FORMATION OF VOIDS IN QUENCHED ALUMINUM DILUTE (Cu, Mg, Si) ALLOYS

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ABSTRACT Experimental study on the effects of solute atoms of copper, magnesium, and silicon on the formation of voids in aluminum dilute alloys is carried out by quenching method. Specimens were heated in helium and hydrogen gases. Hydrogen and carbon atoms are responsible for the nucleation of voids in pure aluminum. To eliminate the hydrogen effects on the formation of voids, we melted aluminum dilute alloys in a highly evacuated vacuum of 10^{-5} Pa. The melting can reduce the hydrogen atoms from alloys. It is found that both voids and faulted loops were observed in quenched Al-0.04at%Mg and Al-0.04at%Si specimens, while only faulted loops were observed in quenched Al-0.04at%Cu specimen by electron microscopy. These results enable one to infer that copper atom has an effect on suppression of nucleation of voids, while magnesium and silicon atoms have an effect on the nucleation of voids in quenched aluminum dilute alloys. A model of process of the formation of voids that were observed in quenched Al-Cu, Al-Mg and Al-Si alloys is discussed.

Keyword: *aluminum dilute alloy, quenching, void, faulted loop, transmission electron microscopy*

1. INTRODUCTON

When pure aluminum is quenched from an elevated temperature below the melting point into icy distilled water, supersaturated vacancies are frozen into the specimen and form aggregates of both faulted dislocation loops of Frank type (referred to hereafter as faulted loops) and voids [1]. Shimomura and Yoshida have found that voids form heterogeneously at vacancy clusters which trap hydrogen atoms [2]. We have recently found that hydrogen and carbon atoms are responsible for the nucleation of voids formation in pure aluminum, only faulted loops are formed in quenched pure aluminum in which dissolved residual gas atoms are removed [3].

Aluminum dilute alloys, for example Al-Cu, Al-Si alloys are used in nano device technology. There are many reports that the mean failure lifetime of aluminum-copper interconnects lines is longer than pure aluminum lines in interconnects of micro-electronic devices [4,5]. It is important to investigate the effects of copper and silicon atoms on the formation of voids. It is the purpose of the present work that investigate the effects of copper, silicon and magnesium atoms in aluminum dilute

alloys on the formation of voids systematically by quenching method. It is found that copper atoms suppress the formation of voids and silicon, magnesium atoms promote the formation of voids in aluminum dilute alloys.

2. Experimental Procedures

Aluminum dilute alloys which are Al-0.04at%Cu, Al-0.04at%Si and Al-0.04at%Mg were employed in this experiment. Due to expect an influence of impurity atoms in specimens , these specimens were melt on an alumina (Al_2O_3) boat in a vacuum of 10^{-5} Pa which is referred to as RM specimens. By melting, hydrogen and carbon atoms contained in the alloys were reduced because a hydrogen have an effect on the nucleation of voids in quenched pure aluminum. After melting, all specimens were rolled down to about 0.05 mm thickness foils in air and rinsed acetone, and annealed at 500℃ for 5 hours in the vacuum of 1×10^{-5} Pa.

Quenching experiments of all specimens was carried out from a high temperature 600°C or 550°C into distilled water of various temperatures [6] . Helium and hydrogen gases were used as a quenching atmosphere. The helium is the low solubility into alloys up to the melting point and hydrogen has an effect on the formation of void.

