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Low void swelling in dispersion strengthened copper alloys under single-ion irradiation

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Abstract

Oxide dispersion strengthened copper (ODS-Cu) alloys GlidCop CuAl15 and CuAl25 were irradiated with Cu^{2+} ions at 573–773 K up to doses of 30 dpa. Void swelling was observed in all specimens irradiated at temperatures ranging from 573 to 673 K. In CuAl15 brazed with graphite at 1083 K, mean grain size was about 800 nm. Voids were observed in grains larger than 1 μm but not in smaller than 500 nm in diameter. The CuAl25 joined with SUS316 by hot isostatic pressing (HIP) at 1323 K had a mean grain size of 60 μm because of a large grain growth during the HIP process and showed large void swelling. Small grain size is effective in suppressing void swelling due to strong sink effects of grain boundaries for the point defects. The present results indicate that joining at high temperatures may reduce the void swelling resistance of GlidCop copper alloys.

1. Introduction

Oxide dispersion strengthened copper alloys (ODS-Cu) GlidCop CuAl15 and CuAl25 are considered as heat sink materials for the ITER divertor and first wall since they have high strength and high thermal conductivity and exhibit low swelling under neutron irradiation [1,2]. These alloys exhibited great swelling resistance under fast neutron irradiations [3–5]. However, in irradiation of welded sample, large degradation of swelling resistance was reported [5]. GlidCop copper alloys have fine grain size and contain a fine dispersion of alumina particles. Furthermore, the microstructure of these alloys is thermally stable up to a relatively high temperature. It is well known that grain boundaries, dislocations, and incoherent interface such as oxide particle and matrix are sinks for vacancies and reduce the magnitude of vacancy supersaturation in matrix [6–8]. On the other hand, hot isostatic pressing (HIP) is useful for the fabrication of divertor and first wall components. For the fabrication, single step HIP has been proposed to minimize thermal effects on material properties. It is expected, however, that bonding at high temperatures might degrade swelling resistance of ODS-Cu. It was therefore decided to investigate the influence of bonding temperature on the swelling behavior of HIP-joined samples of GlidCop copper alloys during irradiation with heavy ions. Results of these investigations are described and discussed in the present paper. For comparison, specimens of pure copper with coarse and fine grains were also investigated. The main results of these investigations are also reported in the following.

2. Experimental

GlidCop alloys CuAl15 and CuAl25 (by SCM metals) were used in this study. CuAl15 with 0.15 wt% Al content in the form of Al_2O_3 particles were joined with graphite by silver brazing at 1073 K. CuAl25 with

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0.25 wt% Al content was joined with SUS316L by HIP at 1323 K. From these samples, 3 mm diameter and 0.2 mm thick TEM discs were prepared for irradiation. The 99.999% pure copper (supplied by Johnson–Matthey Chemical Ltd.) was used as reference material. 3 mm diameter and 0.1 mm thick TEM discs were punched out from the 1000% cold-rolled sheet. Prior to irradiation, these discs were annealed at 873 K for 1.5 h in a vacuum of 1×10^{-4} Pa.

Irradiation was performed with 2.4 MeV Cu^{2+} ion at temperatures ranging between 573 and 773 K using a tandem type accelerator at Kyushu University. Irradiation conditions are shown in Table 1. The depth of the peak damage region during 2.4 MeV Cu^{2+} ion irradiation was estimated to be 400 nm by TRIM code. The damage rate in this region was about 2.5×10^{-3} dpa/s. After irradiation, the specimens were sectioned from the irradiated surface to a depth of 300 nm, which is 100 nm in front of peak damage, and then thinned from the backside by back-thinning method. The thickness of the specimens for TEM observation was typically 200 nm.

Pre-thinned specimens of the brazed CuAl15 and HIPed CuAl25 and pure Cu with fine grains and pure Cu with coarse grains were prepared to investigate the effects of grain boundaries. The thicknesses of specimens were the same as that of the bulk specimens. Twin-jet electro-polishing was performed before irradiation. The mean grain size of CuAl15, CuAl25, pure Cu with fine grain, and pure Cu with coarse grain were 800 nm, 60 μm, 700 nm and 300 μm, respectively. After irradiation, the widths of denuded layer of voids from the surface were measured by TEM. The thickness of the voids denuded layer was measured by thickness fringes under the two-beam diffraction condition. Then, the area of 300 nm from surface was electro-polished for TEM observation.

