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High temperature creep behaviour of 4 mol% yttria tetragonal zirconia polycrystals (4-YTZP) with grain sizes between 0.38 and 1.15 μm

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Abstract

The high temperature mechanical behaviour of 4% mol yttria tetragonal zirconia polycrystals (4-YTZP) with different grain sizes ($0.38 < d < 1.15\ \mu m$) has been analyzed by means of compression creep tests. The working temperature was 1350 °C and the strain rates ranged between 5×10^{-7} and $2 \times 10^{-4}\ s^{-1}$. Experimental results have been fitted to the conventionally accepted creep law for superplastic ceramics. Thus, stress exponents and activation energies have been measured as a function of the grain size. The dependence of strain rate on grain size has also been determined. The experimental data are discussed with respect to the existing theoretical models for these materials.

Keywords: Creep; YTZP polycrystals

1. Introduction

It is a well-known experimental fact that yttria partially stabilized zirconia polycrystals exhibit a remarkable superplastic behaviour at moderate temperatures. Since this crucial phenomenon was reported,[1] a vast scientific effort has been carried out to achieve a further comprehension of the ultimate reasons for such mechanical behaviour. Controlling superplasticity leads to potential applications, which were anticipated at the pioneer times of ceramic superplasticity and are now becoming a reality, like superplastic forming or superplastic joining.[2–7] From a fundamental point of view, there are still some gaps in our common understanding of the intimate mechanisms accounting for superplasticity in YTZP. Although it is an accepted fact that it is a consequence of grain boundary sliding under steady-state conditions, i.e. under microstructural invariance, the accommodation mechanisms are still far from being completely elucidated. More precisely, the grain size dependence of the mechanical response from the nanometric range up to the micrometric one is only partially explained.

All existing models take as starting point the conventional constitutive creep equation[8]:

$$\dot{\varepsilon} = A\frac{Gb}{kT}\left(\frac{b}{d}\right)^{p}\left(\frac{\sigma}{G}\right)^{n} D_0 \exp\left(-\frac{Q}{kT}\right) \quad (1)$$

where $\dot{\varepsilon}$ is the strain rate, A dimensionless constant, G the shear modulus, b the magnitude of the Burgers vector or any characteristic length scale in the material, k the Boltzmann's constant, T the absolute temperature, σ the applied stress, and d is the average grain size. The term D_0 is the frequency factor of an appropriate diffusion coefficient, whose Arrhenius dependence includes the corresponding activation energy Q. This diffusion coefficient is the one for the migrating species involved in the accommodation process. The parameters p and n, respectively the grain size and the stress exponent, as well as the activation energy, Q, and the microstructural changes after deformation are the fingerprints of possible deformation mechanisms.

In the case of YTZP's an empirical law is reported[9] to fit all experimental data between 1250 and 1450 °C (the temperature domain for superplasticity with negligible grain growth), provided the grain sizes are in the range 0.3–0.5 μm. This empirical law has the form:

$$\dot{\varepsilon} = 3 \times 10^{10}\frac{(\sigma - \sigma_0)^2}{Td^2}\ \exp\left(-\frac{460 \pm 40\,\mathrm{kJ/mol}}{RT}\right) \quad (2)$$

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This expression contains a threshold stress σ_0, which is given by[9,10]

$$\sigma_0\,(\text{MPa}) = 5 \times 10^{-4}\,\frac{\exp((120 \pm 30\,\text{kJ/mol})/RT)}{d\,(\mu\text{m})} \quad (3)$$

with σ in MPa and d in μm.

The physical basis for interpreting such a law has been the subject of an intense debate.[11–13] A critical review of the different approaches is reported in literature,[14] as well as the range of validity of this empirical law and its extension to nanostructured YTZP specimens.[15] Quite recently, two theories have been proposed to account for the physical origin of the threshold stress.[15,16] One of them invokes dislocation activity,[16] whereas, the other one is based upon yttrium segregation at the grain boundaries.[17] It seems that the cation segregation to the grain boundaries has revealed as an essential issue for a correct modelling of the mechanical behaviour in this system.

Fundamental explanations are still lacking for some experimental facts, for instance, the reason why the stress exponent seems to depend on the grain size[18] and a possible dependence of activation energy on grain size. Both questions ask for a further insight into the details of the mechanisms controlling superplasticity. The present work tries to provide some clues through an exploration of the changes in stress exponent, activation energy and grain size exponent in different YTZP specimens with different grain sizes ranging between 0.38 and 1.15 μm.

