Some Observations of Grain-Boundary Sliding in Aluminium Bicrystals Tested at Constant Strain Rate and Constant Rate of Stress Increase

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Bicrystals of pure aluminium, aluminium/0.05 wt% and aluminium/0.30 wt% copper have been deformed in shear within the temperature range 350 to 600°C at a constant rate of grip displacement (300 μm/h) and constant rates of increase of grain-boundary shear stress (0.30 to 1.04 g/mm^{-2} min^{-1}). The stress/time, sliding/time and sliding/stress curves for these tests are presented together with metallographic observations. The stress/time curves exhibited changes in the strain-hardening rates which were accompanied by the occurrence of extensive crystal deformation. In many cases, following the change in strain-hardening rate and onset of extensive crystal slip, the extent of grain-boundary sliding was proportional to the shear stress on the boundary. The ratio of the extent of sliding to the grain-boundary shear stress increased with increasing test temperature. The temperature-dependence of the sliding behaviour, as reflected by the sliding/stress results, yielded apparent activation energies of $\sim$ 31 Kcal/mole in the temperature range 600 to 475°C and $\sim$ 8 Kcal/mole in the range 475 to 350°C.

1. Introduction

The relative displacement of grains along their common boundary can make a significant contribution to the creep-deformation of metals and alloys at temperatures greater than $\sim$ 0.5 T_m [1, 2]. It is widely accepted that grain-boundary sliding has an important influence on creep-failure through its association with the initiation and growth of regions of intergranular decohesion [3]. There is also evidence that grain-boundary sliding may play an effective part in the phenomena of superplasticity [4, 5] and dynamic recrystallisation [6] where strain rates can be much greater than those normally observed in "creep".

Constant-stress tests on bicrystals have provided information about the stress and temperature-dependence of sliding and its relationship to crystal creep-deformation, and have also provided valuable leads as to the mechanism of grain-boundary sliding [1, 2]. We can, however, extend our understanding of deformation at

elevated temperatures if tests are carried out in which the stress level is progressively raised during an individual test [7-10]. In this way the broader response to stress level of the sliding and crystal deformation may be established. It is also important when applying basic sliding relationships to be aware of the test conditions involved. For example, the overall grain-boundary sliding, time, stress and crystal-deformation relationships which are observed for constant load tests may change significantly under conditions of constant strain-rate deformation. It was with these points in mind that the present series of tests were carried out in which bicrystals were deformed at constant rates of grip displacement or constant rates of increase of grain-boundary shear stress.

2. Experimental
2.1. Bicrystal Growth
Industrial super pure grade aluminium was used for the production of bicrystals. Spectrographic
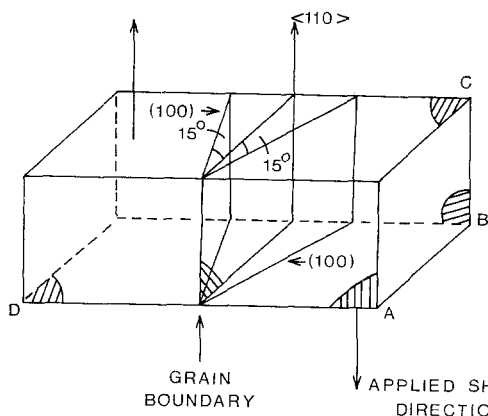
Figure 1 The bicrystal specimen nomenclature and relative crystal orientation.

analysis of the material revealed the presence of the following impurities (in ppm):—$Cu < 50$, $Fe < 100$, $Ni < 50$, $Zn < 100$, $Si\ 100$, $Ti\ 50$. Copper containing < 100 ppm impurities was used to make up aluminium/copper alloys with copper contents of 0.1 and 0.5 wt %. Bicrystals of controlled-orientation, containing macroscopically plane grain-boundaries were obtained from the melt using a modified Bridgman technique with suitably oriented seed crystals. The bicrystal orientation, to within an accuracy of $\pm 2°$, is shown in fig. 1. This orientation involves two sets of {1 1 1} slip planes in each grain being rotated symmetrically about a $\langle 1 1 0 \rangle$ slip vector which is both in the plane of the grain-boundary and parallel to the applied-shear direction.

