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REPORT 1331

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National Advisory Committee for Aeronautics

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REPORT 1331

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By F. N. Rhines, W. E. Bond, and M. A. Kissel

SUMMARY

Grain-boundary displacement, occurring in bicrystals during creep at elevated temperature (350° C), has been measured as a function of the copper content (0.1 to 3 percent) in a series of aluminum-rich aluminum-copper solid-solution alloys. The minimums in stress and temperature, below which grain-boundary motion does not occur, increase regularly with the copper content as would be expected if recovery is necessary for movement. Otherwise, the effects, if any, of the copper solute upon grain-boundary displacement and its rate are too small for identification by the experimental technique employed. It was shown, additionally, that grain-boundary displacement appears regular and proceeds at a constant rate if observed parallel to the stress axis, whereas the motion is seen to occur in a sequence of surges and the rate, to diminish with time if the observations are made perpendicular to the stress axis. This is interpreted as further evidence that grain-boundary shearing occurs within a layer of metal of finite thickness and not by sliding upon a single interface.

INTRODUCTION

In a previous investigation upon grain-boundary creep in bicrystals of pure aluminum (ref. 1) it was shown that displacement at the grain boundary begins only after an induction interval and then proceeds in a spasmodic sequence of surges, alternating with periods of rest. The overall displacement is an approximately linear function of the cube root of the time and increases with temperature according to the Arrhenius relationship, and, over modest ranges of stress, its logarithm is a linear function of the stress. The direction of movement is that of the maximum resolved shear stress in the plane of the grain boundary, irrespective of crystal orientation; but the overall rate of displacement is highly sensitive to crystal orientation, increasing with the angular difference between the active slip systems in the conjugate crystal. Yielding occurs in a zone of metal of substantial depth adjacent to the grain boundary and is obviously heterogeneous in character.

This mode of behavior was explained upon the basis of a slip-recovery process modeled after that which has been employed by McLean (ref. 2) to account for steady-state creep in polycrystalline metals. It was postulated that transgranular slip causes an accumulation of bending energy adjacent to the grain boundary, which accumulation results in an early and localized recovery in a narrow band all along the boundary. The metal, thus softened, yields preferentially and the direction of yielding is parallel to that of the grain boundary. When yielding parallel to the grain boundary has ceased, owing to a loss of coordination of the slip-recovery cycles in the affected zone, there still remains in progress the same transgranular yielding that was present initially. This plastic motion again builds an accumulation of energy at the grain boundary, resulting in another occurrence of coordinated recovery and a new wave of displacement parallel to the grain boundary.

Based upon this view of the mechanism of grain-boundary gliding, it was postulated that the addition of alloying elements should affect the process most markedly in those respects relating to the occurrence of recovery. It was hoped that the observation of the details of spasmodic gliding in an alloy series would shed further light upon the function of alloying elements in depressing the creep rate.

Inasmuch as specialized techniques had been developed for studying grain-boundary creep in aluminum, it seemed best to pursue this study by observing the behavior of an aluminum-rich solid solution. The aluminum-copper system Al-Cu was adopted for investigation and was examined over the composition range of 0.1 to 3 percent Cu. Also, because alloying effects alone were to be investigated, the conditions of observation were limited to a single temperature and range of stress.

With respect to the system Al-Cu, the prediction of the influence of the recovery process upon the occurrence of gliding is clearly substantiated. The effects of dissolved copper upon the detail of spasmodic gliding were found to be too small for positive identification, but some interesting detail concerning the nature of the motions within the active zone along the grain boundary was observed.

This investigation was conducted at the Carnegie Institute of Technology under the sponsorship and with the financial assistance of the National Advisory Committee for Aeronautics.

