MECHANICAL BEHAVIOR OF POLYCRYSTALLINE TUNGSTEN AT ELEVATED TEMPERATURE*

S. L. ROBINSON† and O. D. SHERBY†

The influence of stress, temperature, grain size and subgrain size on the steady state creep rate of polycrystalline unalloyed tungsten has been examined based on the extensive published data available. It was shown that the steady state creep rate of polycrystalline tungsten, and other metals, is given by

$$\dot{\varepsilon} = S\lambda^2 \, \text{Deff} \left(\frac{\sigma}{E}\right)^7$$

where λ = subgrain size, σ = creep stress, E = average unrelaxed elastic modulus, and S is a universal creep constant equal to about 3×10^{40} cm^{-4}. The effective diffusion coefficient, Deff, equals $(D_L f_L + D_D f_D)$ where D_L is the lattice diffusion coefficient, D_D is the dislocation diffusion coefficient, and f_L and f_D are the fraction of atoms participating in lattice and dislocation diffusion respectively. When subgrains form during creep, as in normal behaving materials, $\lambda = k\sigma^{-1}$ (k is a material constant) and the above equation yields the generally observed five power stress law for creep. When subgrains do not form during creep (as in many refractory metals produced by powder metallurgy techniques), λ in the above equation is replaced by the grain size. In this case $\dot{\varepsilon} \alpha \sigma^7$ and $\dot{\varepsilon} \alpha L^2$, predictions in agreement with experimental evidence. The power law breakdown for creep at high stresses can be explained by one of two possibilities: (i) the creation of excess vacancies by plastic deformation which increases D_L in the Deff term, or (ii) the increase in dislocation density with increasing stress, which influences the dislocation diffusion term, $D_D f_D$, in Deff.

COMPORTEMENT MECANIQUE DU TUNGSTENE POLYCRISTALLIN AUX TEMPERATURES ELEVEES

Les auteurs ont examiné l'influence de la contrainte, de la température, de la taille des grains et de la taille des sous-grains sur la vitesse de fluage en régime permanent pour le tungstène polycristallin non allié, en prenant pour base les résultats valables publiés antérieurement. On montre que la vitesse de fluage en régime permanent pour le tungstène polycristallin et pour d'autres métaux est donnée par la relation

$$\dot{\varepsilon} = S\lambda^2 \text{Deff} \left(\frac{\sigma}{E}\right)^7$$

où λ est la taille des sous-grains, σ la contrainte de fluage, E le module élastique moyen non relaxé et S une constante de fluage universelle égale à 3×10^{40} cm^{-4} environ. Le coefficient de diffusion effective Deff est égal à $D_L f_L + D_D f_D$ où D_L est le coefficient de diffusion dans le réseau, D_D le coefficient de diffusion le long des dislocations, et où f_L et f_D sont les fractions d'atomes participant respectivement à la diffusion dans le réseau et le long des dislocations Quand des sous-grains se forment pendant le fluage, comme dans les métaux qui se comportent normalement, $\lambda = k\sigma^{-1}$ (k étant une constante physique) et l'équation ci-dessus donne, comme habituellement pour le fluage, la contrainte à la puissance cinq. Quand des sous-grains ne se forment pas au cours du fluage (comme dans de nombreux métaux réfractaires produits par les techniques de la métallurgie des poudres), λ est remplacé dans l'équation ci-dessus par la taille du grain. Dans ce cas les valeurs prévues pour $\dot{\varepsilon} \alpha \sigma^7$ et $\dot{\varepsilon} \alpha L^2$ sont en accord avec les résultats expérimentaux. Le fait que la loi ne convient plus pour des contraintes de fluage élevées peut être expliqué par l'une des deux possibilités suivantes: (i), la création de lacunes en excès par la déformation plastique, ce qui augmente D_L dans le terme Deff, ou (ii), l'accroissement de la densité des dislocations quand la contrainte augmente, ce qui se répercute sur le terme de diffusion le long des dislocations $D_D f_D$ dans Deff.

DAS MECHANISCHE VERHALTEN POLYKRISTALLINEN WOLFRAMS BEI HÖHEREN TEMPERATUREN

Der Einfluß der Schubspannung, Temperatur, Korngröße und Größe der Subkörner auf die statische Kriechgeschwindigkeit polykristallinen, unlegierten Wolframs wurde an Hand der zahlreichen veröffentlichten Daten untersucht. Es wurde gezeigt, daß die statische Kriechgeschwindigkeit polykristallinen Wolframs und anderer Metalle durch die Gleichung

$$\dot{\varepsilon} = S\lambda^2 \, \text{Deff} \left(\frac{\sigma}{E}\right)^7$$

gegeben ist, wobei λ die Größe der Subkörner, σ die Kriechspannung, E der mittlere, unrelaxierte E-Modul, S eine universale Kriechkonstante der ungefähren Größe 3×10^{40} cm^{-3} ist. Der effektive Diffusionskoeffizient Deff ist gleich $(D_L f_L \pm D_D f_D)$, wobei D_L der Gitterdiffusionskoeffizient, D_L der Versetzungsdiffusionskoeffizient und f_L und f_D die Anteile der Atome, die an Gitterdiffusion bzw. Diffusion entlang Versetzungskernen teilnehmen, sind. Wenn, wie in sich normal verhaltenden Materialien, sich Subkörner während des Kriechens bilden, dann ist $\lambda = k\,\sigma^{-1}$ (k ist eine Materialskonstante) und die oben angeschriebene Gleichung ergibt das allgemein beobachtet σ^5-Gesetz. Bilden sich während des Kriechens keine Subkörner (wie in vielen feuerfesten, pulvermetallurgisch hergestellten Metallen), so wird λ in obiger Gleichung durch die Korngröße ersetzt. In diesem Fall sind die Vorhersagen $\dot{\varepsilon} \sim \sigma^7$ und $\dot{\varepsilon} \sim L^2$ in Übereinstimmung mit experimentellen Ergebnissen. Das Versagen des Potenzgesetzes bei hohen Kriechspannungen hat zwei mögliche Erklärungen: (1) Die Bildung von Überschußleerstellen während der plastischen Verformung hat eine Zunahme von D_L im Deff-Term zur Folge oder (2) die Zunahme der Versetzungsdichte mit zunehmender Schubspannung beeinflußt den Anteil der Versetzungsdiffusion $D_D f_D$ von Deff.

1. INTRODUCTION

Extensive data are now available on the creep behavior of polycrystalline unalloyed tungsten.[1-8] Gilbert et al.[4] have analyzed some of these data and have classified the creep behavior of tungsten into four temperature regions, according to the apparent activation energy for creep. Using activation volume calculations, they assigned probable mechanisms to each temperature region. At high temperatures, that is for $T > 2200°C$ ($T/Tm > 0.65$) they conclude that dislocation climb is not the rate-controlling process on the basis of these calculations. Instead, they suggest that a process involving the nonconservative motion of jogged screw dislocations is rate-controlling. For a number of close-packed metals it has been suggested that dislocation climb is the rate-controlling process for creep above $T/Tm = 0.5$. The above results of Gilbert et al. suggest that perhaps in a more open structure, such as BCC tungsten, this is not the case.

At intermediate temperatures, that is, for $1200°C < T < 2200°C$ ($0.40 < T/Tm < 0.65$), Gilbert et al. suggest that creep occurs by nonconservative motion of jogged screw dislocations, assisted by short circuiting diffusion paths along dislocations and grain boundaries.

It is the purpose of this paper to analyze all the creep data currently available on tungsten but using a different approach from that of Gilbert et al. The apparent activation energy for creep will be corrected for the effects of temperature on the elastic modulus. Then, from the data, the relationship of the activation energy for creep to temperature will be constructed. This relationship will then be compared with the established curves for two f.c.c. metals, copper and aluminum. A stress–creep rate correlation of the type $\dot{\varepsilon}/D_{eff}$ (creep rate over effective diffusivity) against σ/E (stress over modulus) will then be developed. The purpose of this plot is twofold: (i) to determine if the creep of b.c.c. metals differs significantly from the creep behavior of f.c.c. metals, and (ii) to assess further the mechanism of creep of polycrystalline metals over a wide range of temperature.

2. PROCEDURE

The correction of the apparent activation energy for creep for the temperature dependence of the elastic modulus has been described by Ardell et al.[9] The available tungsten modulus data[10,11] are shown in Fig. 1. The single crystal modulus data reported by Armstrong and Brown were used, with the

indicated smooth extrapolation to temperatures above 2200°C.*

The raw data of creep rate as a function of temperature at various stresses for tungsten reported by various investigators[1,4-6] are shown in Fig. 2. Using the elastic modulus described above, a plot of creep rate against stress/modulus was constructed for each set of data. Examples of such plots are shown in Figs. 3 and 4 for the data of Flagella and Gilbert et al. From these curves, the corrected activation energy for creep is readily calculated from the expression

$$Qc]_{\sigma/E} = -Rd \ln \dot{\varepsilon}_s/d(1/T) \qquad (1)$$

where $\dot{\varepsilon}_s$ is the steady state creep rate, R the gas constant, T the absolute temperature, σ the creep stress and E the elastic modulus.