The bicrystal-growth procedure led to a certain amount of zone-refining, and chemical analysis revealed that in the centre portions of each bicrystal the nominally 0.1 and 0.5 wt % copper compositions were 0.04 ± 0.02 wt % and 0.23 ± 0.14 wt % copper respectively.

2.2. The Test Specimens

Specimens for testing were obtained from the centre sections of the as-grown bicrystals by spark-planing and spark-cutting using a Servomet machine on setting 4. This was followed by chemical polishing in a solution of 64 vol % ortho-phosphoric acid, 18 vol % sulphuric acid and 18 vol % nitric acid maintained at a temperature of 85 to 95°C in order to obtain suitable specimen surfaces for electro-polishing. A good mirror finish was obtained by using an electro-polishing solution of 5 parts by volume of ethanol and 1 part of perchloric acid (S.G. 1.48) at 0 to 10°C with an aluminium cathode and a

potential of 10 V. The electro-polishing also removed most of the damage caused by spark-machining since a layer of ~ 0.5 mm was removed from each surface, leaving final specimen dimensions of $\sim 20 \times 5.5 \times 4.5$ mm.

2.3. Testing Techniques

Bicrystal specimens were tested in shear parallel to the grain-boundary and in a direction along the common $\langle 1 1 0 \rangle$ slip direction (fig. 1). The apparatus allowed testing under conditions of constant rate of grip displacement, which approximates to constant strain rate, and constant rate of increase of shear stress. The latter mode was obtained by using a water flow from a constant head to increase the load on the specimen. The specimens were gripped on surfaces C and D (fig. 1), one grip on each side of the grain-boundary. The free space between the grips, which contained the grain-boundary, was ~ 2.5 mm wide. This gripping arrangement resulted in the {1 1 1} slip planes in each crystal being constrained by the grain-boundary, the grips, or both. Temperature control was within $\pm 2°C$ and an atmosphere of pure argon at atmospheric pressure was maintained in the test chamber. Fiducial marks, normal to the grain-boundary, were made on each surface of the specimen before testing by wiping with a paper tissue. Grain-boundary sliding on surface A (fig. 1) was measured during test from the fiducial mark offsets by means of an optical system employing a filar micrometer eyepiece, giving an accuracy of ± 2 μm. Measurements were continued until grain-boundary migration or crystal deformation or a combination of both did not allow the sliding displacement to be measured to the above accuracy. During constant strain-rate deformation the load was continuously monitored on a strip-chart recorder.

3. Results
3.1. Constant Strain-Rate Deformation

Bicrystal specimens of pure aluminium were tested at temperatures from 350°C ($\sim 0.67\ T_m$) to 600°C ($\sim 0.94\ T_m$) and specimens of aluminium/0.05 wt % copper and aluminium/0.30 wt % copper in the temperature range 475 to 575°C at a strain-rate of 300 μm/h. The grain-boundary shear stress, τ/time and the grain-boundary sliding (ϵ_{gb})/time curves are presented in fig. 2 for tests on pure aluminium at 350, 375, 450 and 500°C. The stress/time curves exhibit a diffuse transition in strain-hardening rate, for
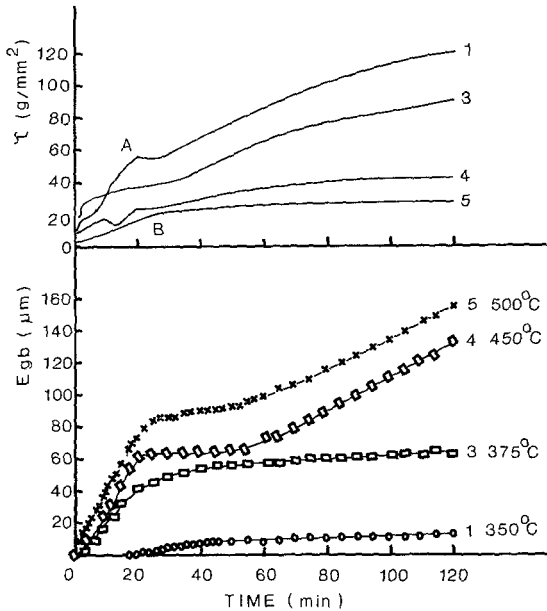
Figure 2 Composite grain-boundary shear stress/time and grain-boundary sliding/time curves for pure aluminium specimens tested at 350, 375, 450 and 500°C. Constant strain-rate = 300 μm/h.