3. Results and discussions

3.1. Microstructure before irradiation

Fig. 1 shows the microstructure of copper alloys before irradiation. The mean grain size of CuAl15 was 800 nm and the number density of cold-worked dislocations varied from grain to grain. But alumina particles was homogeneously dispersed as shown in Fig. 1(d). The range of alumina particle size was 2–800 nm. The median size of alumina particles was ~8 nm, and its number density was 1.5×10^{22} m^{-3}. Coarse particles

Table 1
Irradiation conditions of copper alloys

	Specimen				
	CuA115 brazed (K)	CuA115 (K)	CuA125 HIP (K)	Pure Cu (K)	Pure Cu with fine grain (K)
10 dpa bulk	573–773	673	673	573–773	
30 dpa bulk	573–773	673	673	573–773	
10 dpa film	673		673	673	673

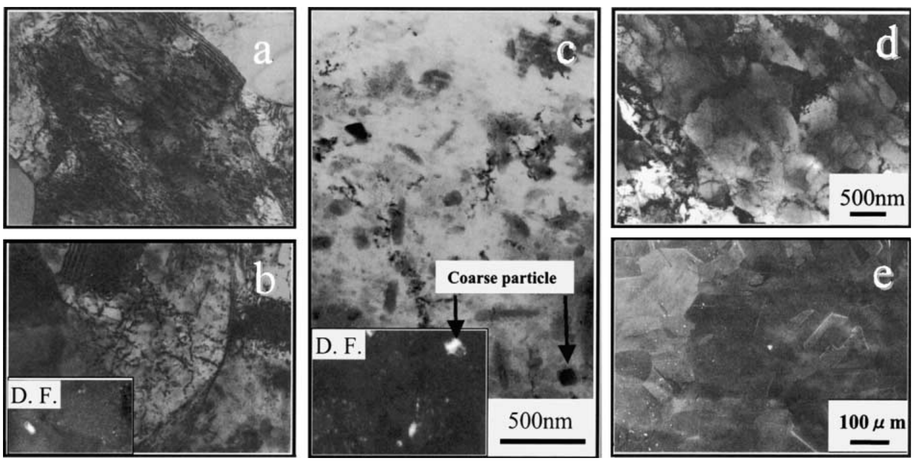

Fig. 1. TEM micrographs of unirradiated CuAl15 (a, d), CuAl15 after brazing (b), HIPed CuAl25 (c), and SEM micrograph of HIPed CuAl25.

over 100 nm in size were also observed in the specimen before and after brazing. TEM observations showed that the samples before and after had similar microstructure (Fig. 1(a) and (b)).

In CuAl25, on the other hand, mean grain size was 60 μm because of grain growth during HIP at 1323 K. Cold-worked dislocations recovered during HIP process as shown in Fig. 1(c). The median size of alumina particles was 8 nm, and its number density was 3.3×10^{22} m^{-3}. The coarse alumina particles were found to be segregated in some regions, probably because of the high HIP temperature. The mean size of these coarse alumina particles was about 100 nm.

3.2. Microstructure after irradiation

In CuAl15, voids were observed in samples irradiated at 573 and 673 K. Void formation was found to be dependent on the grain size. As shown in Fig. 2, in CuAl15, voids formed only in large grains. The void denuded zone along the grain boundary was also observed. The width of denuded zone was about 200 nm at 673 K. Table 2 summarizes the measured size and density of the voids in the three irradiated materials.

In CuAl25, grain growth occurred during HIP process. Due to the large grain size, voids formed in all grains during irradiation. Although the void swelling was less than that in pure Cu, heating during HIP process degraded the swelling resistance of ODS-Cu. Neither voids nor defect clusters were observed in copper alloys irradiated at 773 K.

