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Low-Temperature Deformation Behaviour of Polycrystalline Copper

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Low-temperature plastic flow in copper was investigated by studying its tensile and creep deformation characteristics. The dependence of the flow stress on temperature and strain rate was used to evaluate the thermal activation energy while the activation area was derived from the change-in-stress creep experiments. A value of 0.6 eV was obtained for the total obstacle energy both in electrolytic and commerical copper. The activation areas in copper of three selected purities fell in the range 1200 to 100 b^2. A forest intersection mechanism seems to control the temperature dependent part of the flow stress. The increase in the athermal component of the flow stress with impurity content in copper is attributed to a change in the dislocation density. The investigation also revealed that thermal activation of some attractive junctions also takes place during low-temperature creep. The model of attractive junction formation on a stress decrement during creep, yields a value of 45 ± 10 ergs cm^{-2} for the stacking fault energy in copper.

1. Introduction

The use of copper, both in elemental and alloyed forms, in several engineering applications creates a need for comprehensive information on the deformation properties of this metal at cryogenic as well as high temperatures. Of particular interest is to explore the possibility of developing copper alloys capable of retaining their strength and creep resistance over a wide temperature range. To accomplish this objective a knowledge of the fundamental processes during deformation is necessary, and one often measures the temperature and strain-rate sensitivity of the flow stress, temperature and stress dependence of the creep rate and the like. From measurements of this type it is possible to draw some conclusions about the nature of the thermally activated dislocation processes involved in deformation, and the clues for strengthening will then follow as natural corollaries.

A preliminary investigation was undertaken to determine the tensile and creep behaviour of copper. The temperature and strain-rate dependence of flow stress were obtained for copper of various degrees of purity, thus enabling the determination of activation energy for deformation. Change-in-stress experiments were conducted to evaluate the activation area characterising the obstacle to dislocation motion which can be thermally assisted. In view of the limited information regarding the data on decremental-stress creep experiments on this fcc metal, particular emphasis was placed on understanding the dislocation processes when a small negative stress change was made during low temperature creep.

2. Experimental Procedure

Spectroscopically pure, electrolytic and commercial coppers were used in the current study. Their analyses are presented in table I.

Mechanical testing was carried out on sheet samples of approximately 2 mm thickness and 5 mm width. Gauge length was 4.45 cm for creep samples and 2.55 cm for tensile specimens. Specimens were cut from the same sheet to avoid small differences in grain size and all the specimens were annealed at 875°K for 3 h in a dynamic vacuum of 5×10^{-6} torr. The resulting average grain diameter was 0.02, 0.01 and 0.007 mm in samples of electrolytic, spectroscopic and commercial copper respectively. Each specimen was etched just before testing with a solution consisting of 10 parts $FeCl_3$, 10 parts HCl and 40 parts H_2O.

Tensile tests were conducted in a Hounsfield

TABLE I Analysis of Copper

Element	Concentration		
	Spectroscopic Cu ppm	Electrolytic Cu %	Commercial Cu %
Fe	3	0.009	0.08
Si	1	—	—
Pb	1	0.008	0.05
Ni	2	—	0.06
As	—	—	0.06
Sb	—	—	0.02
S	—	—	0.12
Na	2	—	—
O	100	0.020	0.10
Se	2	—	—
others	4	0.020	0.20

tensometer with facilities for various constant cross-head speeds and accurate load measuring and strain-sensing devices [1]. The dependence of flow stress on temperature in the range of 87 to 525°K was obtained at three strain rates for commercial and electrolytic copper. Spectroscopic copper was tested only at 525°K in an attempt to arrive at the athermal component of flow stress. The load-elongation curves were recorded conventionally up to about 30% strain or until fracture, whichever was earlier.

Creep testing was done in a constant stress tensile creep unit [1] at four selected temperatures on electrolyic copper and at 87°K on commercial and spectroscopic coppers. The transient creep strain was measured by means of a transducer capable of giving a strain sensitivity of $1{\cdot}0^{-5}$. The effect of repeated small changes in stress, both increments and decrements, on creep rate was recorded over a wide stress range in each creep test.