example at points A and B in the tests at 350 and 500°C. This transition was in general accompanied by the formation of coarse slip traces on the specimen surface.* At the same stage in the tests as the change in strain-hardening rate and onset of macroscopic slip, there was a concomitant change in grain-boundary sliding behaviour. For temperatures of 500, 450 and 375°C in fig. 2 sliding almost stopped, but at a later stage in the tests at 500 and 450°C, the sliding rate increased again. On the other hand, in the test at 350°C there was little sliding prior to the change in the strain-hardening rate and the onset of macroscopic slip. These results show that changes in both the character of crystal deformation and grain-boundary sliding can occur in individual tests and it is not surprising therefore that no overall relationship in the form of a direct proportionality between ϵ_{gb} and total deformation was observed.

The influence of the grain-boundary shear stress on the sliding behaviour is demonstrated in fig. 3 for aluminium specimens tested over the temperature range 350 to 600°C and includes the
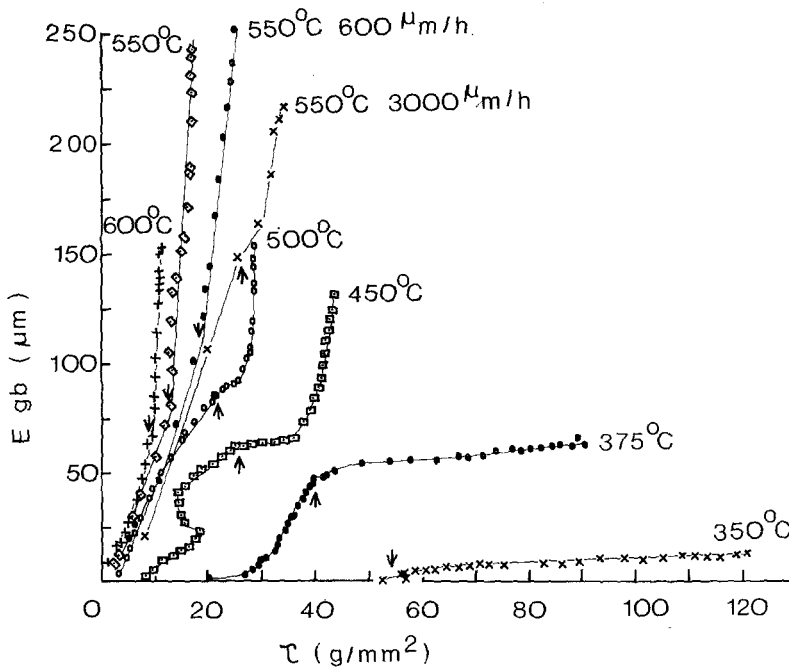

Figure 3 Curves of grain-boundary sliding/grain-boundary shear stress for pure aluminium specimens. Constant strain-rate = 300 μm/h except where otherwise stated. The arrows indicate the general stress-levels when extensive crystal-slip commenced.

*Coarse is here defined as being observable with the optical system used for measuring the grain-boundary sliding displacement. This gave a magnification of × 85.
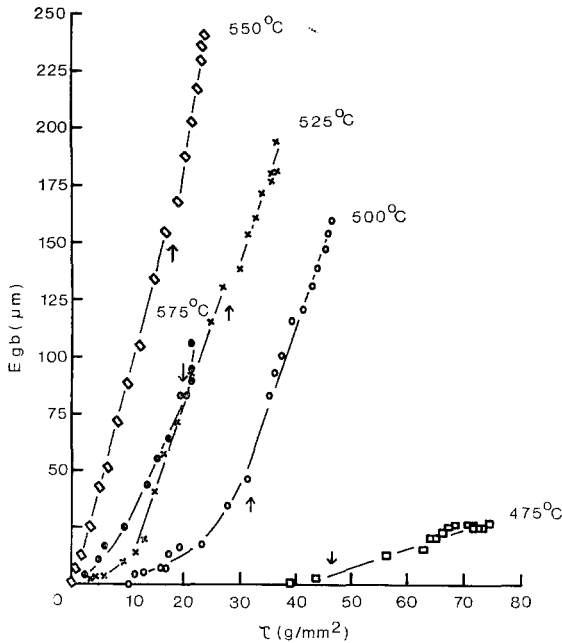
Figure 4 Curves of grain-boundary sliding/grain-boundary shear-stress for aluminium/0.30 wt % copper specimens. Constant strain-rate = 300 μm/h. The arrows indicate the general stress-levels when extensive crystal-slip commenced.

