (s/R)]^2} \right)^{1/2} \\ \times \left(1 + \frac{1}{4[1 + (s/R)]^2} + \frac{3}{4[1 + (s/R)]^4} \right) \\ \times \left\{ 1 + \frac{1}{3} \left[\left(1 + \frac{s}{R} \right)^2 - 1 \right] \right\} \quad (9)$$

Fig. 3 illustrates the variation of normalized stress intensity factor with s/R . Inspection of Fig. 3 indicates that the thermoelastic stress intensity factor (K_t) drops very fast with s/R , reaching an extremely small value at $s/R > 10$. On the other hand, the elastic stress intensity factor (K_a) increases continually with s/R , ultimately reaching the value for an internal circular crack at $s/R \rightarrow \infty$. For the other extreme, when $s/R \rightarrow 0$, the elastic stress intensity factor solution (K_a) approaches zero but the total stress intensity factor (K_{tot}) attains a finite value due entirely to the thermal stress intensity factor (K_t), which becomes the dominant term in controlling the overall criterion for crack extension.

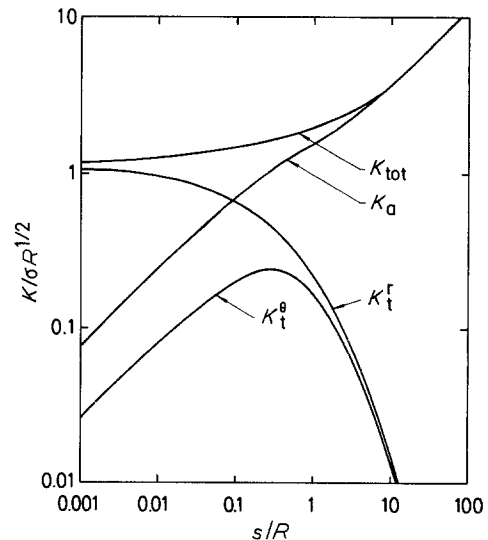


Figure 3 Variation of stress intensity factor (from Equation 9) with s/R .

For a critical condition of crack initiation, $K_{\text{tot}} = K_{\text{IC}} = [2\gamma E/(1 - \nu)]^{1/2}$ and $\sigma = \sigma_f$, the strength of an anisotropic solid is

$$\sigma_f = \frac{1}{\Phi_e} \left(\frac{\pi\gamma E}{D[1 + (s/R)](1 - \nu^2)} \right)^{1/2} - P \frac{\Phi_t}{\Phi_e} \quad (10)$$

where σ_f is the fracture stress, E the Young's modulus, γ the fracture surface energy, D the grain size and $P = 2E \Delta\alpha \Delta T/3(1 - \nu)$ where $\Delta\alpha (= \alpha_{\text{max}} - \alpha_{\text{min}})$ is the thermal expansion coefficient difference and ΔT the temperature difference; also

$$\Phi_e = \left(1 - \frac{1}{[1 + (s/R)^2]} \right)^{1/2} \left(1 + \frac{1}{4[1 + (s/R)^2]} \right) + \frac{3}{4[1 + (s/R)^4]} \left\{ 1 + \frac{1}{3} \left[\left(1 + \frac{s}{R} \right)^2 - 1 \right] \right\}$$

$$\Phi_t = 1 - \left(1 - \frac{1}{[1 + (s/R)^2]} \right)^{1/2} + \frac{1}{2[1 + s/R]^2} \left(1 - \frac{1}{[1 + (s/R)^2]} \right)^{1/2}$$

When using Equation 10, it is important to realize that the residual stress (P) arising from thermal expansion anisotropy refers to a maximum value obtained from the difference between the axes of maximum and minimum thermal expansion. The predicted change of fracture stress (from Equation 10) with grain size is given in Fig. 4. As expected, Equation 10 and Fig. 4 show a strong s/R dependence of fracture strength, with the most severe strength degradation being at small grain sizes. In this context it may be of interest to note also a strong s/R dependence of stress concentration factors (Φ_t and Φ_e). As can be seen from Fig. 5, the ratio Φ_t/Φ_e is the largest at small s/R , whereas at large s/R it approaches zero. This means that the residual stress arising from the thermal expansion anisotropy influences the grain-size dependence