EXPERIMENTAL MATERIALS

Eight binary alloys containing nominally 0.1, 0.2, 0.3, 0.4, 0.5, 1, 2, and 3 weight percent of copper were made from the purest available grades of aluminum (99.955 percent pure) and electrolytic copper, melted together in pure graphite crucibles. A master alloy containing 33.7 weight percent

[1] Supersedes NACA TN 3678, "Influence of Alloying Upon Grain-Boundary Creep" by F. N. Rhines, W. E. Bond, and M. A. Kissel, 1956.

copper was first made and this was then alloyed with pure aluminum to produce the final compositions, which were cast as ingots approximately 2 by 4 by 6 inches. Previous analyses of similar material have indicated that no single impurity above 0.005 percent would be present. The ingot was rolled to a thickness of approximately ⅛ inch and then was treated to produce a coarse-grained material from which bicrystals were cut. The coarse-grained material was made by alternately annealing at a temperature 1° or 2° below the solidus of the alloy, rerolling 1 to 2 percent, and then again annealing until very large grains were produced. In this way grains in excess of 1 inch in diameter were made. From these, tensile bars were cut with areas selected where the grain boundary was straight and each bar cut so that the grain boundary would cross it at an angle of 45° to the longitudinal axis. Thus the test specimen consisted of two crystals differing in orientation and separated by a flat interface perpendicular to the broad surface of the specimen and crossing it at an angle of 45° to its long axis.

The specimen was further prepared by photoengraving upon its broad surface a grid of reference squares, 133 to the inch. This was done by first electropolishing the specimens on all surfaces, next coating one of the broad surfaces with a photosensitive material (such as is used in making photoengraver's plates), exposing it to arc light through a halftone screen, developing it, and finally reetching the specimens with the original electropolishing solution. After the removal of the balance of the sensitive coating, there remained a pattern of squares upon an electropolished surface. Selected squares were used as reference marks for observing the displacement of the two crystals along the grain boundary and also for measuring the elongation of the specimen.

EXPERIMENTAL PROCEDURE

Aluminum-alloy bicrystals thus made were tested in creep under a dead load supplied by a weight suspended from the lower end of each specimen. The bicrystals were enclosed in a tube furnace which maintained the temperature within ±5° C. The reference pattern was observed through a window in the side of the furnace, measurements being taken by means of a long-focus telescope. At three positions along the grain boundary, the horizontal displacement of adjacent portions of the same reference square on the two sides of the grain boundary was measured at regular intervals; see the spans designated "A" in figure 1. At the same time the linear extension of the sample was measured within each grain (spans B_1 and B_2) over a gage length of 3 millimeters and astride the grain boundary over a gage length of 6.3 millimeters (span C). No special atmosphere was used in the furnace, the natural oxide forming upon these alloys affording protection against excessive surface deterioration.

Prior to testing, the orientation of each crystal was determined by the Laue back-reflection method. Also, the photoengraved surface was photographed, so that a comparison could be made between the appearance of the reference pattern before and after testing.

In a typical run, the sample was suspended in the furnace and brought to temperature. A calculated weight was then carefully hung from the sample, and readings were begun

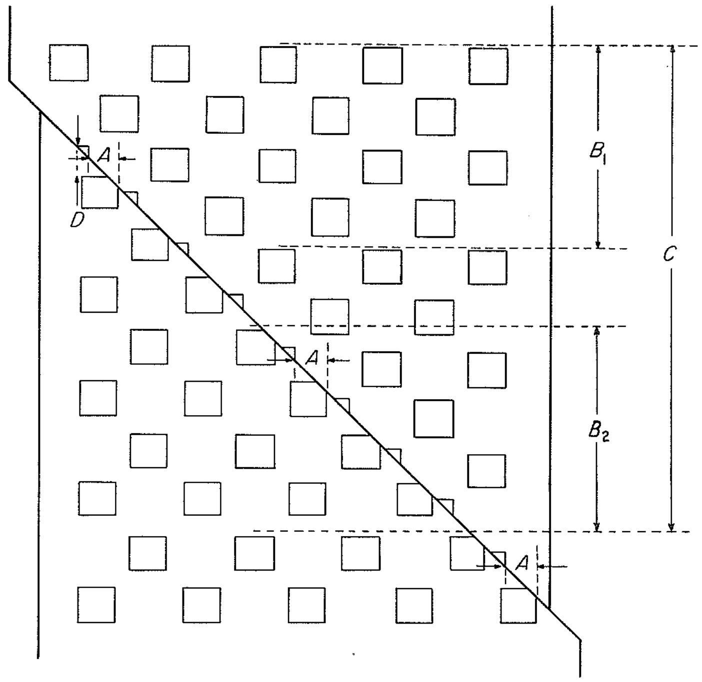

FIGURE 1.—Plan of measurements made upon a bicrystal.

immediately. Frequent readings were continued during the first day of testing, after which the period of observation was reduced to once each day, being thus continued until the test was terminated.