specimens in fig. 2. Two of the tests at 550°C were carried out at strain-rates of 660 and 3000 μm/h. The arrows indicate the general stress levels at which macroscopic crystal deformation commenced, as indicated by the stress/time curves. In the tests at 600, 550 and 550°C (600 μm/h), both before and after the onset of macroscopic slip, there was a direct proportionality between grain-boundary sliding extent and grain-boundary shear-stress, that is, $d\epsilon_{gb}/d\tau$ was constant in both regions. For the tests at 550°C (3000 μm/h) and lower temperatures, $d\epsilon_{gb}/d\tau$ decreased at the onset of macroscopic slip and, in general, subsequently attained a higher value. This may be thought of as an inflexion in the ϵ_{gb}/τ curve. The stress-range over which this inflexion extended increased with decreasing temperature, and also with increasing strain-rate for the three tests at 550°C. The anomalous behaviour of the test at 450°C can be related to a drop in the flow-stress which occurred before the onset of macroscopic slip (fig. 2). There was an overall reduction in $d\epsilon_{gb}/d\tau$ with decreasing temperature and also, for the limited results, with increasing applied strain-rate. The intercepts on the abscissa indicate the higher stress levels which

were required before sliding was detected as temperature was reduced.

Further examples of the approximate linearity between ϵ_{gb} and τ and the change in $d\epsilon_{gb}/d\tau$ which occurred at the transition in strain-hardening rate and onset of macroscopic slip are presented in fig. 4 for aluminium/0.3 wt % copper bicrystals.

3.2. Constant Rate-of-Stress-Increase Deformation

Bicrystals of pure aluminium, aluminium/0.05 wt % copper and aluminium/0.3 wt % copper were tested at varying rates of increase of grain-boundary shear-stress within the temperature range 475 to 575°C. In fig. 5 the sliding/time data for specimens of pure aluminium are presented and the rates of stress increase are given in table I. The arrows in fig. 5 indicate the time at which stress increase was stopped, so that subsequent sliding took place under conditions of constant stress. Results from tests on aluminium/0.05 wt % copper bicrystals are presented in fig. 6. The rates of stress increase are also given in table I. The sliding/time curves in figs. 5 and 6 also represent the sliding/stress behaviour since stress was directly proportional to time. These results show that the major part of the sliding took place with a direct proportionality between
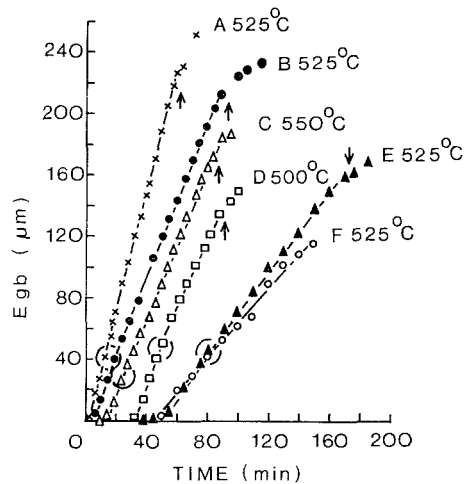

Figure 5 Sliding/time curves for specimens of pure aluminium tested at constant rates of stress-increase as given in table I. The large circles indicate the approximate times when extensive crystal-slip occurred. The arrows indicate the point in test when the stress increase was halted. The origins on the abscissa have been adjusted for clarity and are as follows: A O, B O, C 10, D 20, E 30, F 40.
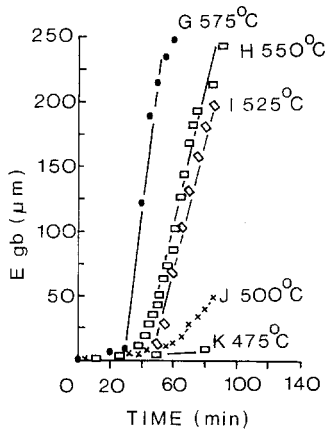
Figure 6 Sliding/time curves for tests at constant rates of stress-increase on aluminium/0.05 wt % copper specimens. The rates of stress increase are given in table I.

