High-Temperature Deformation and Fracture Behavior of Cu-SiO₂ Bicrystals with [011] Twist Boundaries^{*1}

Hiromi Miura, Kazuhide Saijo and Taku Sakai

Department of Mechanical and Control Engineering,
The University of Electro-Communications, Chofu, Tokyo182, Japan

Cu-SiO₂ bicrystals having [011] twist boundaries with different misorientation angles were tensile tested at various temperatures from 473 to 1173K. Most bicrystals fractured intergranularly. As the grain-boundary energy increases, intergranular brittle fracture tended to take place more easily at lower temperatures; the ductility of bicrystals with higher energy boundaries became lower. The elongation to fracture of the intergranularly fractured bicrystals decreased monotonically with increase in temperature. These grain boundary dependent characteristics of deformation and fracture behavior of bicrystals could be explained reasonably by considering the difference in grain-boundary sliding and Schmid factor.

Keywords: Cu-SiO₂ bicrystal, high-temperature deformation, grain-boundary fracture, grain-boundary energy, [011] twist boundary

1. Introduction

Dispersion-hardening is known as strengthening mechanism at high temperature by dispersing fine hard particles in materials. These second phase particles suppress the mobility and annihilation of dislocations and grain-boundary sliding (GBS), and therefore, abrupt decrease in strength is prevented at high temperature. However, these secondary phase particles on the grain boundaries act as an inhibitor of GBS, and provide stress concentration sites to promote the preferential nucleation of voids and cracks. This introduces an earlier grain-boundary fracture, and therefore, loss of ductility (1-3). Therefore,

strengthening by dispersing second phase particles brings two inconsistent mechanical behaviors at high temperature, i.e. preservation of strength and loss of ductility.

It is well known that the occurrence of grain-boundary fracture at high temperature is strongly dependent on the grain-boundary character from the experiments using polycrystals (4,5). However, Raj (6) reported that fracture caused by diffusional cavity growth was slowed down by at least three orders of magnitude in bicrystal specimens compared with that in polycrystal ones. They explained this as the distribution of plastic deformation stress in the bicrystal specimen would be different from that in polycrystal specimen. This suggests that there would be a practical limit for understanding the inherent grain-

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boundary strength from the experiments by using polycrystal specimens. This is because the perfect comprehension of parameters effective for the fracture, such as the distribution of grain boundaries with various characters and triple junctions, is impossible as a matter of fact. Miura et al.⁽²⁾ have investigated the high-temperature fracture behavior of orientation-controlled Cu-SiO₂ bicrystals and showed that grain-boundary fracture took place more easily with increase in grain-boundary energy. They attributed this grain-boundary dependent fracture behavior to GBS. That is, GBS takes place more easily with increase in grain-boundary energy⁽⁷⁻⁹⁾, stress concentration sites are formed at around the grain-boundary SiO₂ particles more easily at lower temperatures. This prompt formation of stress concentration sites promotes the prompt nucleation of voids and cracks, and therefore, leads to the earlier fracture. However, specimens they used were only Cu-SiO₂ bicrystals with [001] twist boundaries. It would be necessary to examine whether their explanation is widely suitable for the other types of grain boundaries. This is just the motivation of the present experiment. In this paper, Cu-SiO₂ bicrystals with orientation-controlled [011] twist boundaries were employed.

II. Experimental

Four bicrystals of a Cu-0.1mass%Si alloy having [011] twist boundaries were grown by the Bridgman method. The misorientation angles were 7°, 20°, 35° and 70.5° (hereafter referred to as the 7°, 20°, 35° and 70.5° bicrystals). According to the recent study by Miura et al.⁽¹⁰⁾, grain-boundary energies of these bicrystals increase in the order of 7°, 70.5°, 20° and 35°. After confirming by the X-ray Laue analysis that the actual misorientation angles were within 1° of the above nominal values, these bicrystals were

Table 1. Size and distribution of grain-boundary SiO₂ particles.