PRECISION OF MEASUREMENTS

Although the equipment used yields measurements of a high order of precision, it was found in previous studies that such accuracy is not required in making the observations of interest in the present research. The plastic motions involved, being inhomogeneous and erratic, give rise to variations in the readings which must be averaged in order to perceive the course of overall gliding. Thus, the fact that displacement can be read within an uncertainty of 0.005 millimeter means that the method is of an order of magnitude more sensitive than is necessary for recording the grain-boundary motion. The same is not true of the overall extension measurements which, for experimental reasons, had to be made over short gage lengths. Here the precision is somewhat less than that in ordinary creep testing, an error of ±0.5 percent being present in extension readings for the elongation of each individual grain. These matters are discussed more exhaustively in an earlier paper (ref. 1).

A variable which is not controlled, in these studies, is the orientation of the grains in the bicrystal. Previously, this had not been found to be limiting, in the measurement of the effects of such external variables as temperature and stress, so long as a sufficient number of duplicate tests were run to permit the orientation effect to be averaged out.

A procedure was later developed for the case of pure aluminum bicrystals which made it possible to work with relatively small numbers of samples of random crystal orientation. This method made use of the observation that the grain-boundary displacement (in unit time) averaged for all orientations D_A is proportional to the displacement D (in unit time) in any one bicrystal divided by the sum of two angles θ and ω

$$D_A = \frac{D}{\theta + \omega} c$$

where the value of the proportionality constant c is, for pure aluminum, approximately 65. Of the two angles, θ is that between the active slip directions in the two crystals of the bicrystal pair, and ω is the angle between the two active slip planes, as measured in the plane of the grain boundary. This method gives the average displacement within 5 or 10 percent, provided that the active slip directions and slip planes can be identified uniquely. Occasionally, more than a single slip system is active in one or both of the crystals, whereupon the method fails altogether.

In applying this correction to readings made upon Al-Cu bicrystals, it was necessary, for lack of information to the contrary, to assume that the proportionality constant c retains its value of 65. Perhaps for this reason, possibly as a result of some other cause, the correction, as applied to the Al-Cu samples, was found less effective than it had been when applied to pure aluminum samples. It has not been possible, with the relatively small number of tests available for each copper concentration, to evaluate the deviation that remains uncorrected; but it is clear that it is larger than that with pure aluminum.

EXPERIMENTAL RESULTS

All of the studies described in this report deal with solid-solution alloys. Although some of the alloys employed are capable, at lower temperatures, of rejecting a second phase ($CuAl_2$), all experiments were conducted at sufficiently high temperature to insure the absence of the precipitate. Concurrent metallographic examination verified the single-phased nature of the test specimens.

Since it was found in an earlier study (ref. 1) that there is a more or less sharp temperature minimum (stress constant and vice versa) below which gliding does not occur, it was desired to establish this limit for the several Al-Cu alloys. This was done by testing one series of alloys under constant stress at a sequence of increasing temperatures until gliding was observed in each and another series at constant temperature but with increasing stress until gliding was observed. In these tests, the temperature and the load, respectively, were increased at 24-hour intervals. The results appear in figures 2 and 3. Some scatter in the readings is to be expected when, as in the present case, the orientations of the bicrystals are not controlled. Despite such scatter, the upward trend, with copper content, of the temperature and stress limits at which gliding occurs is altogether clear.

From these results, conditions of temperature and stress suitable for time-displacement studies were selected. There is an advantage in using a relatively low temperature and stress, because the breadth of the active zone along the grain boundary is kept, thereby, at a minimum and gliding behavior becomes most regular. For tests up to 0.5 percent Cu, a temperature of 350° C and a stress of 200 psi were selected. Some tests run at higher temperatures and stresses contributed nothing additional to the results and are excluded from further mention.

A typical time-displacement plot, for one of the samples tested in the present research, is shown in figure 4. The grain-boundary displacement is plotted as a solid line passing through circles, which represent experimental readings. Also included in the same graph is the concurrent elongation of each of the two grains of the bicrystal, as well as the overall extension measured across the grain boundary and parallel to the direction of loading.