3. Results

The imposed shear strain rate during a tensile test or the shear creep rate, $\dot{\gamma}$, is given by:

$$\dot{\gamma} = P \exp\left(-G/kT\right) \tag{1}$$

where P is the pre-exponential frequency factor, G the thermal activation free energy, k the Boltzmann's constant and T the absolute temperature. The activation free energy, G, is a function of stress and is given by:

$$G = G_0 - \int_0^{\tau^*} A\, b\, d\tau^* \tag{2}$$

where G_0 is the total activation free energy; A, termed the "activation area", is the area swept out by the dislocation during the activation event and τ^*, given by the applied shear stress, τ, minus τ_μ the long-range internal stress is the stress required to by-pass the short-range obstacles to dislocation motion. Determination of the origin of the temperature and strain-rate dependent flow stress τ^*, in metallic structures is facilitated by a knowledge of G and A which can be measured experimentally since [2-4]:

$$G = -k\left[\frac{\delta \ln \dot{\gamma}}{\delta(1/T)}\right]_{\tau^*} \tag{3}$$

and

$$A = \frac{kT}{b}\left[\frac{\delta \ln \dot{\gamma}}{\delta \tau^*}\right]_T \simeq BkT/b \tag{4}$$

where the flow parameter B is given by: $B = (\Delta \ln \dot{\gamma})/\Delta\tau$. In deriving these, the change in entropy of activation is taken as zero and the pre-exponential factor of equation 1 is assumed to remain unchanged with temperature and stress. The data on tensile and creep experiments are utilised to evaluate the above two activation parameters in copper.

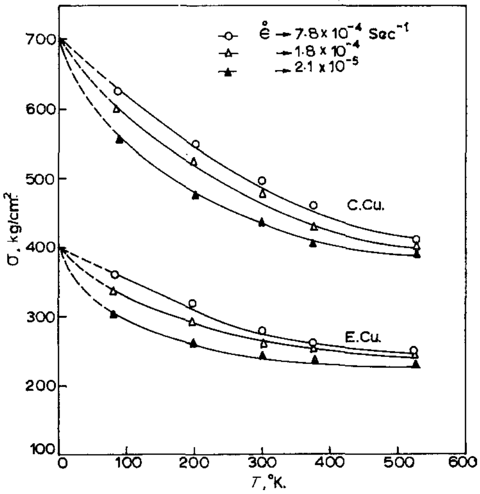

Figure 1 Effect of temperature and strain rate on the flow stress of electrolytic (E) and commercial copper (C).

The results of the tensile tests are summarised in fig. 1, in which is shown the variation of tensile flow stress, σ, with temperature and tensile strain rate, $\dot{\epsilon}$, for electrolytic and commercial copper. The flow stress values, measured at 0.1% strain, are accounted for by the temperature variation of shear modulus [5]. A thermally activated mechanism is seen to control the flow stress at low temperatures. The flow stress at 525° K, the temperature at which strain rate has negligible effect on the flow stress, was considered to be the athermal component of the flow stress, τ_μ. The value of τ_μ as a function of purity of copper is included in table II. Tensile stresses are converted into shear stresses by assuming that $\tau = \sigma/2$. From fig. 1, the value of τ_0/τ_μ, where τ_0 is the flow stress (extrapolated) at 0° K is found to be 1.8 for commercial copper and 1.6 for electrolytic copper.

TABLE II Athermal stress in Copper

Material	Experimental τ_μ kg/cm^2	$\tau_\mu . A_0$ $X\,10^{12}$, kg
Spectroscopic Cu	105	—
Electrolytic Cu	125	117
Commercial Cu	200	110

In view of equation 3 and the data of fig. 1, the thermal activation free energy for plastic flow can be calculated as a function of stress from the slope of $\ln \dot{\epsilon}$ versus $1/T$ plots (since $\delta \ln \dot{\gamma} = \delta \ln \dot{\epsilon}$). In fig. 2 is shown the variation of G with effective stress for commercial and electrolytic coppers. The total activation energy, G_0, required for a glide dislocation to cut through the strain-rate controlling obstacle is found, by extrapolation of the $G - \tau^*$ curve to $\tau^* = 0$, to be $\approx$ 0.6 eV for both the commercial and the electrolytic copper.