Bicrystals	7°	20°	35°	70.5°
Particle radius $r/\mu\text{m}$	0.185	0.194	0.198	0.202
Area fraction of grain-boundary particles A	0.0895	0.112	0.124	0.151
Density of grain-boundary particles $\rho/\mu\text{m}^{-2}$	0.694	0.761	0.720	1.00
$rp/\mu\text{m}^{-1}$	0.128	0.148	0.143	0.202

Fig. 1. Schematic illustration of a tensile specimen.

internally oxidized at 1223K for 24h with a powder pack method. A mixture of Cu (1 part), Cu₂O (1 part) and Al₂O₃ (2 parts) powders was used for this purpose. Dispersed SiO₂ particles were obtained both in the matrix and on the grain boundaries by this internal oxidation treatment. To eliminate the excess oxygen in the copper matrix and on the grain boundary, oxidized specimens were degassed (deoxidized) at 1223K for 6h in a graphite mold in vacuum ($\sim 10^{-3}$ Pa). Although the same internal oxidation and degassing treatments were applied, the size and distribution of the grain-boundary SiO₂ particles were different among the four bicrystals. Table 1 lists the average size and distribution of grain boundary particles in the tested bicrystals. Tensile specimens with 14.8mm gage length and 4x1mm² cross section were spark cut so that grain boundaries aligned at 45° with respect to the tensile direction (Fig. 1). The tensile direction changes depending on misorientation angle, though the primary slip direction is the same and

perpendicular to the grain-boundary plane in all the bicrystals and abutting grains. After the mechanical and electrical polishing, the tensile specimens were finally annealed at 1223K for 0.5h in a graphite mold in vacuum. Tensile tests were conducted in vacuum at various temperatures from 473 to 1023K with an Instron-type testing machine at a strain rate of $4.17 \times 10^{-4} \text{ s}^{-1}$. After the tests, fracture surfaces were observed by scanning electron microscope (SEM).

III. Results

Figure 2(a) and (b) show the series of true stress(σ) vs. true strain(ϵ) curves for 20° and 35° bicrystals, respectively. After the clear yielding, all of the flow curves show notable work hardening with increase in strain and proceed to the fractures with rapid drops in flow stresses irrespective of temperatures and misorientation angles. Flow stress vibration which characterizes the occurrence of dynamic recrystallization can not be observed. And after testing, from optical microscopic observation, occurrence of dynamic recrystallization was not recognized. This means that no dynamic recrystallization took place under the present experimental condition, although some testing temperatures were over $0.5T_m$.

Figure 3 summarizes the fracture strain (ϵ_f) as a function of temperature(T). Open and solid symbols indicate transgranular and intergranular fracture, respectively. For the 7° bicrystals, fracture strain increases with increase in temperature and after reaching maximum at 1073K, it abruptly drops at 1173K. For the 20° bicrystals, fracture strain increases with increase in temperature until 673K and then drops abruptly at 773K. These sharp drops of fracture strains are due to the change in the fracture mode from transgranular to intergranular-mode. For the 35° and 70.5° bicrystals, the fracture

Fig. 2. True stress-true strain curves of (a) 20° and (b) 35° bicrystals deformed at various temperatures.

Fig. 3. Temperature dependence of fracture strain for different bicrystals and volume fractions. Open and solid symbols indicate transgranular and intergranular fracture, respectively.

mode transition takes place relatively at lower temperatures compared with that of 7° and 20° bicrystals. The temperature of fracture mode transition decreases in the order of 7°, 20°, 70.5° and 35° bicrystals. After the occurrence of intergranular fracture mode, fracture strain decreases monotonically with increase in temperature irrespective of the misorientation angles. Different from [001] twist boundaries⁽²⁾, no recovery of fracture strain at higher temperatures were observed.

The fracture strain at the temperature above 700K is a strong function of misorientation angle. For example, at 800K, it decreases in the order of 7°, 20°, 70.5° and 35° bicrystals. This is approximately in good accordance with the previous result of Miura et al.⁽²⁾; grain boundary becomes more brittle with increase in grain-boundary energy. However, the ductility of 20° bicrystal is larger than that of 70.5° one in Fig. 3. This is because of the effect of Schmid factor on fracture behavior, as will be discussed later.