2. Experimental procedure

Fully dense 4 mol% Y_2O_3–ZrO_2 ceramics were fabricated by conventional sintering at 1550 °C during 2 h. Samples were cut into parallelepipeds of (2 mm × 2 mm × 4 mm) and polished for mechanical testing in air and at high temperature. These samples were deformed at constant load (prototype machine),[19] at stresses between 10 and 300 MPa and a temperature of 1350 °C. Several tests have also been performed between 1350 and 1400 °C to determine the activation energy. In order to study the influence of grain size on mechanical behaviour, specimens were annealed at 1550 °C in air for various periods of time to obtain different grain sizes. An analysis of the grain growth laws in these specimens can be found in literature.[20]

The microstructure of the specimens prior to and after deformation was characterized using Scanning Electron Microscopy (SEM). Grain size statistics was obtained using image analysis software (Zeiss Kontron Videoplan) from the scanning electron micrographs, where no less than 350 grains were used for each determination.

3. Results

3.1. Microstructure

Static grain growth annealing of the specimens prior to deformation was carried out at 1550 °C and for different annealing times to obtain a set of specimens with grain sizes ranging from 0.38 up to 1.15 μm.[20] The microstructure of these specimens is shown in Fig. 1, where equiaxed grains can be observed. The grain size histograms follow a lognormal distribution. It is important to emphasize that the "mean grain size" is calculated by the mean intersect line method with no numerical factor.

3.2. Creep tests

A typical plot of strain rate versus strain during creep testing under successive stress values is shown in Fig. 2. The stress exponent and activation energy were calculated through changes in stress or temperature, respectively, at constant microstructure.

The stress exponents vary with increasing stress from 3.5 to an asymptotic value of 2.0 as shown in Fig. 2. This result is common to all the specimens with grain sizes in the interval 0.38–0.60 μm, which are, from now on, denoted type-I specimens. A different behaviour is obtained for specimens with larger grain sizes ($0.75 < d < 1.15$ μm), which we will call type-II specimens. There seems to be a correlation between the stress exponent and the grain size: the larger the grain size, the smaller the stress exponent is, with a negligible influence of the applied stress. This is illustrated by Fig. 3, where a plot of strain rate versus stress is shown for the full set of tested specimens. It is possible to visualize the difference in behaviour between type-I specimens (curved plots) and type-II ones (straight lines). The slopes of those straight lines are the average stress exponents.

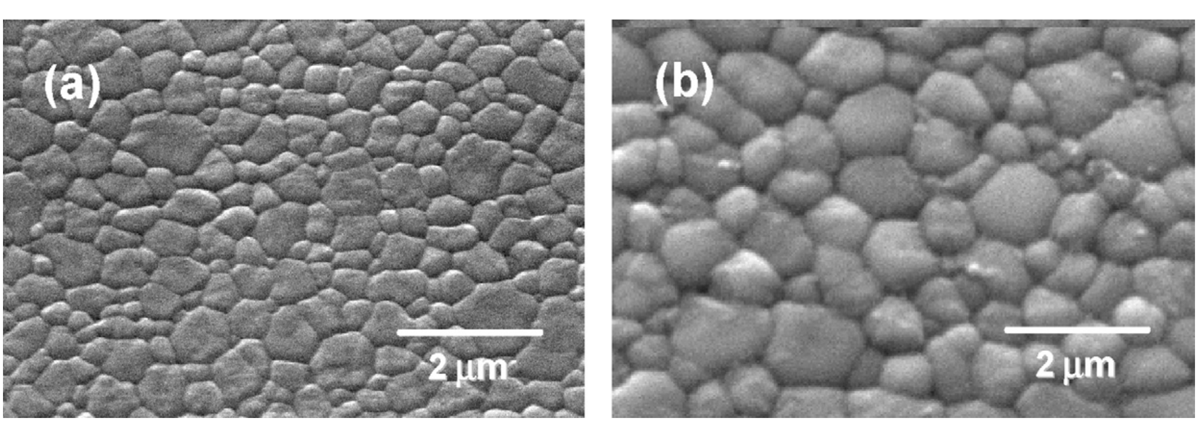

Fig. 1. (a) Micrograph of as-received YTZP specimen with a grain size $d = 0.38$ μm (b) After annealing at 1550 °C, the grain size is $d = 1.15$ μm.

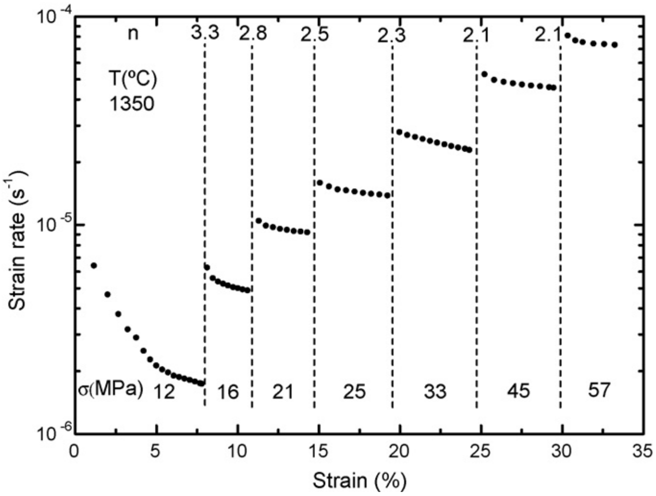

Fig. 2. Creep curve (strain rate vs. strain) for a specimen with $d = 0.38\ \mu m$ (cf. Fig. 1a).

