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Effects of Isothermal Heat Treatment on Microstructural Evolution of Semisolid Al-4Cu-Mg Alloy

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The authors investigated the effects of the isothermal heat-treatment conditions on the microstructural evolution and composition distribution of semisolid Al-4Cu-Mg alloy during isothermal heat treatment. The experimental results show that the microstructural evolution and composition distribution of semisolid Al-4Cu-Mg alloy are controlled by atom diffusion during the isothermal heat treatment process. Grain growth and spheroidization were promoted with the increase of the isothermal temperature and/or the holding time. Moreover, the higher the isothermal temperature, or the longer the holding time, the more segregation constituent elements occurred to the grain boundaries. The low melting structure at grain boundary is greatly affected by Cu. The microstructural evolution in the isothermal heat-treatment process is as follows: recovery, recrystallization, fragmentation, spheroidization, and coarsening. Such fragmentation, spheroidization, and grain growth of coalescence and Ostwald ripening are involved as main mechanisms in the isothermal heat-treatment process.

Keywords aluminum alloy, isothermal heat treatment, mechanism, microstructural evolution

1. Introduction

Among many materials technologies, semisolid metal forming (SSM) is one of the most important technologies in this century for manufacturing near-net-shape components. SSM offers significant processing advantages, such as good surface finish, low energy consumption, and high operating efficiency improvement.[1-3]

Compared with conventional casting and forging technologies, the key feature of SSM is the spheroidal nondendritic microstructure of the semisolid material.[4] A variety of fabrication techniques can help produce semisolid microstructures, including mechanical stirring (MS); electromagnetic stirring (ES); spray deposition (SD); strain-induced, melt-activated (SIMA); and recrystallization and partial melting (RAP). The last two methods are effectively used to obtain desirable microstructures for wrought alloys. These methods involve cold plastic deformation of an alloy to some critical reduction point, followed by heating to a temperature above the solidus of the metal.[5] Therefore, isothermal heat treatment is a most important aspect in controlling the semisolid microstructures in these processes.

At present, studies related to the isothermal heat treatment of alloys for producing the semisolid microstructures have been completed as follows. Kapranos et al.[4] carried out a series of heat-treatment experiments to establish relationships between the fraction of liquid with temperature and grain size, with holding time at a given temperature. Lapkowski[5] studied the influence of process parameters on the resulting microstructure and its applicability to thixoforming. Turkeli et al.[6] investigated the formation of nondendritic structures of 7075 wrought aluminum alloy. They examined the microstructures after heat treatment at different temperatures in the semisolid range. In this case, however, there was no comprehensive discussion about how the microstructure and composition distribution of the alloy evolved during isothermal heat treatment.

In this paper, the effects of isothermal heat-treatment conditions of Al-4Cu-Mg alloy, including isothermal temperature and holding time on the microstructural evolution and distribution of main elements, were investigated. The main mechanisms of microstructural evolution are also discussed.

2. Experimental Procedures

The material used in these experiments was Al-4.10Cu-0.64Mg-0.54Mn-0.37Fe-0.34Si-0.10Zn-0.019Ti, which was hot extruded, solid-solution heat treated, and naturally aged. The semisolid temperature range of this alloy was measured by differential thermal analysis (DTA) and was found to be 514-642 °C.

Cylindrical samples, 15 mm in diameter by 25 mm in height, were compressed to a height reduction of 30% prior to isothermal heat treatment. Heat treatment was carried out in a resistance furnace at temperatures between 580 and 620 °C, for holding times between 2 and 20 min. At the conclusion of the heat-treatment cycle, the samples were quenched in water. The holding time was measured from the moment a sample reached the desired temperature. Cross sections of the samples were cut, polished, and etched in a mixture of HF, HCl, and HNO_3. Subsequently, the quantitative measurements of microstructure were carried out on a Leica LABOR- LUX12MFS/ST (Leica Microsytems AG, Wetzlar, Germany) microscope. The distributions of the major elements were investigated using an AMRAY-1000B (Amray Inc., Bedford, MA) scanning electron microscope (SEM) and a FINDER-1000 (KYKY Technology Development Ltd., Beijing, P.R. China) x-ray energy spectrometer.

