Prismatic Slip in Zirconium Single Crystals at Elevated Temperatures

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Single crystals of Zr oriented favorably for prismatic slip have been deformed in tension over a range of strain rates at temperatures between 473 and 1113 K. A temperature independent plateau is observed between 600 and 800 K and dynamic strain aging occurs in the vicinity of 723 K. The flow stress is temperature dependent both above and below this temperature interval. Plastic flow above 850°K is represented by an equation of the form:

$$\dot{\gamma} = A\tau^n e^{-Q/kT}$$

where $\dot{\gamma}$ is the shear strain rate, A is a constant whose value is 680 ± 20 $(\text{MN/m}^2)^{-4.3}$. The stress exponent $n = 4.3 \pm 0.3$ and the activation energy $Q = 2.05 \pm 0.15$ eV. It is proposed that the high temperature prismatic slip in Zr is controlled by a glide-climb process where the rate of plastic flow is determined by the rate of climb of dislocations.

It is well established now that first order prismatic slip is the primary deformation mode in α-Zr at all temperatures.[1,2] The low temperature rate controlling mechanism,[3] the nature of work hardening,[4] and the effect of interstitial solutes[5,6] on the ease of prismatic slip have been examined earlier using single crystals. The rate controlling mechanism at elevated temperatures has, however, not been investigated systematically in single crystals, although Mills and Craig[5] have measured the critical resolved shear stress and the activation volume at the onset of prismatic slip at temperatures up to 950 K. These authors have also suggested that above 800 K the rate controlling mechanism may be the arrival of vacancies at jogs. Polycrystalline Zr, on the other hand, has been examined in greater detail at elevated temperatures and the results of creep tests[7-9] as well as those of tension[10,11] and compression[12] studies are available.

The aim of the present investigation was to deform in tension oriented single crystals of Zr in prismatic slip at temperatures between 473 and 1113°K, and over a wide range of strain rates, and to identify the dislocation mechanism responsible for plastic flow at elevated temperatures.

EXPERIMENTAL METHOD

Crystal bar Zr in the form of 4.5 mm rods and 22 cm in length was used as the starting material. Single crystals up to 20 cm in length were grown from the melt using an electron beam floating zone technique. Suitable orientations were chosen out of randomly grown crystals. The details of crystal growth and specimen preparation have been described elsewhere.[2,4] The final specimens prior to testing had a reduced gage length of 2.54 cm and a gage diam of 2.2 to 2.4 mm. The interstitial oxygen content of these crystals was assessed in a manner described earlier[2] and was found to be approximately 120 ppm.

Tensile tests were conducted on a floor model Instron. All tests were conducted under a dynamic vacuum of less than 10^{-4} mm of Hg. A quartz tube surrounded the specimen assembly. Resistance heating was used to attain temperatures up to 1113 K and controlled to within ±2 K. It has been established earlier[2] that the interstitial content remains unaffected under the test conditions employed.

RESULTS

The orientations of crystals used in the present investigation were such that a maximum resolved shear stress was applied on the first order prism slip system. With the exception of one specimen tested at 723 K, for which the angle between the tensile axis and the basal plane (x_B) was 53 deg all other specimens had x_B values of less than 26 deg. After fracture, the specimen surface was examined under the optical microscope and the deformation markings were analyzed for a number of specimens using the two surface technique. In each of the crystals examined including the one with $x_B = 53$ deg, only prismatic slip lines were observed between 473 and 1113 K. It has been shown earlier that when x_B is larger than 60 deg basal slip takes place above 850 K. while twinning is observed at lower temperatures.[2] Resolved shear stress vs shear strain curves of a set of specimens deformed at an unresolved initial strain rate of 1.66×10^{-4} s^{-1} between 523 and 1073 K are shown in Fig. 1. Single prismatic slip along the $\langle 11\bar{2}0\rangle$ direction has been assumed in constructing these curves. The curves for 523 and 623 K are similar to those reported earlier.[4] Multiple load drops were observed at 723 K. The absence of twin markings after fracture suggested that multiple load drop is due to dynamic strain aging. For specimens tested above 850 K, the work hardening rate decreased with increasing strain up to approximately 5 to 10 pct deformation, above which the specimen exhibited little or no strain hardening. The flow stress decreased with increasing strain in excess of 0.5. From interrupted tests, it was observed that uniform plastic flow took place over the entire gage length up to a strain of 0.5 above which non-homogenous deformation occurred due to multiple necking.