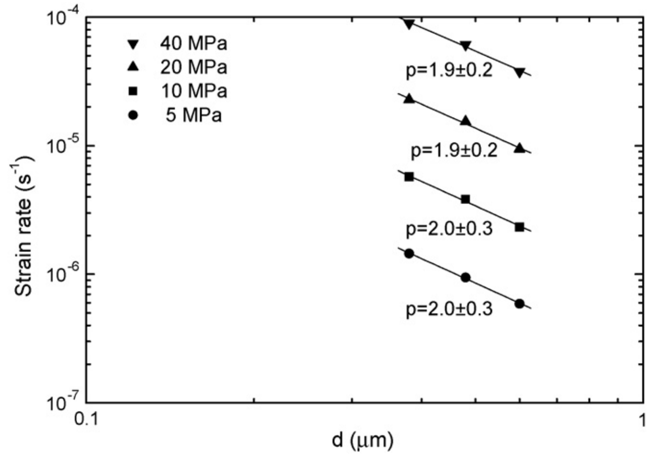

Fig. 4. Strain rate vs. grain size for different values of the applied stress at a constant temperature (1350 °C). Only data for type-I specimens are shown for the sake of simplicity, since a linear plot is expected to fit all data. Nevertheless, a grain size exponent of $p = 2$ is obtained for both types of specimens.

The tendency towards lower stress exponents for larger grain sizes is also displayed.

Concerning the activation energy, our measurements give a value of $Q = 520 \pm 70$ kJ/mol, regardless of the grain size. Thus, no dependence on grain size was obtained.

In order to determine the grain size exponent p, the steady state strain rate is plotted as a function of the grain size for various stresses and temperatures. The results for type-I specimens are shown in Fig. 4. The best fit to these data yields $p = 2.0 \pm 0.3$ for all the type-I samples. Data on type-II samples are not included in this figure because the stress exponent is a function of the grain size and the numerical analysis requires performing a non-linear fit. Such numerical analysis shows that a constant value $p = 2$ is consistent for all the samples within experimental accuracy.

In summary, the grain size exponent and the activation energy do not depend on the grain size. However, a clear-cut dependence of the stress exponent on grain size was found. As will be discussed below, this dependence can be explained in terms of a change in the topological motion of grains.

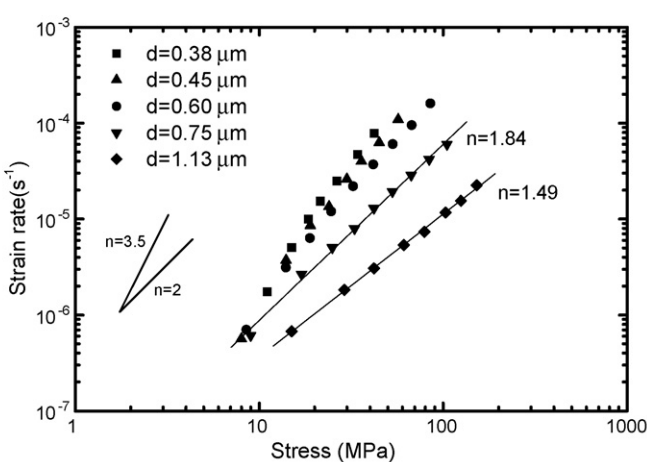

Fig. 3. Strain rate vs. the stress at constant temperature for the full set of investigated samples. Notice that the samples with the smallest grain sizes exhibit a non-linear dependence (type-I behaviour). In contrast, the samples with the largest grain sizes exhibit a linear dependence (type-II behaviour).

3.3. *Microstructural characterization*

No significant grain growth was observed in any of the deformed samples. The grain size distribution analysis revealed a good fit to a lognormal distribution with equiaxial grains. The microstructure of the type-I samples is fully invariant with respect to that of the un-deformed samples. After deformation, no noticeable trace of damage, such as cavitation, could be observed.

Regarding type-II samples, possible changes of the grain size and form factor were carefully investigated, but no grain growth or change in the aspect radio of the grains could be detected.