Figure 4 shows the temperature dependence of fracture stress (i.e. the peak flow stress). Open and solid symbols indicate transgranular and intergranular fracture, respectively. Fracture stress decreases monotonically with increase in temperature in all the bicrystals. Although, the difference in the fracture stress is notable at around the intermediate-temperature range between 600K and 900K, the difference becomes smaller at higher temperatures. The fracture stress decreases in the order of 7°, 20°, 70.5° and 35° bicrystals almost at all the temperatures. This suggests that grain boundary fractures more easily in the order of 7°, 20°, 70.5° and 35° bicrystals. This is in good agreement with the result of the grain boundary dependence of fracture strain in Fig. 3. Intergranular fracture stress tends to be smaller than the transgranularly fractured stress. The intergranular fracture observed is a brittle

Fig. 4. Temperature dependence of peak stress for different bicrystals. Open and solid symbols indicate transgranular and intergranular fracture, respectively.

Fig. 5. Fractured 35° bicrystals at various temperatures.

rupture occurring at low stress.

Fractured 35° bicrystals are shown in Fig. 5 as the typical fractured specimens. At 473K, transgranular fracture accompanied by necking takes place. At above 573K, clear intergranular fractures occur. It is recognized that the fracture strain decreases with increase in temperature.

SEM photographs of grain-boundary fractured surfaces of 35° bicrystals are shown in Figure 6. At 573K, fine dimples

Fig. 6. Scanning electron micrographs of fracture surfaces for 35° bicrystals deformed at (a) 573, (b) 773 and (c) 1073K.

indicating the occurrence of ductile fracture can be seen. At above the intermediate-temperature range, i.e. 773K and 1073K, indication of brittle fracture can be seen. The characteristic of brittle fracture is a relatively flat appearance with elongated voids made by dragging of SiO₂ particles by GBS. Therefore, fracture manner changes from ductile intergranular to brittle intergranular with increase in temperature. The dragged marks at 1073K is longer than those at 773K. This indicates that the amount of GBS

Fig. 7. Scanning electron micrographs of fracture surfaces for (a) 70.5° and (b) 20° bicrystals deformed at 773K.

increases with increase in temperature, although time to fracture becomes shorter with increase in temperature.

Figures 7 show SEM photographs of the fracture surfaces of 70.5° and 20° bicrystals deformed at 773K. The micrograph of fracture surface of 35° bicrystal deformed at 773K has already shown in Fig. 6(b). As can be seen from these micrographs, the appearance of grain-boundary fracture surface becomes flatter with increase in grain-boundary energy. This tendency may seem to be somewhat strange, because fracture strains of 70.5° bicrystals are smaller than those of 20° bicrystals. However, difference in Schmid factor may have affected the grain-boundary fracture, as will be discussed below.

IV. Discussion

As the temperature increases, grain-boundary diffusion becomes more

operative. If the GBS is assumed to be controlled by grain-boundary diffusion(11,12), GBS should occur more easily at lower temperatures in bicrystals with higher energy boundaries(7,9). When GBS occurs, GBS is suppressed by the grain-boundary particles. The blocking develops an internal stress, which acts against the applied shear stress on the particle-free area of the boundary, and eventually stops the sliding. According to Mori et al.(12), the characteristic time τ_L for GBS to reach its equilibrium amount $[u]$ is written as

$$\tau_L = (4/\pi^2)\{(1-\nu)/(2-\nu)\}(\eta/\mu r \rho c) \quad (1)$$

$$[u] = (8/\pi^2)\{(1-\nu)/(2-\nu)\}(\sigma r \rho) \quad (2)$$

where ν : Poisson ratio, μ : shear modulus, η : inherent viscosity of GBS, r : particle radius, ρ : number of particles on a unit area of grain boundary, c : half of the boundary thickness, and σ : applied shear stress parallel to the boundary. Boundary viscosity η is expressed as(13,14)

$$\eta = \eta_0 T \exp(-Q/kT) \quad (3)$$

where η_0 : inherent boundary viscosity, Q : activation energy for GBS, k : Boltzman constant and T : temperature.

