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The influence of grain size on the creep of uranium dioxide

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The creep of uranium dioxide has been investigated as a function of grain size. At high stresses, when creep is controlled by dislocation movement, grain boundaries exert a strengthening effect and this strengthening is correlated with the Hall-Petch equation. The degree of strengthening diminishes with increases in temperature. At lower stresses, when creep is controlled by mass transport, grain boundaries exert a weakening effect owing to the reduction in diffusion path length as grain size is reduced. In this range behaviour is correlated with the Nabarro-Herring equation with stress σ replaced by an effective stress $\sigma_E = \sigma - \sigma_0$ where σ_0 is a threshold stress for diffusional creep associated with the limitation of the ability of boundaries to emit and absorb vacancies. σ_0 appears to decrease as grain size is increased.

1. Introduction

The creep rate of polycrystalline materials is generally not so strongly influenced by changes in grain size as it is by changes in temperature and stress. Nevertheless, the influence of grain size changes can be important although there is relatively little reported work on this aspect especially in ceramic materials.

The region where the influence of grain size is most fully understood is at low stresses when creep is controlled by mass transport from grain boundaries under compressive stresses to those under tensile stresses. In this region, firm theoretical foundations exist which predict creep rate $\dot{\epsilon} \propto 1/d^2$ or $\dot{\epsilon} \propto 1/d^3$ where d is the grain size, depending whether mass transport is predominantly occurring through the lattice [1, 2] or along grain boundaries [3]. The $1/d^2$ dependence arises because the vacancy flux j_V across a grain is proportional to the reciprocal of the diffusion path length which is about equal to d. Since the arrival of each vacancy represents the transport of an atomic volume Ω of material, a layer of material of thickness $\dot{\epsilon}d = j_V\Omega$ builds up per second on tensile boundaries thus giving $\dot{\epsilon} \propto 1/d^2$. This relationship has been verified experimentally in pure metals [4-7] and some oxides [8-11].

When grain-boundary diffusion predominates, the available area for diffusion across any plane is reduced by the factor w/d where w is the grain boundary width and $1/d$ is the length of boundary intersected by a unit plane. Thus, a further factor, w/d, arises in this case giving $\dot{\epsilon} \propto 1/d^3$. Thus a transition from a $1/d^2$ to a $1/d^3$ dependence of creep rate may be expected at lower temperatures or for finer grained material when grain-boundary diffusion should predominate. Such a transition has experimental support [6, 7].

At higher stresses when deformation is controlled by dislocation movement the understanding of the influence of grain size on creep is not so well developed theoretically and is comparatively little studied experimentally. However, it is clear that the influence which boundaries exert is a function of the temperature. At low temperatures boundaries can act as barriers to crystallographic slip [12, 13] and grain-boundary sliding is unimportant, whereas at higher temperatures, the barrier effect of boundaries diminishes and sliding at significant rates becomes possible. Additionally, at higher temperatures, dislocation creation and annihilation may occur more easily at grain boundaries giving rise to lower creep strength. Typical of the experimental studies in this range is that of Barrett *et al* [14] on pure copper at 770K who found that creep rate decreased with increasing grain size up a to grain size of ~ 100 μm. Creep rate remained constant with any further grain size increase. The faster rates at fine grain size were interpreted in terms of contributions by grain-boundary sliding. Similar behaviour has also been observed in Cu-30% Zn at 723K by Dunlop and Taplin [15] who noted that the flow stress was relatively independent of grain size at large grain size but dropped sharply at fine grain size. These observations were again interpreted in terms of grain-boundary sliding. Somewhat different behaviour has been noted however by Garofalo *et al* [16] in an austenitic alloy and by Shahinian and Lane [17] in monel. In these alloys a U-shaped dependence exists between creep rate and grain size with both fine and large grained material showing lower creep strength. The minimum in the curve occurred at a grain size of about 100 μm in both cases. Garofalo *et al* interpreted these observations in terms of the competition between a Hall-Petch barrier hardening effect at lower temperatures which causes an increase in creep strength as grain size is reduced and the increased ease at which dislocations can be created and annihilated at grain boundaries at higher temperatures. This latter process will lead to a decrease in creep strength as grain size is reduced. It has been pointed out, however, by Sherby and Burke [18] that the decreased creep strength of these alloys at larger grain sizes may be associated with differences in solute and precipitate distributions, caused by the various thermal treatments used to obtain a range of grain sizes.