TABLE I Testing conditions for the specimens of fig. 5 (A to F) and fig. 6 (G to K)

Specimen	Test temperature (°C)	Constant rate of grain-boundary shear-stress increase (gm/mm^{-2} min^{-1})
A	525	0.72
B	525	0.52
C	550	0.56
D	500	0.58
E	525	0.30
F	525	0.35
G	575	0.65
H	550	0.50
I	525	0.58
J	500	0.55
K	475	0.60

ϵ_{gb} and τ. Whilst the onset of macroscopic slip resulted in a marked increase in sliding-rate in fig. 6, a similar effect is not apparent in fig. 5 and there is some indication that the sliding rate may have decreased.

3.3 Effects of Temperature and Alloy Content on Sliding

A common link between the two testing modes was the linear behaviour of the sliding/stress curves in the presence of macroscopic slip. If the slope of these curves $d\epsilon_{gb}/d\tau$, is denoted by A then $1/A$ may in accordance with previous work [10] be considered a coefficient of slide-hardening. Values of A for the two types of test are presented in table II. The present results do not allow a definitive interpretation of the effects of copper content on A, but no major effect was apparent. However, the results in table II and figs. 5 and 6 show that in all cases A increased with test temperature. If the temperature-dependence of A is represented by an expression of the form $A = B \exp(- Q/RT)$, where B is a constant, then values of Q may be obtained using the data in table II. The aluminium/copper results in table II gave a value for Q of ~ 31 Kcal/mole over the temperature range 475 to 575°C. The pure aluminium results are consistent with this value for the temperature range 475 to 600°C, but below 475°C the temperature dependence of A was much smaller and a value of ~ 8 Kcal/mole was obtained for Q.

3.4. Metallographic Observations

Although specimens were observed continuously during test, a more detailed metallographic

TABLE II Values of A, the gradients of the sliding/stress curves, in the presence of extensive crystal deformation

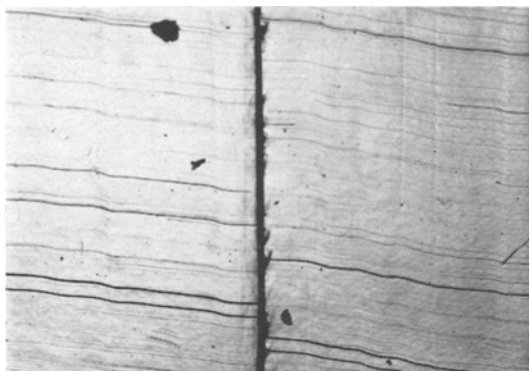| Test temperature (°C) | Pure aluminium | | | 0.05 wt % copper | | 0.3 wt % copper | | T/T_M (°K) |
| | Series 1 | Series 2 | | | | | | |
| | $\dfrac{d\epsilon_T}{dt}$ constant | $\dfrac{d\epsilon_T}{dt}$ constant | $\dfrac{d\tau}{dt}$ constant | $\dfrac{d\epsilon_T}{dt}$ constant | $\dfrac{d\tau}{dt}$ constant | $\dfrac{d\epsilon_T}{dt}$ constant | $\dfrac{d\tau}{dt}$ constant | |
	A (c.g.s. units)	A	A	A	A	A	A	
350	—	1.2×10^{-6}	—	—	—	—	—	~ 0.67
375	—	1.6×10^{-6}	—	—	—	—	—	~ 0.69
400	—	2.4×10^{-6}	—	—	—	—	—	~ 0.72
450	5×10^{-6}	2.9×10^{-6}	—	—	—	—	—	~ 0.78
475	—	4×10^{-6}	—	—	—	2.2×10^{-6}	2.2×10^{-6}	~ 0.80
500	—	8.8×10^{-6}	3.6×10^{-6}	4.2×10^{-6}	3.2×10^{-6}	6×10^{-6}	4.7×10^{-6}	~ 0.83
525	—	—	4.9×10^{-6}	—	1.1×10^{-5}	8.1×10^{-6}	4.7×10^{-6}	~ 0.85
550	3.25×10^{-5}	2.1×10^{-5}	5.1×10^{-6}	—	1.4×10^{-5}	2×10^{-5}	—	~ 0.88
575	—	—	—	—	2.9×10^{-5}	—	6.6×10^{-6}	~ 0.91
600	6.5×10^{-5}	—	—	—	—	—	—	~ 0.94