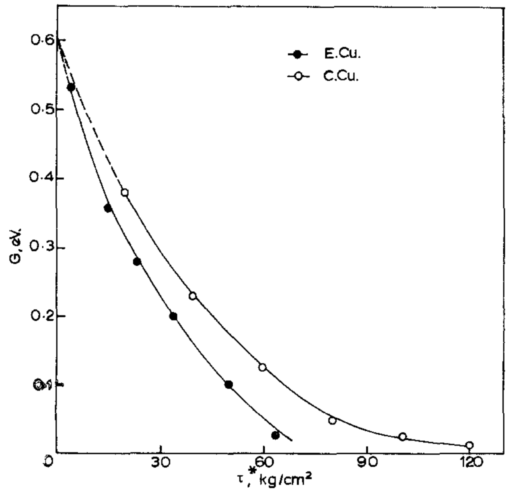

Figure 2 Effect of stress on the thermal activation energy for deformation of copper (E- Electrolytic, C–Commercial).

The data from change-in-stress creep experiments give an estimate of the activation area for the thermally activated deformation process. The typical nature of the creep curves during differential-stress experiments on electrolytic copper at (a) 87°, (b) 197°, (c) 273° and (d) 300° K is shown in fig. 3. The plots clearly show that logarithmic creep is observed at all temperatures over a wide stress range. The above plots are graphically differentiated to arrive at the magnitude of the change in creep rate following stress increments and decrements. The creep rate is plotted in fig. 4 as a function of creep strain, ϵ, and it can be seen from this figure that the values of the change in creep rate on logarithmic scale for decremental stress changes, $[\Delta \log \dot{\epsilon}]_D$, are higher than the corresponding values, $(\Delta \log \dot{\epsilon}]_I$, for an equal incremental stress change.

Fig. 5 presents the effect of stress and the magnitude of the stress change on the flow parameter B during the creep of electrolytic copper at 273° K. These data show that the values of B corresponding to stress increments and their stress dependence are both independent of the magnitude of the stress increment. On the other hand, the values of B corresponding to stress decrements are higher than those obtained for increments and vary with the magnitude of decremental stress, the difference β (fig. 5) being dependent on the applied stress. The higher the stress decrement, the larger is the difference in the B values. Generally this difference, β, between the B values decreases with increasing stress level and becomes negligible beyond a certain stress depending on the magnitude of decremental stress and temperature. Similar results are also observed during creep of spectroscopic and commercial coppers at 87° K.

An examination of the effect of temperature revealed that, β, in electrolytic copper at a given stress, is smaller at higher temperatures and higher at lower temperatures [6]. At room temperature the difference is negligible and stress increments and decrements yield the same flow parameter at a given applied stress.

4. Discussion

The deformation behaviour of copper can best be analysed by examining separately the factors contributing to the thermally activated and athermal components of the flow stress. This will now be done and the effects observed during differential-stress creep experiments will be discussed.