3. RESULTS AND DISCUSSION

A. The temperature dependence of the creep activation energy

The results of the calculation of the true activation energy for creep are shown in Fig. 5, in which the modulus corrected activation energy is seen to vary significantly with temperature. The vertical bars indicate the range of the values due to the nonparallelism of the lines as seen from Figs. 3 and 4. The horizontal bars indicate the temperature range for evaluation of Qc the data point located at the mean temperature.

For temperatures above 2200°C ($0.65\ Tm$) the activation energy scatter band appears to center about $Q = 140$ kcal/mole. At lower temperatures, 1200–2200°C (0.40–$0.65\ Tm$), the activation energy values appear to center around 90 kcal/mole. It would appear that the activation energy values may begin to rise again near 900°C ($0.3\ Tm$), but only a few data points were available at these temperatures to confirm this trend. The activation energy values are expected to fall off linearly with temperature at still lower temperatures, since this has been shown to be the case for copper,[13] aluminum,[14] and iron.[15]

The ratio of Qc to Q_L (where Q_L is the activation

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* Grain boundary relaxation effects apparently cause the increasing curvature with increasing temperature of the polycrystal modulus curve. Thus the smooth extrapolation of the single crystal data appears to be reasonable. The data of Lowrie and Gonas was not used, since their material was polycrystalline and of less than 100% density. In principle the shear modulus on the active slip plane should be used, if a Weertman-like model of the creep process is assumed.[12] However, since tungsten is essentially isotropic, only a small constant factor of error will be involved in using the average Young's modulus rather than the active slip plane shear modulus.
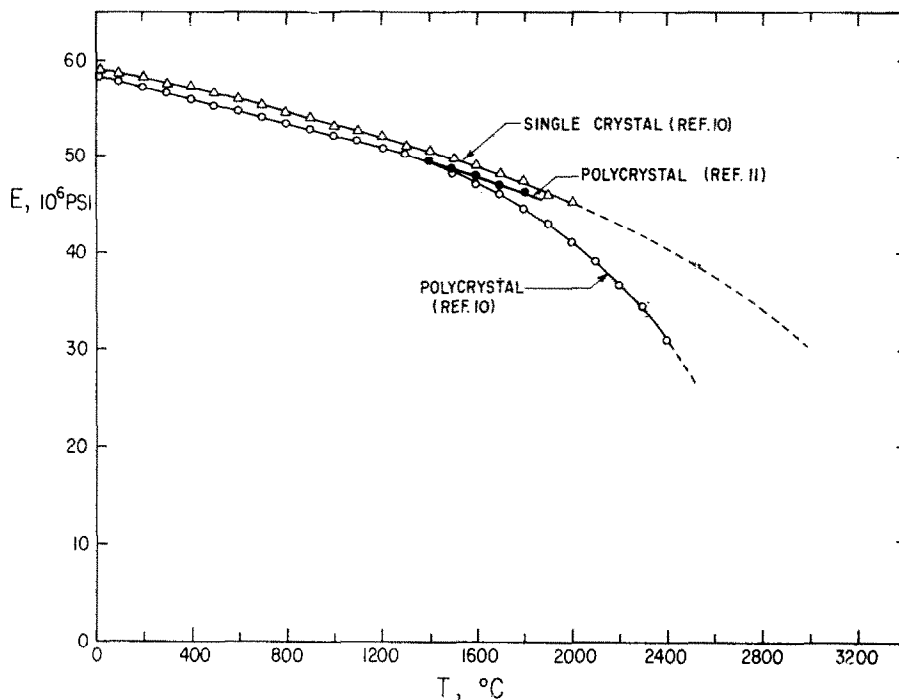
FIG. 1. The influence of temperature on the elastic modulus of tungsten.
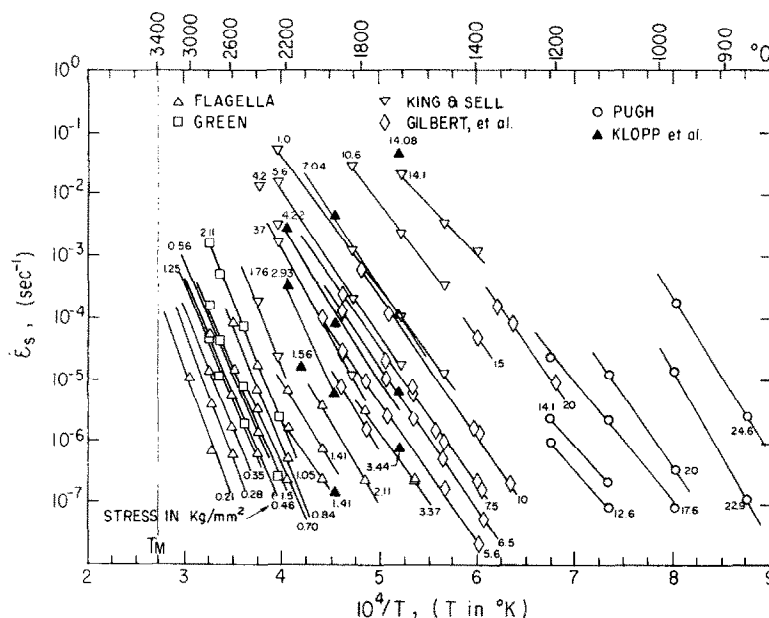

FIG. 2. The influence of stress and temperature on steady state creep for tungsten.

energy for lattice or volume self-diffusion) as a function of homologous temperature has been evaluated for aluminum[14] and copper[16] and such data are shown in Fig. 6. For comparison, a curve has been drawn through the tungsten data of Fig. 5, the values along this curve then being converted to Qc/Q_L ratios, and

the result was plotted on Fig. 6 as the heavy solid line. Here Q_L for tungsten was taken as 140 kcal/mole. The reasons for this choice are noted in the appendix.

From Fig. 6 several features can be noted. The shape of the Qc/Q_L curves for BCC tungsten and FCC copper are very similar over the temperature range
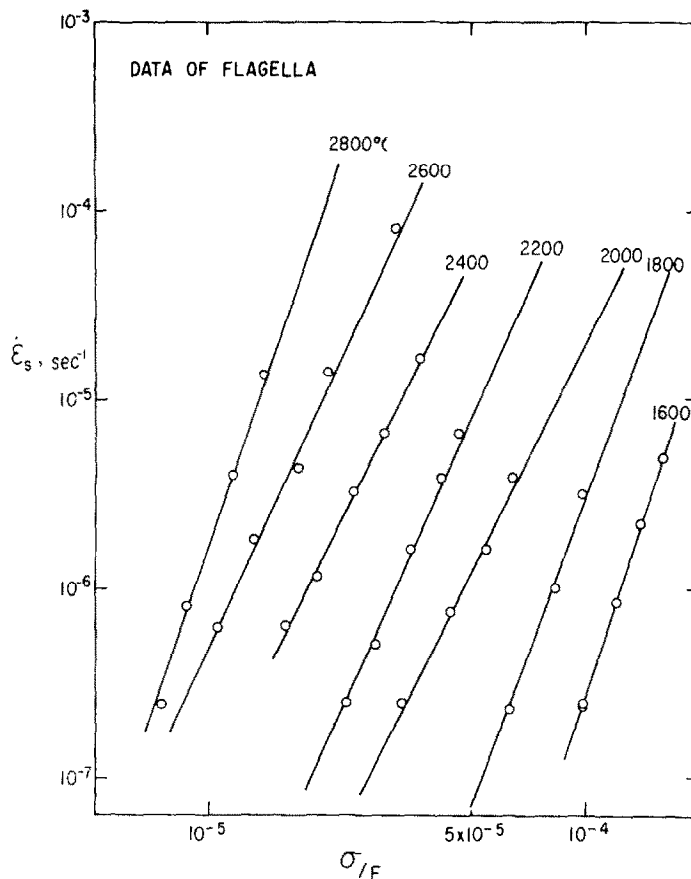
FIG. 3. Steady state creep rate of tungsten as a function of modulus corrected stress.

where both can be compared. When $T/Tm > 0.65$, the ratios of Qc/Q_L are both about one for tungsten and copper. It would appear that lattice self-diffusion is the rate controlling factor in this region. Both curves show a "plateau" region for temperatures below about $0.6Tm$, where Qc/Q_L is about equal to 0.6–0.65. The plateau value for tungsten is the same as the activation energy for dislocation diffusion (where $Q_D = 90,500$ cal per mole[17]) or grain boundary diffusion (where $Q_{GB} = 92,000$ cal per mole).[17] A similar observation has been made on copper by Barrett and Sherby.[16] They suggest that the intermediate temperature plateau in the copper activation energy values may be due to dislocation short circuiting assisting volume diffusion. It is proposed that dislocation short-circuiting may be the rate controlling process for creep of tungsten in the intermediate temperature region. This hypothesis will be tested more fully later.