Selected readings taken from similar graphs, relating to all samples tested at 350° C and 200 psi, are reported in table I. The first two columns of this table identify the samples by number and chemical composition. In the third column are listed values of the angular sum $\theta+\omega$. In all cases there is an interval of time after the application of the stress and before the beginning of gliding; this is termed the "induction period" (see column 4 in the table). The grain-

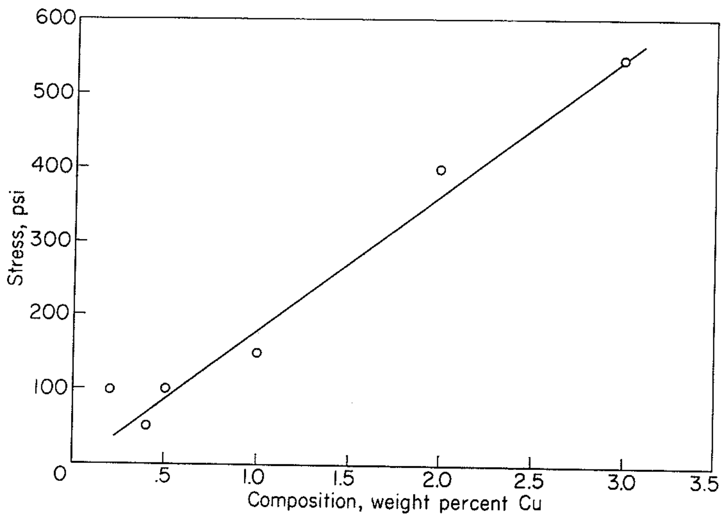

FIGURE 2.—Effect of copper content on stress required to produce a measurable grain-boundary displacement under a constant temperature of 400° C. Interval between stress jumps, 24 hours.

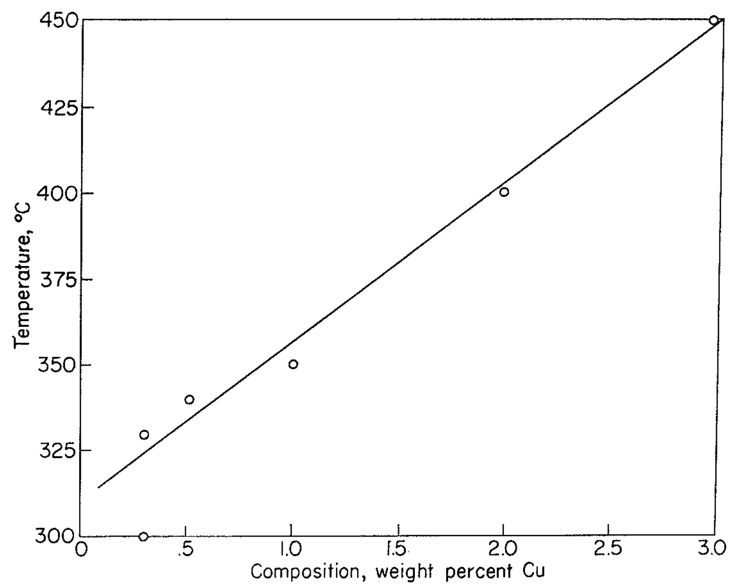

FIGURE 3.—Effect of copper content on temperature required to produce a measurable grain-boundary displacement under constant stress of 400 psi. Interval between temperature jumps, 24 hours.