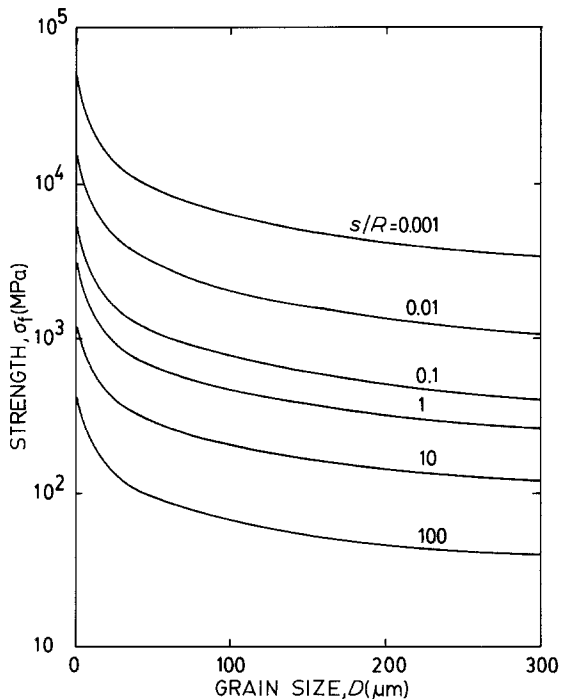


Figure 4 Predicted variation of strength (from Equation 10) with grain size for a given s/R .

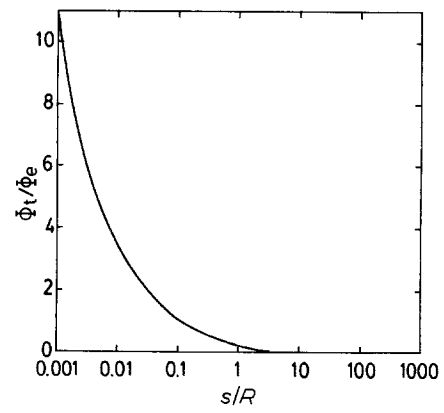


Figure 5 Typical change of Φ_t/Φ_e with s/R in anisotropic ceramics.

of strength only at small s/R values. An important conclusion that can be inferred from this is that, in fine-grained solids in which the pre-existing flaw size (s) is normally larger than the grain size ($s \gg R$), the effect of residual stresses will be negligible.

It can further be shown that for $s/R > 1$, the elastic stress concentration factor (Φ_e) approaches unity and Equation 10 reduces to

$$\sigma_f = \left(\frac{\pi E \gamma}{D[1 + (s/R)](1 - \nu^2)} \right)^{1/2} - P \Phi_t \quad (11)$$

Equation 11 recovers the result obtained elsewhere [13] by applying the same fundamental approach but without taking into account the contribution of elastic stress concentration. Again, as with Equation 10, at large s/R ($s/R > 1$ to 2), Φ_t becomes very small and so the effect of residual stress from thermal expansion anisotropy becomes negligible.

4. Crack initiation and stress relaxation process

For the crack-grain configuration illustrated in Fig. 2, it is self-evident that, during loading, crack surfaces will open and residual stress relaxation will take place. The extent of stress relaxation is determined by the maximum crack opening displacement at distance $C = R + s$ from the crack tip and is given [20] by the expression

$$\Delta = \frac{2(1 - \nu^2)K_I}{E} \left(\frac{R + s}{\pi} \right)^{1/2} \quad (12)$$

where K_I is the stress intensity factor and E is the Young's modulus of the matrix. As expected, Equation 12 shows that the crack opening displacement depends on the crack length ($R + s$), and for long cracks, relatively large openings are required before the crack can propagate.

Assuming no strain-field interaction effects of neighbouring grains, the maximum linear thermal expansion of a grain is

$$\Delta_t = D \Delta\alpha \Delta T \quad (13)$$

where $\Delta\alpha$ and ΔT are as defined earlier. Based on Equation 12, crack initiation will occur when the crack opening displacement reaches a critical value Δ_c . This condition is represented by the expression

$$D \Delta\alpha \Delta T = \frac{2(1 - \nu^2)K_{\text{IC}}}{E} \left(\frac{R + s}{\pi} \right)^{1/2} \quad (14)$$