The deformation markings on two specimens de-

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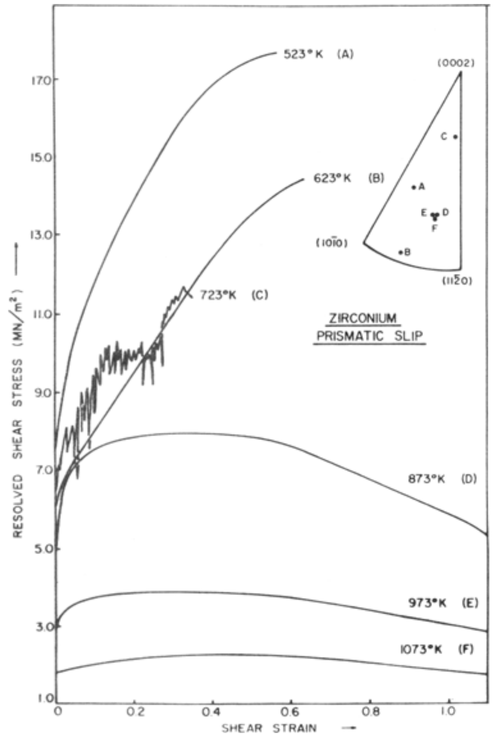
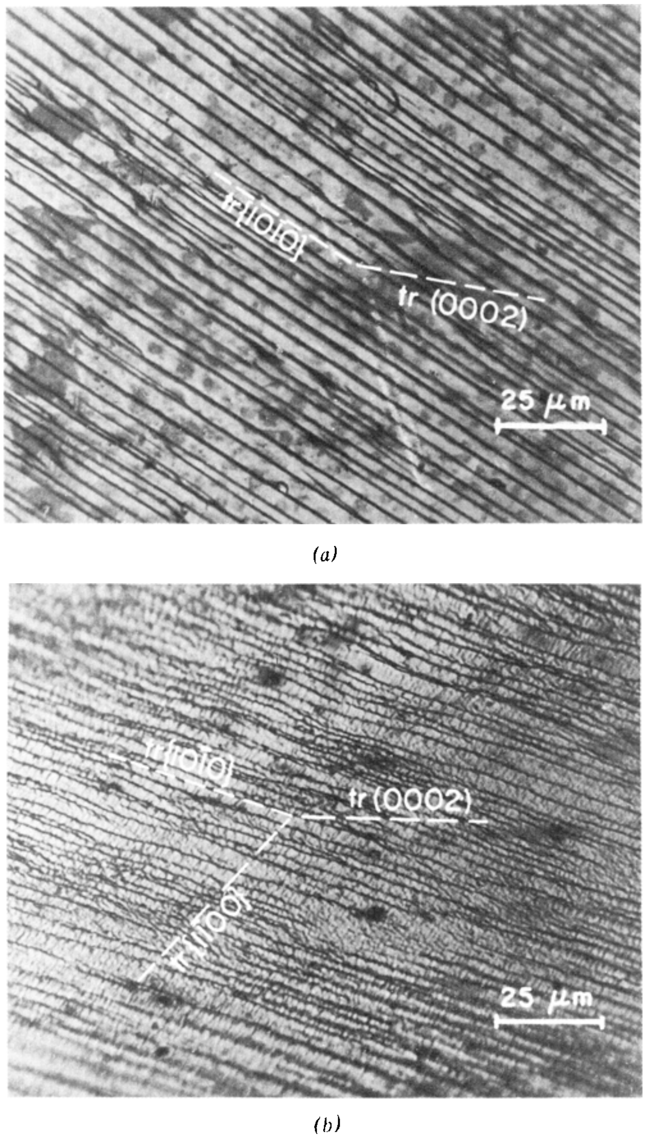