If GBS is controlled by grain-boundary diffusion, η is inversely proportional to the grain-boundary diffusivity. The value at a fixed temperature varies significantly by several orders of magnitude with increase in grain-boundary energy(7-9). On the other hand, the variation of $r \rho$ in different bicrystals is only within a factor two(Table 1). Therefore, the value of τ_L at a fixed temperature abruptly decreases with increase in grain-boundary energy. It depends strongly on the value of η . When GBS occurs, the boundary SiO_2 particles act as an inhibitor of GBS and provide stress-concentration sites to promote the nucleation of voids and cracks. Therefore, it can be reasonably understood that loss of ductility takes place at lower temperatures as the grain-boundary energy increases. Monzen et al.(15) systematically measured the inherent boundary viscosity η_0 and the activation energy Q for GBS of [011] twist

Fig. 8. Changes in calculated τ_L with temperature for each grain boundaries.

boundaries. Substituting their results for η_0 and Q the value of $r \rho$ (Table 1) and $\mu = 4.6 \times 10^{10} \text{ Pa}$, $\nu = 0.34$, $c = 2.5 \times 10^{-10} \text{ m}$ into eq.(1) and (3), τ_L is calculated as a function of temperature T . τ_L for each grain boundary is plotted as a function of temperature in Figure 8. Provided that low energy boundary such as 7° boundary would hardly slide(7-9), τ_L value for 7° boundary can be expected to be much larger than those of the others. τ_L value decreases in the order of $7^\circ \gg 70.5^\circ > 20^\circ > 35^\circ$ at a fixed temperature. Therefore, stress concentration, which introduces to the early fracture, easily takes place in the same order with the above.

If the occurrence of brittle fracture is a simple function of the ease of GBS, grain-boundary fracture should take place more easily, and therefore, ductility should decrease with increase in grain-boundary energy. However, our experimental results show that 70.5° boundary is more brittle than 20° boundary. This means that simple interpretation of the brittle fracture only by GBS is insufficient. Some cavity formation mechanisms due to the plastic deformation have been proposed; (i)

dislocation pile-up against the grain boundary(16-18), (ii) stress concentration associated with the GBS at grain-boundary steps which are formed by slip deformation in matrix(19). The effect of Schmid factor on the fracture of different bicrystals should, therefore, be considered in such cases. Schmid numbers for 7°, 20°, 35° and 70.5° bicrystals are 0.43, 0.45, 0.48 and 0.50 increasing with increase in misorientation angle. In particular, primary slip direction is perpendicular to the grain boundary ([011] direction) in the present [011] twist boundaries. Therefore, the pile-up of dislocation against grain boundary and the formation of grain-boundary steps must takes place more easily by smaller deformation with increase in Schmid number or misorientation. Therefore, it can be thought that smaller fracture strains of 70.5° bicrystals than those of 20° bicrystals is due to the ease of dislocation pile-up against the grain boundary and the boundary step formation to promote the stress concentration site.

Miura et al.(2) have reported the occurrence of clear intermediate-temperature embrittlement in Cu-SiO₂ bicrystals with [001] twist boundaries. They explained the observed recovery of ductility at higher temperatures as a result of stress relaxation by the Cu/SiO₂ interfacial diffusion. However, no recovery of ductility at higher temperature was observed in this study. This is probably due to the difference in the distribution of grain-boundary particles between [001] twist and [011] twist boundaries, although the chemical composition is the same. That is, the density, radius and area fraction of grain-boundary particles in [011] twist boundaries are smaller than those of [001] twist boundaries, and therefore, GBS could not be effectively suppressed in [011] twist boundaries. GBS continued with the dragging of grain-boundary particles to form elongated voids in such condition. In fact, Miura and Sakai(3) reported that suppression of GBS became impossible with

decrease in the density of grain-boundary particles even in the same type of grain-boundary plane. Grain-boundary viscosity η , which controls the GBS, decreases abruptly with increase in temperature, and therefore, the amount of GBS also increases. Figures 6(b) and (c) show the clear evidence of the increase in the amount of GBS with increase in temperature. Therefore, we can conclude that recovery of ductility at higher temperature did not occur in [011] twist boundaries in this experiment.

V. Conclusions

High-temperature deformation and fracture behavior of Cu-SiO₂ bicrystals with [011] twist boundaries were systematically investigated. The results are summarized as follows.

1. The occurrence of fracture mode transition from transgranular to intergranular with increase in temperature causes the abrupt decrease in fracture strain.
2. Fracture strain due to grain-boundary fracture decreases monotonically with increase in temperature.
3. As the grain-boundary energy becomes higher, grain-boundary fracture occurs at lower temperature with lower fracture strain.
4. Grain-boundary dependence of fracture behavior can be reasonably understood by the occurrence of stress concentration site induced by grain-boundary sliding.
5. Schmid factor also influences the occurrence of grain-boundary fracture.

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