The intermediate temperature plateau in aluminum is only different from copper or tungsten in that it occurs at a lower value of T/Tm, that is, it exists from 0.25 to $0.40Tm$. It has been suggested that the mechanism of creep for aluminum in this range is

due to cross-slip of screw dislocations.[18] The value of Qc/Q_L, however, is equal to 0.7 which is nearly the same as that found for Cu and W. It is thus possible that an alternate explanation for the low temperature plateau in aluminum is also due to lattice diffusion assisted by dislocation diffusion. No data are available, however, on the dislocation (or grain boundary) diffusivity in aluminum to test this hypothesis. The temperature and stress dependence of the dislocation density as well as the magnitude of the dislocation diffusivity are important factors in influencing the shape and position of the two plateaus. The nature of these problems will be discussed more fully in subsequent sections.

For BCC metals in the temperature region $0.25 < T/Tm < 0.50$, Conrad has suggested that the most likely rate-controlling mechanism is the interaction of dislocations and interstitial atoms.[15] Gilbert et al. have discussed available data on the activation energy of self diffusion of carbon, nitrogen and hydrogen in tungsten. They conclude that the interaction of interstitials and dislocations is probably not rate-controlling in this temperature region, since the activation energy for the diffusion of these impurities is
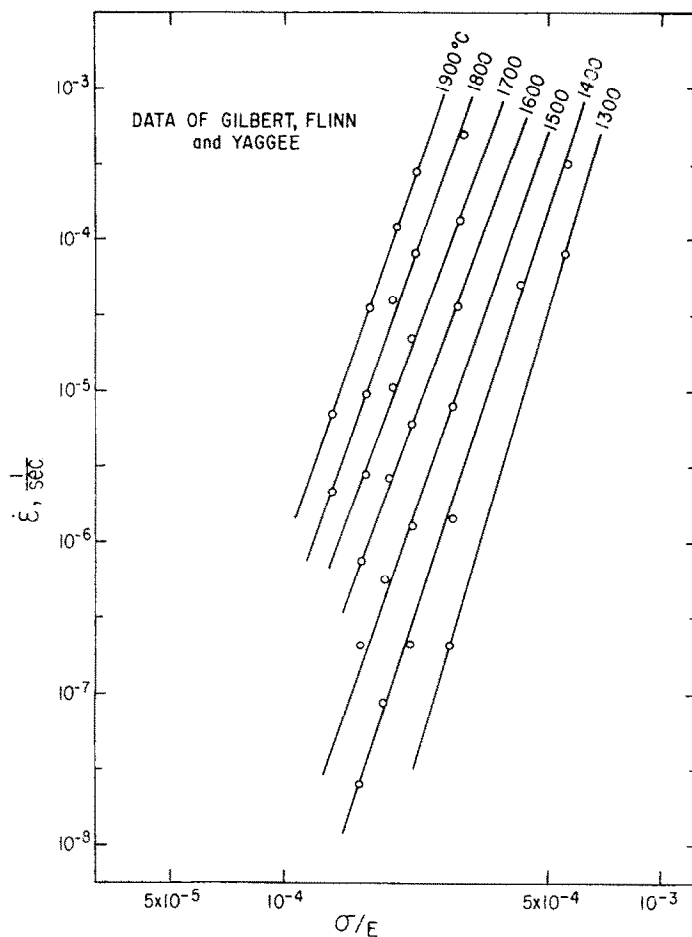
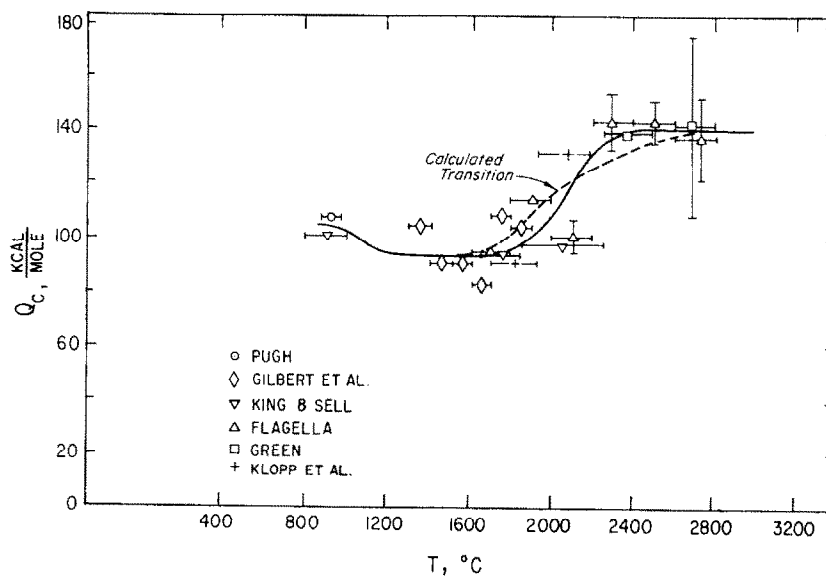FIG. 4. Steady state creep rate of tungsten as a function of modulus corrected stress.

FIG. 5. The modulus corrected activation energy for creep of tungsten as a function of temperature.
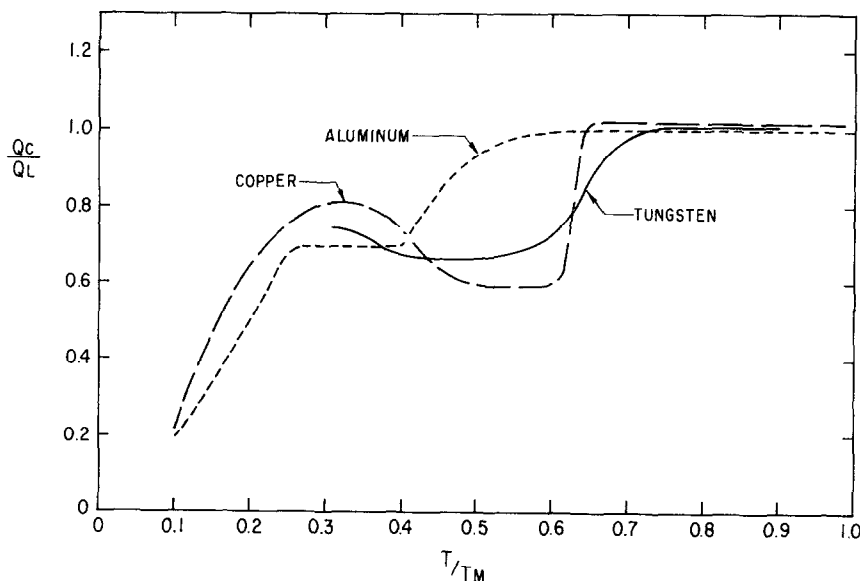
FIG. 6. Q creep/Q self-diffusion ratio as a function of homologous temperature for aluminum, copper and tungsten.

much smaller than the observed creep activation energy. We are in agreement with the position taken by Gilbert *et al.* on this point.

B. *The stress–creep rate correlation*

(i) *Creep at temperatures where lattice diffusion is controlling* (>0.65Tm). It has been shown that creep data at various temperatures and stresses can be correlated by plotting the creep rate divided by the diffusivity against stress divided by modulus.[19] As was established from Fig. 6, creep is lattice diffusion controlled at temperatures above 0.65Tm, and therefore the use of the lattice diffusivity is appropriate in this temperature region. Some diffusivity data for tungsten are documented in Fig. 7 and tabulated in Table 1. The relationship $D_L = 5.6 \exp (-140,000/RT)$ cm²/sec was chosen. The indicated dashed line in Fig. 7 shows the temperature dependence of the lattice diffusivity described above. The choice of this value is discussed in the appendix and is essentially that selected by Kreider and Bruggeman in their analyses of dislocation diffusion in tungsten.[17]

The correlation of creep rate/lattice diffusivity ($\dot{\varepsilon}/D_L$) against stress/modulus (σ/E) for data obtained at temperatures above 2200°C (0.65Tm) is shown in Fig. 8. As can be seen the data fall into two separate groups. A good correlation, within a scatter of a factor of two in $\dot{\varepsilon}/D_L$ at a given stress is obtained through about five orders of magnitude of $\dot{\varepsilon}/D_L$. For each curve the data appear to obey the power law expression

$$\frac{\dot{\varepsilon}}{D_L} = B \left(\frac{\sigma}{E}\right)^n \qquad (2)$$

where B is a structure constant and n is about 5 for curve A and about 7 for curve B. At high values of $\dot{\varepsilon}/D_L$, about $\dot{\varepsilon}/D_L > 10^9$, it would appear that the relation breaks down for the data represented by curve A. The power law breakdown at $\dot{\varepsilon}/D > 10^9$, in fact, has been shown to be typical for pure materials that exhibit $n = 5$.[25,26] In this respect, the tungsten behavior given by curve A appears to be typical of pure metals in general. The data of Green[1] given by curve B, indicates that his material is significantly stronger than that of Flagella[7] and King and Sell[6] (curve A). The reason for this discrepancy probably lies in the purity of the materials.