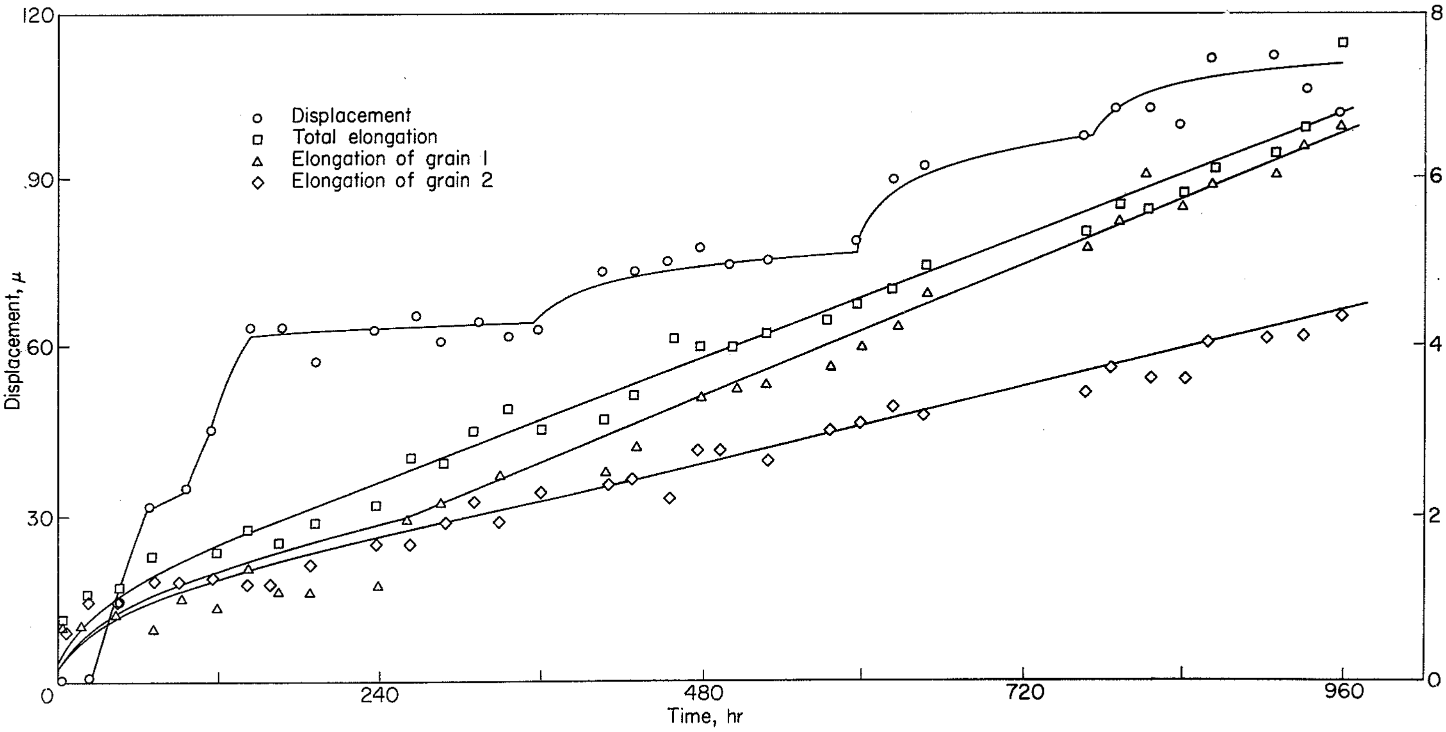

FIGURE 4.—Typical time-displacement plot. Specimen 2D (0.4 percent Cu). Temperature, 350° C; stress, 200 psi.

boundary displacement curve rises in a series of terraces; the first complete terrace is called the first cycle of gliding and the amount of the displacement therein is shown in the fifth column of the table. Because the amount of this displacement is sensitive to the orientation relationships obtaining in the bicrystal, a correction has been applied (column 6) in the manner which has already been described. The total displacement in 125 hours is shown in column 7, with these values corrected for orientation in column 8. In the ninth and tenth columns are given, respectively, the initial slopes of the displacement curve and these values corrected for orientation. The eleventh column gives the duration of the rest period at the crest of the terrace, as measured from the cessation of rapid gliding to the beginning of the next active period. The last three columns refer to the elongation of the sample, reporting elongation in the direction of the applied stress, as measured within each grain and astride the grain boundary during the time period only of the first cycle of gliding.

In a separate set of experiments, using pure aluminum bicrystals, a new study has been made of the effect upon the result of the direction in which grain-boundary movement is measured. Two measurements were made at each reference point, such as *A* in figure 1. One measurement, as previously, was made perpendicular to the direction of the applied stress (span *A*); the other was made parallel to the applied stress (span *D*) using the same bisected reference square and taking the two kinds of readings at the same time.