Relationship between the measured void volume and grain size is shown in Fig. 3. The measured grains were chosen randomly. Under this irradiation condition, voids formed in coarse grains larger than 1 μm but not in grains smaller than 500 nm. This result indicates that

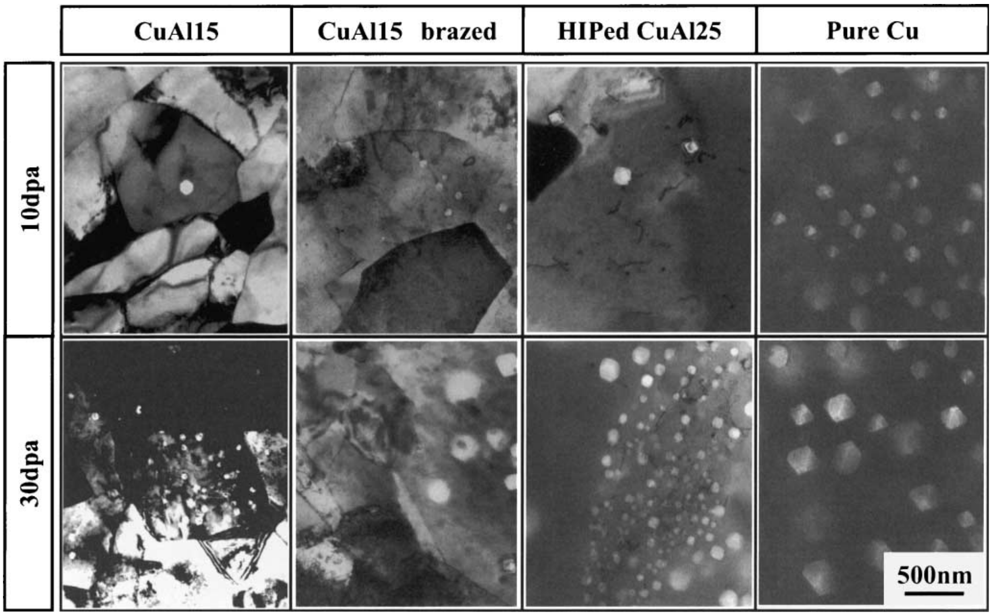

Fig. 2. TEM micrograph of irradiated bulk copper and copper alloys by 2.4 MeV Cu^{2+} at 673 K.

Table 2
Microstructural parameters for irradiated copper and copper alloys to 30 dpa at 673 K

Parameter (units)	Dose (dpa)					
	Pure Cu		CuA115 brazed		CuA125HIP	
	10	30	10	30	10	30
Average grain size d_g (μm)	300		0.8		60	
Denuded zone with W_D (nm)	300		150		250	
Mean void diameter d_v (nm)	220	260	60	130	100	150
Local void density (10^{18} m^{-3})	5.6	6.6	300	150	25	30
Local void volume $(\Delta V)_l$ (%)	3.2	6.1	<0.5	<6.6	0.8	3.7

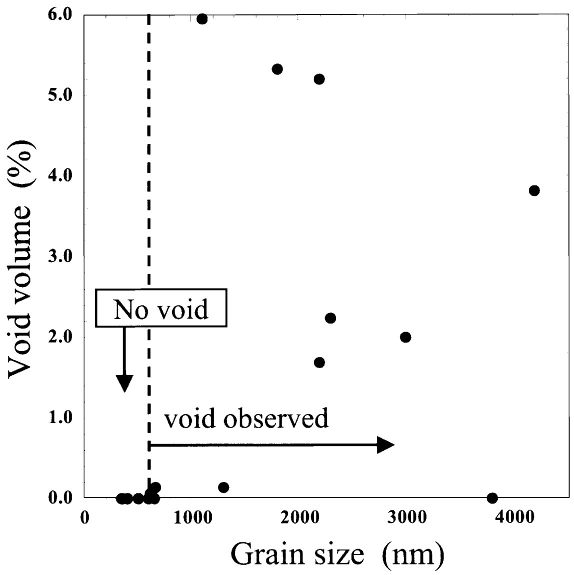

Fig. 3. Relationship between void volume and size of a grain in CuAl15 specimens irradiated at 673 K to 30 dpa.

small grain size causes suppression of void formation. The width of the void denuded layer along the surface was about 300 nm in the thin film irradiation of pure Cu with coarse grains at 673 K. In CuAl25, the width of denuded layer was about 250 nm at 673 K as shown in Table 2. Since the supersaturation of vacancies is low, voids do not form in the denuded zone. Assuming that the sink strength of grain boundary is almost the same as that of the surface, whole interior of the grain will be considered as denuded zone for a grain smaller than 500 nm in diameter.