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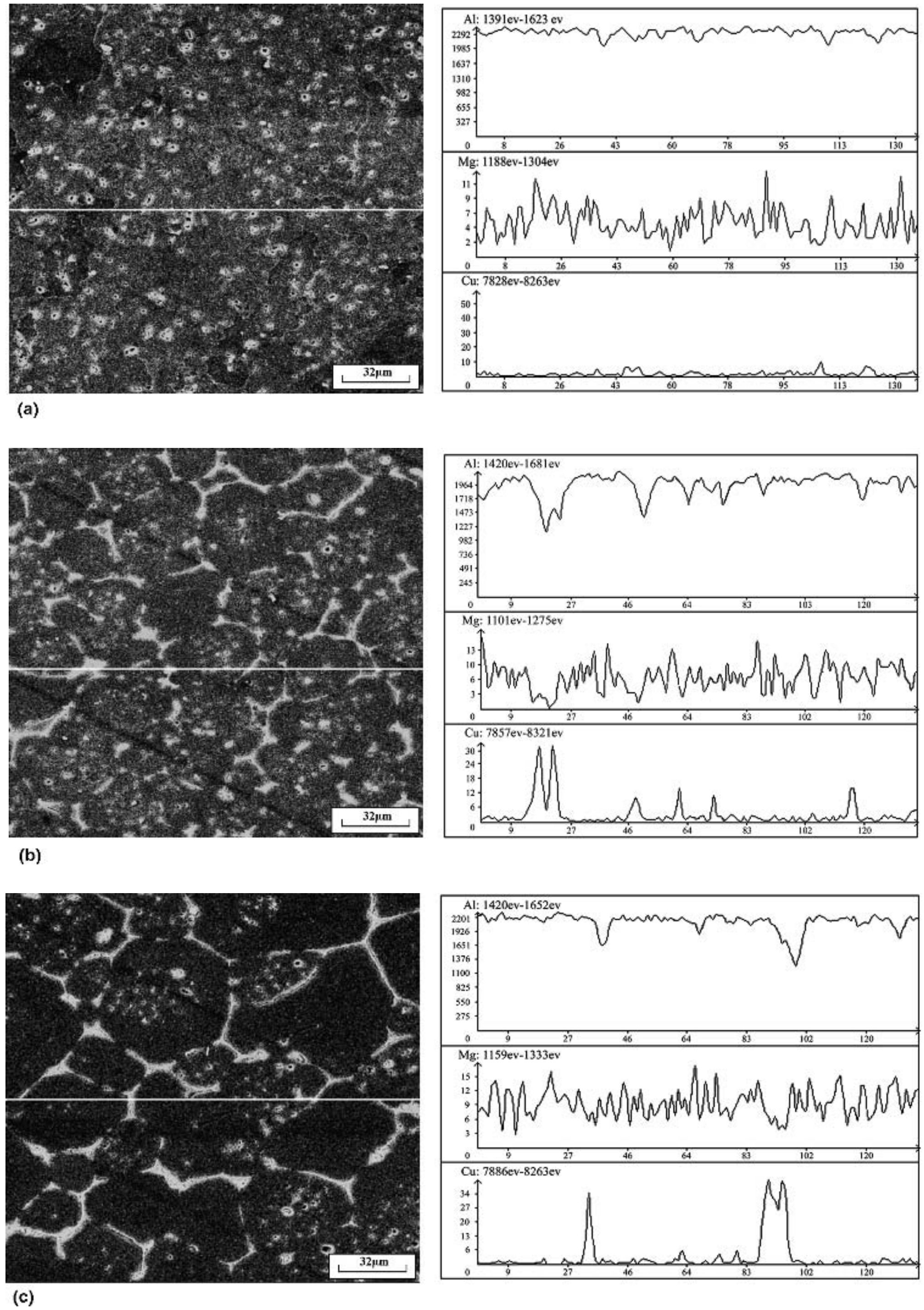
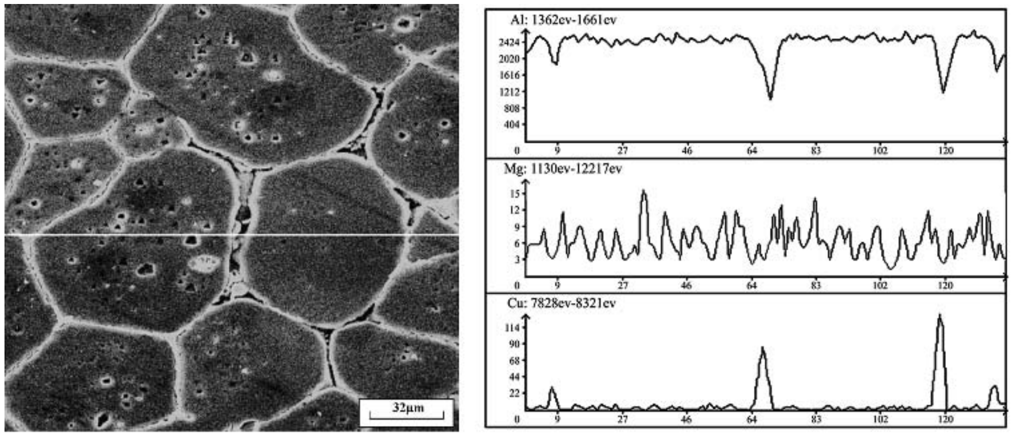