4. Discussion

A separate treatment is required for the two types of mechanical responses corresponding to type I and type II specimens. Regarding type-I behaviour, an accurate analysis of the stress exponent versus strain rate in terms of a threshold stress shows that this exponent is independent of the strain rate. Indeed, such threshold stress-induced effects had been reported in literature. The threshold stress for each specimen of type-I has been calculated according to Eq. (2), and a plot of the strain rate versus the effective stress (the applied stress minus the threshold stress) yields a good fit to straight lines with a slope of 2 (Fig. 5). Such a behaviour is the one predicted by Eq. (3) and the model[9,10] for fine-grained specimens.

For type-II specimens, a different approach must be adopted. It is obvious that threshold stress effects must have no significant influence on the mechanical response, since the threshold stress is inversely proportional to the grain size and reaches negligible values for all type-II specimens. The origin of the smooth dependence of the stress exponent with the grain size must have

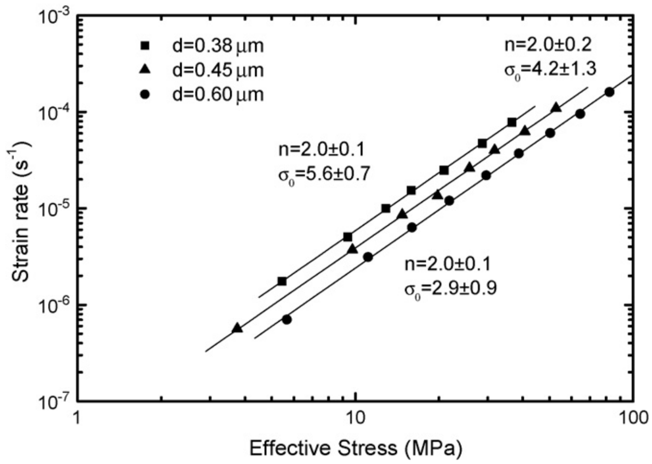

Fig. 5. Strain rate vs. the effective stress for type-I samples. The fit to a parabolic dependence is fairly good, in agreement to a model developed in the literature.[9]

a fundamental basis and two alternative explanations can be proposed.

Grain boundary sliding obviously becomes more and more limited with increasing grain size, and higher stresses are required to obtain the same strain rates. These conditions favor diffusional creep with respect to grain boundary sliding. As a consequence, the actual deformation mechanism is likely to be a superposition of grain boundary sliding and diffusional creep, the latter being all the more important as grain size increases. Since diffusional creep is characterized by a stress exponent $n = 1$, intermediate values between 1 and 2 are expected and are, indeed, experimentally obtained. This explanation should, however, entail some changes in grain shapes during deformation, which is in full contradiction to the present observations.

The second possible explanation is as follows. It is well known that grain boundary sliding is compatible with a stress exponent equal to 1 in other systems than the present one, particularly in metals. The Ashby–Verrall model[21] makes strong assumptions on the grain boundaries shape and predicts such a value for superplastic deformation. The conditions leading to any of the two values $n = 1$ or 2 for the superplastic response are unknown. There are examples showing that $n = 1$ in NiO polycrystals with grain sizes between 9 and 21 μm,[22] and in UO_2 with grain size between 2 and 10 μm[23]; both of these ceramics deform by grain boundary sliding. In the case of YTZP ceramics, values of $n = 1.5 \pm 0.3$ have been reported for a grain size of 1.8 μm,[18] whereas, for the grain sizes between 0.3 and 0.6 μm, probably the most widely studied YTZP ceramics, values of n between 2 and 5 have been reported.[9] Quite recently, Tekeli et al.[24] has reported stress exponent values to be 1 in TiO_2-doped 8 mol% Y_2O_3 stabilized ZrO_2 under tension and compression. The grain size value of their specimens is over 1 μm, according to their displayed micrographs. Several authors have found similar behaviour in YTZP specimens, their references being collected by this last paper.[24]

These results show that a same mechanism can be characterized by different n values, suggesting that only grain dynamics may be dependent on grain size. A strong support to this statement arises in the present work from the fact that p and Q exhibit no dependency on the grain size. The values $p \approx 2$ and $Q \approx 520$ kJ/mol are in full agreement with those reported in the literature, within the experimental uncertainty, for lattice cationic diffusion as the accommodation mechanism for plasticity.[9,14] This is clear evidence that no change in mechanism is occurring in our set of samples: the accommodation mechanisms are the same, but the grain dynamics is modified for the larger grain sizes. Indeed, grain boundary profiles like the one described in the model by Ashby and Verrall may occur more likely when the grain sizes increases than when it decreases. At present, there is no theoretical analysis explaining how grain topology can influence the stress exponent values, a task that is now under development.

5. Conclusions

A set of 4 mol% YTZP specimens with different grain sizes have been crept at high temperatures within the temperature and strain rate domain in which superplastic behaviour is expected to occur. The mechanical results show a dependence of the stress exponent versus the grain size, whose origin may be linked to a change in grain dynamics for larger grain sizes.

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