Fig. 1—Resolved shear stress vs shear strain curves of crystals tested between 523 and 1073 K. The specimen axes are plotted in the unit triangle.

formed to a shear strain of 0.5 at 673 and 1073 K are shown in Fig. 2. Coarse primary slip lines with a few cross slip traces are seen at 673 K. At the higher temperature of 1073 K, the slip lines are finer and more wavy. The spacing between the primary slip bands at a shear strain of 0.5 in specimens tested at 673 and 1073 K were roughly 3 μm and 2 μm respectively. Particularly coarse slip bands were observed in specimens which exhibited serrated plastic flow when tested in the neighborhood of 723 K.

The critical resolved shear stress (CRSS) which was taken at the first deviation from linearity (roughly a plastic strain of 10^{-4}) is plotted against temperature in Fig. 3. Some of the published data obtained earlier on comparable purity material at low temperatures[4] are also included for comparison. The CRSS decreases with increasing temperature up to approximately 600 K, remains relatively temperature insensitive between 600 and 800 K and decreases rapidly thereafter up to 1113°K. The allotropic transformation hcp $\rightleftharpoons$ bcc in Zr takes place at 1135 K. A replot of the temperature-CRSS relationship is shown in Fig. 4, where log τ is plotted against $1/T$. A straight line describes well the data above 850 K.

In order to examine the effect of a wide range of strain rates on the flow stress at a fixed temperature, two crystals were deformed at cross head speeds between 8.34×10^{-2} cm/s and 8.34×10^{-6} cm/s. The push button speed selector of the Instron was employed for instantaneous change in cross head speed. Since there is practically no work hardening at temperatures of 973 and 1073 K and at strains between 0.1 and 0.5 (Fig. 1), it was observed that the stress required for flow at a certain rate was almost exactly reproduced if this rate was re-established after some intermediate rates of cross head motion. This enabled a determination of the flow stress at different strain rates using the same specimen. The results of such experiments are shown as flow stress vs cross head speed on a log-log plot in Fig. 5. The data points for each test temperature are best described by a straight line. Note that the slopes of the two straight lines for 973 and 1073 K in Fig. 5 are the same.



(a)

(b)

Fig. 2—Prismatic slip lines on specimens deformed to a shear strain of 0.5 (*a*) tested at 673 K, (*b*) deformed at 1073 K.

The strain rate effect was further examined between 773 and 1113 K by cycling crystals between cross head speeds of 8.34×10^{-5} cm/s and 8.34×10^{-4} cm/s keeping the test temperature fixed. The strain rate was changed instantaneously using the push button speed selector and deformation continued until the flow stress became nearly independent of strain, at this point the cross head speed was changed again. The nature of the transients on strain rate change are shown in Fig. 6. At 823 K, an increase in cross head speed from 8.34×10^{-5} cm/s to 8.34×10^{-4} cm/s resulted in an instantaneous increase in flow stress followed by a gradual decrease in work hardening rate which extended over considerable strain. At tempera-

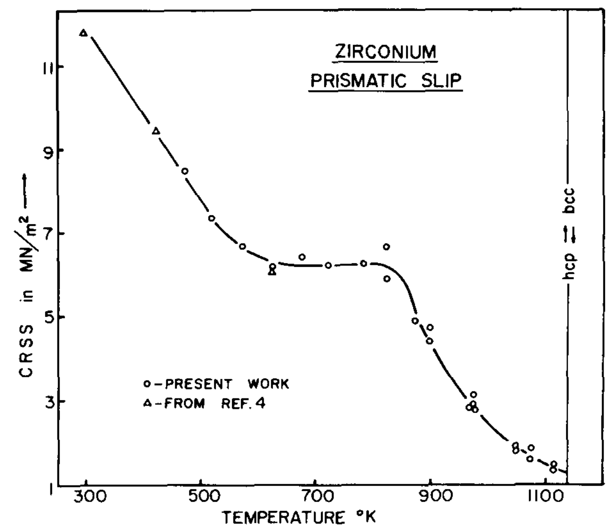


Fig. 3—The critical resolved shear stress vs temperature.

