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Grain-size dependence of microcrack initiation in brittle materials

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In this paper, the energy criterion is extended to predict the relationship between the temperature of microcrack initiation (T_{MB}) caused by thermal stressing in brittle materials and the grain size. The relation can be extrapolated to room temperature to provide an estimate of the critical grain size. When the relation was compared to literature data, it was found that: (1) the predicted inverse square root relation of T_{MB} to grain size is satisfied; (2) the room temperature intercept on the grain-size axis agrees well with the measured critical grain sizes. Also presented is a graphical method, based on the proposed relation, by which an engineering estimate of the critical grain size may be made from a minimal set of data.

1. Introduction

During heating and cooling, thermal expansion anisotropy creates stresses between the grains of a non-cubic polycrystalline material [1–3]. For brittle materials such as ceramics, cooling from the sintering temperature often creates sufficiently large stresses to cause internal microcracking [4]; Microcracking can also result from differing thermal expansion coefficients for the phases in a multiphase polycrystalline body or from crystalline phase transformation [5, 6]. This paper, however, will only be concerned with single-phase materials in which phase transformations do not occur.

Upon thermal cycling, materials with thermal expansion anisotropy show hysteresis in linear expansion [7–10], elastic moduli [8–13], strength [14, 15], and thermal diffusivity [16]. The anomalous behaviour is explained by the opening of microcracks during cooling and by crack closure during heating.

A variety of microcracked materials have been studied. Several investigators have showed that thermal expansion anisotropy leads to internal fracture in polycrystalline rutile [17, 18] and graphite [9, 19]. Hunter and co-workers [8, 11–13, 20] studied microcracking in Nb_2O_5, Eu_2O_3, Eu_2O_3–HfO_2, Eu_2O_3–Ta_2O_5, Gd_2O_3, and HfO_2. Kuszyk and Bradt [21] and Cleveland [22, 23] investigated the effect in various pseudobrookites, including $MgTi_2O_5$, Fe_2TiO_5, and Al_2TiO_5.

Hunter [8, 11, 12], Bradt [21, 23], and other investigators [24–26] report that there is a critical size in microcracked materials. It is evident that polycrystalline materials with grains smaller than the critical size do not microcrack while specimens with grains larger than the critical size do.

Currently, the critical grain size is one of the few empirical ways of characterizing the microcracking nature of a given material. Cleveland and Bradt [23] have proposed a method of calculating critical grain size which relies on a single data value per material – the critical grain size – to verify their theoretical work. Thus, if the critical grain size for Fe_2TiO_5 is known to be 3 μm, for example, then the entire model is judged on how well it predicts that single value.

In this paper, the energy criteria of Kuszyk and

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Bradt [21] and the equation proposed by Cleveland and Bradt [23] are extended to predict the temperature of microcrack initiation as a function of grain size over a range of grain sizes. The relation can be extrapolated to room temperature to provide an estimate of critical grain size. The results are compared with published data.

2. Discussion

Consider a thermally anisotropic polycrystalline material at some high temperature where stress relaxation processes are rapid enough to make the body stress-free. As the body is cooled below the temperature (T_0) at which the internal stresses are no longer relieved by such processes as grain-boundary sliding, the internal stresses increase as a result of the thermal expansion anisotropy and crystallographic misorientation across grain boundaries. The stored elastic strain energy will continue to increase on cooling until at some temperature (T_{MB}) the microcracking process begins. When microcracks begin to form some of the stored elastic energy will be converted to the fracture surface energy. It is logical to assume that microcracking will begin at grain boundaries of greatest thermal expansion anisotropy.

Bradt and co-workers [21–23] have addressed the above problem and developed the following expression:

$$(gs)_{\mathrm{cr}} = [14.4\gamma_{\mathrm{f}}/(E\Delta\alpha_{\max}^2\Delta T^2)], \qquad (1)$$

where the constant 14.4 is a function of the modelled grain's geometry, γ_{f} is fracture energy, E is Young's elastic modulus, $\Delta\alpha_{\max}$ is $(\alpha_{\max} - \alpha_{\min})$ where α refers to the single crystal thermal expansion coefficients, and ΔT is the temperature change of interest.

Although the above equation is based on arguments that pertain to the point (temperature) at which microcracks begin, Bradt and co-workers choose to only evaluate the equation at room temperature. By letting $\Delta T = T_0 - T_{\mathrm{RT}}$ where T_0 is defined above and T_{RT} is room temperature they could estimate the critical grain size of the material.

Equation 1 can be extended to relate the temperature of microcrack initiation to grain size. If $\Delta T = T_0 - T_{\mathrm{MB}}$ where T_{MB} is the temperature on cooling where microcracking begins then $(gs)_{\mathrm{cr}}$ in Equation 1 becomes the grain size (gs) of the specimen and Equation 1 can be rewritten as:

$$[T_0 - T_{\mathrm{MB}}] = \left(\frac{14.4\gamma_{\mathrm{f}}}{E\Delta\alpha_{\max}^2}\right)^{1/2}(1/gs)^{1/2} = C(1/gs)^{1/2}, \qquad (2)$$

where C is a constant depending on the material parameters and grain geometry of the specimen under study.