A typical result is shown in figure 5. The circles represent displacement readings measured parallel to the stress direction; the squares, readings measured perpendicular to the stress direction. As has been observed in almost all bicrystals studied, the displacement measured perpendicular to the stress axis proceeds in a series of distinct terraces, and the rate of displacement diminishes progressively with time. Scatter in the readings is small at first and increases with the duration of the test. The readings taken parallel to the stress axis, however, exhibit a scatter but no indication of terraces. Moreover, the rate of displacement, as measured parallel to the stress axis, becomes constant after the first few days of testing.

DISCUSSION

If the grain-boundary gliding is indeed a type of slip-recovery process, as has been proposed, it should be operative only within that temperature range wherein recovery occurs at a sensible rate. The minimum recovery temperature of any metal is expected to rise with solid-solution alloying, the effect being most pronounced with the first addition of the alloying element and the recovery temperature increasing thereafter at a diminishing rate as more of the solute is added. As had been anticipated, the minimum temperature for grain-boundary gliding, at a given stress level, was found to rise with the addition of copper to aluminum (fig. 3).

For the want of sufficient evidence to direct otherwise, a straight line has been drawn through the plotted points in figure 3; actually, it would be possible to draw a parabolic curve that would fit the points almost as well. The readings represent, for a randomly oriented bicrystal pair, that temperature at which the induction period is less than 24 hours. Since the length of the induction period has been shown to be sensitive to crystal orientation, scatter was expected in the measurements. Thus, it appears that the observations confirm qualitatively that the minimum gliding temperature varies with alloying, as does the minimum recovery temperature.