To investigate the effect of dislocation and alumina particle on void swelling, the number density of dislocations and alumina particles were measured. Although dislocation density varied considerably from grain to grain (1.2×10^{13}–1.9×10^{14} m^{-2}), voids formed even in the grains with high dislocation density. This result indicates that there is no clear correlation between the suppression of void volume and dislocation density. As regards the effects of alumina particles, the void volume decreased with increasing number density of alumina particles at 10 dpa, which shows that alumina particles contribute to suppression of void volume. However, no clear correlation was found at 30 dpa, indicating that the effect of alumina particles on the suppression of void volume is not strong.

Bright and dark field images of irradiated HIPed CuAl25 of the same region are shown in Fig. 4. Cold-worked dislocations do recover during the HIP process. Although the dispersion of alumina particles is not homogeneous, voids do not form in the region where alumina particles are present in high density. These results suggest that alumina particles act as sinks for point defects. Identical HIPed joints of CuAl25/SUS316L was also irradiated by neutrons at 566 ± 19 K to a dose level of 4.6 dpa [9]. Recovery of dislocations and grain growth were also observed in the regions close to the interface. Moreover, in this region, alumina particles were not observed. This resulted in prominent growth of void.

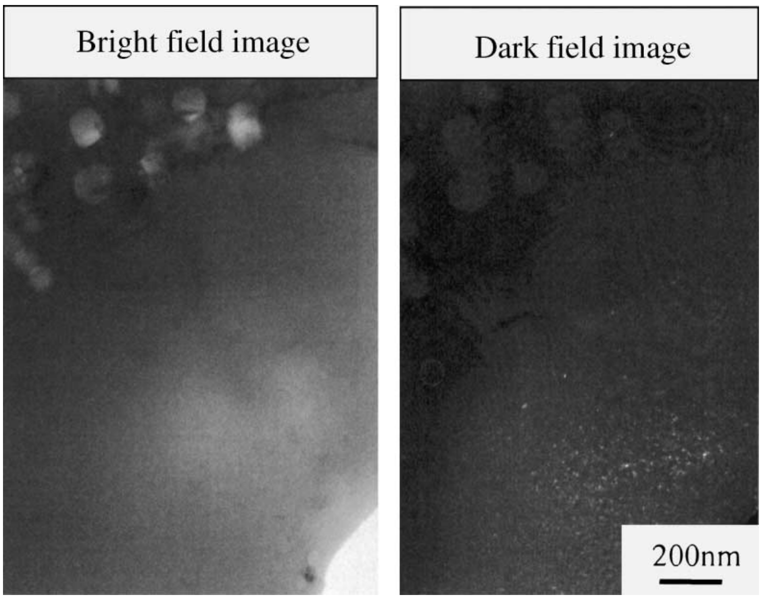

Fig. 4. Micrographs of HIPed CuAl25 irradiated 30 dpa at 673 K.