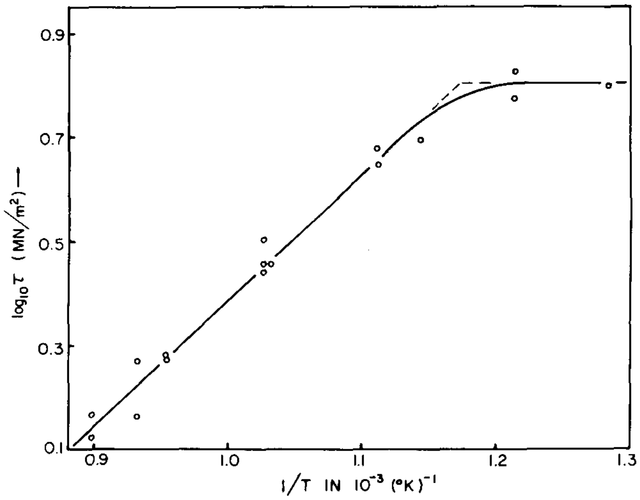


Fig. 4—Log τ vs $1/T$. Deviation from linearity occurs at temperatures below approximately 850 K.

tures between 898 and 1113 K, the transients were short and a steady state of plastic flow with a zero work hardening rate was established over less than 5 pct shear strain, thus making it practical to evaluate the steady state flow stress ratio for a change in strain rate by a factor of ten. At 773 K, no change in flow stress was detected when strain rate was changed. The flow stress ratio is plotted against strain in Fig. 7. Although experimental data above 0.5 pct strain have been plotted in Fig. 7, these will be excluded from the interpretation of results due to the non-homogeneous plastic flow taking place at these strains. The ratio τ_1/τ_2 in Fig. 7 does not show a systematic variation with temperature or with strain up to 0.5 and may be described by a scatter band between 1.66 and 1.79.

DISCUSSION

The temperature and strain rate dependence of the flow stress shown in Figs. 4 and 5 may be described by an equation of the form:

$$\gamma = A\tau^n \exp(-Q/kT) \qquad [1]$$

where

$\dot{\gamma}$ is the strain rate,
A is a constant,
n is another constant referred to as the stress exponent,
Q is the activation energy,
k is Boltzmann constant, and
T is the temperature in K.

Eq. [1] is often used to describe the high temperature deformation of metallic materials and the constants Q and n are helpful in identifying the rate determining process.

The slope of log τ vs log $\dot{\gamma}$ for the two straight lines in Fig. 5 is 0.232 which yields a value of $n = 4.3$. Also from Fig. 4

$$\frac{Q}{nk} = \frac{d \ln \tau}{d(1/T)} = 5.48 \times 10^3 \text{ K} \qquad [2]$$

which gives $Q/n = 0.474$ eV.

Using a value of $n = 4.3$, one obtains

$$Q = 2.05 \text{ eV}$$

The stress exponent may also be determined using the steady state flow stress ratio plotted in Fig. 7. Taking the logarithm Eq. [1] may be rewritten as:

$$\ln \dot{\gamma} = \ln A + n \ln \tau - Q/kT$$

Therefore for strain rate change experiments at constant temperature:

$$\ln\left(\frac{\dot{\gamma}_1}{\dot{\gamma}_2}\right) = n \ln\left(\frac{\tau_1}{\tau_2}\right) \qquad [3]$$

For $\dot{\gamma}_1/\dot{\gamma}_2 = 10$, *i.e.*, the ratio of cross head speeds 0.05 cm/min and 0.005 cm/min and the corresponding $\tau_1/\tau_2 = 1.66 - 1.79$, n turns out to be 4 – 4.6. The corresponding Q is 2.05 ± 0.15 eV. This value of activation energy for plastic deformation is in the same range as that for self-diffusion in α-Zr which has been reported to be between 1.98^{13} and 2.26^{14} eV in the temperature interval 923 through 1123 K.

