Direct Observation of Internally Oxidized Cu-Al Alloys Deformed in Tension

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The dislocation configuration and density in internally oxidized Cu-Al alloys deformed in tension have been investigated by means of transmission electron microscopy. It was found that the extension eventually led to the formation of small size cells through the following process: After the formation of loops around the particles, the interaction of loops with other dislocations caused tangled dislocations, and the combining of the tangled dislocations formed a cell structure. In the interaction between particles and dislocations were found two types of the loops. This indicates that the dislocation proceeds not by shearing particles but by making cross slip. The deformation-induced substructure exhibited a remarkable increase in dislocation density.

It is proposed therefore that the major contribution to the work hardening in Cu-Al₂O₃ alloys results from the reduction in dislocation spacing in the forest and that the flow stress is the one required to cut through the forest dislocations formed around the particles.

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I. Introduction

It has been established lately that the fine dispersed particles which impinge the movement of dislocation enhance the yield strength and raise the recrystallization temperature of the dispersion strengthened alloys^{(1)~(6)}. A great deal has been reported on the mechanical properties of the alloys, including creep-rupture and tensile strength. But there are very few fundamental studies on the dislocation distribution and interaction between dispersed particles and dislocations in these alloys except the recent work on Cu base alloys by Lewis⁽⁷⁾ and on Al alloys by Ansell⁽⁸⁾.

The experimental results available show that the cell size is greatly reduced during the plastic deformation owing to the presence of the dispersed particles. From the direct observation that loops were left around the SiO₂ particles in deformed Cu-SiO₂ alloys, Ashby⁽⁹⁾ proposed that they were formed by the dislocation bypassing the particles by the cross slip mechanism as proposed by Hirsch⁽¹⁰⁾. On the other hand, from a direct observation of deformed Al-Al₂O₃ alloys, Ansell considered that the dislocation would bypass the Al₂O₃ particles by shearing them.

The present paper deals with the direct observation of the Cu-Al₂O₃ alloys deformed in tension with respect to the dislocation configuration, interaction of dispersed particles with dislocations and the work hardening due to the presence of fine dispersed Al₂O₃ particles.

II. Experimental Procedure

Highly pure electrolytic copper and aluminum were used as the matrix and alloying element, respectively. Table 1 shows the chemical analysis of the specimen

Table 1 Chemical analysis of the prepared specimen.

Specimen	Al (wt%)	Al ₂ O ₃ (vol%)
Cu-0.2 Al	0.225	1.02

prepared. The value in vol% represents the volume percentage of the Al₂O₃ when the added aluminum wholly changed to the oxide. A high frequency vacuum furnace was employed for melting the alloy and specimen was prepared into a plate 0.2 mm in thickness by ordinary forging and rolling.

Internal oxidation was carried at 1000°C by the Cu pack method after Rhines⁽¹¹⁾. Tensilon was used for the specimen extension and the strain rate was kept at a constant speed of 5 mm/min throughout this experiment. Specimens for transmission electron microscopy were cut from the extended tensile specimens suffering different amounts of strain. The initial thinning of the cut specimen was carried out with the chemical reagent composed of an equal part of phosphoric, nitric and acetic acid. The final thinning was done with an electrolyte of an equal part of phosphoric acid and glycerin and a small amount of water, while cooling in ice water to control the rate of thinning following a modified Bolman method⁽¹²⁾. Specimens were examined by a transmission electron microscope operated at 100 kV and equipped with a tilting device. Measurement of the dislocation densities was carried by the method developed by Keh⁽¹³⁾. According to this method, the dis-

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location density D is determined from the number of intercepts with two sets of parallel lines in a cross grid, using following equation

$$D = \left(\frac{n_1}{L_1} + \frac{n_2}{L_2} \right) \frac{1}{t},$$

where t stands for the specimen thickness, L_1 the total length of the parallel lines of one set, L_2 the other, n_1 and n_2 the number of intercepts with these two sets of parallel lines respectively.