Equation 2 can be rewritten as:

$$T_{\mathrm{MB}} = -C(1/gs)^{1/2} + T_0. \qquad (3)$$

Equation 3 predicts that if T_{MB} is plotted versus $(1/gs)^{1/2}$, the result is a straight line.

Three points should be noted about the above analysis. First, it is assumed that enough stress and potential sites for microcrack initiation are present. Second, it follows from the energy basis of the model that it is a necessary, but not necessarily sufficient condition for fracture. However, the results of Davidge and Green [27], Rice and Pohanta [28], and Cleveland and Bradt [23], as well as the results discussed below, show that this approach is reasonable. Third, it is assumed that T_0 is independent of grain size. This assumption is appropriate within experimental error and is suggested by the data of other investigators [11, 12, 29] (see discussion below).

T_{MB} can be inferred from the Young's modulus versus temperature curve for a microcracked material (see Fig. 1). Specimens below the critical grain size have a linear Young's modulus versus temperature relation on heating and cooling between room temperature and some elevated temperature, where the modulus goes from an unrelaxed to relaxed state.

In contrast, when the grain size exceeds the critical grain size the Young's modulus versus temperature curve exhibits hysteresis and on cooling the curve departs from linearity at some temperature (T_{MB}) above room temperature. This departure has been interpreted as the beginning of microcracking [12, 30]. A review of the data in the literature suggests that T_{MB} is a function of the grain size of the specimens [11, 12], in support of Equation 3.

In addition to Young's modulus, other properties such as linear thermal expansion [21] and thermal diffusivity [16] suggest a T_{MB}–grain size relation.

3. Comparison with literature data

The following literature data supports the above discussion.

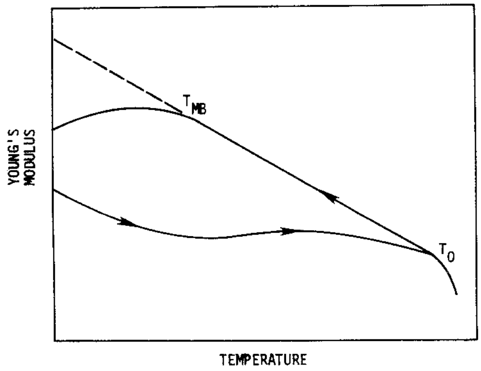

Figure 1 Young's modulus versus temperature (schematic).

A study of monoclinic Eu_2O_3 and Eu_2O_3–HfO_2 has been reported by Suchomel and Hunter [11] in which they measured the temperature dependence of Young's modulus for specimens of grain sizes below the critical size of 8 μm and up to a maximum of 153 μm. They added up to 10 mol% HfO_2 to the Eu_2O_3 as a grain-growth inhibitor, but only the lower HfO_2 content samples of 0, 2, and 4 mol% HfO_2 additions were chosen as trial data for the model. This selection allowed a reasonable number (eleven) of different grain sizes to be included as data without including compositions whose HfO_2 content might appreciably alter the mechanical and thermal properties of Eu_2O_3.

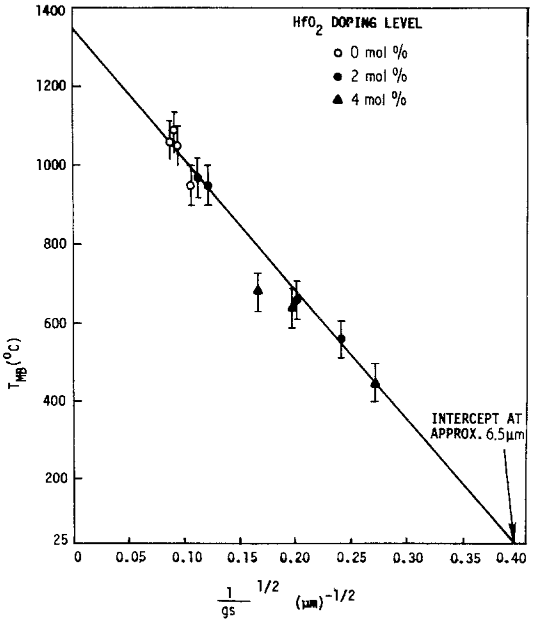

Figure 2 T_{MB} versus $(1/gs)^{1/2}$ for Eu_2O_3 and Eu_2O_3–HfO_2. Data taken from Young's modulus versus temperature study by Suchomel and Hunter [11].