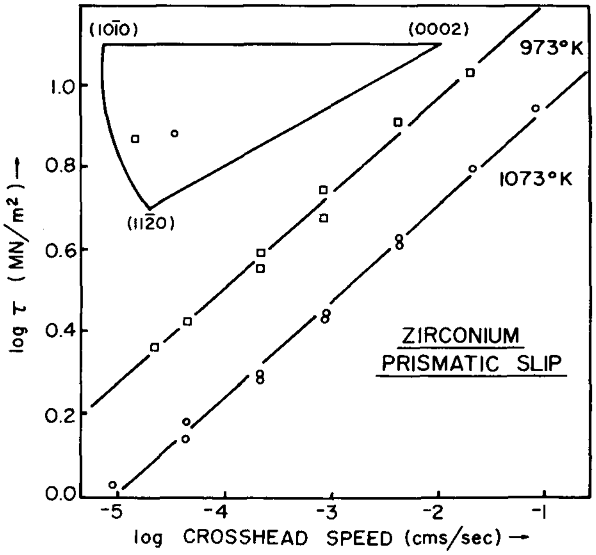


Fig. 5—Flow stress vs cross head speed plotted on a log-log scale for test temperatures of 973 and 1073 K. The specimen orientations are shown in the unit triangle.

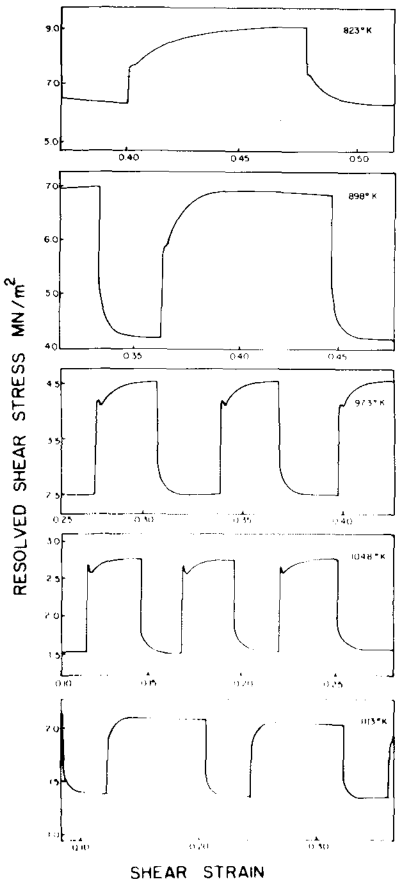


Fig. 6—The nature of transients on cycling the cross head speed between 8.34×10^{-4} and 8.34×10^{-5} cm/s. Note that a steady state has not been reached for the specimen tested at 823 K.

A. The Constant A

In order to evaluate the constant A, the data of Fig. 5 are replotted as $(\dot{\gamma})^{1/4.3}$ vs τ as shown in Fig. 8. The shear strain rate was obtained by multiplying the tensile strain rate by a factor of two. A straight line passing through the origin describes a set of data obtained at a fixed temperature. From Eq. [1], it is readily seen that the slope of the straight line in Fig. 8 is equal to $Ae^{-Q/kT}$. Substituting $Q = 2.05$ eV, one therefore obtains $A = 680 \pm 20$ $(\mathrm{MN/m^2})^{4.3}$.