The specimen thickness was measured from the width of slip trace⁽¹⁴⁾ and by the following method. The size and the number of the dispersed particles were measured on the photograph and their volume fraction was calculated. By comparing it with the fraction determined from the chemical analysis the thickness was estimated. During the thinning process, there is the possibility that the dislocations are free to move away, but in this case the dislocations pinned at the particles are considered difficult to move. As the dislocation density increases rapidly with deformation, it becomes very difficult to identify the individual dislocations in the tangled area. For this reason, the dislocation density of the highly extended specimen was not measured.

III. Results and Discussion

Fig. 1 shows a stress-strain curve for the internally oxidized tensile specimen. The proportional limit of

Fig. 1 Stress-strain curve of an internally oxidized tensile specimen for direct observation.

the alloy strengthened by the internal oxidation is fairly large, while that of the pure Cu is fairly small. For comparison of the two, the residual strain was employed instead of the ordinary strain for the strengthened alloy. Each point on the stress-strain curve represents the residual strain and the stress of the specimens prepared for transmission electron microscopy.

Photo. 1 shows the microstructure of the alloy as

internally oxidized. It is observed that the formed Al_2O_3 particles are uniformly distributed in the specimen with the particle radius of $140 \text{ \AA}$ and the inter-particle spacing of 0.2μ . The X-ray diffraction of the dispersed particles showed that they consisted mainly of

Photo. 1 Microstructure of Cu-Al alloy as internally oxidized.

crystalline $\gamma\text{-Al}_2\text{O}_3$. As reported⁽¹⁵⁾ by Ashby, the dispersed Al_2O_3 particles are likely to form a platelet different from the round shape of SiO_2 particles observed in internally oxidized alloys. At the both ends of the particles are observed a marked diffraction contrast effect, with a contrast free area at the center. This marked diffraction contrast is explained in terms of the elastic strain field⁽¹⁶⁾ existing at the interface between the matrix and the particles. The cause of the elastic strain field at the interface may be due to the coherency between the matrix and the particles or to the difference of the thermal expansion coefficient between them. In this case this effect is mainly attributed to the latter from the fact that the size of the Al_2O_3 particles is large, the Moiré patterns are not observed around the particles and the specimens were cooled down rather rapidly from the elevated temperature.

In Photo. 2 are shown the interfaces between the matrix and the dispersed particles, acting as dislocation

Photo. 2 Interfaces between the matrix and the dispersed particles, acting as a dislocation source.

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sources by giving a slight strain to the thin specimen. Photo. 3 shows the typical impingement effect of the particles upon dislocation movement.

Photos. 4 and 5 show the microstructures of the specimen, respectively, when extended by 0.89% and 2.8

single slip plane bypasses particles by bowing out between them and eventually leaving loops around the particles. Hirsch⁽¹⁰⁾ proposes, on the other hand, that the jogged loop be left by the cross slip mechanism. As cross slip takes place in Cu at room temperature, no

Photo. 3 Impingement effect of the particles upon dislocation movement.

Photo. 5 Microstructure of the specimen extended by 2.8%.

Photo. 4 Microstructure of the specimen extended by 0.89%.

%. A remarkable increase in dislocation density is observed in spite of the lower deformation. As a distinct feature, the loops appearing around the particles by the 0.89% extension and the loops extending in the

clear Orowan type loop has been observed yet. In the Mg-MgO system where the cross slip does not occur, however, the presence of an Orowan type has been reported⁽¹⁸⁾. As already mentioned, Ashby has observed a jogged loop left around the dispersed particles of SiO₂. Ansell considers from a direct observation that the Al₂O₃ particles are sheared when the dislocations bypass them, while he observed the loops around the particles which appeared to be formed by cross slip. Koda⁽¹⁹⁾ and other investigators⁽²⁰⁾ have confirmed that the dislocations pass through the precipitated Al-Cu or other systems. In the dispersion strengthened Cu alloys, however, there is little possibility that the dispersed particles are cut through when the dislocation advances. There is no evidence of the distortion of the Al₂O₃ platelet in this experiment, as in Cu-SiO₂ alloys⁽⁷⁾. It is more likely that the jogged loop is formed by the Hirsch mechanism in Cu alloys where the stacking fault energy is fairly large.

