Hardening Behavior in Aged Al-4%Cu-0.3%Mg Alloys with 0.5 and 2%Ag Additions

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The precipitation characterization of Al-4 mass% Cu-0.3 mass% Mg alloy with additions of 0.5 and 2 mass% Ag during aging treatments was carried out by transmission electron microscopy (TEM) and microhardness measurements. TEM observation of aged samples showed that the Ω phase is the dominant phase and it is in coexistence with the θ' phase. Additionally, the Ω phase has the morphology of polygonal prism (almost hexagonal) with flat interfaces and a thin thickness. The variation of the cube of mean radius of precipitates, r^3 , followed a linear relationship, as predicted by the Lifshitz, Slyosov and Wagner (LSW) theory for diffusion-controlled coarsening in both alloys. The coarsening process of the Ω phase was slower in the aged Al-4 mass% Cu-0.3 mass% Mg-2 mass% Ag alloy. The decrease in hardness seems to be related to the coarsening of the Ω phase and the loss of coherency. [doi:10.2320/matertrans.M2009233]

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1. Introduction

Al-Cu-Mg alloys belong to the group of heat treatable alloys. The precipitation hardening of this type of alloys is caused not only from precipitates themselves, but also from the coherent elastic strain of matrix phase provided by coherent precipitates. These can act as obstacles to the motion of dislocations producing an increase in mechanical properties.^{1,2)} Therefore, the volume fraction, size, distribution, morphology, and composition of these precipitates are directly related to the mechanical properties.³⁾ Additionally, the precipitates must be thermodynamically stable and they need to be resistant to the coarsening process at the service temperature. This can be obtained if the precipitates are coherent and coplanar with the matrix phase in order to achieve microstructural stability.⁴⁾

Al-Cu-Mg alloys, such as 2124, 2219 or 2618 are currently used for applications where good specific strength and heat resistant capability up to 423 K are required.⁵⁾ Typical applications are structural components in supersonics aircraft and automobiles.⁵⁾ The additions of small amounts of other elements to these alloys modify the precipitation kinetic and improve its performance at high temperatures (≈ 473 K).⁶⁾ The addition of Ag to Al-Cu-Mg alloy system with a high Cu/Mg ratio increases significantly the mechanical properties and it decreases the effect of the overaging, material degradation.⁷⁾ The improvement of the mechanical properties is mainly attributed to the fine and uniform dispersion of thin hexagonal-shape plate-like precipitates, designated as the Ω phase, which precipitates uniformly on the $\{111\}$ planes of the matrix.⁸⁾ The Ω phase is semicoherent with the Al-rich matrix.⁹⁾ This phase has also shown to be relatively stable at high temperatures.¹⁰⁾ The crystalline structure of the Ω phase has been the subject of several studies. At first, it was proposed¹¹⁾ to have a chemical composition similar to that of the θ -Al₂Cu phase with a monoclinic structure. Later, it was described¹²⁾ as being hexagonal, although the most

widely accepted¹³⁾ structure of the Ω phase is orthorhombic with lattice parameters $a = 0.496$ nm, $b = 0.859$ nm and $c = 0.848$ nm. Teleshov *et al.*¹⁴⁾ reported that a high Cu : Mg ratio of about 16 with 0.6% Ag increased considerably the strength of artificially-aged pressed strips at room and high temperatures. Additionally, the size and aspect-ratio evolutions of the Ω phase have been only reported for the plate-shape precipitates (thickening) and the results showed an increase in size with aging time.¹⁵⁾ Besides, there is no report of the coarsening process of the Ω phase and its effect on the hardening behavior in this type of alloys. It is also analyzed the effect of an addition of about 2 mass% Ag on the precipitation behavior. A maximum addition of about 0.75 mass% Ag has been reported in Al-Cu-Mg alloys.⁸⁾ Thus, the purpose of this work is to study the effect of Ag additions (0.5 and 2%) on the aging behavior and growth kinetics of precipitation in the artificially-aged Al-4% Cu-0.3% Mg alloys.