For comparison purposes there has been included, as the

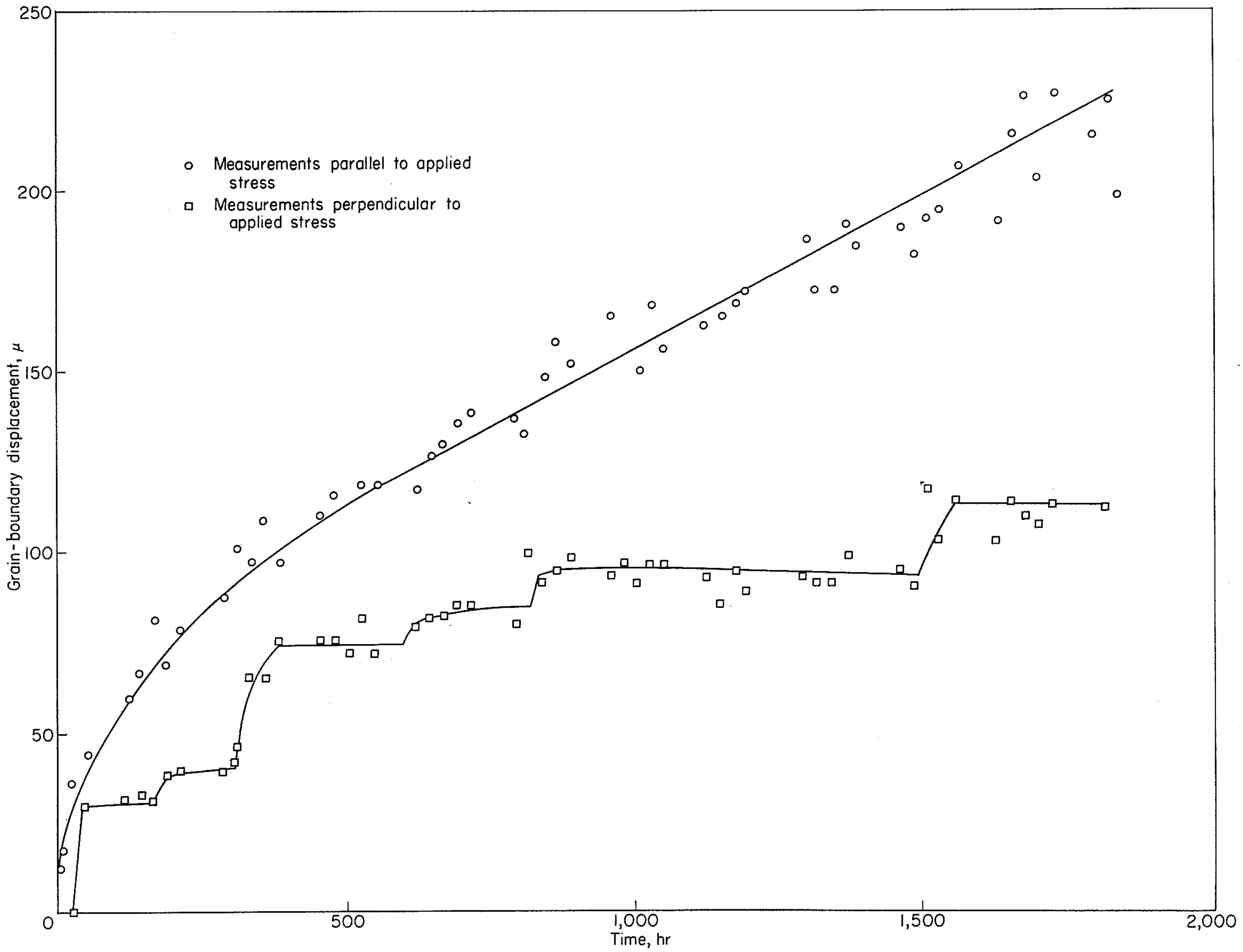

FIGURE 5.—Time-displacement plots, with measurements made parallel and perpendicular to applied stress.

first line in table I, a series of values derived from measurements made previously (ref. 1) upon pure aluminum. Since no tests had been run on pure aluminum bicrystals at 350° C and 200 psi, it was necessary to interpolate between readings that had been taken at 300° C and 400° C with a stress of 200 psi. Few such measurements were available, so that the values shown for zero percent copper in table I have about the same degree of uncertainty as those derived from the alloyed samples themselves.

A small, though distinct, trend toward a diminishing overall rate of creep with rising copper content can be seen in the last three columns of table I; that is, the elongation values decrease with increasing copper content. This is to be compared with a scarcely perceptible change in the total grain-boundary displacement in 125 hours (columns 7 and 8) that can be considered to be diminishing with alloying only if the high value for zero percent copper is considered more reliable than the low values found for the smallest copper addition.

A similar situation exists with respect to the detail of grain-boundary gliding. The trends shown are too small to merit credence. This is true of the induction period (column 4), the displacement in the first cycle (columns 5 and 6), the initial rate of displacement (columns 9 and 10), and the length of the first rest period (column 11). It might be inferred from this, coupled with the apparently definite decrease in the overall creep rate with alloying, that the contribution of grain-boundary creep to gliding is diminishing with alloying. That such a conclusion is not justified, however, is shown by a comparison of the copper content with the ratio of grain-boundary migration to overall extension:

Percent Cu	$\frac{\text{Grain-boundary displacement}}{\text{Total elongation}} 100$
0	97
.1	25
.2	59
.4	61
.5	118

It must be concluded, therefore, that the effects, if any, of dissolved copper upon grain-boundary displacement are too small for detection by the experimental method employed.

Of considerable interest is the new observation relating to the influence of the axis of measurement upon the appearance of grain-boundary gliding (fig. 5). The two curves presented in figure 5 differ both in their general form and in their detail, although they were derived from simultaneous measurements taken at right angles to each other at the same reference markers astride the same grain boundary. Were gliding to occur by shear upon the grain boundary as an interface, it seems inevitable that the two kinds of measurements should produce nearly identical results.

It is concluded, accordingly, and in conformity with previous deductions (ref. 1), that gliding does not occur upon an interface. It occurs rather as a complex plastic motion in a thin layer of metal adjacent to the grain boundary. Extension parallel to the stress axis is always in progress in this region and at a rate somewhat exceeding that of the general extension of the grains themselves. This is shown by the fact that the "total elongation" (column 14 in table I) usually exceeds the elongation of the individual grains (columns 12 and 13). Meanwhile, there is in progress a superimposed, spasmodic adjustment parallel to the grain boundary. It is the latter motion alone that results in an observable shearing of markers that cross the grain boundary.

Since various observers of grain-boundary gliding have, in some cases, detected the motion by a shearing of marker lines that crossed the boundary, while, in other cases, they measured the extension occurring between reference points astride the boundary, it is not surprising that somewhat inconsistent results have been obtained. Evidently, the two kinds of observations measure different quantities.