A careful observation of the loops shown in Photos. 4

Fig. 2 Mechanism of the loop formation associated with two particles.

slip direction rapidly increases by the 2.8% extension. Two mechanisms have been proposed to explain the formation of a dislocation loop around the particles: According to Orowan⁽¹⁷⁾, the dislocation confined to a

and 5 indicates that they can be classified into two types; one associated with a single particle and the other

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with two particles. The loop associated with two particles appeared at the early stage of deformation, while the loop with a single particle after a certain extent of deformation. The formation of the former may be explained on the basis of the mechanism proposed by Hirsch. At the early stage of deformation, as shown in Fig. 2, the dislocation bows out between the particles A, B and C, D when the interparticle spacing between A, B and C, D are fairly large compared with that between B and C. Deformation continues successively by the double cross slip around the particles due to the stress concentration effect induced by their existence. As the segments of the dislocations cross slipped over, the particles have the screw components of the opposite sign which eventually attract each other and annihilate to form the loops. Thus the loops left surround the particles at the both ends. If this loop were formed by slip on the same plane instead of cross slip, dislocation segments of the opposite sign would attract and annihilate because of the small distance to leave only one loop for each particle. Nemoto⁽²¹⁾ observed a similar phenomenon in Al-Ag alloys. This loop formation is considered to correspond to the later stage of the so-called "ribbon slip" which is initiated by the dislocation passing through the relatively large spaced particles. Further deformation increases the formation of a loop surrounding a single particle as pointed out by Ashby.

Photo. 6 Microstructure of the specimen extended by 7.9%.

Photo. 7 Microstructures of the specimen extended by 14.3% and the pure Cu by 30%.

Photo. 6 shows the microstructure of the specimen when extended by 7.9%. As the deformation goes on,

the loops around the particles interact with other dislocations to form tangled areas and thus their dislocation density is increased. The microstructure of the 14.3% extended specimen is shown in Photo. 7. With a remarkable formation of the cell structure, the cell size reduces to the mean value of 0.2μ which is fairly small compared with that of 1μ of the extended Cu shown in the same figure. The dispersed particles are likely to promote the formation of cell walls which combine the densely tangled dislocations around them. In Table 2 are shown the measured dislocation densities of the extended specimens and the polycrystalline Cu foil⁽²²⁾. It is noticed that with increasing strain, the dislocation density increases much more rapidly in the dispersion strengthened alloys. Especially, the dislocation density

Table 2 Effects of deformation upon the dislocation density of internally oxidized Cu-Al alloy and pure Cu.

(a) Internally oxidized Cu-Al alloy

Residual strain in elongation pct.	Measured dislocation density (lines/cm ²)	Mean dislocation density (lines/cm ²)
[0.89]	1.92×10^{10}	2.29×10^{10}
	2.22×10^{10}	
	2.23×10^{10}	
	2.33×10^{10}	
	2.48×10^{10}	
	2.55×10^{10}	
2.8	3.50×10^{10}	3.86×10^{10}
	3.58×10^{10}	
	3.62×10^{10}	
	4.20×10^{10}	
	4.38×10^{10}	
7.9	4.75×10^{10}	4.96×10^{10}
	4.82×10^{10}	
	4.85×10^{10}	
	4.85×10^{10}	
	5.12×10^{10}	
	5.37×10^{10}	

(b) Pure Cu

Elongation pct.	Mean dislocation density (cm/cm ³)
1.0	0.19×10^{10}
8.0	1.50×10^{10}
17.5	3.00×10^{10}
30.0	5.7×10^{10}

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is very large at the early stage of deformation which can explain the high work hardening rate characteristic

of this alloy.

