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Internal Strain Energy and the Strength of Brittle Materials

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The theory of Clarke for the formation of grain boundary cracks in anisotropic polycrystalline materials, is re-examined in the light of recent experimental data. The theory predicts correctly the conditions for the formation of grain boundary cracks of length similar to a grain dimension. However, the theory cannot be used to explain the experimentally observed strength/grain size and strength/irradiation dose relationships, for example for BeO. The theory supposes that the process controlling catastrophic fracture is the growth of a crack from a grain boundary pore with an energy absorption rate corresponding to the grain boundary surface energy of $\sim 10^3$ erg/cm^2. In practice, the process controlling catastrophic fracture is the subsequent growth of a crack from a grain dimension, with a higher energy absorption rate corresponding to an effective surface energy of $\sim 10^4$ erg/cm^2.

1. Introduction

The factors controlling the strength of brittle materials are far from clear. Fracture usually occurs in a catastrophic manner by the rapid growth of a crack from some flaw in the material. The initial growth of the crack is governed by the Griffith criterion and occurs when the increase in surface energy, as the crack grows an infinitesimal amount, can just be supplied by a decrease in elastic energy of the specimen plus any work done by the applied stress if the specimen is allowed to elongate. Subsequent crack growth is generally catastrophic because the energy release rate as the crack grows increases much more rapidly than the energy absorption rate due to the formation of new surface. The condition of instability is given by the well-known Griffith equation,

$$\sigma = (2E\gamma/\pi c)^{\frac{1}{2}} \qquad (1)$$

where σ is the fracture strength of the material, E is Young's modulus, γ the effective surface energy, and c is a flaw size related to the structure of the material. Of these variables only the elastic constants are generally available.

This paper is concerned with the strength/grain size and strength/irradiation dose behaviour of single phase polycrystalline ceramic oxides and BeO in particular. Although BeO is inherently a complex material, owing to its anisotropic nature, a study and discussion of BeO does throw light on to the general problem of the strength of ceramics.

2. Some Effects of Crystal Anisotropy

An important consequence of crystal anisotropy is that internal strains are set up around grain boundaries. Two cases are of particular practical importance: the strains produced during cooling after a hot pressing fabrication route, and the strains produced by neutron irradiation. When a freshly fabricated body cools down, strains arise because of differential thermal contraction between various crystallographic axes. At high temperature the strains can be relieved by mechanisms such as plastic flow or grain boundary sliding, but at lower temperatures the strains become "frozen-in" and they build up as the temperature falls further. In BeO, which has hexagonal symmetry, a and c axis contractions of respectively 10.2×10^{-3} and 9.3×10^{-3} occur on cooling from 1020 to 20° C [1]. In the case where the a and c axes of neighbouring grains are parallel, the maximum differential grain boundary strain occurs and equals 0.9×10^{-3} at 20° C, assuming that strain relaxation processes are operative only above 1020° C. Similarly, as a result of neutron irradiation, a and c axis expansions are respectively ~ 0.5 and $3.0 \times 10^{-23}\, \phi$ where ϕ is the neutron dose in neutron/cm^2 $\geqslant 1$ MeV and $\phi \leqslant 10^{20}$ neutron/cm^2 [2]. A maximum strain of $\sim 2.5 \times 10^{-3}$ is thus produced by a dose of 10^{20} neutron/cm^2, and this will be additive to the above thermal strain.

Clarke [3] has analysed the effects of internal strain on the fracture strength. The model, fig. 1, assumes that fracture initiates at a grain boundary pore of diameter c_0 and travels along the grain boundary between A and B where the grain boundary strain is ϵ. The grain size is $2l$. Energy for the growing crack is supplied from two sources: the internal strain field round the boundary, and the strain field due to the applied stress σ. The crack commences to grow from the pore when

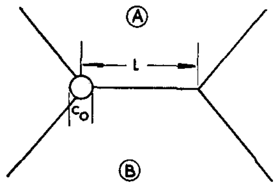


Figure 1 Model for fracture initiation at a grain boundary pore. (After Clarke [3]).