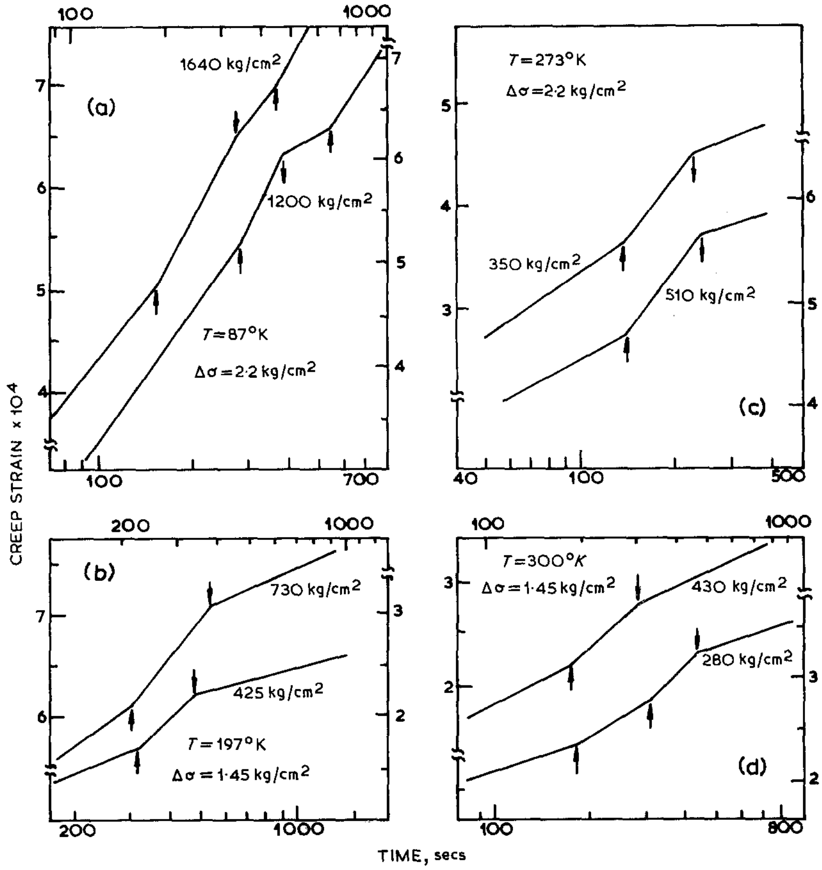

Figure 3 Creep strain in electrolytic copper as a function of time at different temperatures.

4.1. The Origin of τ^*

In fcc metals, the mechanism of thermally activated deformation may be one of the following [2]:

(i) Intersection of forest dislocations,
(ii) Non-conservative motion of jogs, and
(iii) Suzuki locking.

A knowledge of G_0 and A will be extremely useful in deciding the rate-controlling dislocation process. The activation area can be evaluated from the flow parameter values. It will be shown later in discussion that the B values obtained by incremental stress changes during creep are more reliable. Hence utilising the B values for stress increments in creep tests conducted at 87° K, the activation areas evaluated from equation 4 are shown in fig. 6 for copper of the three selected purities. The values obtained from fig. 2 $[A = -(1/b)(\delta G/\delta\tau^*)]$ are in fair agreement with those from creep data at low stresses.

Mechanisms such as dislocation pinning by solute atoms and Peierls-barrier overcoming can be discarded since the activation area is not as strongly dependent on purity (fig. 6) as is required for the former and it never fell below 100 b^2 as is the characteristic of the latter. The inapplicability of Suzuki locking can be easily demonstrated by considering the value of τ_0/τ_μ, a measure of the temperature-sensitivity of the flow stress. If solute atoms segregate to the stacking faults and widen them, only a weaker dependence of flow stress on temperature will be exhibited. However, commercial copper has a stronger temperature dependence of flow stress; the value of τ_0/τ_μ is 1.8 as against 1.6 for electrolytic copper. Moreover, Suzuki locking will require higher energies for constriction of split dislocations and their continued glide. One may then expect a higher value for the total activation energy G_0 in commercial copper. But the total activation energies determined experimentally in the present work are almost the same