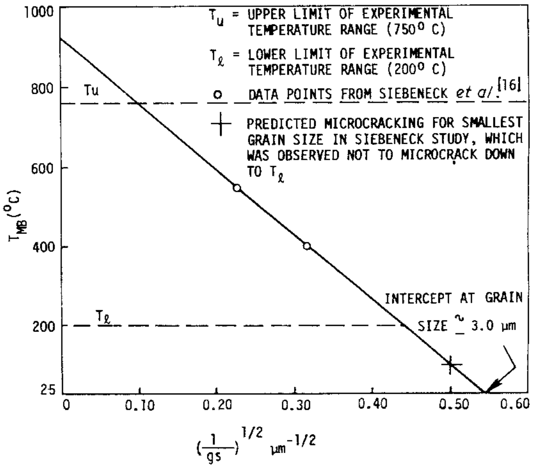

Figure 3 T_{MB} versus $(1/gs)^{1/2}$ for Fe_2TiO_5. Data taken from thermal diffusivity versus temperature study by Siebeneck *et al.* [16].

Suchomel and Hunter reported that the addition of HfO_2 did not appreciably alter the thermal expansion of Eu_2O_3.

Fig. 1 gives a plot of T_{MB} versus $(1/gs)^{1/2}$ for Suchomel and Hunter's data. T_{MB} was inferred from the temperature on the Young's modulus versus temperature cooling curve where the modulus just deviates from what is expected for a non-microcracked body. Error bars represent the estimated ± 50° C uncertainty in the graphical method of determining T_{MB} for each grain size. The data fit the general linear form of T_{MB} versus $(1/gs)^{1/2}$ well.

If the curve in Fig. 2 is extrapolated to room temperature, the grain-size intercept is approximately 6.5 μm. According to the equation, 6.5 μm should be the lower size limit of the grains that crack upon cooling to room temperature. This is essentially the definition of critical grain size, and the 6.5 μm value found from the graph agrees well with Suchomel and Hunter's measured value of 8 μm.

The results of the thermal diffusivity of Fe_2TiO_5 by Siebeneck *et al.* [16] was used also to test the relation. Grain sizes for annealed specimens were obtained from Cleveland [22]. Siebeneck *et al.* interpreted an observed drop in the diffusivity versus temperature on cooling curve as an indication of the onset of microcracking for that grain size. The results of the Siebeneck *et al.* study are shown in Fig. 3, again in terms of T_{MB} versus $(1/gs)^{1/2}$. The smallest grain size (approximately

4 μm) failed to crack at 200° C, the lower limit of the experimental temperature range. This agrees with the graph in Fig. 3, which predicts that a 4 μm grain-sized sample (indicated by + in Fig. 3) should not begin cracking until about 100° C. Again, when the curve is extrapolated to room temperature, the graph gives a critical grain size estimate of 3 μm. Cleveland [22] measured a critical grain size of 3 μm for Fe_2TiO_5.

Thus, in the two studies used to test the proposed relation, it was found that: (1) the predicted linear relation of T_{MB} versus $(1/gs)^{1/2}$ is satisfied; (2) the predicted critical grain size agrees well with the measured critical grain sizes.

4. Graphical method for "engineering estimate" of critical grain size

If the proposed relation holds, then the following graphical method for estimation of critical grain size may be of use.

Two specimens of different grain size are thermally cycled while measuring Young's modulus.* The two temperatures at which microcracking begins (T_{MB}) for the samples are then plotted versus $(1/gs)^{1/2}$ and the resulting straight line extrapolated to room temperature. The critical grain size can then be read directly from $(1/gs)^{1/2}$ intercept value corresponding to room temperature (see Figs. 1 and 2).

Equation 1 proposed by Bradt and Cleveland requires prior knowledge or measurement of γ_f, Y, and $\Delta\alpha_{max}$ for a material to predict the critical grain size. Although modulus values are often available in the literature, γ_f must often be measured for the material under study. This is because γ_f varies with testing techniques, material preparation, etc. The value of $\Delta\alpha_{max}$ requires a high-temperature X-ray diffraction measurement. However, often in the literature only the average value of the thermal expansion coefficients are available, so the engineer would be obliged to do the X-ray diffraction measurements in order to determine $\Delta\alpha_{max}$.

Thus, based on the linear T_{MB} versus $(1/gs)^{1/2}$ relation developed in this paper, an engineering estimate of the critical grain size may be made from a minimal set of data.

5. Conclusions

The energy criteria of Bradt and co-workers was extended to predict a linear relation between the temperature at which microcracking commences and the inverse square root of the grain size for a polycrystalline brittle material. The relation can be extrapolated to room temperature to provide an estimate of the critical grain size. Good agreement was obtained between the results predicted by the proposed model and literature data. Moreover, a graphical method for estimating the critical grain size, based on the proposed relation, was presented.

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* Two specimens would be the minimum needed. A greater number of specimens would yield greater accuracy. Also, in addition to Young's modulus, any other parameter that shows microcrack-induced hysteresis (shear modulus, linear expansion, thermal diffusivity, etc.) could be used.

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