JOURNAL OF MATERIALS SCIENCE 8 (1973) 1759–1764

Creep in non-ductile ceramics

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The effect of a transition in creep behaviour in non-ductile ceramics (i.e. those with limited slip systems available) from diffusion controlled creep, to a mechanism involving non-viscous grain-boundary sliding and localized crack propagation is examined. Localized crack propagation is considered as a transition between diffusional creep and instantaneous fracture, and the fracture strength is used as a guide to predict the conditions necessary for the onset of this high strain-rate creep mechanism. In this way the variation in stress, temperature and grain size dependencies of creep rate reported in the literature for these materials may be explained and experimental evidence in support of the present hypothesis is presented.

1. Introduction

It has been generally accepted that creep in non-ductile ceramics is a diffusion controlled phenomenon at low stresses. Creep mechanisms, proposed by Nabarro [1], Herring [2] and Coble [3], invoke the creation of excess vacancies at grain boundaries under tension, and the diffusion of these vacancies to vacancy depleted compression boundaries. The difference in the mechanisms results from the two possible diffusion paths for the vacancies: lattice diffusion (Nabarro-Herring, $\dot{\epsilon} = (13.3\ D_L\ \Omega/kT)\ (\sigma/G^2)$ or grain-boundary diffusion (Coble, $\dot{\epsilon} = 47.1\ w\ D_B\Omega/kT)\ (\sigma/G^3)$), where $\dot{\epsilon}$ is the strain-rate, σ the stress, Ω the volume of the rate limiting species, D_L and D_B the lattice and grain-boundary diffusion coefficients at temperature T, w is the width of the grain boundary, G the grain size and k the Boltzmann constant.

These mechanisms have been cited to describe the creep behaviour in many ceramics where stress exponents, n, of unity, and activation energies similar to those for ionic self-diffusion, have been experimentally determined. However, stress exponents slightly greater than unity ($1.1 < n < 1.7$) are frequently reported, and Heuer *et al* [4], who have reviewed the literature in this field, have produced direct evidence to show that non-viscous grain-boundary sliding is associated with such stress exponents in alumina. These authors suggested that a transition from diffusional creep to non-viscous grain-boundary sliding was occurring in the grain size range investigated. However, the variation in stress exponent made rigorous interpretation difficult.

Stress exponents of 2 and above have been found (e.g. [5-9]) at high stresses or large grain sizes, or in porous materials. Poteat and Yust [10] reported variations in stress exponent with temperature for thoria from $n = 1.04$ at 1430°C to $n = 1.59$ at 1790°C. Warshaw and Norton [11] observed higher activation energies for creep in large-grained alumina compared with finer grained material. In a ductile ceramic, these phenomena could be explained by a transition from diffusional creep to a mechanism involving the glide and climb of dislocations, but in materials with a limited number of slip systems (e.g. alumina) another mechanism must be sought. In this paper we examine the interrelated effects of stress, grain size and temperature and also the role of localized crack propagation as a stress relieving mechanism that accommodates grain-boundary sliding. A distribution of flaws will be present in a polycrystalline ceramic (e.g., residual porosity, grain boundaries emerging at surfaces) and creep deformation can multiply them. There will exist for the local stress conditions a critical flaw size above which localized crack propagation will occur – as opposed to catastrophic crack propagation and failure.

2. Effect of stress on the creep rate

We will assume, since there is ample supporting evidence, that at low stresses viscous flow is predominant and $\dot{\epsilon} \propto \sigma$. Under these conditions, stress concentrations, arising from fabricational defects (e.g. pores) or from defects produced during creep, will not be sufficiently high to promote crack propagation, and stress relief occurs by diffusional mechanisms. Crack propagation would generally occur when sustained deformation raised the stress at the initiation point to a critical value. Such local stresses would be reached at more localities at higher applied stresses, and localized crack propagation would become an important stress relieving mechanism. This would allow increasing proportions of non-viscous creep strain to occur with increasing stress. The slope, n, of the stress-strain-rate curve would gradually increase with stress up to the fracture stress of the material, when the strain-rate would be extremely high, Fig. 1 (there would thus be a transition stress when n became > 1). The lines in Fig. 1 are spaced assuming that $\dot{\epsilon} \propto G^{-2}$ but this is not critical to the argument that follows. Fig. 2 shows how some previously reported work may be fitted to curves of this sort, the fit being particularly good for the results of Hensler and Cullen [6] and Evans [7]. Experimental scatter, combined with measurement over a limited stress range may result in the assumption of a second straight line regime at higher stresses, with $n > 1$, as previously reported [5-9], instead of the two regimes we propose, in which the $n = 1$ regime gives way to a regime in which $n > 1$ and increases with increasing stress.