$$\sigma = [\mathrm{E}\{2\gamma_b - \mathrm{E}\epsilon^2(l - c_0)/12(1 - \nu^2)\}/\pi c_0]^{\frac{1}{2}} \quad (2)$$

where E is Young's modulus, γ_b the grain boundary surface energy and ν Poisson's ratio. Equation 2 is essentially the Griffith equation (equation 1) with the surface energy term reduced by an amount equal to the strain energy release rate associated with the grain boundary strain, i.e. the right-hand term in the curly brackets. The analysis also predicts [5] that spontaneous cracking, in the absence of an applied stress, will occur when

$$2\gamma_b = \mathrm{E}\epsilon^2(l - c_0)/12(1 - \nu^2)$$

or

$$\epsilon \simeq (24\gamma_b/\mathrm{E}l)^{\frac{1}{2}} \quad (3)$$

Equation 3 is consistent with two experimental observations [6]: the spontaneous cracking observed in BeO of grain size $\geqslant 120\mu$m [7], and the neutron irradiation doses required to produce cracking in BeO of various grain sizes [8, 9]. A value for γ_b of $\sim 10^3$ erg/cm^2 is indicated in accord with expectations [10].

For grain sizes $\leqslant 100$ μm equation 2 determines the fracture stress provided that the subsequent fracture surface energy for cracks $> l$ does not exceed γ_b. If higher surface energies are required a stress greater than σ may be required to continue the fracture process; this point is discussed below. For grain sizes $\geqslant 100$ μm the strength does not fall to zero as indicated by equation 2, but is assumed to be given by $\sigma = (2\mathrm{E}\gamma_b/\pi l)^{\frac{1}{2}}$ i.e. that expected with a crack length equal to l and surface energy γ_b. These ideas have been applied to explain the form of the experimental strength/grain size relationship in BeO [3].

A similar equation

$$\frac{\sigma_i}{\sigma} = \left\{\frac{2\gamma_b - \mathrm{E}\epsilon_i^2(l - c_0)/12(1 - \nu^2)}{2\gamma_b - \mathrm{E}\epsilon^2(l - c_0)/12(1 - \nu^2)}\right\}^{\frac{1}{2}} \quad (4)$$

where σ_i is the fracture strength after irradiation and ϵ_i is the sum of the irradiation-induced strain and the thermal strain, is proposed to explain strength/neutron irradiation dose curves for various grain sizes [4].

It will be shown below that, in the light of recent experimental data, it is incorrect to use equation 2 in explaining strength/grain size data and equation 4 in explaining strength/irradiation dose data.

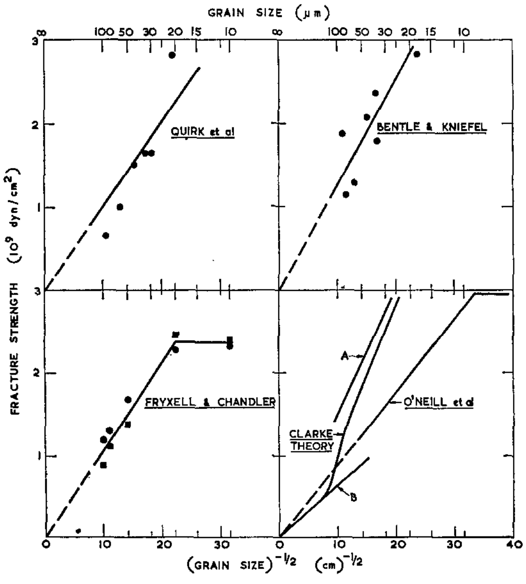


Figure 2 Strength/grain size data for polycrystalline BeO: Quirk *et al* [11], Bentle and Kniefel [12], Fryxell and Chandler [13], O'Neill *et al* [14], theory of Clarke [3]. (The large number of data points of O'Neill *et al* have been omitted for reasons of clarity).

3. The Strength/Grain Size Relationship

Four sets of representative experimental data are given in fig. 2 for various BeO samples of $>95\%$ theoretical density. The top two sets of data can be represented by a single expression of the form $\sigma \propto (2l)^{-\frac{1}{2}}$; in addition, the lower sets of data show a constant fracture stress at small grain sizes. The form of the expression at the larger grain sizes suggests that the strength is controlled by a Griffith type equation with the crack size c proportional to the grain size. However, there are two unknowns in the Griffith equation: the crack size and the effective surface energy. The problem in interpreting the strength/grain size relationship is in choosing correct values for these unknowns.

Clarke [3] uses a crack size equal to the pore size, assumed to be $l/5$, and a γ value of 10^3 erg/cm^2. Following equation 2 the strength at small grain sizes, where the amount of grain boundary strain energy is small, is given by the upper line (A) in fig. 2. But at large grain size the strength should follow the lower line (B), given by a crack size of l and $\gamma = 10^3$ erg/cm^2, since cracks can grow to a grain dimension aided by the grain boundary strain energy at a stress lower than the fracture stress. The strength according to equation 2 is shown as the curve from line B approaching asymptotically line A.