No concrete theory has been established to account for the work hardening of dispersion strengthened alloys. Lewis⁽²³⁾ stated that the work hardening in this system could not be explained on the basis of Fisher's mechanism. But recently Ashby proposed that the cell structures formed during deformation contribute to the work hardening of the dispersion strengthened alloys.

As for the f.c.c. metals, several theories have been proposed to explain the work hardening behavior^{(24)~(27)}. From the results of direct observations, the present author considers that the high dislocation density especially at the tangled areas has a large effect upon work hardening as in pure Cu. From the fact that dislocation pile-up was not observed in this experiment, the long range pile-up theory may not be applied. Instead, the forest theory proposed by Hirsch or Saada is likely to be applied. According to the forest theory, the flow stress is given as a stress to cut through the dislocations in the dense forest region and is inversely proportional to the dislocation spacing.

$$\text{Therefore } \tau = \alpha Gb/l_f,$$

where l_f denotes the dislocation spacing in the forest regions, G the shear modulus, b the Burgers vector and α the constant. Since N_f stands for the dislocation density in the forest regions, l_f is proportional to $N_f^{-1/2}$. Supposing that there is the relation $N_f = mN$ for the dispersion strengthened Cu-Al₂O₃ alloys as proposed for the deformed pure Cu and Ag by Bailey, the flow stress may be given by the following equation^{(22) (28)}:

$$\tau = (Gb / 2 \pi k) (mN)^{1/2}.$$

Here N is the average dislocation density, m the constant, $k=1$ for a screw, $k=1-\nu$ for an edge dislocation and ν the Poisson's ratio.

The plotting of τ/Gb versus the square root of the

Fig. 3 Plotting of (τ/Gb) against $N^{1/2}$.

average dislocation density in Fig. 3 shows a linear relationship predicted by the forest theory where τ is taken as the 1/2 of the tensile stress. As the solute element is oxidized in the course of internal oxidation, it is considered that the solute hardening contribution to the flow stress does not occur. Hence the straight line can be drawn passing through the origin in Fig. 3. In Cu-Al₂O₃ alloys dislocations form the jogged loops around the particles at the later stage of ribbon slip due to their presence. Further deformation causes the dislocation tangling by the interaction of loops with other dislocations and eventually builds up the cell structure of small size. Through the deformation process, the spacing of the dislocations in the dense forest region decreases keeping the relation of $N = mN_f$. Thus it is confirmed that the flow stress in the dispersion strengthened Cu-Al₂O₃ alloys is the one required to cut through the dense forest dislocations around the particles.

Since the value of m calculated from the slope in Fig 3 is 12 over 5 found for deformed Cu⁽²²⁾, it is considered that dislocations are more closely spaced in the dense forest region of the Cu-Al₂O₃ alloy than pure Cu.

IV. Conclusion

By means of transmission electron microscopy, the microstructure of the internally oxidized Cu-Al alloy deformed in tension was mainly investigated. The results obtained are summarized as follows:

(1) Owing to the presence of the dispersed particles, the dislocation density is very high at the early stage of deformation which explains the high work hardening rate at the initial stage.

(2) During the deformation, the dislocation proceeds not by shearing particles but by making cross slip to leave loops around the particles.

(3) With increasing deformation, two types of loops are formed, one is associated with a single particle, and the other with two particles depending upon the inter-particle spacing. The formation of loops seems to correspond to the later stage of "ribbon slip".

(4) The increment in dislocation density is much larger than that of the pure Cu, due to the presence of dispersed particles.

(5) The work hardening behavior of Cu-Al₂O₃ alloy is determined by the configuration of dislocations in the dense forest regions and the flow stress is given as the one required to cut through them.

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