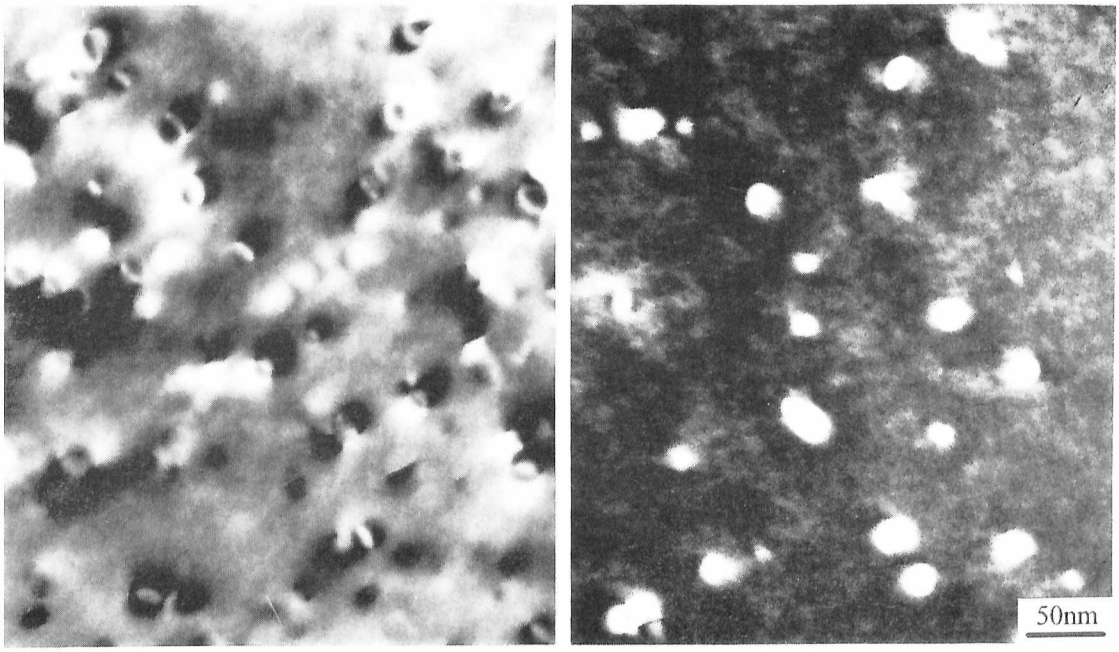
Quenched specimens were electron polished in liquid that consisted of 20% perchloric acid (60%) and 80% methanol at room temperature, The observation were carried out with 120 kV by a JEOL-2000EX transmission electron microscope. Both number density and size of voids and faulted loops were measured from TEM photographs, and the vacancy concentration is calculated with which measured number density and size of faulted loops and voids.

3. Results

To investigate the effects of copper, silicon and magnesium atoms on the formation of voids in aluminum dilute alloys, the specimens of Al-0.04at%Cu, Al-0.04at%Si and Al-0.04at%Mg were quenched from 600℃ or 550℃ to 0℃ and 30℃ in helium and hydrogen gases atmosphere, respectively.

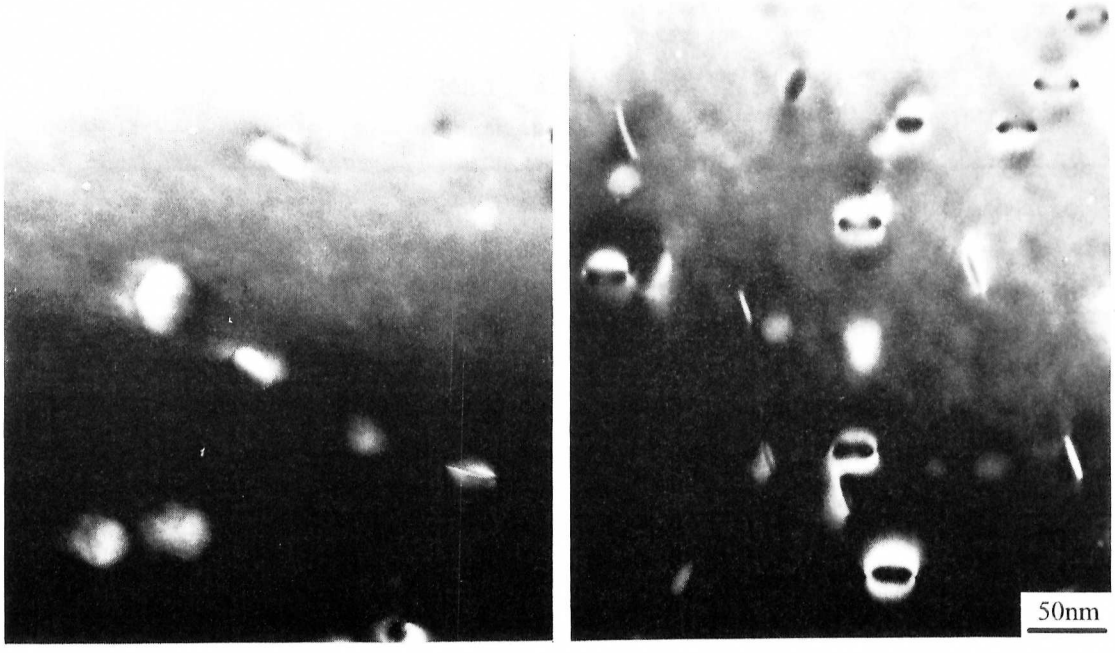
Fig. 1 show TEM photographs of vacancy aggregates which were observed in quenched Al-0.04at%Cu specimen in helium gas atmosphere as shown in Fig. 1 (a), and in hydrogen gas atmosphere as shown in Fig. 1 (b). The hexagons are faulted loops in TEM photographs. However, only faulted loops were observed in quenched Al-0.04at%Cu specimens.