It is interesting to compare at this juncture the values of activation energy and stress exponent arrived at in earlier studies of plastic deformation of Zr. Mills and Craig[5] deformed single crystals and determined activation volumes of 200 to $1000b^3$ and activation energies of 0.8 to 3 eV between 800 and 950 K. Their study was limited to the onset of plastic flow and the stress exponent was not determined. Polycrystalline Zr samples were compressed between 898 and 1098 K over a range of constant strain rates by Luton and Jonas[12] and the flow stress was found to obey a strain rate dependence of the type described in Eq. [1]. The stress exponent n was found to be 4.6 at yield and 4.5 at steady state, while $Q = 2.43 \pm 0.13$. As noted by these authors,[12] the activation energy obtained was slightly higher than that for self-diffusion. Luton and Jonas have argued that at yield the dislocation density is likely to be low, therefore the applicability of recovery through classical climb models leading to annihilation is unlikely. A process of glide involving thermal activation over local obstacles was considered, and it was concluded that the nature of interaction was such that the back stress had both short and long range components. The atomistic nature of the barrier and the mechanism of the interaction process have not been dealt with by these authors.

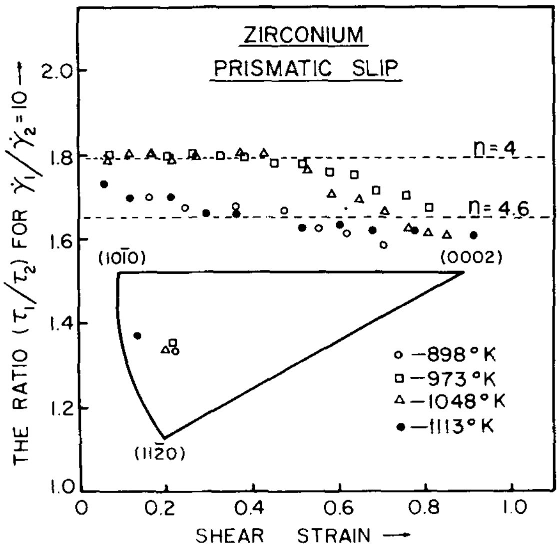


Fig. 7—The steady state flow stress ratio τ_1/τ_2 plotted against shear strain for a change in strain rate by a factor of 10. The data for test temperatures between 898 and 1113 K may be described by a scatter band in the flow stress ratio of between 1.66 and 1.79. The corresponding stress exponent n lies between 4 and 4.6. Note non-homogeneous plastic flow occurs above a shear strain of 0.5.

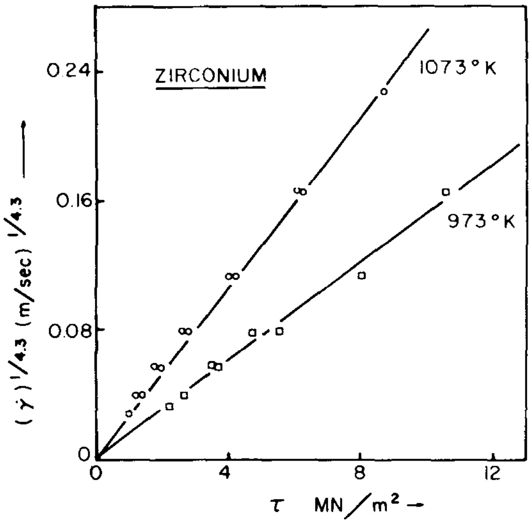


Fig. 8—Data of Fig. 5 replotted as (shear strain rate)$^{1/n}$ vs flow stress. Note both straight lines pass through the origin.