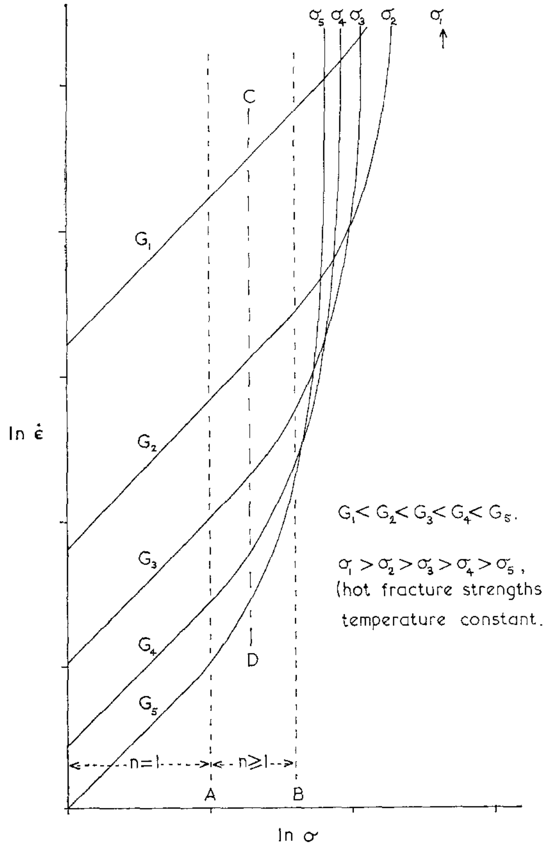

Figure 1 Predicted strain-rate/stress relationship for different grain sizes.

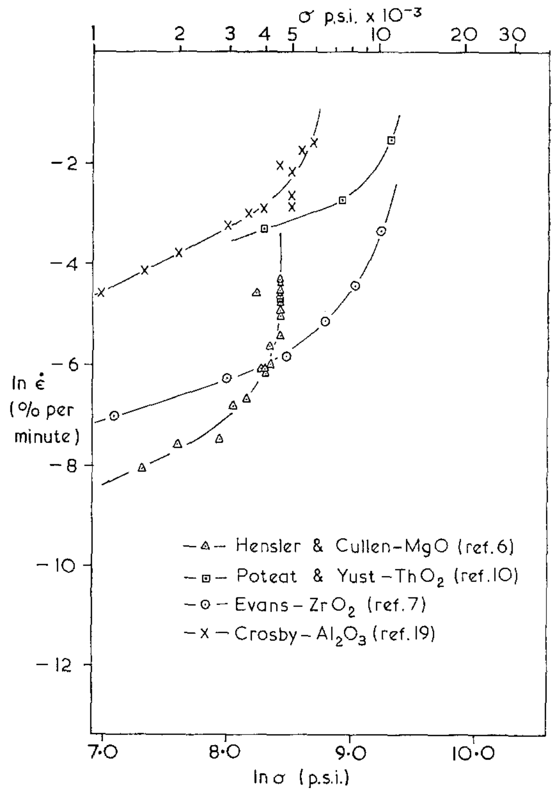

Figure 2 Some strain-rate/stress relationships for previously reported work.

At the transition stresses the microcracks will propagate in an unstable manner, since propagation would be initiated by a stress concentration just in excess of the critical value, and the crack would generally be halted after traversing one boundary facet or one grain. For an *intergranular* crack the resolved stresses would not generally be sufficiently high for transgranular propagation and the resolved stresses along other boundary facet orientations may be too low for continued propagation in an intergranular fashion. For a *transgranular* crack, propagation across a grain boundary without a change in crack direction would in general be energetically unfavourable because of the difference in crystallographic orientation of the crystals. The resolved forces acting on alternative, energetically favourable transcrystalline propagation directions would be too small to propagate the crack, since the initial crack direction would usually be dictated by the severest stress system. Further propagation would occur only when deformation by other processes had created sufficient stress concentrations in an energetically favourable crack direction.

Many such cracks may operate simultaneously throughout the specimen during creep at high stresses. As some cracks are arrested, deformation by diffusional creep or grain-boundary sliding may produce the stress concentrations necessary to propagate other cracks in other parts of the specimen. Thus, a steady state creep process is possible by a combination of boundary sliding, diffusional mechanisms, and localized crack propagation. When the density of cracks in the specimen reaches a limiting value, crack linking will lead to accelerating creep rates and eventual failure.