Fig. 2 are TEM photographs of vacancy aggregates which were observed in quenched Al-0.04at%Si specimen as shown in (a), and Al-0.04at%Si(RM) specimen as shown in (b) from 600℃ to 30℃ in helium gas atmosphere. The hexagons are faulted loops, and very small squares are voids in TEM photographs. Both faulted loops and voids were observed in quenched Al-0.04at%Si specimen and Al-0.04at%Si(RM) specimen. In the same way, both faulted loops and voids were observed in quenched Al-0.04at%Mg specimen as shown in Fig. 3 (a), and Al-0.04at%Mg(RM) specimen as shown in Fig. 3 (b) from 600℃ to 30℃ in helium gas atmosphere. It is clear that silicon and magnesium atoms concerned in the formation of voids.



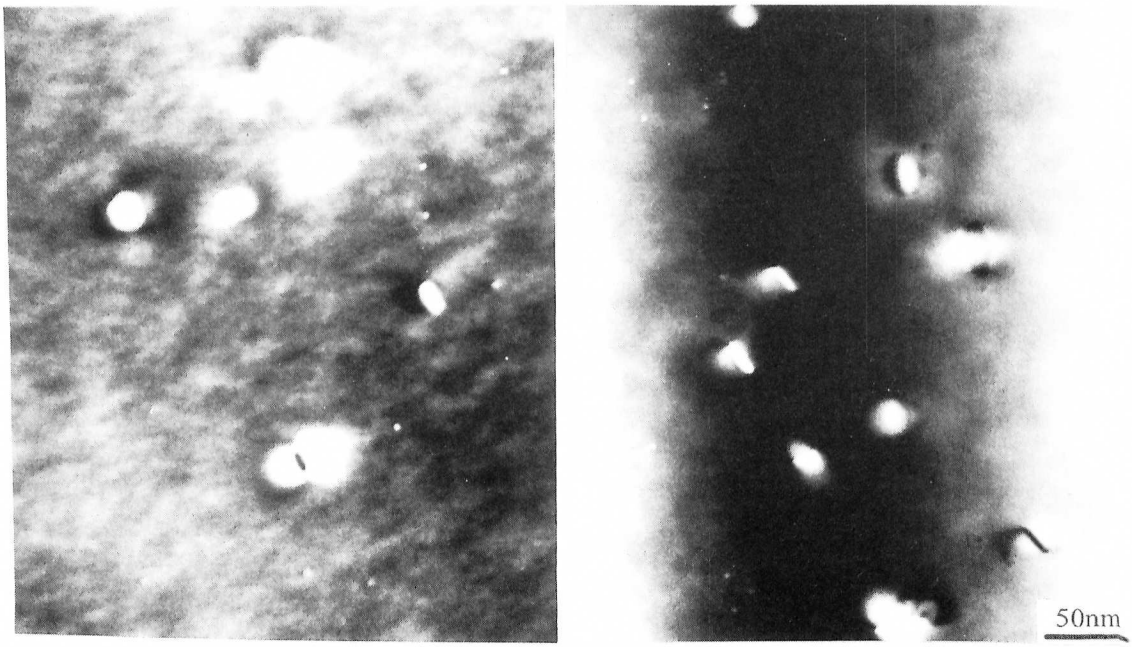
(a) (b)

FIG. 1 TEM photographs of only faulted loops were observed in quenched Al-0.04at%Cu from 600℃ to 30℃ in: (a) helium gas atmosphere, (b) hydrogen gas atmosphere.



(a) (b)

FIG. 2. TEM photographs of both faulted loops and voids which were observed in quenched: (a) Al-0.04at%Mg, (b) Al-0.04at%Mg(RM) from 600℃ to 30℃ in helium gas atmosphere.



(a) (b)

FIG. 3. TEM photographs of both faulted loops and voids which were observed in quenched: (a) Al-0.04at%Mg, (b) Al-0.04at%Mg(RM) from 600℃ to 30℃ in helium gas atmosphere.

Table. I Number of densities and average sizes of vacancy aggregates observed in quenched aluminum dilute (Cu,Si,Mg) specimens. All specimens were quenched form 600°C to 30°C in helium and hydrogen gases atmosphere. After melting in a vacuum of 10^{-5} Pa on an alumina boat, the specimens are marked as RM.