Several studies of creep on polycrystalline Zr have been undertaken in the past. Warda *et al*[15] have concluded that minor changes in interstitial content result in a substantial variation of the creep behavior. Values of stress exponent ranging between 4.8 and 13.8 depending on test variables were obtained in the temperature interval between 723 and 823 K by these authors.[15] Bernstein[7] investigated creep in polycrystalline Zr at temperatures between 793 and 893 K and obtained two values of n. The steady state creep at low stresses was characterized by a stress exponent of one and $Q = 1.3$ eV, while at high stresses n had a value of 6 to 7 and $Q = 2.44$ eV. The interpretation offered by Bernstein was that at low stresses creep is controlled by grain boundary diffusion, while at higher stresses bulk diffusion controls the creep. Ardell and Sherby[8] have studied steady state creep in polysrystalline Zr at temperatures between 933 and 1118 K. The low stress region was reported to be associated with $n = 7.5$, while at higher stresses n had a value of 4.7 and $Q = 2.08$ to 2.38 eV. Ardell and Sherby concluded from their study that the observations could be rationalized on the basis of either a glide controlled creep process or a diffusion controlled process. Furthermore, these authors suggested that on the basis of a glide climb model, plastic deformation will be glide controlled at low stresses due to a rapid rate of diffusion in α-Zr, while at high stresses diffusion will be the rate determining process.

B. Comparison with Theories of High Temperature Plastic Flow

The high temperature prismatic slip of Zr above 850 K is described by a single activation energy term (Eq. [1]) implying that only one dislocation mechanism is rate controlling. Also the activation energy is independent of the applied stress which suggests that all mechanisms for which the activation energy is stress sensitive are not applicable.

The observed activation energy which is similar to that for self-diffusion in α-Zr and a stress exponent of 4.3 ± 0.3 are in good agreement with the theory of steady state plastic flow, where the strain is derived due to glide, but the strain rate is controlled by dislocation climb. An early formulation of the model put forward by Weertman[16] is as follows:[17]

$$\dot{\gamma} = \frac{C\tau^{4.5} Do \exp(-Q/kT)}{\mathrm{b}^{1/2} N^{1/2} kTG^{3.5}} \qquad [4]$$

where

Do is the diffusion constant,
b is the Burgers vector,
G is the shear modulus,
N is the density of dislocation sources per unit volume of material, and
C has a value of approximately 2.5.

It was assumed[16] in deriving Eq. [4] that pairs of edge dislocations of opposite sign on different parallel slip planes are blocked from passing each other, thereby leading to the formation of pile ups between the blocked dislocations and the sources. During steady state deformation, the blocked dislocations which lead the pile ups are assumed to climb and annihilate each other. Eq. [4] has been substantiated experimentally for pure metals at low stresses to a greater extent than any other theoretical relationship, although dislocation pile ups of the magnitude required by the theory have not been experimentally observed.[17, 18]

Subsequently, Weertman[19] has analyzed the model of climb controlled glide without the need for the formation of pile up. Edge dislocations from neighboring sources overlap to form dipoles which in turn collapse through climb. The steady state strain rate so developed is given by:

$$\dot{\epsilon} = \{\alpha\beta^{4.5} Do/\mathrm{b}^{3.5}\alpha^* \log(d/\mathrm{b})\}(\pi/N)^{1/2}(\sigma/G)^{3.5} \cdot (\sigma\Omega/kT)\, e^{-Q/kT} \qquad [5]$$

where

$\dot{\epsilon}$ is the tensile strain rate,
σ is the tensile stress,
d is the spacing between parallel slip planes of neighboring sources,
Ω is the at. vol,
α is a constant = 0.5,
α^* is another constant = 6, and
β has values between 1 and 2.

Substituting for α, α^*, β, $\log d/\mathrm{b} = 10$, and $\Omega = 0.7\mathrm{b}^3$,[19] Eq. [5] may be rewritten in shear stress-strain form:

$$\dot{\gamma} = C' \frac{\tau^{4.5} Do \exp(-Q/kT)}{\mathrm{b}^{1/2} N^{1/2} kTG^{3.5}} \qquad [6]$$

where

$C' = 0.03$ for $\beta = 1$

and

$C' = 0.8$ for $\beta = 2$.