3. Effect of grain size

Under constant temperature conditions the fracture strength (σ_f) of ceramics generally obeys a Petch [12] or Orowan [13] relationship with grain size, i.e. $\sigma_f = \sigma_0 + k(\bar{G})^{-\frac{1}{2}}$ where $\bar{G}$ is the average grain size, and k and σ_0 are constants, with $\sigma_0 = 0$ for Orowan behaviour and $\sigma_0 > 0$ for Petch behaviour. A similar relationship has been proposed by Knusden [14], where $\sigma_f = k\ (\bar{G})^{-\alpha}$. Carniglia [15, 16] has critically reviewed the application of these equations to ceramic materials and found that the Orowan-Petch equation more closely predicts the grain size – fracture strength relationship. In both equations, the fracture stress decreases with increasing grain size and, since we postulate that the change in stress dependence of creep rate involves localized crack propagation, the transition regime should move to lower stresses at larger grain sizes. Fig. 1 shows how the log strain-rate versus log stress curves for various grain sizes may appear over a wide range of stress, and demonstrates how the stress exponent may vary with grain size when a limited stress range AB is examined. Slight experimental scatter could make the curved portions appear as straight lines with good correlation coefficients. The results of Warshaw and Norton [11], who reported stress exponents of unity at small grain sizes, but at large grain sizes found a stress exponent of 4 (which Coble and Guerard [17] later associated with grain-boundary separation and not transgranular slip) may be interpreted in this manner.

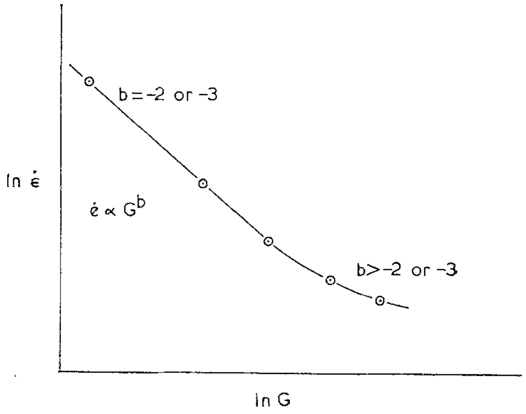

Figure 3 Expected grain size dependence of strain-rate in the transition regime.

When a non-ductile ceramic deforms in such a non-viscous fashion, erroneously low numerical values of the grain size exponent of creep rate may be obtained. A stress level corresponding to the line CD in Fig. 1 for instance gives a log $\dot{\epsilon}$ versus log G curve as in Fig. 3. Here the numerical value of the gradient decreases with increasing grain size at large grain sizes. Experimental scatter could make such a curve appear to fit a straight line with a lower grain size exponent than that for a diffusion controlled process. Under certain conditions of stress and grain size the decrease in grain size exponent could lead to strain-rates independent of grain size.

The currently accepted models for diffusional creep give grain size exponents of -2 (lattice diffusion) or -3 (grain-boundary diffusion). An exponent between these values could, therefore, indicate a combination of the two diffusional mechanisms and/or the introduction of localized crack propagation. A grain size exponent numerically less than 2 would be a strong indication of the crack propagation mechanism supervening in non-ductile ceramics.

4. Effect of temperature

For a diffusion controlled mechanism the creep rate is strongly temperature dependent, and obeys the relationship $\dot{\epsilon} \propto e^{-\Delta H/RT}$, where ΔH = activation energy for creep (or diffusion). The introduction of a non-diffusional creep accommodation mechanism would alter the temperature dependence of creep rate, which will be governed by the effect of temperature on the rate allowed by the new mechanism. An increase in temperature could slow down the rate of crack propagation because of an increase in the proportion of energy absorbed by plastic flow at the crack front (although in ceramic materials with limited slip systems this may not be significantly altered). However, the rate of accommodation of boundary sliding should not alter appreciably since the reduction in accommodation by crack propagation would be compensated by the increase in accommodation by plastic flow at the crack tip. The temperature dependence of strain-rate would then be controlled largely by the variation in sliding rate with temperature, which will be governed by the variation in intrinsic viscosity of the boundary regions with temperature. Thus, one would expect an increase in the strain-rate with increasing temperature owing to non-viscous boundary sliding and localized crack propagation. In the transition region, diffusional mechanisms will also be important, and the total strain rate will equal the sum of the components due to each mechanism. This would lead to the apparent activation energy increasing with T beyond the transition region.