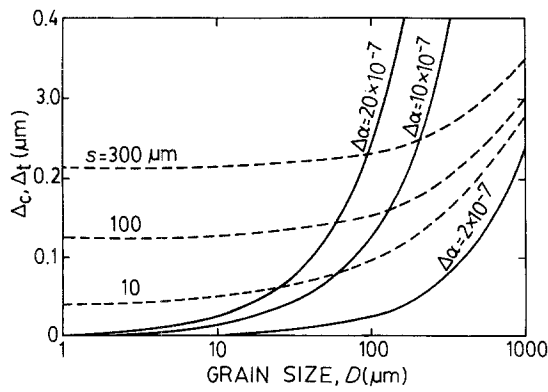


Figure 6 Variation of (---) critical crack opening displacement (Δ_c) and (—) linear thermal expansion (Δ_t) with grain size and pre-existing flaw size (s).

where K_{IC} is the single-crystal or the grain-boundary fracture toughness, depending on the location of a crack tip. Fig. 6 illustrates the change of a critical crack opening displacement (Δ_c) and the maximum linear thermal expansion of the grain (Δ_t) as a function of grain size, pre-existing flaw size (s), and the $\Delta\alpha$ value. Due to the different grain-size dependences of the linear thermal expansion of the grain (Δ_t) and the critical crack opening displacement (Δ_c) (linear compared with square-root), Fig. 6 shows that there is a region of grain sizes in which the opening of the crack is larger than the total linear thermal expansion of the grain diameter, and the residual stress will be completely relieved on loading prior to crack initiation. When the total linear thermal expansion of the grain becomes equal to the critical crack opening displacement, $\Delta_t = \Delta_c$, the central grain in Fig. 2, which was originally subjected to a compressive stress, will now be subjected to uniform tension. If, at this stage, the stress intensity factor at some pre-existing flaw located within the grain (A) becomes larger than at the annular flaw, (s), crack extension may first occur in the grain rather than from the annular or radial flaw.

The critical grain size (D_c) for crack extension based on crack opening displacement can be obtained by solving Equation 14 for D . This procedure yields (positive solution of quadratic equation)

$$D_c = \frac{(1 - \nu^2)^2 K_{IC}^2}{\pi E^2 (\Delta\alpha \Delta T)^2} + \left(\frac{(1 - \nu^2)^2 K_{IC}^2}{\pi^2 E^4 (\Delta\alpha \Delta T)^4} + \frac{4(1 - \nu^2)^2 K_{IC}^2 s}{\pi E^2 (\Delta\alpha \Delta T)^2} \right)^{1/2} \quad (15)$$

5. Discussion and comparison with experiments

So far, in interpreting the data on grain-size dependence of strength in polycrystalline, single-phase ceramics, the practice has been to assume that the flaw size is equal to the grain size. Based on this assumption, it follows that the grain-size exponent from the strength against grain-size plot should always be -0.5 , regardless of the material and the grain-size range. However, the experimental data reported for a wide variety of polycrystalline ceramics have shown

that the grain-size exponent can vary from a very small negative value of $-1/7$, for a polycrystalline magnesia, to a very large negative value of -1 , for a polycrystalline beryllia [1]. This wide variation of grain-size exponents was attributed to variations in the degree of thermal expansion anisotropy. Magnesia, which is known to be the most isotropic material, has the lowest grain-size exponent and exhibits weak grain-size dependence of strength, at least in the grain-size range between a few micrometres to $400 \mu\text{m}$ [21]. Almost identical results were reported for a polycrystalline FeO also known to be a highly isotropic material [22]. These results clearly demonstrate that the grain boundary itself does not necessarily represent the barrier for crack extension, and that the inherent flaw size is likely to be determined by the other microstructural features such as micropores, second-phase particles, etc. Non-cubic, polycrystalline solids, on the other hand, are known to exhibit varying degrees of thermal expansion anisotropy and show a strong grain-size dependence of strength.

Since the primary objective in the present paper is to develop the relationship between the strength and the grain size in non-cubic solids, we will first concentrate on a polycrystalline alumina known to possess a moderate degree of anisotropy. Fig. 7 illustrates the predicted and measured variations of strength with grain size. Before discussing the results of Fig. 7 it is considered appropriate at this point to briefly examine the condition of crack initiation as defined by Equation 3. Inspection of Equation 3 shows that the thermal stress intensity factor of a solid containing a single grain with a lower coefficient of thermal expansion is a strong function of grain size, and for a given residual stress P and the fracture toughness K_{IC} the crack initiation will occur when the grain size reaches a critical