The comments to follow are centered on an attempt to understand the difference in behavior of the two sets of materials shown in Fig. 8. The resulting conclusions are important in our development of a phenomenological relation for creep of polycrystalline solids to be described in a later section. In general, powder metallurgy products are less pure than the corresponding vacuum melted products. It is our observation that refractory metals prepared by powder metallurgy techniques generally exhibit seven power law behavior[1,3,4,7] whereas refractory metals prepared by vacuum arc-casting or electron beam melting exhibit five power law behavior.[2,7,27,28] The latter would be considered "normal behaving materials" since most metals are known to exhibit this stress dependence at high temperature.[19] The only exception to the above generalizations appears to be the data of King and Sell;[6] their powder metallurgy tungsten creep data fall on the so-called normal behaving curve A. King and Sell noted that their
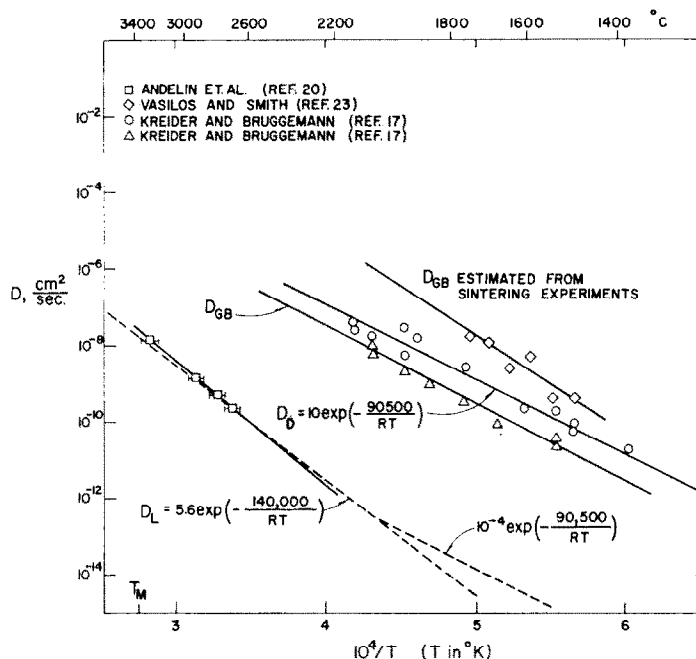
FIG. 7. Lattice, dislocation and grain boundary self-diffusivities in tungsten. D_L and D_D as shown were used for creep analyses.
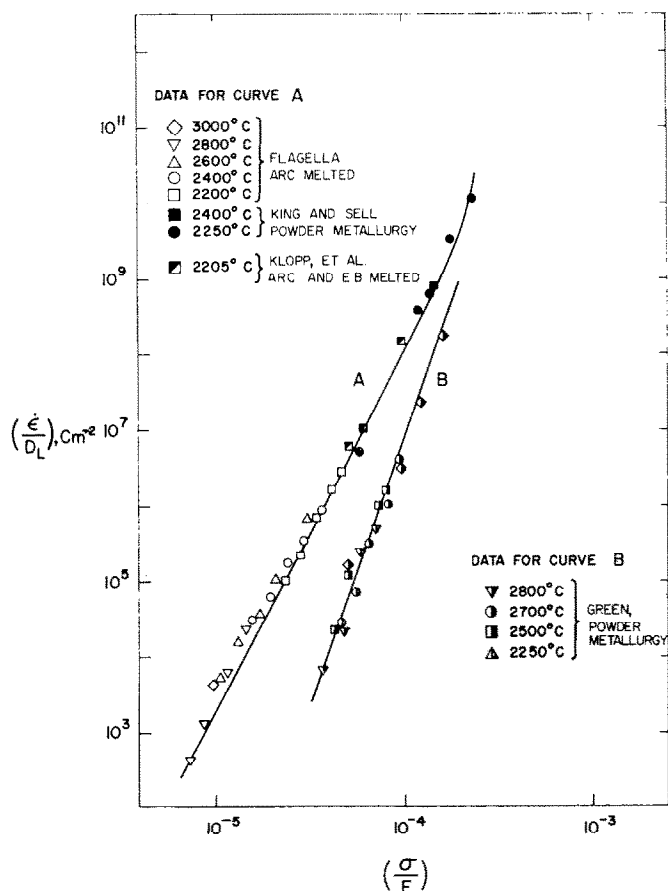

FIG. 8. The stress–creep rate correlation for tungsten at temperatures above $0.65 Tm$.

powder metallurgy samples were prepared by a proprietary process which resulted in "superior purity" tungsten.

We believe the difference in behavior between the two sets of materials shown in Fig. 8 is due to the influence of grain size and subgrain size on creep. From metallographic observations of Flagella[29] and Green[30] and their colleagues it appears that the refractory metals that exhibit five power law behavior develop subgrains during creep whereas materials that exhibit seven power law behavior do *not* develop subgrains during creep. Subgrains are known to be important barriers to creep and a material prepared with fine subgrains will creep slower than the same material containing coarse subgrains.[25] The steady state subgrain size, however, is a unique function of the creep stress generally following the relation $\lambda \alpha \sigma^{-1}$ where λ is the subgrain size and σ is the creep stress.[25] Such materials generally do not exhibit a strong grain size dependence on steady state creep rate.[31] This might be expected since the contribution of sub-grain boundary barriers to creep would overshadow the fewer barriers associated with the original grain boundaries (in general, subgrains are an order or two smaller in magnitude than the grain size). In agreement with this suggestion, Klopp et al.[2] found only a small grain size effect on the steady state creep rate of arc-cast electron beam melted tungsten. In contrast, Sutherland and Klopp[3] Taylor et al.[8] found a large effect of grain size on the steady state creep rate of tungsten prepared by powder metallurgy methods. The results revealed that the finer the grain size the lower the creep rate. This would be expected if grain boundaries are barriers to dislocations and no subgrains form in such powder metallurgy products. There were no inversions in the creep rate–grain size relation even at very fine grain sizes where grain boundary shearing may be expect to be important and could reverse the trend observed. Apparently, the impurities lodged at boundaries inhibit grain boundary shearing and in general failure occurred by cavitation at boundaries at right angles to the tensile direction (very little cavitation is observed after creep in arc melted tungsten).

From the above discussion we are now prepared to propose the following suggestion. The steady state creep rate might be proportional to the area swept out by a dislocation from its creation to its destruction. It is reasonable to assume that dislocations are terminated at grain boundaries or subgrain boundaries. In this case, it might be expected that the creep rate would be proportional to the area swept out per dislocation, or $\dot{\varepsilon} = \lambda^2 f(\sigma, T)$ for materials that form

subgrains (λ = subgrain diameter) and $\dot{\varepsilon} = L^2 f(\sigma, T)$ for materials that do not form subgrains (L = grain dia.). According to this consideration, $\dot{\varepsilon} L^{-2} = f(\sigma)$ at constant temperature for non-subgrain forming materials. Experimental confirmation for the proposed concept is shown in Fig. 9 for powder metallurgy tungsten creep data of Sutherland and Klopp,[3] Green[1] and Flagella.[7] The grain size difference for the various lots studied was over a factor of five so that the convergence of the data into a fairly narrow band is most encouraging. Furthermore, the resulting slope of the curve in Fig. 9 reveals that $\dot{\varepsilon} L^{-2}$ is proportional to about the seventh power of the stress, giving added confirmation for the seven power law behavior of the powder metallurgy samples shown in curve B in Fig. 8. The ideas developed here on the possible influence of grain size and subgrain size on creep will permit us to develop later a phenomenological explanation of the five and seven power law behavior observed in refractory metals.