2. Experimental techniques

The material used in this investigation was reactor fuel grade UO_2 produced by UKAEA. It was received in the form of 14 mm right cylindrical pellets having a 5 mm hole down the centre. The as-received grain size was 7 μm and the residual porosity ~ 3%. Various grain sizes were obtained by performing grain growth anneals on a pellet and machining out creep specimens after various times. This technique was adopted rather than annealing pre-machined specimens in order to minimize surface contamination during annealing. The annealing furnace atmosphere was hydrogen and temperatures of 1873 and 1973K were used. No systematic differences in behaviour could be attributed to differences in the temperature at which grains were grown.

The measurement of the compressive creep rate of these specimens was performed in a hydrogen atmosphere in a way that has been previously described [19] and will not be repeated here. At higher stresses (> 70 MN m^{-2}) and faster creep rates ($> 10^{-2}$ h^{-1}) it was more convenient to use an "Instron" machine fitted with a high temperature compression testing facility. In this apparatus it was necessary to use an argon atmosphere since heat losses from the element in hydrogen created difficulties in temperature control.

Grain sizes were measured after testing using standard metallographic techniques and grain sizes quoted are mean linear intercepts.

3. Results and discussion

The relationship between creep rate and stress for UO_2 at 1623K is shown in Fig. 1 for material of three different grain sizes. Two regimes of creep behaviour exist depending on stress. At high stresses creep rate depends strongly upon stress, typical of deformation controlled by dislocation movement. In this region, in sharp contrast to observations on metals, the fine grained material has the greatest creep strength. At lower stresses, the stress index n ($= \partial \log \dot{\epsilon}/\partial \log \sigma$) tends towards unity as diffusional creep predominates. In this region, the coarse grained material has the greatest creep strength. At still lower stresses, for the finest grained material it should be noted that n deviates from unity and creep rates fall to low values. In a previous paper [19] it was shown that this deviation could best be interpreted in terms of a σ_0 term, such that in the diffusional creep range, $\dot{\epsilon} \propto (\sigma - \sigma_0)$, where σ_0 is a threshold stress below which diffusional creep does not contribute to strain, and takes the value of ~ 7MN m^{-2}. The origin of this term was considered to arise because of some limitation of the ability of grain boundaries to act as sources and sinks for vacancies.

The influence of grain size upon creep rate is also shown in Fig. 2 where creep rate at 1623K interpolated from Fig. 1 is plotted against grain size for different values of applied stress. Again this figure demonstrates the strengthening effect of grain boundaries in the dislocation creep region (at 70 MN m^{-2}) and the weakening effect in the diffusional creep region (at 15 MN m^{-2}). At this latter stress the slope ($\partial \log \dot{\epsilon}/\partial \log d$) $= -2$ demonstrating the validity of the predicted relationship $\dot{\epsilon} \propto 1/d^2$. At stresses

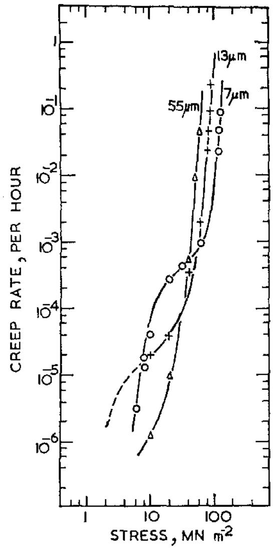

Figure 1 Creep rate-stress relationship at 1623K for uranium dioxide of various grain sizes.