Specimens	T_q (°C)	Quenching atmosphere	Faulted loops		Voids		Calculated
			Density (cm^{-3})	size (nm)	Density (cm^{-3})	size (nm)	C_v
Al-0.04at%Cu	600	He	1.23×10^{15}	17	---	---	2.03×10^{-4}
	600	H_2	5.73×10^{14}	19	---	---	1.13×10^{-4}
Al-0.04at%Si	600	He	2.16×10^{14}	28	0.64×10^{14}	7.2	1.29×10^{-4}
	600	H_2	4.56×10^{13}	35	5.01×10^{14}	5.4	1.47×10^{-4}
Al-0.04at%Si (RM)	600	He	4.20×10^{14}	29	0.91×10^{14}	5.2	2.10×10^{-4}
	600	H_2	4.34×10^{14}	27	2.91×10^{14}	5.1	2.26×10^{-4}
Al-0.04at%Mg	600	He	9.23×10^{13}	84	2.32×10^{14}	7.0	4.76×10^{-4}
	600	H_2	5.33×10^{12}	64	1.33×10^{14}	7.1	0.84×10^{-4}
Al-0.04at%Mg (RM)	600	He	1.44×10^{13}	100	0.98×10^{14}	8.3	1.60×10^{-4}
	600	H_2	1.59×10^{13}	84	0.29×10^{14}	7.8	0.82×10^{-4}

Table 1 shows that number of densities and average sizes of vacancy aggregates observed in quenched Al-0.04at%Cu, Al-0.04at%Si and Al-0.04at%Mg specimens from 600°C to 30°C in helium and hydrogen gases atmosphere. Vacancies concentrations calculated from these data is smaller than the calculated equilibrium values [8].

4. Discussion

Impurity atoms studies by Y. Shimomura et al. reported that hydrogen atom are responsible for the formation of voids in quenched pure Aluminum [3,7]. Hydrogen atom seems to easily nucleate on the formation of voids in quenched pure aluminum. In this work, only faulted loops are observed in quenched Al-0.04at%Cu specimen in helium gas atmosphere as shown in Fig. 1(a), even if it is quenched in hydrogen atmosphere as shown Fig. 1(b). The results suggested that copper atom seems to suppress the action of hydrogen atom on the formation of voids in aluminum copper alloy. We guess that such copper atom can trap hydrogen atom, that hydrogen atom can not nucleate on the formation of void.

To investigate the effect of silicon and magnesium atoms on the formation of voids in aluminum dilute alloys. First, the Al-0.04at%Si and Al-0.04at%Mg specimens were quenched in helium gas atmosphere, respectively. Both faulted loops and voids were observed in quenched these specimens, as shown in Fig. 2(a), 3(a). Of course, both faulted loops and voids were observed in all Al-0.04at%Si, Al-0.04at%Mg specimens quenched in hydrogen atmosphere as shown in Table 1. By melting, hydrogen and carbon atoms were reduced to be rule out the effects on the formation of voids. However, both faulted loops and voids still were observed in quenched these specimens, as shown in Fig. 2(b), 3(b). It is clear that silicon and magnesium atoms are responsible for the formation of voids. These results suggested that silicon and magnesium atoms have an effect of promotion on the formation of voids in aluminum dilute alloys.

We have investigated that the effects of impurity atoms (in this case, hydrogen and carbon atoms) for the formation of voids, as the results it is suggested that either of hydrogen and carbon atoms have an effect on the nucleation of voids, in quenched pure aluminum as written in detail at Ref. 3. By this work, it may be thought that silicon and magnesium atoms have an effect as the same as hydrogen atom, which promoted the nucleation of voids in quenched Al-0.04at%Si, Al-0.04at%Mg specimens. When copper atom is existence, impurity atoms of hydrogen and carbon atoms lose the effect on the formation of voids in quenched Al-0.04at%Cu specimen. It seems that hydrogen and carbon atoms are unable to nucleate on the formation of voids because which be trapped by copper atom. This seems to may be explained that why copper atom has an effect and silicon, magnesium atoms have no effect on suppression of the formation of voids in quenched aluminum dilute alloys.

5. Conclusion

5.1 It is clear that silicon and magnesium atoms have an effect on the nucleation of voids in quenched aluminum dilute alloy as well as hydrogen and carbon atoms. By this result, it is better

that silicon and magnesium atoms are avoid in aluminum dilute alloy due to reduce the nucleation of voids.

5.2 Copper atom has an effect on suppressing the formation of voids in quenched aluminum dilute alloy. It may be thought that such hydrogen atom are trapped by copper atom, that voids can not nucleate. We suggest that the addition a very small quantity of copper atom into aluminum dilute alloy is efficient to suppress the formation of voids.

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