Fig. 1 SEM micrograph and line scans for the samples at various isothermal heat-treatment conditions (magnification 900×): **(a)** The origin Al-4Cu-Mg alloy, **(b)** 580 °C, 5 min, **(c)** 600 °C, 5 min

3. Results and Discussion

3.1 Effect of Isothermal Heat Treatment on Microstructural Evolution and Composition Distribution

Figure 1 shows SEM micrographs and chemical composition line scans of the distribution of the major elements in the alloy after isothermal heat treatment at different temperatures and holding times. Cu and Mg are the major alloying elements in the Al-4Cu-Mg alloy and exist in the form of θ ($CuAl_2$) and S (Al_2CuMg) precipitates. From the SEM micrograph in Fig. 1(a), it can be seen that the white Cu-rich particles are dispersed homogeneously in the Al matrix.



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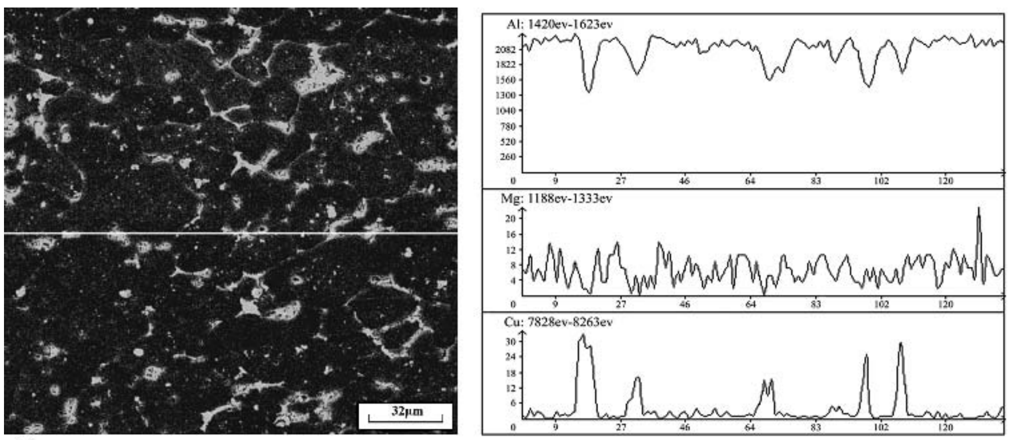

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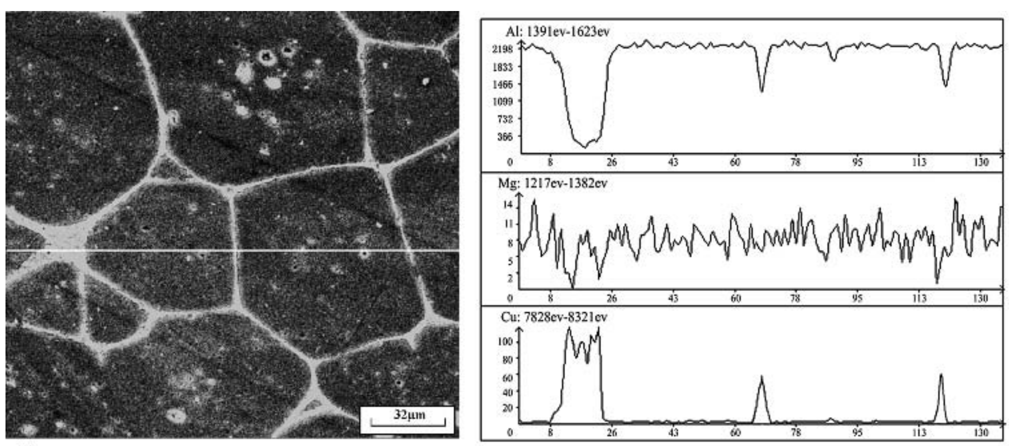

(f)