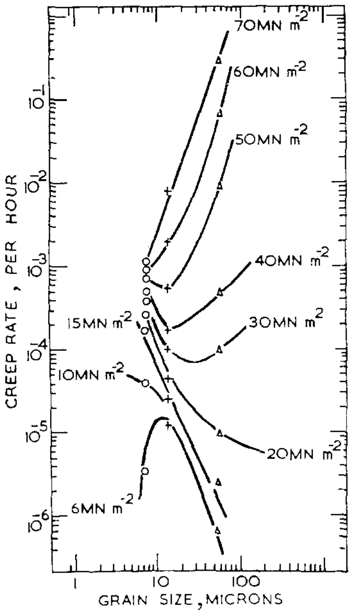

Figure 2 Creep rate-grain size relationship at 1623K for uranium dioxide at various stresses.

lower than 15 MN m^{-2} the form of the log $\dot{\epsilon}$ – log d curve again changes owing to the influence of the σ_0 term. It was not possible to assess fully the influence of grain size on σ_0 owing to difficulties in performing creep tests at very low stresses in the present apparatus. However, we anticipate σ_0 decreasing with increasing grain size as the dotted extrapolation of the data for the 13 µm material indicates in Fig. 1.

Whilst it is easy to understand the influence of grain size upon diffusional creep rate in terms of variation in the diffusion path length with grain size, it is not so easy to understand the influence on dislocation creep rate. Since creep strength increases with decreasing grain size in this latter region, behaviour cannot be explained in terms of boundary sliding contributions nor by the increased ease at which dislocation creation and annihilation may occur at boundaries. Thus it appears that under these experimental conditions grain boundaries are acting as barriers to crystallographic slip. In such a case, fine grained material contains more barriers to slip and will have a higher creep strength. This type of strengthening is described by the Hall-Petch equation [12, 13]:

$$\sigma_F = \sigma_i + K_y\, d^{-\frac{1}{2}}$$

where σ_F is the flow stress σ_i a friction stress for dislocation movement in regions of crystal far away from a boundary and K_y is a constant which is a measure of the effectiveness of the boundary as a barrier to slip. The form of the equation can be arrived at as follows.

Consider a slip band in a grain meeting the boundary and exerting a stress concentration in the next grain. The slip band may be considered to behave like a crack of length d and the stress concentration at a distance x into the next grain is $\sim (\sigma - \sigma_i)\sqrt{(d/x)}$ where σ is then applied stress. If the stress required to start slip at position x is σ_c the $(\sigma_F - \sigma_i)\sqrt{(d/x)}$ must be $\geqslant \sigma_c$ where $\sigma = \sigma_F$ is the applied stress needed to propagate slip across the boundary; that is the flow stress. This is the Hall-Petch equation with $K_y = \sigma_c\sqrt{x}$. In order to correlate the present high stress results with this expression the flow stress at a creep rate of 2.3×10^{-2} h^{-1} is plotted against $d^{-\frac{1}{2}}$ in Fig. 3. The original three points from Fig. 1 are plotted with some additional points obtained for individual specimens at 1623K and two other runs were performed at 1873 and 1373K. This figure shows that the present high stress results are in good agreement with the Hall-Petch equation. (It should be noted that the form of Fig. 3 is not sensitive to $\dot{\epsilon}$; if σ_F at $\dot{\epsilon} = 2.3 \times 10^{-1}$ h^{-1} is plotted the line at 1623K moves vertically only by about 15 MN m^{-2}.) The decrease in the K_y term with increasing

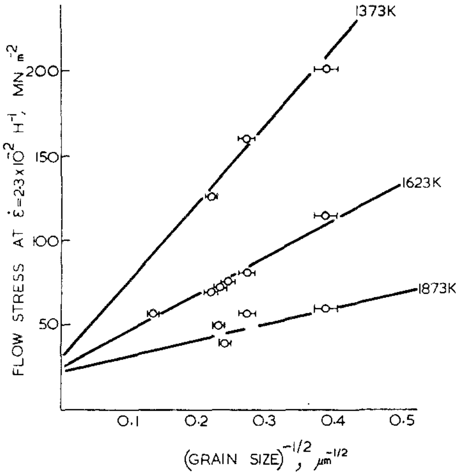

Figure 3 Flow stress-grain size relationship at various temperatures.