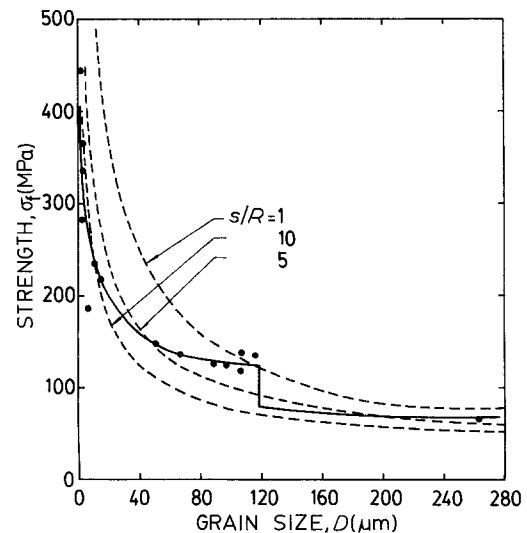


Figure 7 Comparison of predicted effects of grain size on strength with (●) data obtained for polycrystalline alumina [21]. Although fracture is expected to occur from the largest flaw size (largest $R + s$), predictions are based on the average grain size. The present model shows that fracture will occur when either anisotropic stress or crack length reaches a critical value. Due to the fact that the internal stresses are dependent on the grain orientation and the stress-field interaction effects, the fracture may not necessarily occur from the largest flaw size.

value D_c . According to Equations 10 and 11, before the grain size reaches a critical value there will be a region of continual decrease of s/R as the grain size increases, provided that the inherent flaw size remains independent of grain size. Although the inherent flaw size may vary from sample to sample due to different fabrication conditions, observations made on a number of polycrystalline ceramics indicate that the fracture normally originates from pre-existing flaws which are usually different from the average grain size [1, 23, 24]. The difference between the average grain size and the inherent flaw size appears to be greatest in the case of isotropic polycrystalline ceramics such as MgO [21] and FeO [22]. For example, an increase of grain size from approximately 6 to 365 μm has led to a minor change of inherent flaw size, namely 6 to 37 μm , indicating no real connection between the grain size and the flaw size [13]. In anisotropic, polycrystalline titania, on the other hand, a much smaller difference between calculated critical crack size (from the Griffith equation*) and measured grain size has been reported [1]. Furthermore, in the same material, cracks were found to terminate frequently at the centre of a grain rather than at the grain boundaries.

Inspection of Fig. 7 shows that the most severe degradation of strength occurs at small grain sizes, below $\sim 20 \mu\text{m}$, and as the grain size increases the strength levels off, becoming almost independent of grain size up to 120 μm size. When the grain size reaches approximately 120 μm , the strength appears to drop abruptly to a lower value and thereafter it remains almost unchanged with further increase of grain size. It is of interest here to point out the coincidence of the grain size at which the sudden drop in strength occurs and the critical grain size for spontaneous cracking normally observed in polycrystalline alumina [5, 25].

In correlating the experimental data for the grain-size dependence of strength with predicted variation from Equation 10, a constant s/R was first assumed (dashed lines in Fig. 7). However, at constant s/R , good correlation between predicted and measured strength is found only at small grain sizes (below approximately 20 μm) and at large s/R values, $s/R = 10$. Particularly poor correlation is obtained at small grain sizes and at small s/R values. When no change of pre-existing flaw size with grain size is assumed ($s = \text{constant}$), a continual increase of grain size leads to an equivalent decrease of s/R resulting in a levelling off of the strength, as shown in Fig. 7 (solid line). When the grain size reaches a critical value of $\sim 120 \mu\text{m}$, a spontaneous crack extension occurs causing an instantaneous increase of s/R and a corresponding drop in strength. The new crack length is now sub-critical and the grain size must be increased to a new value before the crack again moves instantaneously. At this point, the residual anisotropic stress will be relaxed leaving essentially a stress-free solid. Further increase of grain size, beyond the critical size, has little effect on the strength in accordance with the predictions.