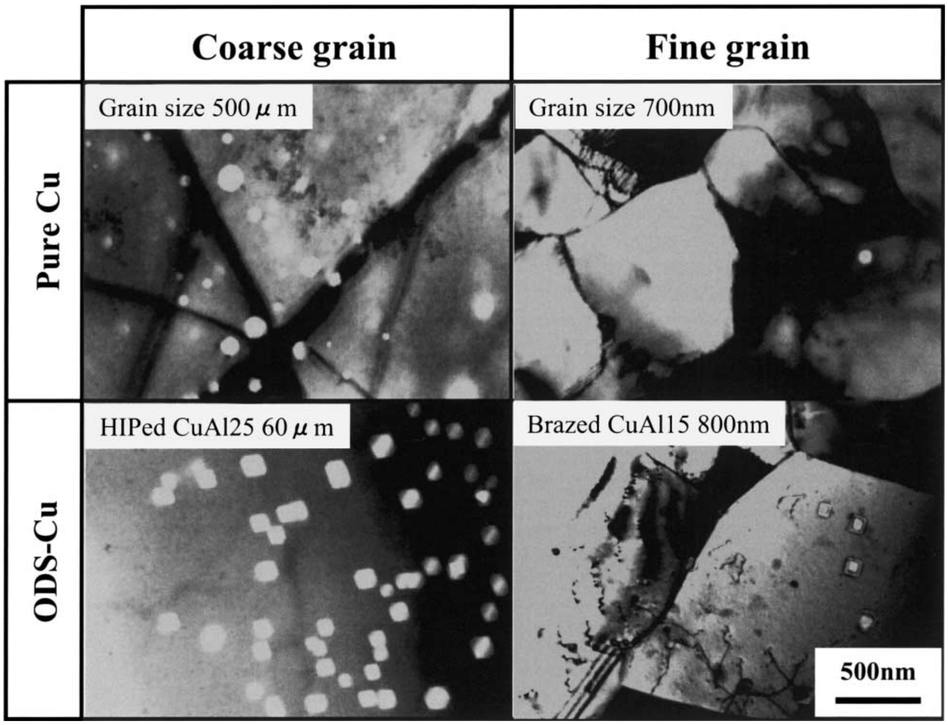

Fig. 5. TEM micrographs show the dependence of irradiated microstructure of pre-thinned pure Cu and ODS-Cu on grain size to 10 dpa at 673 K.

In ODS-Cu alloys, the presence of alumina particles (in high density) prevents grain growth even at high temperature of 1273 K. To know the effects of grain boundary, thin film specimens were irradiated at 10 dpa. The specimens were prepared using the same procedure as the one used for the bulk specimens. Fig. 5 shows the microstructure of the thin film specimens irradiated at 673 K. In pure Cu with coarse grains, brazed CuAl15 and HIPed CuAl25, no difference in microstructure was found between thin foil and bulk specimens. In pure Cu with fine grains, void volume was less than that in the pure Cu with coarse grain. The denuded zone along grain boundaries was similar to that in the CuAl15. It also indicates that small grain size is essential for suppressing the void volume in copper alloys [6–8].

3.3. Influence of joining temperature

To evaluate the global void swelling, we convert the local void swelling into average void swelling. For a spherical grain of diameter; (d_g), the average swelling; $(\Delta V)_{av}$ will be [8]

$$(\Delta V)_{av} = (\Delta V)_1 (d_g - 2W_D/d_g)^3, \tag{1}$$

where W_D is the void-denuded zone width. In CuAl15, there was no influence of brazing on the microstructure. The average swelling volume of CuAl15 is less than 0.5% at 673 K, 30 dpa. On the other hand, in the CuAl25 joined by HIP at 1323 K, the mean grain size was 60 μm. Therefore, the grain boundary sink strength in the CuAl25 alloy was considerably lower than that in the CuAl15 alloy. Therefore, even though the CuAl25 contains more alumina particles than CuAl15, swelled 0.8% at 10 dpa and 3.7% at 30 dpa. It is suggested that the reduction of point defects which flow to grain boundary enhances the void swelling in the grain interior. Moreover, void swelling of CuAl25 was lower than that of pure Cu. This result suggests that alumina particles also contributes to the reduction in void swelling.

Since HIP process causes grain growth, it is necessary to optimize the temperature where the swelling resistance of material is not degraded and grain growth does not occur. Sato et al. [10] studied HIP procedure of CuAl25 and the SUS316L at temperature ranging from 1253 to 1323 K, and no big difference in the strength of the joined interface was detected. According to the results of SEM observations on joint's interface of the HIPed specimen of SUS316L and ODS-Cu at 1253, 1303 and 1323 K, grain growth occurred only in the sample joined at 1323 K and the grain size was twice of that in the material before joining [11]. It is necessary to optimize the temperature range where grain growth is not prominent.

4. Conclusions

Specimens of CuAl15, CuAl25, and pure copper irradiated with heavy ions were investigated using transmission electron microscopy. Results may be summarized as follows:

(1) Size and density of voids depend on grain size, number density of alumina particle and dislocation density, and are greatly influenced by the temperature of the HIPing process.
(2) Grain boundaries in ODS-Cu act as main sinks for vacancies, and suppresses void formation. Alumina particles have less effects on void swelling than grain boundaries in ODS-Cu. Alumina particles also play a role in suppressing void swelling indirectly by preventing grain growth above the recrystallization temperature of Cu.
(3) The HIP process at 1323 K causes grain growth in ODS-Cu and degrades its swelling resistance. The development of HIPing procedure which causes no grain growth is needed.

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