Fig. 1 cont. SEM micrograph and line scans for the samples at various isothermal heat-treatment conditions (magnification 900×):**(d)** 620 °C, 5 min, **(e)** 600 °C, 2 min, **(f)** 600 °C, 20 min

During isothermal heat treatment in the semisolid temperature range, the diffusion of atomic species is closely associated with the isothermal temperature. For the samples held for 5 min at 580, 600, and 620 °C, the distribution of Al, Mg, and Cu changed in different ways (illustrated in Fig. 1b-d). With an increase of the isothermal temperature, the Cu segregation increases to the grain boundaries, and the number of Cu-rich particles decreases in the grain interior. The reason for this situation is that the differential diffusion velocities for Cu and Al atoms in the intragranular and intergranular regions of grains are enhanced by the increase in the isothermal heat-treatment temperature. Therefore, few Cu-rich particles can be found in the grains in Fig. 1(d), due to the almost complete diffusion to the boundaries at 620 °C. According to the line-

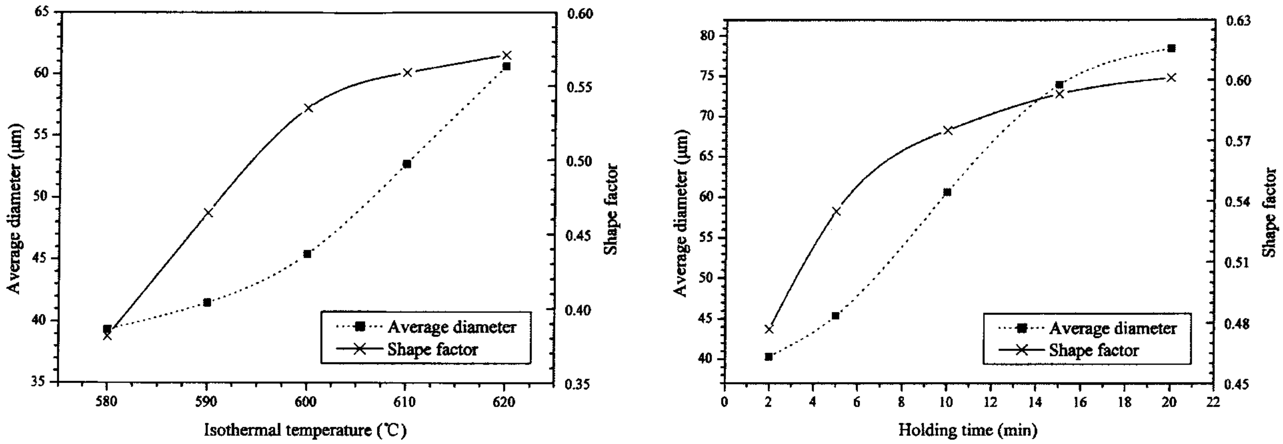

Fig. 2 Effects of isothermal heat-treatment conditions on the average grain size and the shape factor: **(a)** isothermal temperature and **(b)** holding time

scan analyses for Mg in Fig. 1(b-d), the Mg atoms were uniformly distributed throughout the matrix, and this situation is almost unaffected by the isothermal heat treatments.

The distributions of the major elements affected by the holding time at 600 °C are shown in Fig. 1(c) and (e and f). It was observed that the longer the holding time, the more segregation of Cu to the grain boundaries. After isothermal heat treatment at 600 °C for 20 min, the Cu atoms have so seriously segregated to the grain boundaries that some pools of low melting point eutectic have formed. In this case, only a few Cu-rich particles are dispersed in the Al matrix. The effect of different isothermal heat-treatment conditions on the distribution of Mg is negligible.

One of the unique features for the semisolid Al-4Cu-Mg alloy is the spheroidal microstructure, as shown in Fig. 1. After the isothermal heat treatment at 580 °C for 2 min, a fine spheroidal grain structure appears in the deformed sample, but the grain shape is irregular (Fig. 1b). As the isothermal temperature increases, the grain boundary gradually becomes thick and rounded, corresponding to an increase in the liquid fraction at the grain boundaries. The grains have spheroidized extensively; they are uniformly wetted by the thick liquid film when heat treated at 620 °C (Fig. 1d). The effect of holding time on the shape and size of the grains is similar to that found by changing the isothermal temperature. Figure 1(f) shows the microstructure of the deformed sample after 20 min at 600 °C. The coarsened microstructure comprises low-curvature solid interfaces in contact with a matrix of interconnected liquid.