2. Experimental Procedure

Al-4 mass% Cu-0.3 mass% Mg-X ($X = 0.5$ and 2 mass% Ag) alloys were prepared from pure elements keeping constant a Cu/Mg ratio of about 13 : 1 and melted using an electrical-resistance furnace in a silicon carbide crucible under an argon gas atmosphere. Chemical analysis of alloys was carried out using atomic absorption spectroscopy (Perkin Elmer AA400). Samples of $10 \times 10 \times 3$ mm were encapsulated in Pyrex tubes under an argon gas atmosphere and then homogenized at 798 K for 612 ks (7 days) and subsequently quenched in ice water. The homogenized and solution treated samples were aged at temperatures of 423 K, 473 K and 523 K for times from 600 s to 720 ks for both alloys. Vickers hardness (HV) measurements were determined with a FUTURE TECH equipment using 10 indentations for each sample and a load of 200 g. The aged samples were subsequently prepared by the twin-jet electropolishing

Table 1 Chemical composition of alloys.

Alloy	Cu	Mg	Ag	Al
Al-4 mass% Cu-0.3 mass% Mg-0.5 mass% Ag	3.78	0.32	0.55	Balance
Al-4 mass% Cu-0.3 mass% Mg-2 mass% Ag	3.87	0.31	2.31	Balance

Fig. 1 Aging curves of (a) 0.5Ag and (b) 2Ag alloys aged at different temperatures.

Fig. 2 BF-TEM micrograph and its electron diffraction pattern in the solution treated 2Ag alloy.

technique at 218 K with an electrolyte composed of 22 vol% nitric acid and 78 vol% methanol and observed with a conventional transmission electron microscope (TEM) JEOL-2000FXII at 200 kV, equipped with EDS analysis.

3. Results

The chemical compositions of Al-4 mass% Cu-0.3 mass% Mg-0.5 mass% Ag and Al-4 mass% Cu-0.3 mass%Mg-2 mass%Ag alloys are shown in Table 1. The chemical compositions are close to those corresponding to the nominal one. Thus, the prepared alloys were designated as 0.5Ag and 2Ag alloys.

3.1 Aging curves

Figures 1(a) and (b) show the aging curves for the 0.5Ag and 2Ag, respectively, alloys aged at 423, 473 and 523 K. It is observed a slower hardening kinetic in the aging of both alloys at 423 K than those at higher temperatures. The peak hardness was higher and achieved in shorter aging times

in the aged 2Ag alloy, Fig. 1(b). The faster precipitation kinetics can be attributed to its higher supersaturation and the higher peak hardness can be related to the higher volume fraction of precipitates corresponding to a higher supersaturation.¹⁶⁾ The highest hardness was determined to be about 145 and 155 HVN in the 0.5Ag and 2Ag alloys aged at 423 K for 720 ks and 90 ks, respectively. This hardness peak is higher than that of 110 HVN reported¹⁷⁾ in an Al-4 mass% Cu-0.3 mass% Mg alloy aged at 423 K. Additionally, a hardness peak of 140 VHN was determined in an Al-4 mass% Cu-0.3 mass% Mg-0.75 mass% Ag alloy aged at 453 K. Thus, the maximum peak hardness of present work is slightly higher than those reported in similar alloys. Nevertheless, it seems that an addition of Ag as high as 2 mass% is not effective to increase considerably the alloy hardness.

3.2 Precipitation characterization

Bright-field (BF) TEM micrograph and its corresponding selected-area electron diffraction (SAED) pattern are shown in Fig. 2 for the 2Ag alloy after solution treatment. This

Fig. 3 BF-TEM micrographs of the (a) 0.5Ag and (b) 2Ag alloy aged at 523 K for 90 and 180 ks.

Fig. 4 (a) and (b) DF-TEM images using $2/3(220)$ reflection of (c) electron diffraction pattern of the Ω phase in the 0.5Ag alloy aged at 473 K for 36 ks.

micrograph presents the absence of any precipitates and corresponds to a supersaturated solid solution α_{ss} , where some dislocations are only present. The electron diffraction pattern corresponds to the $[011]$ zone axis of the α matrix. A similar behavior was observed to occur in the other alloy composition.