The shape of the log $\dot{\epsilon}$ versus $1/T$ curve may also be deduced by considering the known effect of temperature on the fracture strength of a ceramic, i.e. a reduction in strength with increasing temperature. Thus, the transition in stress dependence of strain-rate should occur at lower stresses at higher temperatures since we invoked localized crack propagation as an accommodation mechanism that allows of increased strain-rates, and the log $\dot{\epsilon}$ versus log σ curves for various temperatures may be represented as in Fig. 4a. A stress level represented by the line AB in Fig. 4a would give the temperature dependence shown in Fig. 4b. If the temperature were restricted to this transition regime, and a straight line relationship were assumed, a measured value of activation energy higher than that for a diffusion controlled process would be observed. It seems likely that the higher activation energies observed by Warshaw and Norton [11] for large grain alumina (185 kcal mol^{-1} compared with 130 kcal mol^{-1} for fine grained material) resulted from measurements taken in this transition region. This is supported by the high stress dependence (n = 4) for the large grained alumina, and later work by Coble and Guerard [17].

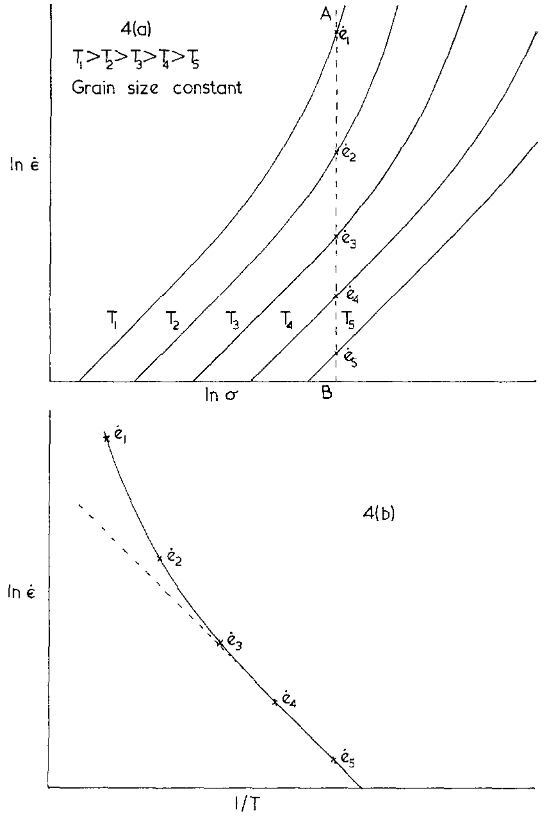

Figure 4 Possible variation in measured activation energy with temperature.

5. Effect of porosity or second phase

The presence of pores or a weaker second phase reduces the hot fracture strength of a ceramic. Pores act as stress concentrators and localized crack propagation could occur at lower values of applied stress than in a fully dense material. This would explain the results of Fryer and Roberts [18], who observed stress exponents of 2 and 3 for two aluminas with porosities 8 to 9% and 34 to 35% respectively. They also observed activation energies of 120 and 185 kcal mol^{-1} respectively for the two materials, and this difference can also be explained by the ideas presented here.

A microcrack would propagate more easily in or around a weak second phase particle than in the neighbouring stronger phase, and this too would lead to accommodation of sliding at lower stress values. A stronger second phase material would reduce the proportion of transcrystalline cracking, but initiation at triple points would still lead to the development of grain-boundary cracks. If the bonding at the phase interface were weaker than the grain boundaries in the single phase material, then localized crack propagation could again occur at lower stress values.

6. Experimental evidence for localized crack propagation

In addition to the evidence in favour of localized crack propagation already reported in the literature, further evidence came to light during our work on the creep of alumina. Both pure and two-phase nickel-doped alumina were used in the study and further material and experimental details are reported elsewhere [9, 19]. Metallography revealed an association between high stress exponents and localized crack propagation [7, 9, 19], and evidence of boundary sliding in the form of boundary corrugations.

