Steady-State Creep of α-Zirconium at Temperatures up to 850 °C

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Cumulative zirconium creep data over a broad range of stresses (0.1 to 115 MPa) and temperatures (300 °C to 850 °C) were analyzed based on an extensive literature review. Zirconium obeys traditional power-law creep with a stress exponent of approximately 6.4 over strain rates and temperatures usually associated with the conventional “five-power-law” regime. Thus, dislocation climb, rather than the often assumed glide mechanism, may be rate controlling. Power-law breakdown occurs at values of \( \varepsilon_0/D \) greater than approximately 10⁹ cm⁻², consistent with most traditional five-power-law materials. The creep rate of zirconium at low values of \( \sigma/G \) varies proportionally to the applied stress. The rate-controlling mechanism(s) for creep within this regime is unclear. A grain-size dependency may exist, particularly at small (<90 μm) sizes, suggesting a diffusional mechanism. A grain-size independence at larger grain sizes supports a Harper–Dorn mechanism, but the low observed activation energy (~90 kJ/mol) is not consistent with those observed at similar temperatures at higher stresses in the five-power-law regime (270 kJ/mol) where creep is also believed to be lattice self-diffusion controlled. The stress dependence in this regime is not consistent with traditional grain-boundary sliding mechanisms.

I. INTRODUCTION

ZIRCONIUM alloys are commonly used as a cladding material to contain nuclear fuel at elevated temperatures. Knowledge of the creep properties of zirconium and zirconium alloys is, therefore, very important. Creep of zirconium and zirconium alloys has often been described as “anomalous.” Researchers often report that zirconium and its alloys never reach true steady-state creep (for example, References 1 through 3). It has also been reported that the stress exponent [4,5,6] and activation energy [4,5] change continuously with stress, which is not reflective of climb control as most other “pure” metals within the five-power-law regime. Many interpretations have been offered explaining the creep behavior of zirconium. Some have suggested that creep is dislocation climb controlled in the “intermediate stress” regime corresponding to the five-power-law regime, [7–10] as in other metals and alloys, while others maintain that creep is dislocation glide controlled [4,5]. Still others suggest several different controlling mechanisms within the five-power-law regime depending on stress and temperature. [6,11] The creep rate of zirconium at stresses below those associated with five-power-law creep varies nearly linearly with stress. It has been suggested that creep in this regime is controlled by a combination of Harper–Dorn creep for large grain sizes [12,13] and diffusional (Coble [9,10,12–15] or Nabarro–Herring [9,15]) creep for small grain sizes. Ruano et al. [16] suggested that creep at low stresses and relatively small grain sizes is controlled by a grain-boundary sliding mechanism.

These conclusions often reflect the analysis of limited data. Cumulative zirconium creep data will be presented here based on an extensive literature review that includes data often not included in earlier analyses. New interpretations of results will be presented and compared with previous interpretations.

II. RESULTS AND DISCUSSION

A. General

In the past, a variety of explanations have been presented to describe the creep behavior of zirconium over a range of stresses and temperatures. These analyses [4,5,6] suggest several different controlling mechanisms depending on the temperature and stress. These interpretations were often developed to explain the creep behavior observed in individual creep studies. However, when comprehensive data are analyzed, more reliable, and often different, interpretations emerge.

Data from various studies are presented in Figure 1, which is a plot of the diffusion coefficient compensated steady-state strain (creep) rate \( \sigma/G \) vs the modulus-compensated creep stress \( \sigma/G \). The steady-state strain rate is additionally (conventionally) normalized by \( k, G, b, \) and \( T \). [17] Values for the modulus \( G \) of zirconium were calculated from

\[
G(T) = -0.0203 \cdot T(\degree C) + 36.27 \text{ (GPa)}
\]

based on the data presented by Koster et al. [18] Data in the moderate \( \sigma/G \) (five-power-law) and high \( \sigma/G \) (power-law breakdown) regimes are normalized by a diffusion coefficient with an activation energy, \( Q \), of 270 kJ/mol and a (typical) pre-exponential, \( D_0 \), of \( 5 \times 10^{-4} \text{ m}^2/\text{s} \). Data in the low \( \sigma/G \) regime (bottom of graph) are normalized by a...
diffusion coefficient with an activation energy of 90 kJ/mol and a pre-exponential of $5 \times 10^{-3}$ m$^2$/s. A discussion of the selection of activation energies is given subsequently. Data at stresses above the transition from low to moderate $\sigma/G$ reported by Prasad et al.\cite{9} and Bernstein\cite{10} are also included with the low $\sigma/G$ data that correspond to the right-hand ordinate in Figure 1. These show the transition between the two regimes (although the activation energy at $\sigma/G$ values above the transition should increase to 270 kJ/mol, which would result in a slope consistent with the other data at intermediate stresses corresponding to the left-hand ordinate).