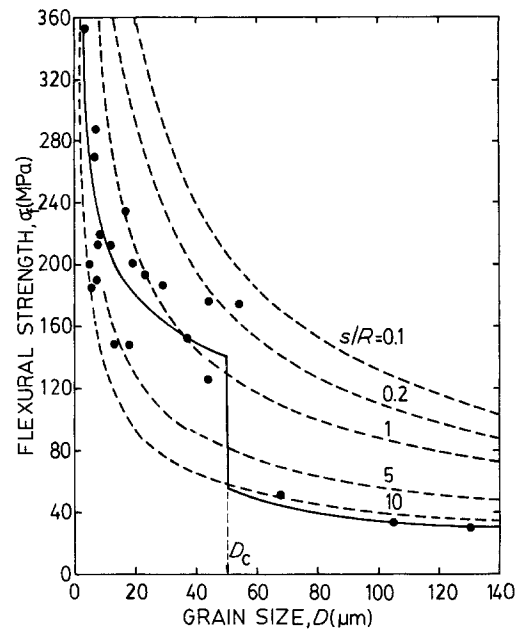


Figure 8 Comparison of (●) experimental data for the grain-size dependence of strength in polycrystalline TiO_2 [1] with predicted effects of grain size on strength.

Due to limited data points and the considerable scatter in Fig. 7, it was difficult to accurately predict the critical grain size at which an instantaneous degradation of strength occurs. In spite of this a reasonably good agreement is found between the grain size for instantaneous degradation of strength from Fig. 7 ($D_c \sim 120 \mu\text{m}$) and the critical grain size for spontaneous cracking ($D_c \sim 150 \mu\text{m}$) determined by direct observations [5, 25]. Similar general trends were observed in a highly anisotropic, polycrystalline titania known to exhibit a strong grain-size dependence of strength. For comparison, Fig. 8 shows the predicted and measured variations of strength with grain size in polycrystalline TiO_2 . Assuming no change of pre-existing flaw size with grain size, a continual increase of grain size leads to an adequate decrease of s/R , resulting in a strength degradation at a decreasing rate (solid line in Fig. 8). At approximately 50 μm grain size, an instantaneous drop in strength occurs and this appears to coincide with a phenomenon of spontaneous cracking normally observed in polycrystalline titania [1, 5]. At this stage a crack is initiated and, due to kinetic energy, will increase its length to a value which is larger than the grain size, leaving s/R normally much larger than the grain size ($s/R \gg 1$). Any subsequent increase in grain size beyond the critical size will have a minor effect on a material's strength, simply because the contribution of grain size to the total crack length is also small.

Inspection of Figs 7 and 8 shows that the changes of strength with grain size in polycrystalline alumina and titania exhibit the same trend, but the instantaneous drop in strength at a critical grain size appears to be appreciably larger in titania samples. Although the exact nature of this phenomenon is beyond the scope of the present theoretical analysis, the extent of post-initiation crack propagation and thus the level of

*It should be noted that the critical crack size calculated from the Griffith-type equation refers to a total crack length equal to $C = R + s$ in the present model.

strength degradation at critical grain size is believed to be associated with the kinetic energy of the crack and the phenomenon of fast fracture.

The lack of equivalence between the grain size and the pre-existing flaw size has an added consequence in that the grain-size refinement below the inherent flaw size in cubic, isotropic solids may not have any beneficial effect on their strength. Unlike cubic ceramics, where the inherent flaw size responsible for fracture is independent of grain size, in non-cubic ceramics both the pre-existing flaw size and the grain size must be controlled.

As shown in the previous section, an alternative approach for predicting the critical grain size for spontaneous cracking in anisotropic solids, and therefore the critical grain size for an instantaneous drop in strength, is to employ a crack opening displacement concept. The incidence of stress relaxation that is liable to occur when an external stress is applied may be obtained by comparing the critical crack opening displacement (Equation 12) and the linear thermal expansion of a grain (Equation 13) as a function of grain size. This is done in Fig. 9 for polycrystalline titania by substituting appropriate values of $E = 30 \times 10^4$ MPa, $K_{IC} = 2.5$ MPa m^{1/2}, $\nu = 0.22$, $\Delta T = 1200^\circ$ C and $\Delta\alpha = 19 \times 10^{-7}$ ($^\circ$ C)⁻¹ in Equation 10. Examination of Fig. 9 shows that, for flaw sizes (s) in the range 10 to 100 μ m, the grain size at which the critical crack opening displacement becomes equal to the total linear thermal expansion of the grain is between 18 and 46 μ m. These values compare quite favourably with the critical grain size for spontaneous cracking in polycrystalline TiO₂ determined by other means [1, 5]. For example, Kichner and Gruver [1] reported the commencement of microcracking at $D = 20$ to 35 μ m as determined by direct optical observations, and McPherson [26] detected the first microcracks at $D > 8$ μ m by using X-ray techniques. These and the earlier observations of Charvant and Kingery [27], showing some cracking in their TiO₂ with $D < 30$ μ m, are reasonably consistent with the predictions given in Figs 8 and 9.