(ii) *Creep at all temperatures above 0.4 Tm.* In a previous section, it was suggested that dislocation short-circuiting might be the creep-rate controlling process in the intermediate temperature plateau region ($0.40 < T/Tm > 0.65$). If this is the case, we may then describe the creep rate in terms of the effective diffusivity over that temperature range.

Therefore, following a method suggested by Hart,[32] we can write for the effective diffusion coefficient involving only lattice diffusivity and dislocation diffusivity, namely

$$\text{Deff} = D_L f_L + D_D f_D \qquad (3)$$

where D_L is the volume diffusivity, (equal to $Do_L \exp - Q_L/RT$), D_D is the dislocation diffusivity (equal to $Do_D \exp - Q_D/RT$), and f_L and f_D are the fraction of atom sites associated with each diffusion process. The stress–creep rate relation would then be in the form of $\dot{\varepsilon}/\text{Deff}$ as a function of σ/E.

The diffusion coefficient along dislocations in tungsten as reported by Kreider and Bruggeman[17] is $D_D = 10 \exp - 90,500/RT$ (see Fig. 7 and appendix). In order to calculate f_D it is necessary to know the typical dislocation density at the creep stresses used in the correlation between Qc and T shown in Fig. 5. Since it is our contention that the contribution of lattice diffusivity and dislocation diffusivity is about equal at the transition temperature in the $Qc-Tm$ correlation, one can calculate f_D by equating $D_L f_L$ to $D_D f_D$ at 2000°C (Fig. 5). The term f_L is essentially unity and f_D is calculated to equal 10^{-5}. The effective
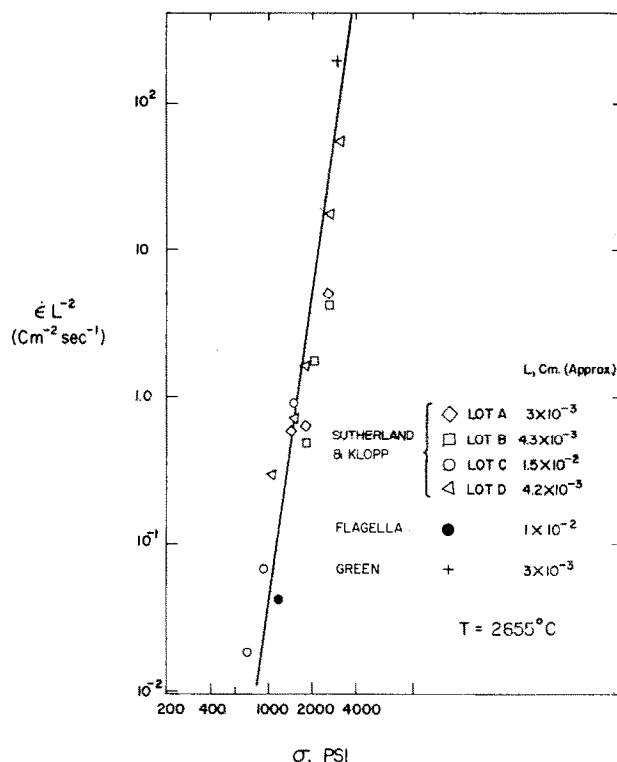
FIG. 9. The influence of grain size on creep of powder metallurgy tungsten samples, suggesting that the creep rate is proportional to grain diameter squared and to the seventh power of the stress.

diffusivity can now be described by the equation

$$\text{Deff} = 5.6 \exp\left[-\frac{140,000}{RT}\right] + 10^{-4} \exp\left[-\frac{90,500}{RT}\right].$$

$$(4)$$

The dislocation density ρ, appropriate to the value of f_D selected to correlate with the creep data can be calculated. The value of ρ is given by

$$\rho = \frac{f_D N}{n}$$

$$(5)$$

where N = number of atoms per cm^2 and n = number of atoms participating in dislocation short-circuiting at a dislocation site. Shewmon[33] suggests that n about equals 10 for the case of static dislocations (it may be higher for the case of dynamic dislocations). With $N = 10^{15}$ and $f_D = 10^{-5}$, this yields $\rho = 10^9$ dislocations/cm^2. This is the dislocation density we expect to see for creep specimens tested at stresses in the vicinity of $\sigma/E = 10^{-4}$ and higher. Although data are not yet available in the literature for the dislocation density in tungsten creep samples, it is encouraging to note that for iron–silicon[34] and for iron[35] values of ρ for specimens crept at $\sigma/E = 10^{-4}$ range about 10^8 to 5×10^8 cm^{-2}.

It is now possible to correlate all of the creep data for tungsten above $0.5Tm$ by means of equation (4) using Deff. The stress–creep rate correlation for data obtained at temperatures above $0.5Tm$ is shown in Fig. 10. The correlation obtained appears to be at least as good as that shown in Fig. 3. It seems quite probable therefore, that the effective diffusivity is the creep rate controlling factor at temperatures above $0.5Tm$.

As was seen in Fig. 8, the data again appear to separate into two distinct groups. One group, given by curve A in Fig. 10 exhibits about 5 power law behavior, and breakdown of the power law at $\dot{e}_s/$ Deff $= 10^9$, i.e. typical pure metal behavior. The other group shown in curve B, exhibits 7 power law behavior and does not show power law breakdown behavior which is not typical of pure metals; it may be typical of materials that do not develop subgrains. The excellent correlation shown in Curve B for different investigators probably represent materials that have about the same grain size. As mentioned in the earlier section, other parallel lines with $n = 7$ are obtained for different grain sizes.

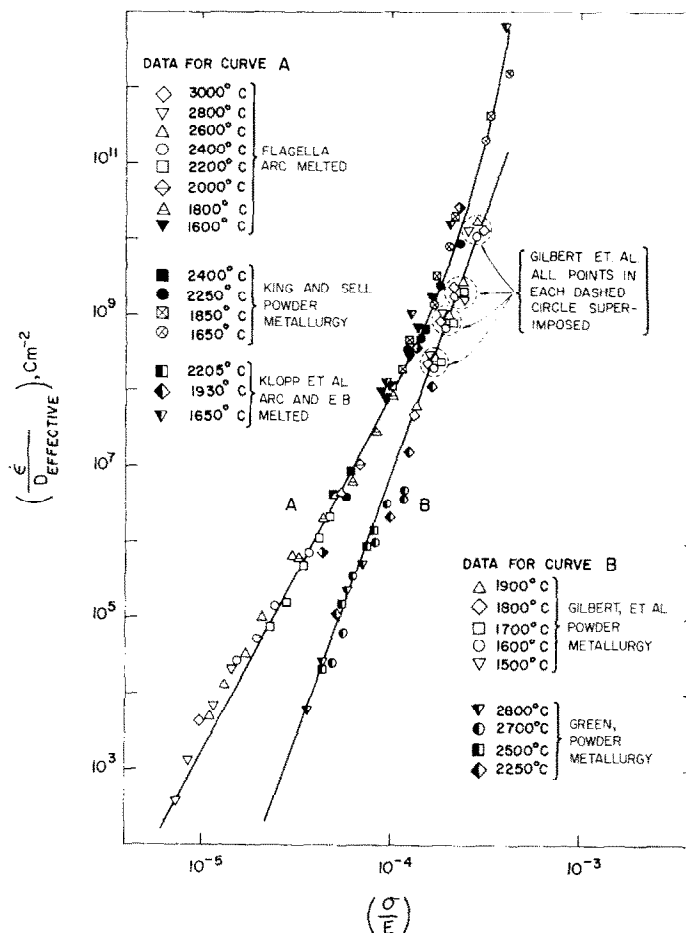The correlation of Fig. 10 indicates that diffusion

FIG. 10. The stress–creep rate correlation for tungsten at temperatures greater than $0.5\,Tm$, showing the separation of data into two distinct types according to purity and method of preparation.

in the lattice and along dislocation lines is the rate-controlling process for creep at temperatures above $0.5\,Tm$. Therefore, the temperature dependence of the creep rate should be directly related to the temperature dependence of the effective diffusivity. Since $\dot{\varepsilon}\,\alpha\,\mathrm{Deff}$, we can calculate Qc, the creep activation energy, by differentiating equation (3), namely

$$Qc = -\frac{R\,d\ln\dot{\varepsilon}}{d\,1/T}\bigg|_{\sigma/E} = -\frac{R\,d\ln\mathrm{Deff}}{d\,1/T}$$

or

$$Qc = \frac{Do_L f_L Q_L \exp-\dfrac{Q_L}{RT} + Do_D f_D Q_D \exp-\dfrac{Q_D}{RT}}{Do_L f_L \exp-\dfrac{Q_L}{RT} + Do_D f_D \exp-\dfrac{Q_D}{RT}}.$$

(6)

Using the values given in equation (4), the indicated dotted line in Fig. 5 is calculated. The results are indeed most encouraging, although the temperature dependence of the calculated Qc is somewhat more gradual than the experimental curve. This difference is in fact not surprising. Equation (6) contains the term f_D which was assumed to be a constant but, in fact, it is variable with the creep stress. Since most creep investigators test at combinations of creep stress and temperature which give experimentally convenient creep rates, the creep stress generally decreases for increasing temperature. Since $\rho \propto \sigma^{\delta}$ where δ may be two or three,[34] ρ and hence f_D may decrease moderately rapidly with increasing temperature. The result of the dislocation density stress dependence would then be to sharpen the calculated transition, so that it is more nearly in accord with the observed transition. In the general case, therefore, the temperature dependence of the effective diffusivity to describe the creep rate would be slightly more complex than that given by equation (4).