A comparison of the empirical Eq. [1] and the theoretical Eq. [6] gives:

$$A = \frac{C'Do}{\mathrm{b}^{1/2} N^{1/2} kTG^{n-1}} \qquad [7]$$

Experimentally, $n = 4.3$ has been obtained. Substituting $C' = 0.03$, $T = 1000$°K and the appropriate value of G[20] and the experimental value of A, one obtains:

$$N = 1.67 \times 10^{24}(Do)^2 \text{ in. m}^{-3} \qquad [8]$$

A reliable value of Do for Zr, however, is lacking. Lyashenko *et al*[13] have reported a value of $Do = 5.9 \times 10^{-6}$ m^2/s, which when substituted in Eq. [8] gives $N = 4 \times 10^9$ cm^{-3}. Using $Do = 5.6 \times 10^{-8}$ m^2/s as reported by Flubacher,[14] one obtains $N = 3 \times 10$ cm^{-3}. However, Kidson[21] has noted that the value of Do reported by Flubacher is lower than that representing bulk diffusion. Therefore, the value of N, the dislocation sources per unit volume should be between 4×10^9 and 3×10^5 cm^{-3} with the lower limit being too low due to the low value of Do. These values of N are reasonable for crystals deforming under stresses of the order of 10^{-4} to 10^{-5} G, as is the case in the present work.

C. Jogged Screw-Dislocation Model

It has been pointed out by Weertman[19] that a jog segment in a screw dislocation is a segment of an edge dislocation. Since the mobility of edge segments is lower than that of screws, it is argued that the dragging of such a segment through arrival of vacancies is in effect equivalent to the kinetics of dislocation annihilation considered in the model whose final mathematic form is Eq. [5]. Therefore, the jogged screw dislocation model need not be considered as a separate case from climb of edge dislocations.

D. High Temperature Prismatic Slip of Other Hcp Materials

Relatively few investigations of prismatic slip at elevated temperatures have been carried out using singl crystals. All earlier investigations have been with single crystals of materials like Zn,[22] Mg[23], and Mg + 12 at. pct Li alloy.[24] In all these materials basal slip is the primary deformation mode, whereas prismatic slip is a secondary mode of plastic flow. Such is not the case with Zr whose primary deformation mode is prismatic slip. In Zn[22] and Mg,[23] the elevated temperature prismatic slip is thought to be controlled by Friedel cross slip process, whereas a diffusion controlled climb process similar to that of Zr has been reported for Mg + 12 pct Li alloy single crystals.[24]

E. The Dislocation Sub-Structure

Under conditions of steady state plastic flow, *i.e.*, steady state creep under constant stress and deformation with zero work hardening rate in constant strain rate experiments, it has frequently been observed that a characteristic cellular distribution of dislocation develops.[25] An increase in strain rate decreases the cell size with an associated increase in the flow stress. In experiments involving a change in the imposed strain rate the new steady state structure is not developed instantaneously. The requirement of time or strain is essential for such a process. Furthermore since the steady state of plastic flow at elevated temperatures is associated with thermally activated processes the attainment of the steady state structure would be easier the higher the test temperature. The observations of Fig. 6 are consistent with this idea. In Fig. 6 a steady state was not reached at 823 K, while the transients became shorter with an increase in the test temperature.

Direct experimental evidence for the development of cellular structure is still lacking in the present work. Transmission electron microscopy of thin foils or an examination of the dislocation etch-pit pattern would be required to study the sub-structure.

CONCLUSIONS

1) The critical resolved shear stress for prismatic slip in Zr single crystals containing approximately 120 ppm oxygen decreases with temperature up to 600 K, a temperature independent plateau is observed between 600 and 800 K followed by a decreasing CRSS at elevated temperatures.

2) Dynamic strain aging has been observed at temperatures in the vicinity of 723 K.

3) Prismatic slip above 850 K is thermally activated and the strain rate is determined by a diffusion controlled process.

4) The results are in good agreement with a model of climb controlled glide.

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