In creep tests on large grained pure alumina ($\bar{G}$ = 45 μm) at 3000 psi. (20.7 MN m^{-2}) and at temperatures from 1575 to 1700°C, there was considerable scatter in the results from different specimens. This shows that a large grain size, with the consequent bigger local cracks that can occur, makes the strain-rate particularly structure sensitive. Tests, wherein single specimens were strained at a series of temperatures in this range, gave little scatter in the log $\dot{\epsilon}$ versus $1/T$ plots and, therefore, support this argument. The temperature range was extended up to 1800°C, to see how the introduction of localized crack propagation affected the activation energy for creep at higher temperatures (Fig. 5). The data for the temperature range 1650 to 1800°C gave an activation energy for creep of 153 ± 15 kcal mol^{-1} by a least mean squares fit. However, when the temperature ranges 1650 to 1725 and 1725 to 1800°C are considered separately, activation energies of 122 ± 24, and 209 ± 24 kcal mol^{-1} respectively, are obtained. The value for the lower temperature range agrees, within experimental error, with our previous results for alumina [9] and with those generally obtained for creep in alumina. The value for the higher temperature range is nearly twice this figure, and emphasizes a change in creep mechanism. The data are probably best fitted (as in Fig. 5) by a curved line at high temperatures with a straight line emerging beyond the transition zone at intermediate temperatures. The general pattern of these results confirms the temperature dependence predicted in Section 4.

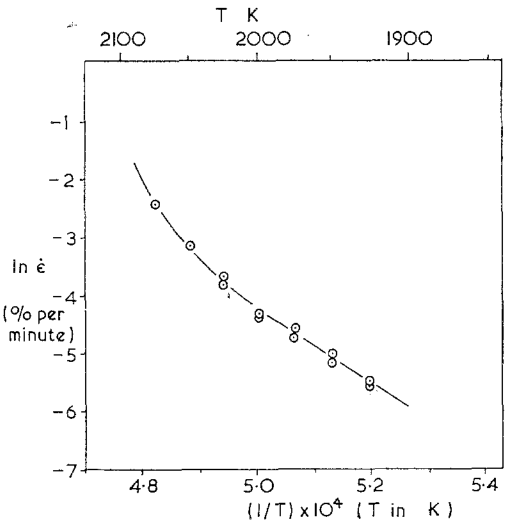

Figure 5 Temperature dependence of strain-rate for 45 μm alumina at 3000 psi.

The two-phase nickel-doped specimens used in this study were particularly susceptible to localized crack propagation, especially at large grain sizes when the weaker, spinel, phase allowed crack propagation at much lower stresses than the corresponding pure specimens. This may have caused the high values of activation energy (185 kcal mol^{-1}) previously reported [9].

Localized crack propagation can lead to erroneous values of temperature corrected strain rate ($Z = \ln\dot{\epsilon} + \Delta H/RT$) when an average activation energy value is employed, as illustrated by a plot of Z versus $1/T$ in Fig. 6. If a single creep mechanism is dominant over the temperature range investigated this should be a straight line parallel to the temperature axis. However, there is a marked deviation at higher temperatures from a constant value of Z and the use of this parameter outside the diffusion controlled creep regime is not, therefore, an acceptable method of temperature correction.

7. Conclusions

1. Localized crack propagation in non-ductile ceramics could accommodate significant amounts

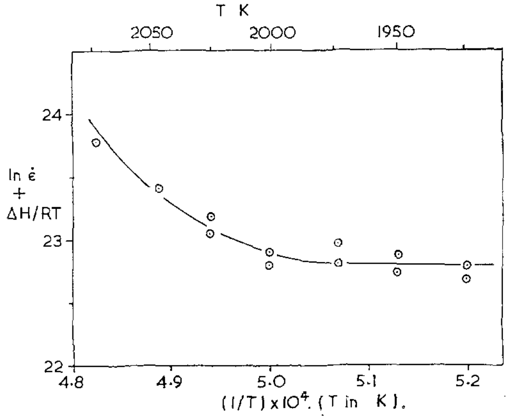

Figure 6 Effect of temperature on the "temperature-corrected" strain-rate in the transition regime, for 45 μm alumina at 3000 psi.

of non-viscous grain-boundary sliding during creep.

2. This mechanism may be extremely important during creep at high stress or temperatures, and in large grained, two-phase, or porous materials.

3. If the creep strain-rate increases in a continuous manner with increasing stress up to the strain-rate at the hot fracture strength of a ceramic, then the wide variations in stress exponents reported in the literature can be explained.

4. Accommodation of grain-boundary sliding by localized crack propagation can result in high measured values of activation energy for creep. This is substantiated by our own work and by results reported in the literature.

5. When localized crack propagation occurs the grain size exponent in the creep rate equation may be numerically lower than the value expected for a diffusion controlled mechanism.

6. The application of an average activation energy value in the determination of the temperature corrected strain-rate, over a range of temperatures where localized crack propagation occurs, can give misleading results.

7. More work is needed to determine experimentally the shapes of the creep rate curves over wide ranges of stress, grain size and temperature. This must involve many individual tests in the high strain rate regime, where specimen structure may cause considerable scatter in the results.

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Received 10 May and accepted 25 June 1973.