C. Mechanism of creep in polycrystalline metals, with special reference to tungsten, above 0.4Tm

The analyses made on various grades of poly-crystalline tungsten at low (0.4–0.65Tm) and at high (>0.65Tm) temperature permits us to suggest two phenomenological relations for creep of polycrystalline metals. For those polycrystalline metals that do not form subgrains the steady state creep rate is given by

$$\dot{\varepsilon} = S'L^2 \, \mathrm{Deff} \left(\frac{\sigma}{E}\right)^7 \qquad (7)$$

where S' might be a universal constant. The value of S' derived from the tungsten data yields 7×10^{40} cm^{-4} (calculated from the data given in Fig. 9, equation (4) and Fig. 1).

For the case of polycrystalline metals that do form subgrains during creep, we propose that the steady state creep rate, in the power law region, is expressed by the same type of relation as given by equation (7), namely,

$$\dot{\varepsilon} = S\lambda^2 \, \mathrm{Deff} \left(\frac{\sigma}{E}\right)^7 \qquad (8a)$$

where S might be a universal constant for pure metals.

Equation (8a) reduces to the more familiar five power law relation when it is remembered that λ in general is given by $\lambda = k\sigma^{-1}$. Substituting this expression into equation (8a) yields

$$\dot{\varepsilon} = S'' \, \mathrm{Deff} \, \frac{\sigma^5}{E^7} \qquad (8b)$$

where $S'' = Sk^2$ with k being a constant for a given metal but generally varying from material to material.

There are a number of important aspects with regard to equation (8a). This equation was derived from a purely phenomenological approach by examining various creep data. It has, however, considerable theoretical justification since both the λ and Deff terms are reasonable from a dislocation glide or climb viewpoint. The high stress dependence (seven power) has not yet been assessed by theoretical models for creep. It is possible to estimate the value of S in equation (8a) from the tungsten data and knowledge of the sub-grain size at a given σ/E. An approximate value of λ can be determined from a photomicrograph reported by Conway and Flagella,[27] for arc melted tungsten samples crept at 1800°C and $\sigma = 3500$ psi ($\lambda \cong 4 \times 10^{-3}$ cm). At this temperature $E = 47.5 \times 10^6$ psi, yielding $\sigma/E = 7.36 \times 10^{-5}$. From Fig. 10, $\dot{\varepsilon}/\mathrm{Deff}$ at this stress is estimated to be 2×10^7 cm^{-2}. Thus $S = 10^{41}$ cm^{-4}. Additional values on subgrain size variation with stress reported by

Bartram and Rau[36] yield similar values of S. It is possible to calculate S values for creep data for other pure polycrystalline solids that develop subgrains.[25] These other data, which represent among the few metals for which subgrain sizes have been measured during creep (aluminum, sodium chloride, alpha iron, nickel and copper) yield $S \cong 1 \times 10^{40}$ cm^{-4}. Although there is an order of magnitude difference between the average S for other materials and the S calculated for tungsten, we are tempted to assume that S may be (almost) a universal constant equal to about 3×10^{40} cm^{-4}.

Thus, we believe that equation (8a) will permit a prediction of the steady state creep rate of pure polycrystalline metals if the subgrain size, effective diffusivity and unrelaxed average elastic modulus are known. Although Deff and E can usually be obtained from the literature or occasionally estimated, λ at the present time cannot be calculated by contemporary dislocation theories and one must resort to a creep test to obtain λ. As mentioned earlier, however, it seems to be fairly well established that $\lambda = k\sigma^{-1}$ (at temperatures above 0.4Tm) so that if a single λ is determined at a given stress for a given material, then one is able to estimate the creep rate at any other stress and temperature (provided again that Deff and E are known). Another interesting possibility with equation (8a) is that creep tests may provide a facile method of determining Deff (probably within a factor of 2) since it is relatively easy to do creep tests and to determine the subgrain size. This method may be especially valuable to determine Deff (as influenced only by lattice and dislocation diffusion) at low temperatures (0.4–0.6Tm) where diffusion experiments are difficult to perform mostly because of the long times of exposures needed to make reliable measurements of the depths of diffusion penetration.

The proposed equation for creep of pure metals [equation (8a)] does not contain a stacking fault energy term as was proposed in studies on the influence of stacking fault energy on creep of pure metals and solid solutions.[31,38] It is possible that the influence of stacking fault energy on creep is primarily due to its influence on the subgrain size developed at a given stress; thus, it is an experimental fact that smaller subgrains are developed in a material with a low stacking fault energy (like copper) than in a material with a high stacking fault energy (like aluminum) when compared at the same value of stress over modulus.

One further point is worthy of note. The constant S' for materials that do not form subgrains [equation (7)] has about the same value as the constant S

for materials that form subgrains [equation (8a)]. This result would suggest that subgrain boundaries which are considered to consist of discrete dislocations with accompanying long range forces, are about equivalent barriers to plastic flow as random grain boundaries. In the latter boundaries, long range forces may not exist, although, for the case of tungsten at least, they may contain a high concentration of impurity atoms. One general problem exists, however, for the case of materials that develop subgrains. That is the problem of their stability during creep. If a material is prepared with a very fine subgrain size and then crept at a given stress where the equilibrium subgrain size is coarse, the subgrains will grow (with an accompanying accelerating stage of creep) until the stable coarse subgrain size is obtained. On the other hand, fine grain size materials prepared by powder metallurgy methods, although they can be stable, are oftentimes considerably less ductile than the corresponding material prepared from the melt.[7]

The steady state creep expression given by equation (8a) differs in several respects from contemporary creep theories. The two most commonly quoted theories are those of Weertman, where creep is controlled by dislocation climb in a glide and climb process,[12] and of Barrett and Nix where creep is controlled by the glide of jogged screw dislocations.[34] A recent theory by Nabarro[39] involves a creep process controlled by dislocation climb without accompanying glide. All three theories predict that the steady state creep rate should be proportional to the lattice diffusivity (with one exception in the case of the Nabarro theories). The Weertman theory predicts that the creep rate should be proportional to $\sigma^{4.5}$ in the power law range, and the Barrett–Nix theory also predicts a power law relation if the mobile dislocation density, ρ, is proportional to σ^{δ} (as appears to be the case for an Fe–3% Si alloy). Nabarro's theory predicts $\varepsilon = K\sigma^n$ where n can equal 1, 3 or 5 depending on whether grain boundaries are sources and sinks for vacancies, dislocations are the sources and sinks or whether core diffusion is rate-controlling.

None of the three theories of creep consider the possible influence of subgrain size on creep and none utilize the concept of an effective diffusion coefficient involving lattice and dislocation short-circuit diffusion (within the same stress range).

Contemporary creep theories suggest that the elastic modulus contribution to creep is smaller than that predicted by the phenomenological [equation (8a)], where $\dot{\varepsilon} \alpha E^{-7}$. The Weertman theory predicts $\dot{\varepsilon} \alpha E^{-3.5}$ whereas the Nabarro theory predicts $\dot{\varepsilon} \alpha E^{-2}$ when $n = 3$ and $\dot{\varepsilon} \alpha E^{-4}$ when $n = 5$.

The seven power law stress dependence of the creep rate given by equations (7) and (8a) is not easy to explain and needs considerable further theoretical attention. Barrett has discussed the stress dependence of high temperature creep[40] and notes that glide theories predict that the creep rate is given by

$$\dot{\varepsilon}_s = A\rho \sinh \frac{\sigma V}{kT} \qquad (9)$$

where V is the activation volume, k is Boltzmann's constant and A is a material constant. Barrett proposes that $\rho \alpha \sigma^{\delta}$ and $\delta = 2\sigma/(\sigma - \sigma_0)$ where σ_0 is a friction stress. This approach gives ρ a large stress dependence when $\sigma \cong \sigma_0$, and hence the creep rate also takes on a large stress dependence. A power law for the creep rate can thus be predicted where n can equal any large number including the value seven. It is to be pointed out, however, that such a relation cannot explain the validity of $\dot{\varepsilon} \alpha \sigma^7$ for the wide stress range observed (Figs. 8 and 10) unless σ_0 is assumed to be some complex function of $\dot{\varepsilon}$ and T.