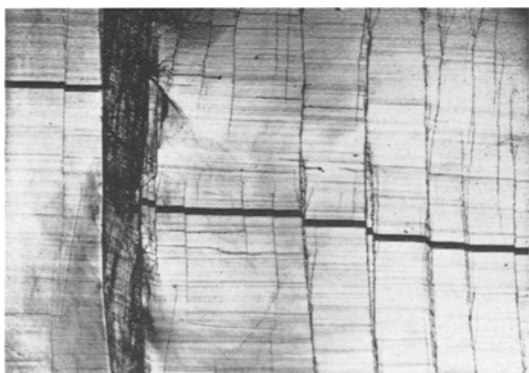Figure 7 Region of the grain-boundary on surface A of a pure aluminium specimen tested at 300 μm/h and 375°C (× 65).

Figure 8 Region of the grain-boundary on surface A of a pure aluminium specimen tested at 600 μm/h at 550°C (× 50).

examination was performed after test. From these latter examinations it was observed that the nature of the crystal slip traces changed with test temperature. At the lower temperatures broad slip bands were observed (fig. 7), but at 500°C it was difficult to detect any slip traces. Above 500°C extensive slip took place on narrow slip bands and cross-slip was particularly evident (fig. 8). Weinberg [11] observed a change in grain-boundary sliding characteristics of aluminium tricrystals at ~ 425°C and suggested that this was associated with the operation of a secondary slip system, possibly of the {100} type. In the present work there was no evidence of slip taking place on planes other than the {111} type.

4. Discussion

The general form of the stress/time curves for the constant strain-rate tests was similar to that reported by Bush et al [9], with the exception that a sharp "yield" was not observed at the onset of extensive crystal slip (fig. 2). It should be noted, however, that in the tests carried out by Bush et al the strain rates (295 to 29 500 μm/h) were, in almost all cases, in excess of those in the present tests. During deformation prior to the occurrence of "yield", Bush et al reported linear sliding/time and stress/time behaviour, the sliding rate approximating to the applied rate of straining. The limited observations in fig. 3 for the initial parts of the tests at 550 to 600°C are consistent with the proportionality between sliding and stress implied above. Neither the present results, nor those of Bush et al, support the conclusions of Walter and Cline [8] who found that the initial sliding rate, $d\epsilon_{gb}/dt$, was proportional to grain-boundary shear stress.

The bicrystal orientations and testing geometry used by Bush et al permitted unconstrained crystal slip, and as a consequence, the extent of strain-hardening following "yield" was relatively small. This, however, was not the case for the present experiments, nor for previous work of Halliday et al on zinc bicrystals [10] where sliding took place in the presence of extensive crystal deformation combined with significant strain-hardening due to the constraining effects of grain-boundary and grips. In the experiments of Halliday et al, the shear stress on the grain boundary was found, as in figs. 3 and 4, to be approximately proportional to the extent of grain-boundary sliding. It is interesting to note that in constant-stress tests performed by Gifkins et al [12] on polycrystalline aluminium and other metals following extensive secondary creep deformation, the extents of sliding on individual grain-boundaries were directly proportional to the components of the applied stress resolved along those boundaries. This relationship between sliding and stress is further confirmed by the present results obtained from constant-rate-of-stress increase tests (figs. 5 and 6). From the data for the tests at 525°C in fig. 5 we find that the rate of sliding is proportional to the rate of stress increase, $d\tau/dt$ (fig. 9). Whilst the proportionality between ϵ_{gb} and τ has been demonstrated over wide ranges of stress and temperature, the relationship between $d\tau/dt$ and $d\epsilon_{gb}/dt$ is drawn from a limited range of test variables.

The apparent activation energy for grain-boundary sliding at temperatures of 475°C and above was approximately 31 Kcal/mole for both pure aluminium and aluminium containing up to
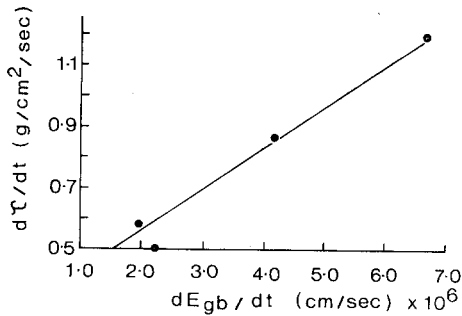
Figure 9 The rate of sliding in the presence of extensive crystal slip versus the rate of stress increase for the tests at 525° C shown in fig. 5.