Also of interest is the fact that a scatter in the readings (fig. 5) sets in earlier in the measurement parallel to the stress axis. It had been noted previously, in measurements perpendicular to the applied stress, that scatter begins to appear after the first few cycles and thereafter increases with the subsequent cycles becoming less and less distinct. This condition in the readings parallel to the stress axis would seem to indicate that only a portion of this extension is derived from yielding parallel to the grain boundary. This was, of course, predicted by the mechanism previously proposed.

CONCLUSIONS

Studies of grain-boundary gliding in a series of aluminum-copper alloys (0.1 to 3 percent copper) have indicated the following conclusions:

1. The minimum temperature and the minimum stress for gliding increase progressively with the copper content. It is concluded from this that recovery is necessary for the progress of gliding.
2. The overall creep rate diminishes somewhat with increasing copper content in Al-Cu bicrystals; but the influence of copper content upon the rate and detail of grain-boundary gliding, if any, is concealed by the much larger effect of the variation of the rate, from sample to sample, with difference in the crystallographic orientation relationships from sample to sample.
3. Grain-boundary yielding, as measured parallel to the applied stress, progresses linearly and without perceptible surges, whereas gliding, as measured perpendicular to the applied stress, is spasmodic and diminishes with time. This observation is believed to prove that grain-boundary gliding occurs not as a shear upon an interface but as a complex plastic motion within a layer of metal adjacent to the grain boundary.

Carnegie Institute of Technology,
Pittsburgh, Pa., *June 24, 1955.*

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TABLE I

SAMPLES TESTED AT 350° C UNDER A STRESS OF 200 PSI

Test designation	Percent of copper	Angle $(\theta+\omega)$, deg	Induction period, hr	Displacement in first cycle, μ		Displacement in 125 hr, μ		Initial rate of displacement, μ/hr		Length of first rest period, hr	Percent elongation, grain 2, during first cycle	Percent elongation, grain 1, during first cycle	Percent total elongation during first cycle
				As read	Corrected	As read	Corrected	As read	Corrected				
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)	(12)	(13)	(14)
Average [a]	0	65	--------	--------	90	--------	180	--------	--------	87	1.9	1.7	3.1
2A	0.1	61	24	76	81	80	85	3	3.2	130	11.0	1.5	7
6A	.1	36	48	28	52	30	54	1.2	2.2	85	1.5	1.5	2
7A	.1	108	.5	130	78	120	72	1.8	1.1	90	1	.5	2
Average	0.1	--------	24	78	70	77	70	2	2.2	102	4.5	1.2	3.7
1B	0.2	44	7	33	49	50	74	2	3	48	2.0	2.0	2.5
3B	.2	30	7	24	52	37	80	1.4	3	48	.7	.5	2
Average	0.2	--------	7	29	50	44	77	1.5	3	48	1.4	1.2	1.7
1D	0.4	29	24	45	100	55	123	1	2.2	145	1.0	0.7	2.0
2D	.4	50	24	35	45	45	59	1	1.3	95	1.5	1.5	2.0
3D	.4	50	6	30	39	60	78	3	3.9	50	1.0	1.0	1.5
4D	.4	62	.5	40	42	60	63	2	2.1	50	2.0	1.5	2.0
5D	.4	73	.5	60	88	80	71	3	2.7	60	2.0	1.5	2.0
6D	.4	37	5	27	47	50	87	2	3.5	60	1.5	.5	1.0
Average	0.4	--------	10	40	60	58	80	2	2.6	77	1.5	1.1	1.7
1E	0.5	33	22	45	89	50	98	1	2	40	0.5	1.0	1.5
2E	.5	53	7	55	67	55	67	1	1.2	100	.3	1.0	1.5
5E	.5	52	.5	45	56	65	78	2	2.5	70	.5	1.0	1.0
6E	.5	37	24	45	79	50	88	1	1.7	80	----------	1.0	----------
Average	0.5	--------	13	48	73	55	83	1.3	1.9	72	0.4	1.0	1.3

[a] Derived from measurements made previously on pure aluminum (ref. 1).

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