Two comments must be made about this curve. Although the shape of the curve agrees quite well with the earlier data of Quirk *et al*, there is no general agreement when all the recent data are considered. Furthermore the key assumption in Clarke's interpretation, that the appropriate value of the effective surface energy for cracks to grow a distance more than l is still 10^3 erg/cm^2, is not valid. It is known from work of fracture tests [15, 16] that the appropriate value for the effective surface energy of BeO, when integrated over the whole fracture face, is $\sim 10^4$ erg/cm^2. Furthermore we have measured the strain energy release rate at the instant of fracture by a fracture mechanics technique [17], applied to a notched bar deformed in bending, and obtain values for BeO of $\sim 10^4$ erg/cm^2. It seems therefore that the correct value of γ to use for the initiation of the controlling fracture process is 10^4 and not 10^3 erg/cm^2. Some of the reasons for this order of magnitude difference have been discussed [6, 15, 18] and these reflect the increased difficulty of fracture as the initial crack spreads into neighbouring grains and grain boundaries. Thus although internal strain may enable a crack to grow to a grain dimension this is not necessarily the process controlling fracture because the stress may then have to build up to satisfy the higher value of γ.

Carniglia [19] suggests that the critical crack size to use is approximately the grain size. Accepting that the data can be expressed as $\sigma = (2E\gamma/\pi c)^{\frac{1}{2}}$, where c is the critical flaw size, then substituting the value $\gamma = 10^4$ erg/cm^2 indicates that the flaw size is from 1 to 3 grain diameters. In principle it should be possible to detect such cracks in specimens stressed to near the fracture stress but before macroscopic fracture occurs.

4. The Strength/Irradiation Dose Relationship

The objection to the use of equation 4 to explain irradiation behaviour is the same as above, *viz* that the growth of a crack to a grain dimension is not the process controlling the fracture strength. Representative data for neutron irradiated BeO of four grain sizes is given in fig. 3. The dose to produce a fall off in strength increases with decreasing grain size. The doses at which microcracking is first observed is also indicated. The structure of the 70 μm material after three doses is shown in fig. 4 and cracks of roughly a grain dimension occur after a dose of 3.5×10^{19} neutron/cm^2. It is important to note that this cracking is observed at doses where there is