It is apparent from Figure 1 that a cumulative plot developed from all zirconium creep data appears to be fairly consistent with typical creep behavior for Class II (M) metals and alloys.\cite{17} At low $\sigma/G$, the data in Figure 1 have a constant slope (stress exponent, $n$) of approximately 1.1, perhaps indicative of Harper-Dorn or diffusional creep. At intermediate values of $\sigma/G$, the data have a constant stress exponent of approximately 6.4, which is within the range of so-called five-power-law creep. At values of compensated strain rates above approximately $10^{-4}$, zirconium reaches power-law breakdown, where the stress exponent then increases with increasing $\sigma/G$. This value corresponds to a value of $\dot{\varepsilon}_{\text{ss}}/D$ of $10^9$ cm$^2$/s, consistent with the value suggested by Sherby and Burke\cite{19} to correspond to power-law breakdown for traditional five-power-law metals. (Figure 1 includes all data that the authors were aware of, except one set (of four) of data presented by Gilbert et al.,\cite{16} as it appeared inconsistent with other data reported by Gilbert and co-workers; Siethoff and Ahlborn\cite{11} also noted this apparent inconsistency. Also, only the final steady-state values reported by Warda et al.\cite{20} (who suggested “early”

dynamic strain-aging induced steady-state creep of zirconium, prior to a final steady state) are plotted. The strain-aging effects, when they are observed, occur at relatively low strains (or creep time) and are considered within the transient (primary) creep regime. Finally, normalized data were omitted in cases where values for normalizing factors were not reported.)

B. Moderate $\sigma/G$ (Five-Power-Law) Regime

In pure metals and Class II (or M) alloys, there is an established, largely phenomenological relationship between the steady-state strain rate, $\dot{\varepsilon}_{\text{ss}}$, (or creep rate) and stress, $\sigma_{\text{ss}}$, for steady-state five-power-law creep:

$$\dot{\varepsilon}_{\text{ss}} = A_0 \exp \left[ \frac{-Q_c}{kT} \right] \left( \frac{\sigma_{\text{ss}}}{G} \right)^n$$  \[2\]

where $A_0$ is a constant, $k$ is Boltzmann’s constant, $Q_c$ is the creep activation energy, $G$ is the shear modulus, and $n$ is the stress exponent, which varies from approximately 4 to 7.\cite{17} The zirconium data in the moderate $\sigma/G$ regime of Figure 1 indicate a constant stress exponent of approximately 6.4, typical of five-power-law creep. The data reported by Ardell and Sherby\cite{4} and MacEwen et al.\cite{5} appear to indicate a nonconstant stress exponent. These data appear slightly “curved” in Figure 1. Ardell and Sherby found that the stress exponent decreases with increasing stress, while MacEwen et al. reported that the stress exponent increases with increasing stress over a similar range of $\sigma/G$. It is not clear what caused this (different) behavior, however, both sets of data fit well within the scatter reported by other investigators. Other investigators did not observe such behavior in zirconium. It appears that some of the lower stress data reported by MacEwen et al. may fall into the one-power-law regime, possibly explaining, at least partially, the lower reported stress exponents at lower stresses. Ardell et al., however, did not observe a transition to the one-power-law regime in their data.

The apparent activation energy for creep in the five-power-law regime is approximately 270 kJ/mol and appears independent of stress and temperature. This value of activation energy was found to best condense the data onto nearly a single line. It has been suggested that the activation energy for creep of $\alpha$-zirconium decreases with increasing stress.\cite{14,15} However, no such general trend is apparent when examining data from various authors over a wide range of temperatures (400 °C to 800 °C).

An activation energy of 270 kJ/mol falls within the range of self-diffusion activation energies reported in the literature, which range from approximately 88 to 315 kJ/mol.\cite{21–32} It is also fairly close to what is considered to be the “intrinsic” (free from impurity and other “extrinsic” effects) self-diffusion activation energy, reported by Hood\cite{30} as approximately 315 kJ/mol. Thus, at least initially, it appears that creep is self-diffusion (dislocation-climb) controlled rather than dislocation glide controlled as suggested by Ardell and Sherby\cite{4} and MacEwen et al.\cite{5}.

A major weakness in this argument is that, at least with the purity of available zirconium, the self-diffusion activation energy does not appear constant with temperature. The self-diffusion activation energy at low temperatures (below...