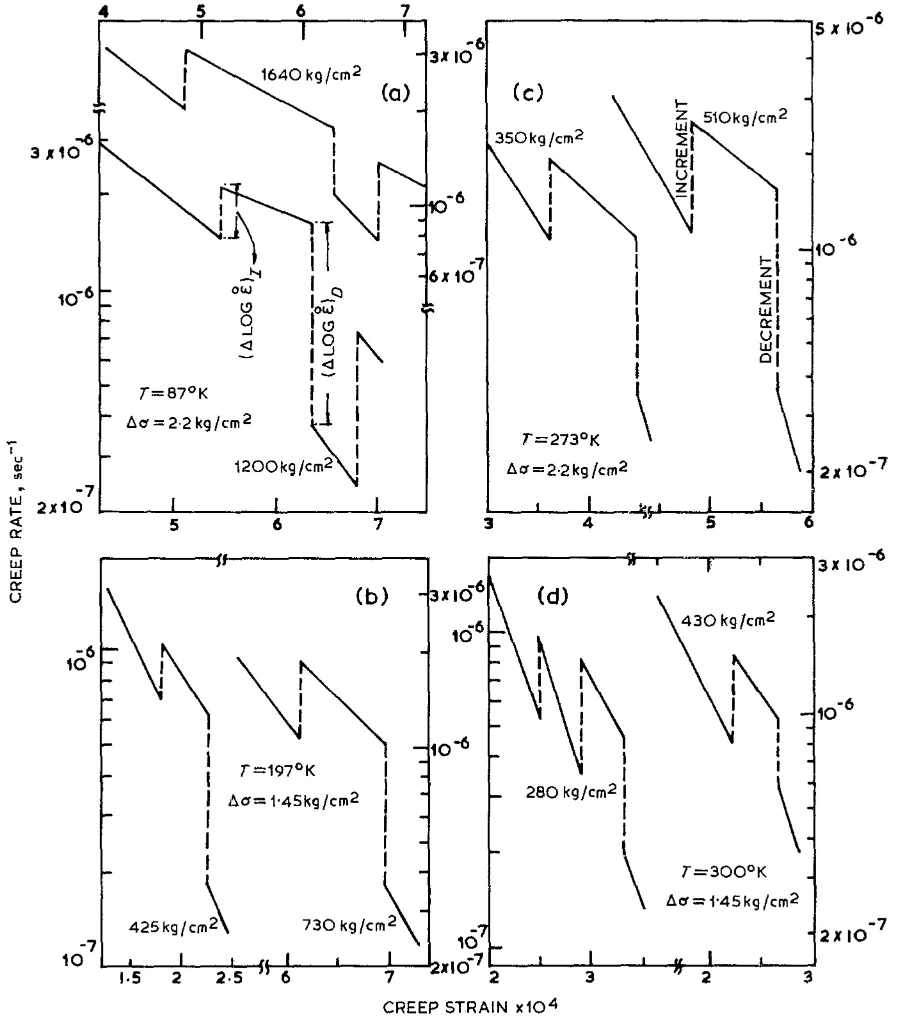

Figure 4 Effect of stress increments and decrements on the creep rate of electrolytic copper.

namely, 0.6 eV (fig. 2) for commercial and electrolytic copper.

The mechanism of non-conservative motion of jogs is not normally accepted for the thermally activated plastic flow of pure metals at low temperatures, as argued by Mott [7]. However, it may be important in somewhat impure metals where the point defect (vacancy) that is formed at the jog moving non-conservatively gets bound to the solute atom present near the dislocation. This decreases the probability of the dislocation jumping backward and a net forward movement is possible, unlike in a pure metal. This may also result in a higher temperature dependence in commercial metal which is observed in the present investigation. However, the activation energy for such a mechanism will be the formation energy of vacancies which is generally expected to vary with the impurity content of the metal, according to the Lomer equation [8]. In the present investigation with commercial and electrolytic coppers, the same value (0.6 eV) was however obtained for G_0, in spite of the wide composition difference between them. This leads to a lack of support for the mechanism based on non-conservative motion of jogs.

All the experimental data can however be accounted for on the basis of the intersection model. The activation areas in copper fall in the range 1200 to 100 b^2, as is to be expected for the

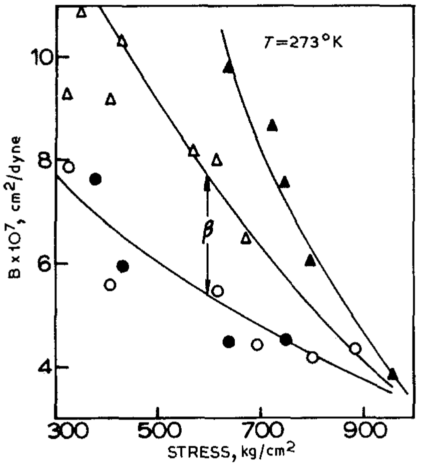

Figure 5 The variation of the flow parameter B with stress in electrolytic copper at 273° K.

Incr.		Decr.
○	$\Delta\sigma = 2.2$ kg/cm^2	△
●	$\Delta\sigma = 3.1$ kg/cm^2	▲

intersection mechanism [2]. The activation area can be written as the product of the average spacing, L, of the forest dislocations and the activation distance, D. It may be concluded from the following considerations that the activation distances are almost the same in electrolytic and commercial copper: (i) If the activation distance in commercial copper is higher (may be due to Suzuki locking) than in electrolytic copper, then the activation areas in commercial copper will be higher. Also a lower dependence of flow stress on temperature will be exhibited. Both are contrary to the experimental observations. (ii) The total activation energy, G_0, for intersection mechanism in fcc metals is twice the sum of jog formation energy and extended-dislocation constriction energy. The constant value of 0.6 eV obtained for G_0 indicates almost the same constriction energies and hence the same activation distances in electrolytic and commercial copper. The slightly higher activation areas in electrolytic copper and spec-pure copper compared with those in commercial copper are then a result of the possible presence of a higher dislocation density (and hence lower L) in the latter. There is only a minor difference in the impurity contents of electrolytic and spectroscopic copper. Hence the activation areas in spectroscopic copper are not significantly higher than in electrolytic copper.