0.30 wt % copper. This value compares favourably with those of Bush *et al* [9] which varied between 29.9 and 38.6 Kcal/mole for high-angle aluminium bicrystals, and is close to those for self-diffusion (33 Kcal/mole) and high-temperature creep (33 to 36 Kcal/mole) in aluminium [13]. In the lower temperature range however (< 450°C) a much smaller activation energy was observed (~ 8 Kcal/mole). Similar values of the activation energy for sliding in this temperature range have been reported [14, 15]. The implications of these low activation energies are not clear. Such relatively low values are observed for creep, but at extremely low temperatures and are associated with dislocation-glide processes.

The sliding/time curves for short-term constant load tests can show continuous decreases in $d\epsilon_{gb}/dt$ and this has been associated with "slide-hardening" [16, 17]. The sliding/time curves in fig. 5 show this decrease in $d\epsilon_{gb}/dt$ when the stress increase was stopped and the specimens were effectively subjected to constant stress conditions. Whilst the activation energies reported for grain-boundary sliding in constant-stress tests are similar to those obtained from the present results, there is no simple correlation between the stress-dependence for sliding in the different types of test. It must be assumed that some form of slide-hardening accompanied bicrystal deformation in the present series of tests. The nature and extent of the slide-hardening which took place when the stress levels were progressively increased and the corresponding changes in the deformation characteristics of the bicrystals may be different from the constant-stress case. It may well be that such differences in slide-hardening behaviour do not allow the sliding-rates in the present results to be related to the applied stress by the simple power law

commonly obeyed in constant-load tests.

5. Conclusions

In tests on aluminium and aluminium/copper bicrystals at a constant rate of grip-displacement and constant rates of increase of grain-boundary shear-stress, the extent of grain-boundary sliding in the presence of extensive crystal deformation was in many cases proportional to the shear-stress on the grain-boundary. The ratio of grain-boundary sliding to shear-stress increased with increasing test temperature and this temperature-dependence of the sliding behaviour was reflected in apparent activation energies of ~ 31 Kcal/mole (600 to 475°C) and ~ 8 Kcal/mole (475 to 350°C).

Acknowledgements

References

1. R. N. STEVENS, *Met. Rev.* **11** (1966) 129.
2. R. L. BELL and T. G. LANGDON, Interfaces Conference, Melbourne (Butterworths, 1969), p. 115.
3. D. MCLEAN, Reports on Progress in Physics **29** (1966) 1.
4. T. H. ALDEN, *Trans. ASM* **61** (1968) 559.
5. D. LEE, *Acta Metallurgica* **17** (1969) 1057.
6. M. D. HALLIDAY, N. B. W. THOMPSON, and C. J. BEEVERS, *Met. Sci. J.* **3** (1969) 88.
7. A. W. MULLENDORE and N. J. GRANT, *Trans. Met. Soc. AIME* **227** (1963) 319.
8. J. L. WALTER and H. E. CLINE, *ibid* **242** (1968) 1823.
9. R. H. BUSH, A. N. LORD, W. D. NIX, and R. A. HUGGINS, Report No. 64-9, Dept. of Materials Science, Stanford Univ., California.
10. M. D. HALLIDAY, C. A. P. HORTON, and C. J. BEEVERS, *Met. Sci. J.* **3** (1969) 195.
11. F. WEINBERG, *Trans. AIME* **212** (1958) 808.
12. R. C. GIFKINS, A. GITTINS, R. L. BELL, and T. G. LANGDON, *J. Mater. Sci.* **3** (1968) 306.
13. F. GAROFALO, Fundamentals of Creep and Creep Rupture in Metals, (MacMillan, New York, 1965), p. 82.
14. M. KATO, *Trans. Japan Inst. Met.* **10** (1969) 215.
15. G. MIMA and T. OKA, *ibid* **8** (1967) 32.
16. R. L. BELL, N. B. W. THOMPSON, and P. A. TURNER, *J. Mater. Sci.* **3** (1968) 524.
17. C. A. P. HORTON, N. B. W. THOMPSON, and C. J. BEEVERS, *Met. Sci. J.* **2** (1968) 19.