BF-TEM micrographs for the 0.5Ag and 2Ag alloys aged at 523 K for 90 and 180 ks, respectively, are shown in Figs. 3(a) and (b), respectively. It can be observed the coexistence of θ' and Ω phases and the Ω phase is the dominant one. It can be seen that the distribution of the Ω phase is uniform for both samples. The volume fraction of Ω precipitates in 2Ag alloy is higher than that in the 0.5Ag alloy. The Ω phase was observed in 2D with two shapes. One is plate-shaped with polygonal outlines and the other one corresponds to laths. The θ' phase also shows a lath-shape morphology, but the θ' precipitates are wider than the Ω precipitates. On the other hand, the broad faces of the plate of the Ω phase are aligned in the $\langle 111 \rangle$ matrix directions, while the broad faces of the plate of the θ' phase

are aligned in the $\langle 100 \rangle$ matrix directions, see Fig. 3. This alignment has been attributed to a rapid clustering of Mg and Ag atoms which occur during, or immediately after quenching facilitating the nucleation of Ω on the $\{111\}_{\alpha}$ planes.^{1,2)}

Figures 4(a)–(c) shows the dark-field (DF) TEM micrographs and electron diffraction pattern of the 0.5Ag alloy aged at 473 K for 36 ks. This figure confirms that the precipitates of the Ω phase have a polygonal morphology with flat interfaces, Fig. 4(a) and lath-shape, Fig. 4(b). The lath-shape is observed at the radial direction of the Ω phase and the polygonal morphology at the axial direction.

3.3 Coarsening behavior of the Ω phase

DF-TEM observation of the coarsening evolution of the Ω phase during aging is shown in Figs. 5 and 6(a)–(d) for both alloys aged at 523 K. It can be noticed a gradual increase in size of precipitates with aging time. Additionally, it can be observed that the volume fraction of precipitates decreases with aging time. The polygonal morphology with flat

Fig. 5 DF-TEM micrographs of 0.5Ag alloy aged at 523 K for (a) 3600 s, (b) 18, (c) 36 and (d) 90 ks.

Fig. 6 DF-TEM micrographs of 2Ag alloy aged at 523 K for (a) 3600 s, (b) 18, (c) 90 and (d) 180 ks.

interfaces is maintained for 18 ks of aging in both alloys. Further aging causes the flat interfaces become rounded (see arrows in Figs. 5(d) and 6(d)). This behavior is an indicative of the coherency loss between the Ω precipitates and the

Fig. 7 Plot of r^3 vs. t for 0.5Ag and 2Ag alloy aged at 473 and 523 K.

matrix. This fact seems to be related to the decrease in hardness after aging at 523 K for 18 ks. The loss of coherency took place more rapidly in the aged 0.5Ag alloy because of the faster diffusive process related to the lower solute content.¹⁸⁾

The volume of precipitates was calculated from the average area of the Ω precipitates and the thickness of the thin face. Then, the mean radius of precipitates was determined in both alloys. Figure 7 shows the plot of the cube of mean radius, expressed as $r^3 - r_0^3$, for the Ω precipitates against the aging time, t . The points of this plot give a good fit to a straight line which means that the coarsening of precipitates follows the LWS theory for diffusion controlled growth,⁹⁾ that is, the coarsening process can be described by the following equation:

$$r^3 - r_0^3 = kt \quad (1)$$

The rate constant k can be obtained from the slope of straight lines in Fig. 7. These values are also shown in Fig. 7. The values of rate constant k suggests that the coarsening process is slower in the aged 2Ag alloy than that corresponding to the other composition alloy. This behavior seems to be related to the slower diffusion process for this alloy composition due to its higher content of solutes.¹⁸⁾

4. Conclusions

A study of the precipitation process was carried out in the aged 0.5Ag and 2Ag alloys and the conclusions are summarized as follows:

- (1) The precipitation kinetics process was slower in the aged 2Ag alloy which permitted to obtain a higher hardness.
- (2) The coarsening process was slower in the aged 2Ag alloy which permitted to keep its hardness for longer aging times.
- (3) The Ag addition up to 2 mass% to the Al-4 mass% Cu-0.3 mass% Mg alloy caused a slightly increase in hardness, but increase considerably its coarsening resistance in the aged alloy.

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