temperature shown in Fig. 4 indicates that boundaries are becoming less effective barriers to slip and would appear to provide no obstacle to slip at $(T/T_m) \sim 0.7$ when K_Y extrapolates to zero. Above this temperature it may be anticipated that creep strength may decrease with decreases in grain size as observed for metallic systems. It should be noted that in UO_2 grain boundaries are exerting a profound strengthening effect in dislocation creep up to higher values of T/T_m than in metals. For example, in Cu [14] and Cu-30% Zn [15] grain boundaries exert a weakening effect even at $T \sim 0.5\ T_m$.

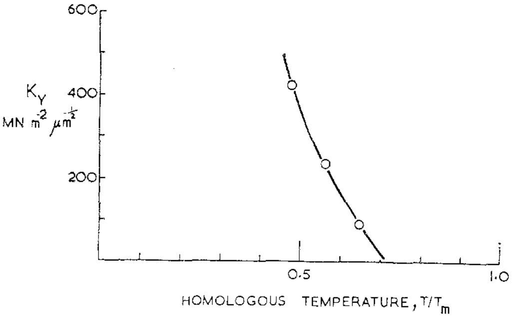

Figure 4 Variation of the K_Y term in the Hall-Petch equation with homologous temperature.

3.1. The effect of grain size on σ_0

The existence of a threshold stress for diffusional creep may be expected if grain boundaries do not act as perfectly efficient sinks and sources for vacancies [20]. In this case all the work done by the applied stress does not solely drive the vacancy flux but some goes to drive the interface reaction. Two theoretical proposals exist which predict the likely dependence of σ_0 upon d. Burton [21] suggests that if a grain boundary dislocation climb source is responsible for the process of vacancy emission/absorption then at sufficiently fine grain size this dislocation will be pinned between points a distance d apart. Then a stress $\sigma_0 \sim 2E/\mathbf{b}d$ where E is the line energy of the defect and $\mathbf{b}$ the Burger's vector must be exceeded in order for the climb source to operate. Taking $E \sim 0.5\ \mu\mathbf{b}^2$ where μ is the shear modulus ($\sim$ 100 GN m^{-2}), $d = 7\ \mu m$ and $\mathbf{b} = 2.5 \times 10^{-10} m$ gives $\sigma_0 \sim 3.5$ MN m^{-2} in reasonable agreement with the experimentally observed value of 7 MN m^{-2}. However, as discussed previously by Burton and Reynolds, σ_0 also has a temperature dependence much stronger than can be explained by the above model which is thus unsatisfactory.

A further proposal by Ashby and Verrall [22] which also predicts $\sigma_0 \propto 1/d$ considers the configuration of groups of grains undergoing creep by mass transfer processes. They consider the deformation process occurring by a grain-boundary sliding process with diffusional accommodation across grain corners. It should be noted that in this model the diffusional path length is less than but proportional to d and thus a $\dot{\epsilon} \propto 1/d^2$ relationship is predicted but at a faster rate than the Nabarro-Herring equation. In the model, the σ_0 arises because of the work needed to overcome increases in grain boundary area during the relative translation of one grain past another. It predicts $\sigma_0 = 0.72\ \gamma/d$ where γ is the grain-boundary energy. This value of σ_0 is very small, however, and again does not predict a strong temperature dependence.

4. Conclusions

The presence of grain boundaries in UO_2 has a strengthening or weakening effect in the region of $0.5\ T_m$ depending upon the stress level.

At high stresses, the presence of boundaries has a strengthening effect which may be interpreted in terms of the path length which dislocations can move before meeting a barrier (the boundary). This type of strengthening diminishes at higher temperatures.

At lower stresses, when deformation occurs by stress-directed diffusion, the presence of boundaries has a weakening effect which can be interpreted in terms of the diffusion path length from boundaries which act as vacancy sources to those which act as vacancy sinks.

A threshold stress for diffusional creep exists which is probably associated with the limitation of the ability of boundaries to act as vacancy sources and sinks. This threshold stress is higher for fine grained material.

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