The power law breakdown at high value of $\dot{\varepsilon}/D_{eff}$ observed for tungsten is typical of that observed in other metals. It appears to be an experimental fact that this occurs at $\dot{\varepsilon}/D_{eff} \cong 10^9$.[19,20,25] Both the Weertman and the Barrett–Nix expression for creep predict a power law breakdown at high stresses. The physical explanation for this occurrence is the same for both models and involves the presence of a high vacancy concentration above the equilibrium number; both theories deal with dislocations as source and sinks for vacancies. It is relatively simple to introduce a similar concept to the phenomenological equation (8a) to predict a power law breakdown at high stresses. The approach is to consider the lattice diffusion coefficient, D_L in the Deff term. Here $D_L = n_v D_v$ where n_v is the vacancy concentration and D_v is the diffusion coefficient of vacancies. Under creep conditions $n_v = n_\tau + n_\varepsilon$ where n_τ is the equilibrium vacancy concentration and n_ε is the vacancy concentration produced during creep deformation. The value of n_ε is dependent on the rate of vacancy generation and the life-time of the vacancy; each factor in turn is probably a function of the creep stress and temperature. Although it is not our intention to do so at this time, it is possible to develop an expression for n_ε with σ/E which could qualitatively explain the power law breakdown at high stresses.

Another possible explanation of the power law breakdown may be associated with the influence of stress on dislocation density which in turn would influence the dislocation diffusivity term in Deff.

Some simple estimates can be made from this contribution. The dislocation density change with stress has been studied extensively for the case of an iron–3% silicon alloy by Barrett and Nix.[34] It has been shown that the relation between the density of free dislocations and creep stress for this alloy can be given[41] by $\rho = \rho_0 e^{B\sigma}$ where $\rho_0 =$ dislocation density at zero stress and B is a material constant (equal to 3.54×10^{-4} psi^{-1} for the Fe–3% Si alloy). One can make some calculations for the case of tungsten to determine if this approach appears feasible for explaining the power law breakdown. For simplicity, let us calculate the creep rate–stress relation for the low temperature range, say 1600–1900°C where Deff would be dominated by the term $D_D f_D$. Thus, in this case, equation (8b) becomes

$$\dot{\varepsilon} = \frac{S''}{E^7} D_D f_D \sigma^5. \tag{10}$$

Over the short temperature interval, 1600–1900°C, it is reasonable to assume that the elastic modulus is constant ($E_{1750°C} = 48.0 \times 10^6$ psi), thus $S''/E^7 = 3.36 \times 10^{-11}$ cm^{-2} psi^{-5}. From equation (5) $f_D = 10^{-14} \rho = 10^{-14} \rho_0 e^{B\sigma}$. Thus, equation (10) can be rewritten as

$$\frac{\dot{\varepsilon}}{D_D} = [3.36 \times 10^{-25}] \rho_0 e^{B\sigma} \sigma^5. \tag{11}$$

The units for $\dot{\varepsilon}/D$ are in cm^{-2} when ρ is expressed in dislocations per cm^2 and σ in psi. Now, we need only estimate ρ_0 and B to attempt to fit the experimental data. In developing the exact expression for the effective diffusion coefficient given in equation (4) we concluded that $\rho = 10^9 = \rho_0 e^{B\sigma}$ at $\sigma/E \cong 1 \times 10^{-4}$. At low values of the creep stress $e^{B\sigma} \cong 1$ and $\rho = \rho_0$. At the highest stress used in the tungsten creep study ($\sigma = 20,000$ psi) the creep rate is faster by a factor of 100 than a linear extrapolation of the power law region predicts. Thus $e^{B \times 20,000} = 100$ or $B = 2.3 \times 10^{-4}$ psi^{-1}. From this one calculates $\rho_0 = 3.3 \times 10^8$ dislocations per cm^2.

A comparison of the $\dot{\varepsilon}/D$ vs. σ relation for tungsten as predicted by means of equation (11) and the actual "low temperature" data reported by Flagella[7] and by Klopp et al.[2] is shown in Fig. 11. The predicted curve in Fig. 11 illustrates a more or less power law region at low stresses and an upsweep region at high stresses. These results suggest that the power law breakdown can be explained by the increasing influence of dislocation short-circuit diffusion as the dislocation density increases with increasing stress.

Although there may seem to be a great deal of flexibility in manipulating equation (8a) to fit in with the experimental data, this is not really the case. There are basically three constants that can be altered, namely, ρ_0 and B in the dislocation density term and n, the number of atoms around each dislocation site participating in dislocation short circuiting diffusion. The values of ρ_0 and B must be reasonable and certainly the values obtained for tungsten are realistic (comparable to experimental values obtained for Fe–3% Si). The experimentally determined value of ρ_0 for tungsten, however, may well be lower than that used to make the correlation shown in Fig. 11 (i.e. $\rho_0 < 3.3 \times 10^8$ cm^{-2}). If this becomes a reality then n will have to be increased above the value of 10 atoms in order to obtain the good correlation shown in Figs. 8, 10 and 11. The number 10 for n was chosen by Shewmon[33] in his study of static diffusivity as influenced by dislocation short-circuiting. Perhaps for moving dislocations n may be an order of magnitude higher. Furthermore, there is a hint that n may be higher than 10 for the case of static dislocations. Thus, if the effective diffusivity for tungsten as measured by Neuman and Hirschwald (Table 1) is due primarily to dislocation short circuiting, n can be readily calculated. On the assumption that $\rho = 10^9$ cm^{-2}, $n = 40$, and for $\rho = 10^8$ cm^{-2}, $n = 400$. From these calculations it can be seen that rather high values of n are obtained for a reasonable value of dislocation density. It is anticipated that further studies will shed light on this important question. When the significance and value of n is clarified, it may be possible to calculate D_D from creep experiments.

4. SUMMARY AND CONCLUSIONS

The influence of stress, temperature, grain size and subgrain size on the steady state creep rate of polycrystalline unalloyed tungsten has been examined based on the extensive published data available.[1–8] It is found that for temperatures above 2200°C ($0.65 Tm$) the modulus corrected creep activation energy was about 140 kcal/mole, while for temperatures 1200–2200°C (0.40–$0.65 Tm$) it was about 90 kcal/mole. The first value is near to that reported experimentally for tungsten lattice self diffusion, while the second is equal to the activation energy for dislocation diffusion.

It is shown that the steady state creep rate of unalloyed polycrystalline tungsten is proportional to an effective diffusion coefficient involving lattice and dislocation short-circuit diffusion. That is, $\dot{\varepsilon} \alpha$ Deff $\alpha (D_L f_L + D_D f_D)$. Here, D_L is the lattice diffusion coefficient, D_D is the dislocation diffusion coefficient,
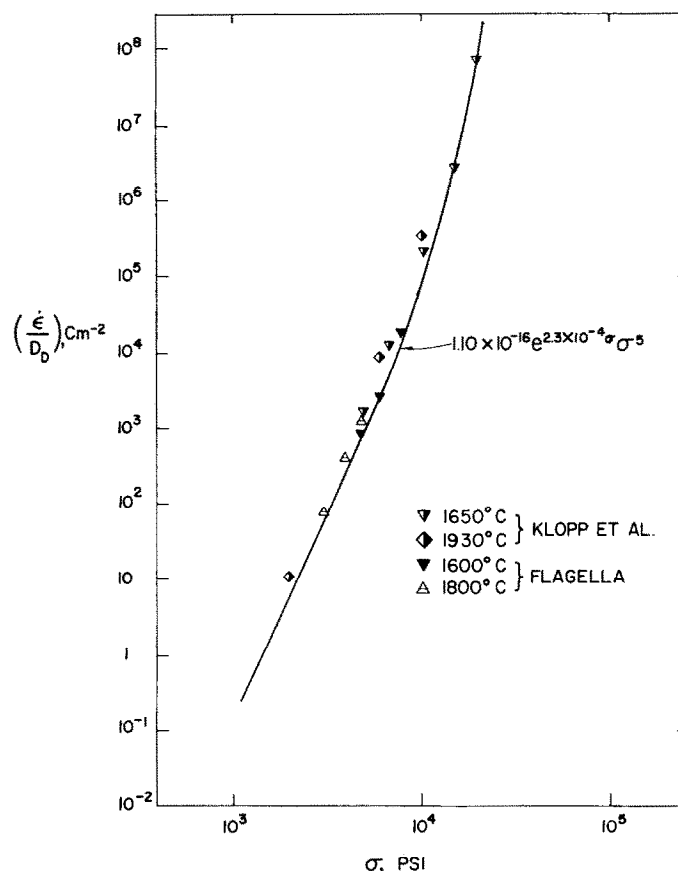
FIG. 11. Comparison of the creep behavior of arc melted and electron beam melted tungsten at low temperatures (1600–1900°C) with equation (11). The resulting correlation suggests that upsweep behavior at high stresses may be due to the increase in dislocation density with increasing stress, which influences the dislocation diffusivity term in Deff.