As Equation 14 and Fig. 9 show, below approximately 18 to 46 μ m grain size the critical crack opening is higher than the total linear thermal expansion of the grain. In this region, the residual stress relaxation will

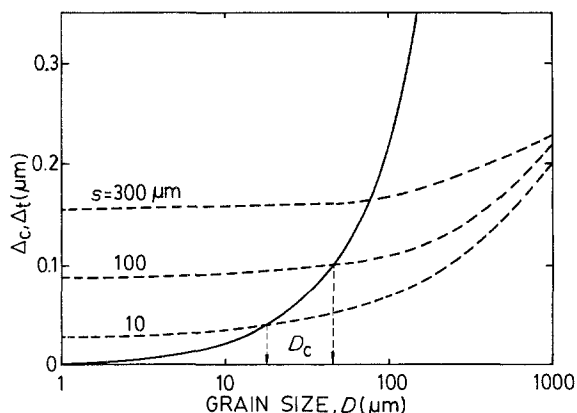


Figure 9 Variation of (---) critical crack opening displacement (Δ_c) and (—) linear thermal expansion (Δ_t) with grain size in polycrystalline TiO₂.

take place simultaneously with opening of the crack, followed by pure elastic extension until the critical crack opening is reached and catastrophic fracture ensues.

In a coarse-grained material, $D > 46$ μ m, the total linear thermal expansion of the largest grain, or of the most favourably oriented grain (Δ_t), is larger than the critical crack opening displacement and a spontaneous crack extension will occur on cooling from high temperature prior to stressing. This process will be followed by residual stress relaxation, leaving an almost stress-free structure. Hence, in both grain-size regions, below and above D_c , a part or all of the residual stresses will be relieved during loading and the overall strength of a solid will be governed mostly by the pre-existing crack length and the grain size as given by the Equation 14.

Clearly the crack opening displacement and therefore the residual stress relaxation process is also determined by the grain orientation and the level of constraints imposed by the neighbouring grains. For example, while the external loading may relieve the residual stresses in those grains under compression, it may simultaneously develop tensile stresses in the neighbouring grains. Nevertheless, the above analysis serves to indicate the importance of crack opening displacement in predicting the critical grain size for spontaneous cracking, and also in interpreting the data on grain-size dependence of strength.

These general findings regarding the role of residual stresses and grain-size dependence of strength appear to be of similar, if not identical, nature in interpreting the data on grain-size dependence of fracture energy. Recent comprehensive fracture studies in cubic ceramics have shown no variation of fracture energy with grain size [28], similar to the grain-size dependence of strength. Non-cubic ceramics, on the other hand, exhibit a rather strong grain-size dependence of fracture energy (γ), with γ passing through maxima that are typically 100 to 400% of the values at fine or large grain size. This variation of fracture energy with grain size was attributed to microcracking induced by the combined effects of the applied stress and the residual stress generated by the thermal expansion anisotropy [28].

6. Conclusions

A micromechanical model is presented to describe the grain-size dependence of strength in anisotropic polycrystalline ceramics. The model considers that a spherical cavity possessing an annular flaw is occupied by a grain of equivalent size. Based on this crack-grain assembly, the stress intensity factor and therefore the fracture strength of a polycrystalline solid is found to be governed by the residual stress concentration arising from thermal expansion anisotropy and the elastic stress concentration which is, in turn, determined by the flaw to grain size ratio. According to the present model, at small grain sizes the inherent flaw size is normally larger than the grain size, whereas at large grain sizes the inherent flaw size (s) is essentially independent of grain size. Before reaching a critical grain size for spontaneous cracking,

the increase of grain size forces s/R to decrease, resulting in a strength degradation at a decreasing rate. When a critical grain size for spontaneous cracking under an applied stress is reached, an instantaneous increase of crack length occurs followed by a decrease of strength in the same fashion. After initiation, the extent of crack propagation before arrest, and thus the severity of strength degradation due to spontaneous cracking, is shown to be governed by the degree of thermal expansion anisotropy and the grain-size distribution.

An alternative criterion for crack extension based on crack opening displacement has also been developed, and tested against experimental data on spontaneous cracking in anisotropic ceramics such as TiO_2 and Al_2O_3 . In addition to predicting the critical grain size for spontaneous cracking, the crack opening displacement concept provides an effective mechanism for stress relaxation operating in the course of external loading.

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