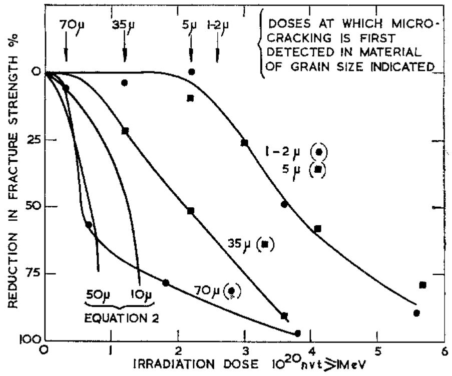
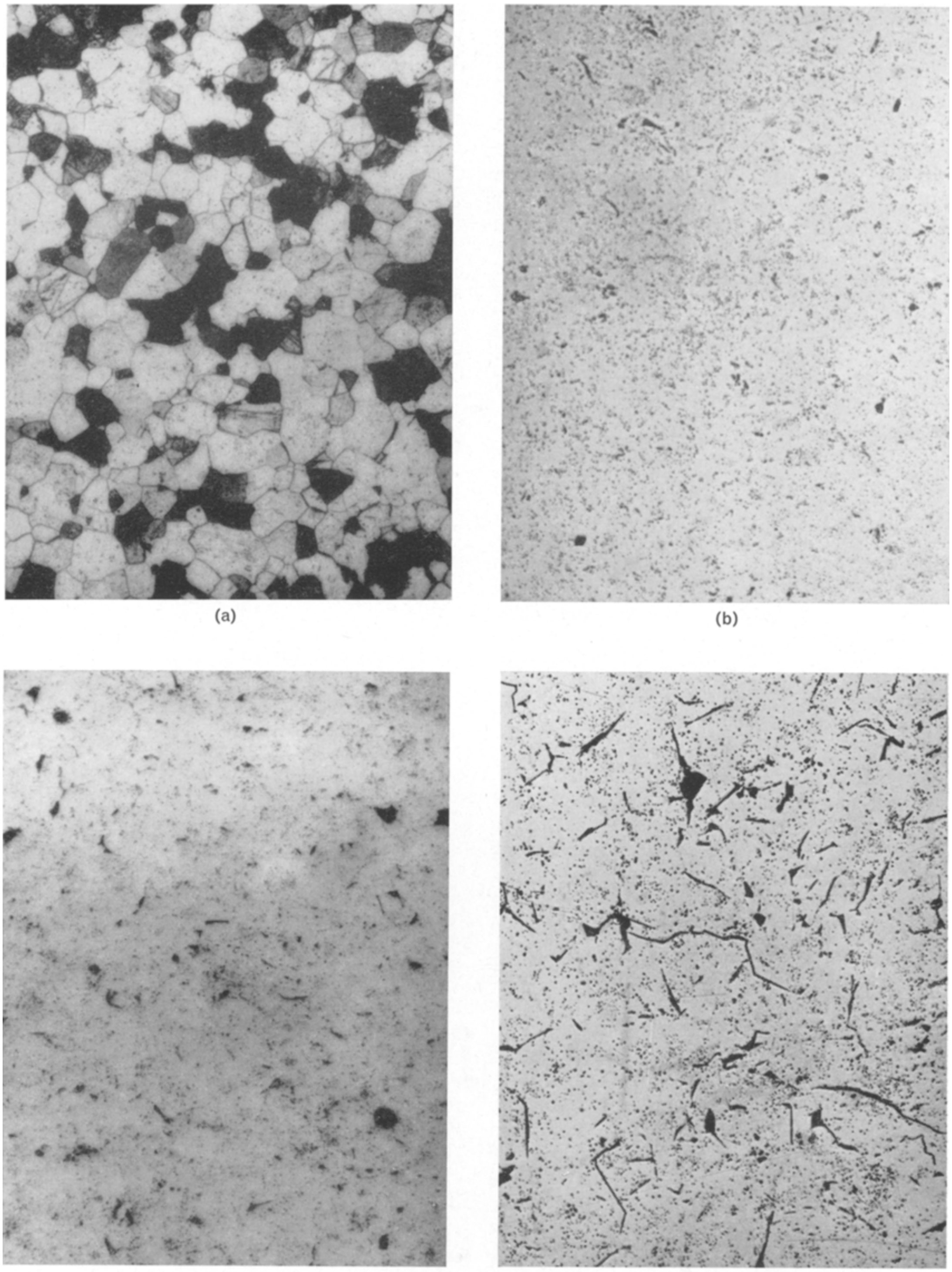


Figure 3 Strength/irradiation dose data for polycrystalline BeO. 70 μm: present work; 35, 5 μm: Collins [9]; 1-2 μm: Hanna *et al* [8]; theoretical curves for 50 and 10 μm [4].



(a) (b) (c) (d)

Figure 4 Structure of 70 μm BeO ($\times$ 90). (a) Unirradiated, etched; (b) unirradiated; (c) irradiated, 3.5×10^{19} neutron/cm^2 $\geqslant$ 1 MeV, cracks of grain size dimension present; (d) irradiated 7.0×10^{19} neutron/cm^2 $\geqslant$ 1 MeV, cracks of several grain dimensions present.

little reduction in strength. Equation 4 implies a marked fall off in strength before cracking occurs and the equation is thus inapplicable. Moreover, the equation predicts a fall in strength at doses that are too low; strength/irradiation dose plots for grain sizes of 10 and 50 μm are given for comparison with the experimental data. The theoretical curve can be fitted to the experimental data by choosing a value for the grain size, if this is unknown, as was done previously [6]. This is clearly inadmissible.

5. Conclusions

(i) Grain boundary strains occur in anisotropic materials, such as BeO, and these lead to grain boundary cracking. The model of Clarke predicts successfully the strain at which cracking first occurs in material of a given grain size.

(ii) There is no evidence to suggest that these internal strains or subsequent cracks up to a grain dimension in length lead to a reduction in strength. This is because the growth of a crack to a grain dimension is not the process that controls macroscopic fracture.

(iii) The growth of a crack, with $\gamma \sim 10^3$ erg/cm^2, to a grain dimension is a necessary prelude to fracture since this produces a Griffith crack of length $\sim l$. Macroscopic fracture, however, occurs only when the Griffith equation is satisfied with $\gamma \sim 10^4$ erg/cm^2.

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