TABLE 1. Volume, grain boundary and dislocation self diffusion in tungsten

	Temp. range (°C)	Do (cm²/sec)	Q (kcal/mole)	Investigator	Comments
Volume	2000–2700	0.29	120.5	Danneberg[21]	Polycrystal wires used
	1287–1453	6.3×10^7	135.8	Vasilev and Chernormorchenko[24]	Results reveal diffusion coefficient that are orders of magnitude higher than in other investigations
	2676–3228	42.8	153.1	Andelin et al.[20]	Single crystal data
	1740–2100	1.80×10^{-3}	93.1	Neumann and Hirschwald[22]	Polycrystals. The low temperatures of measurement suggest that an effective diffusion coefficient was measured involving grain boundary and dislocation short circuiting
Grain boundary	1300–1750	3×10^4	110.7	Vasilos and Smith[23]	
	1450–2200	3.73	92.0	Kreider and Bruggeman[17]	
Dislocation	1400–2200	10	90.5	Kreider and Bruggeman[17]	

and f_L and f_D are the fraction of atoms participating in lattice and dislocation diffusion respectively. By using a reasonable value for f_D (based on the presence of about 10^9 dislocations per cm^2) it was possible to show that the steady state creep rate of polycrystalline tungsten is given by

$$\dot\varepsilon = S\lambda^2 \, \mathrm{Deff} \left(\frac{\sigma}{E}\right)^7 \qquad (8a)$$

where λ = subgrain size, σ = creep stress, E = average unrelaxed elastic modulus and S is a constant equal to about $10^{41} \, cm^{-4}$. The value S compares favorably with S values obtained for other metals (within an order of magnitude) and may be considered a universal creep constant.

It appears to be a general observation that the subgrain variation with creep stress follows $\lambda = k\sigma^{-1}$, where k is a material constant. Substituting this expression into the above equation yields the usually observed five power law relation, namely

$$\dot\varepsilon = S'' \, \mathrm{Deff} \, \frac{\sigma^5}{E^7} . \qquad (8b)$$

where $S'' = Sk^2$.

In powder metallurgy tungsten samples subgrains do not form during creep. In this case λ in equation (8a) is now equal to the grain size L. Since L does not change with the creep stress, equation (8a) predicts that $\dot\varepsilon \propto \sigma^7$ and $\dot\varepsilon \propto L^2$. Both predictions are in agreement with the behavior of powder metallurgy tungsten samples. Similar trends are observed in molybdenum.

The power law relation for creep breaks down at high stresses, specifically at $\dot\varepsilon/\mathrm{Deff} > 10^9 \, cm^{-2}$. There are two possible explanations for the higher creep rate than a linear extrapolation predicts at high stresses. One possibility is due to the mechanical creation of vacancies at high stresses which will increase the lattice diffusion coefficient term in Deff. The second possibility is that the increase in dislocation density with increasing stress (at high stresses) will influence the dislocation diffusion term (i.e. $D_D f_D$) in Deff.

The phenomenological creep expression, equation (8a), is reasonable from contemporary understanding of dislocations and their influence on high temperature mechanical properties. The λ^2 term can be related to the area swept out by a dislocation from its creation to its elimination at the subgrain boundary. The Deff term may represent the rate limiting process of dislocation climb of edge dislocations or of motion of jogged screw dislocations. Neither the theories of Weertman,[12] or of Barrett and Nix,[34] as well

as the more recent theory of Nabarro,[39] take into account these two factors. Although all of the three creep theories predict power law behavior at low stresses, the value of the exponent is always less than seven (typically between 3 and 5).

It would appear that the creep behavior of tungsten, a b.c.c. metal, is analogous to the behavior of other crystalline metals. Equation (8a) appears applicable to the creep of other pure materials (Al, Cu, α-Fe, Ni and NaCl). The breakdown of the power law for creep for tungsten occurs at the same value of $\dot\varepsilon/\mathrm{Deff}$ as for other metals.[25,26] There is, however, a disparity in the Qc/Q_L vs. T/Tm curves between the W and Cu data and the Al data. That is, the transition from $Qc/Q_L = 1.0$ to $Qc/Q_L = 0.7$ occurs at $0.45Tm$ for Al (Fig. 6) rather than at $0.65Tm$ as observed for the other two metals. If the transition in aluminum is attributable to the same reason used to explain the W and Cu data it can be explained by either (i) a very low D_D for aluminum, (when compared on a homologous temperature basis), (ii) a low value of dislocation density for aluminum at the transition temperature, or (iii) a very low value of n, the number of atoms contributing to dislocation short circuiting at a given dislocation site.

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APPENDIX

Data on lattice, grain boundary and dislocation self diffusion in tungsten are shown in Fig. 7. Most of the data reported for diffusion in tungsten are summarized in Table 1. The lattice diffusivities reported by Danneberg,[21] and by Vasilev and Chernormorchenko[24] are not shown in Fig. 7, as they are not considered indicative of the true value of the lattice diffusivity. Andelin *et al.*[20] have discussed the difficulties associated with these two values of the volume diffusion coefficient.

The long-dashed line shown in Fig. 7 was taken as the volume self diffusion coefficient of tungsten. It is represented by $D = 5.6 \exp(-140{,}000/RT)$ cm²/sec. The reasons for this choice, rather than selecting the equation of Andelin *et al.* ($D = 42.8 \exp(-153{,}100/RT)$ cm²/sec) are discussed below. Extensive evidence has indicated that for most pure metals, Q creep and Q self diffusion have about the same value. Therefore we have assumed that the modulus-corrected creep activation energy for tungsten at high temperature, 140 kcal/mole (Fig. 5), is a probable value for the activation energy for lattice self-diffusion. The chosen value of $Q_L = 140$ kcal/mole is in agreement with empirical formulas developed for prediction of self diffusion activation energies. The formula of Sherby and Sinmad[42] gives 138 or 146 kcal/mole, depending upon whether the valence of tungsten is taken as 5 or 6. (It would seem unlikely that the valence of tungsten is greater than 6.) The formula of Nachtrieb and Handler[43] based on knowing the latent heat of melting, yields $Q_L = 139$ kcal/mole. Thus we favor the selected value of $Q_L = 140$ kcal/mole over the experimentally calculated value of $Q_L = 153.1$ kcal/mole reported by Andelin *et al.*

The diffusivity curve we selected was made to fall within the data of Andelin *et al.* (see Fig. 7) yielding a D_0 value of 5.6 cm²/sec. This value of D_0 is considerably less than the value given by Andelin *et al.* of 42.8 cm²/sec. It is to be pointed out that most values of D_0 for b.c.c. metals are 10 cm²/sec or less.[42] For this reason, and for the above arguments regarding the value of Q chosen, we believe our expression for the lattice self diffusion coefficient of tungsten is more correct than the one given by Andelin *et al.* Quite independent of our analyses, Kreider and Bruggeman,[17] in their studies on grain boundary and dislocation diffusion used an equation for the lattice coefficient ($D_L = 7.5 \exp -141{,}000/RT$) very nearly equal to the one we selected for analyses of the creep data.

Two different lines are shown in Fig. 7, representing grain boundary self-diffusion in tungsten. The lower curve is that obtained by Kreider and Bruggeman from direct tracer experiments. The other results obtained by Vasilos and Smith were deduced from a study of sintering kinetics utilizing a theory of Coble for calculating D_{GB}. The higher grain boundary diffusion coefficient obtained from sintering studies would suggest that the theory of Coble may not take into account all the rate-controlling factors occurring

during sintering (this conclusion is, of course, based on the assumption that the grain boundary diffusion data by Kreider and Bruggeman is correct).

The dislocation diffusion coefficient is slightly higher than the grain boundary diffusion coefficient obtained by tracer methods (Fig. 7) but with essentially the same slope. This correlation is supported by a similar result found for silver where D_D was slightly greater than D_{GB} but the same activation energy described both types of diffusion.[44] In the original publication by Kreider and Bruggeman they erroneously calculated $(D_0)_D$ as 0.1 cm² per sec rather than 10 cm² per sec as used in our studies. This came about from an accidental error by the authors in their use of the Turnbull and Hoffman equation for calculating D_D.