The variations of the average diameter and shape factor of grains with isothermal temperature are shown in Fig. 2(a). The average diameter and shape factor were measured as 39 μm and 0.38, respectively, for the sample held at 580 °C for 5 min. The shape factor is defined as $4\pi A/P^2$, where P is the average perimeter and A is the average area of the grains. This shape factor varies between zero, for grains having very elongated structure in one dimension, and one, for grains having a circular cross section.[7] With an increase of the isothermal temperature, the velocity of grain growth initially increases slowly, but then speeds up rapidly. However, the shape factor of the grains initially increases rapidly with the increase in isothermal temperature, but then slows down. The average diameter and shape factor are 61 μm and 0.57 for the microstructure after isothermal heat treatment at 620 °C for 5 min. Figure 2(b) plots the average diameter and shape factor as a function of holding time. The velocity of grain growth increases rapidly with the increase in holding time, up to 15 min. From that point (Fig. 2b), the variation of shape factor with time is similar to that observed in Fig. 2(a). In the case of a holding time in excess of 10 min at 600 °C, the shape factor value increases slowly because most grains are already spheroidized.

3.2 Main Mechanisms of Microstructural Evolution

After being compressed by a height reduction of 30%, the Al-4Cu-Mg samples have stored considerable deformation energy related to crystal defects such as vacancies and dislocations. During isothermal heat treatment in the semisolid temperature range, static recovery takes place first, leading to subgrain formation from gliding and climbing dislocation in the intragranular region of grains. Recrystallization then proceeds within the subgrains as a result of the lowering of the stored energy from deformation. Accompanying the recovery and recrystallization processes, crystal defects and low melting point elements move to the high-angle boundaries. With an increase of the isothermal temperature above the solidus temperature, the low melting eutectic phases at the high-angle boundaries melt and penetrate the region between the high-angle boundaries, causing further fragmentation to the new grains.

The evolution of grain shape is essentially a diffusion-controlled process that occurs during isothermal heat treatment. The new grains grow and spheroidize as a result of the migration of the grain boundaries under the driving force that comes from the decrease in the interfacial free energy. The spheroidal grains with a large surface area will grow to reduce the energy in the liquid-solid system if the atoms have the ability to diffuse. The semisolid microstructures generally possess irregular boundary shapes with different curvatures. According to solidication thermodynamics, the equilibrium melting point is connected with interface energy. The smaller the curvature radius is, the lower the equilibrium melting point.[8] Therefore, when the sample is heated above the solidus temperature, the concave parts of the spheroidal grains, encircled by the liquid at the boundary, are easier to melt than the convex areas. In addition, Al atoms concentrate preferentially in the convex areas of the grains, from which the Cu atoms discharge to deposit at grain boundaries at the same time.

It can be seen clearly from Fig. 1(e) that eutectic liquid is not able to wet all grain boundaries during the initial stage of the isothermal heat treatment. The main mechanism of grain growth is coalescence, where several grains congregate to form a large one. When the holding time is 5 min, the eutectic liquid almost wets all grain boundaries, and the coarse grains evolve (Fig. 1c). Moreover, the longer the holding time, the larger the coarse grains are. The main mechanism of grain growth is Ostwald ripening, which is a competitive process whereby large grains grow as the small ones dissolve. There are several diffusion paths for Ostwald ripening, such as the energy difference across a curved interface, volume diffusion, grain boundary diffusion, and dislocation diffusion.[9,10] That is to say, grain coarsening is controlled by these different diffusion paths at the latter stage of the isothermal heat treatment. Therefore, the long holding time at a given isothermal temperature will lead to grain coarsening, but the effect is minimal for holding time up to 15 min (Fig. 2b). Considering the distribution of Cu and Mg, it is possible that the particle-inhibited grain coarsening is not dominant for the semisolid Al-4Cu-Mg alloy.

4. Conclusions

Microstructural evolution and composition distribution in the semisolid Al-4Cu-Mg alloy are controlled by atom diffusion during isothermal heat treatment in the liquid-solid temperature range. Grain growth and spheroidization are promoted with increasing isothermal temperature and/or holding time. Moreover, the higher the isothermal heat-treatment temperature, or the longer the holding time, the more opportunity for grain-boundary segregation of constituent elements. The low melting eutectic at the grain boundary is greatly affected by Cu concentration.

During isothermal heat treatment in the semisolid temperature range, the process of microstructural evolution in the deformed sample proceeds in the following sequence: recovery, recrystallization, fragmentation, spheroidization, and coarsening. The main mechanisms involved during isothermal heat treatment of this alloy are fragmentation, spheroidization, and grain growth, leading to coalescence and Ostwald ripening.

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