An examination of the effect of impurities on

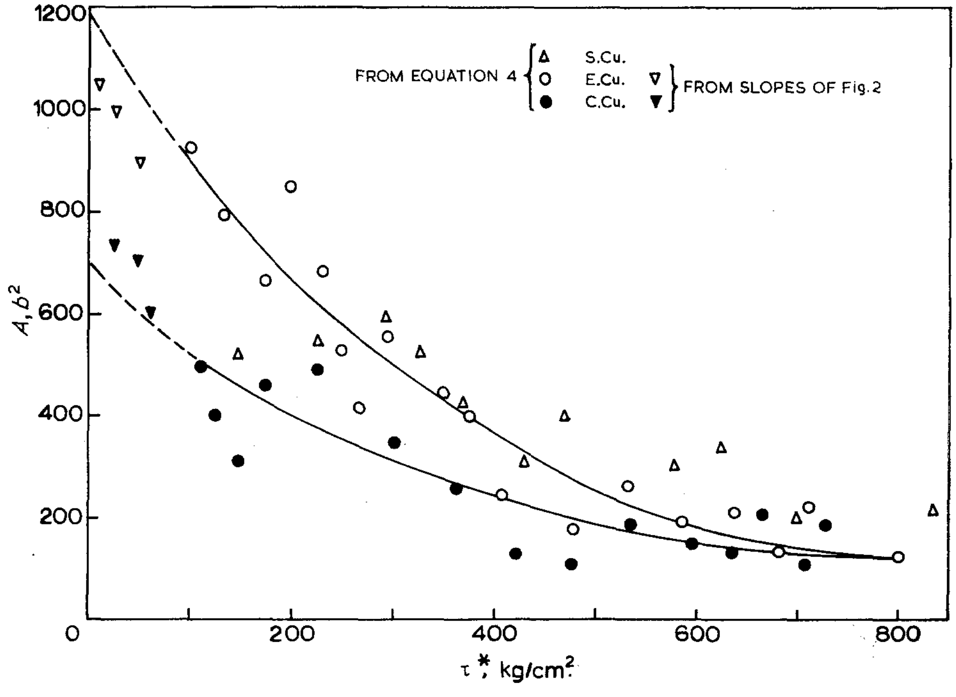

Figure 6 Dependency of activation area in copper on effective stress and purity (S – spectroscopically pure; E – Electrolytic; and C – Commercial).

the $\sigma - T$ curve (fig. 1) also points to the conclusion that forest dislocations are the rate-controlling barriers in copper. The slope of the $\sigma - T$ curve at temperatures lower than say 400° K, is higher for commercial copper. This, in terms of the theory of thermally activated glide, means that the density of the short-range obstacles is increased in commercial copper, a conclusion arrived at from considerations of the activation areas too. Thus an increase in the value of τ_0/τ_μ does not necessarily reflect a change in the low-temperature deformation mechanism. It is also to be noted that the value of G_0 (0.6 eV) obtained in the current work compares well with 0.67 eV obtained by Evans and Flanagan [9] for copper and Cu-Si solid solutions where intersection mechanism was found to be rate-controlling, and with 0.7 eV estimated by Thornton and Hirsch [10]. From these considerations it appears that the same intersection mechanism controls the low-temperature deformation behaviour in copper of different purities.

4.2. The Origin of τ_μ

It is seen from table II that the temperature-independent part of the flow stress increases with the amount of impurity elements present in copper. The athermal stress arising due to long-range stress field can be affected by one or more of the following interactions involving the solute atoms and dislocations: (i) a change in dislocation density, (ii) Suzuki interaction (chemical interaction), (iii) short-range ordering and (iv) elastic and modulus interactions. The following analysis will show that the increased athermal stress with impurity content in copper can almost be completely accounted for on the basis of increased dislocation density. The long range stress field, τ_μ, is related to the dislocation spacing in fcc metals as follows [10, 11]:

$$\tau_\mu = \alpha b \, \mu / L$$

where α is a constant which depends on the arrangement of dislocations and their character and μ is the shear modulus. The above equation can be rewritten [10] in the following form:

$$\tau_\mu = \alpha \, \mu \, b \, D / A_0 \tag{5}$$

where A_0 is the extrapolated activation area at zero effective stress. The activation distance D for intersection mechanism is twice the stacking-fault width and can be taken as 10 Burgers vectors after Seeger [11].

Ignoring minor variations in μ with purity, it can be seen from equation 5 that the product $\tau_\mu A_0$ should be constant. Using the value of A_0 from fig. 6, the quantity $\tau_\mu A_0$ is calculated for electrolytic copper and commercial copper. The values indicated in table II are, as expected, almost the same. The A_0 values for electrolytic and spectroscopic copper appear to be very close (fig. 6). This is reflected in the observed τ_μ values (table II) for these two purities, which do not differ much. Taking $\mu = 4.5 \times 10^{11}$ dynes/cm^2 and $D = 10b$, the value of the constant α is calculated to be 0.03 which is of the right order of magnitude [12].

4.3. Low-Temperature Creep Behaviour

The available literature [13-15] indicates that small incremental and decremental stress changes during low temperature creep of cph metals result in the same value of flow parameter, B, unlike the observation made on copper in this work. The fact that the B values for copper obtained from stress decrements are dependent on the working stress and the magnitude of stress decrement (fig. 5) implies that whatever be the process responsible for such a discrepancy, it is stress dependent. On the other hand, different magnitudes of stress increments resulted in the same B values: the dislocation processes involved during incremental creep are thus stress insensitive. This phenomenon was investigated in greater detail on several fcc metals and the results were reported in an earlier publication [6]. Briefly, the difference in the B values is thought to arise from a change in the number of elements participating in the activation event when a stress decrement is made as against the absence of any such change subsequent to a small stress increase during creep. The absence of this effect in cph metals has led to the postulation that the strong tendency for formation of attractive junctions on a stress decrement in fcc metals might cause a change in the number of thermally activatable dislocation segments [16-18]. The experimental observations on the effect of stress and temperature on β showed satisfactory coincidence with the predictions based on an attractive junction model. The suggested model permitted an evaluation of the stacking-fault energy from a knowledge of the change in the frequency factor and the flow stress ratio τ_0/τ_μ. Essentially the calculation involves the computation of the width of the barrier to the glide dislocations from the magnitude of the change in frequency factors for at least two stress decrements at a major stress level during creep. The width of the barrier, D, in the direction of the Burgers vector is approximated to the stacking-fault width. The calculated fault width in copper is 19Å or about 7 Burgers vectors. This compares well with the value of $12b$ for an edge dislocation and $5b$ for a screw dislocation in copper quoted by Seeger [11]. Taking this as the equilibrium width, the stacking fault energy (*SFE*) is calculated from the equation:

$$SFE = \mu a^2/24\pi D \tag{6}$$

where "a" is the lattice parameter of copper. This yields, taking μ for copper as 4.5×10^{11} dynes/cm^2, a value of 45 ± 10 ergs/cm^2 for the *SFE* of copper which is in good agreement with that reported in the literature [19, 20] as obtained from other methods. The value of 35 ± 10 ergs/cm^2 for the *SFE* of copper presented earlier [6] was based on the use of 1.2 for τ_0/τ_μ quoted by Thornton and Hirsch [10].

5. Conclusions

(i) The activation areas in copper fall in the range 1200 to $100b^2$, decreasing with stress.

(ii) The total activation energy for plastic flow of copper is 0.6 eV and is almost independent of purity.

(iii) The intersection of glide and forest dislocations controls the strain rate at low temperatures, and purity appears to have little effect on the rate-controlling barriers in this metal.

(iv) The variation of the temperature-independent part of the flow stress with purity in copper can be understood in terms of a change in the dislocation density.

(v) The stacking-fault energy of copper calculated on the basis of the model of attractive-junction formation during decremental creep is 45 ± 10 ergs/cm^2.

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