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Structure evolution of AlSi6.5Cu2.8Mg alloy in semi-solid remelting processing

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The transformation from solid to liquid and the structure evolution of primary α crystals of AlSi6.5Cu2.8Mg alloy were investigated in semi-solid remelting condition. Results show that the structure evolution of the alloy consists of three stages: liquid increasing, composition change of liquid from eutectic to hypoeutectic, and uniformity of hypoeutectic liquid. The α crystals grow in forms of the coalescence and the collision-amalgamation in the first and second stages and in the form of Ostwald ripening in the third stage. It was found from statistical analysis that 1) the power exponent in the parabolic function relation between α crystal diameter and remelting time changes with remelting temperatures, 2) the exponents with respect to remelting temperatures of 573°C, 578°C and 584°C are 4.004, 2.38 and 1.84, respectively, and 3) convection intensity in the semi-solid melt is a main factor to influence these exponents. It is found in this investigation that there is another mechanism of coalescence growth, which is responsible for the high exponent at low temperature. And at high temperature, the minimum value of the exponent is 1 (due to) Ostwald ripening growth. © *2001 Kluwer Academic Publishers*

1. Introduction

After nearly 20 years investigation and development, thixoforming is successfully used in products, specially, in automobile and aircraft and spacecraft parts [1–3]. The process of this technique consists of: 1) preparation of semi-solid rheological slurry into ingot; 2) remelting the ingot to obtain thixotropical slurry by controlling remelting temperature between solidus and liquidus and holding time; and 3) casting or forging the slurry into a component under pressure.

It is very important to accurately control the remelting temperature and holding time to obtain proper thixotropy properties for a specific alloy owing to its specific remelting characteristics of solid-liquid transforming and structure evolution of α crystals. In recent years, structure evolution of AlSi7Mg alloy was extensively investigated [4–6]. In the present work, attention was focused on experimental investigation of structure evolution of AlSi6.5Cu2.8Mg alloy during solidification and the effects of some factors on the evolution were studied as well.

2. Experiment description

2.1. Materials for remelting

The compositions of AlSi6.5Cu2.8Mg alloy in weight percentage are 6.5% for Si, 2.8% for Cu, and 0.2% for Mg, balanced with Al. The liquidus and solidus of the alloy are 590°C and 567°C, respectively, by theoretical *calculation* [7]. The alloy was melted at 640°C and mechanically stirred when temperature dropped to 600°C and stirred continuously to 580°C using a ϕ20 mm diameter agitator. Then the melt was heated back to 590°C at a rate of 0.5°C/min under stirring condition. Rotating rate of the stirring was 900 r/min and a shear rate in the melt was 105 S^{-1}. The melt was poured into a ϕ50 mm diameter graphite mould to get ingots for thixotropy remelting after 60 minutes stirring at 590°C.

2.2. Semi-solid remelting procedure

During semi-solid remelting, the ingot was heated in a resistance furnace controlled by a KSY silicon rectifier, with which the variation of temperature was controlled within ±1°C. Remelting temperatures were set to 570°C, 573°C, 578°C and 584°C, respectively, which correspond to solid fractions (fs) of 87%, 74%, 52% and 26% by theoretical calculation [7]. Raw materials cut off from the ingot were individually heated at the selected temperatures for 15 min, 30 min, 45 min, 60 min, 75 min, 120 min and 240 min, and then quenched in water for semisolid structure observation.

2.3. Structure observation and α crystal size measurement

Metallographic specimens were made from the quenched ingots and their structures were observed with an optical microscope. Mean diameters of spherical α crystals in the specimens were measured with this microscope.

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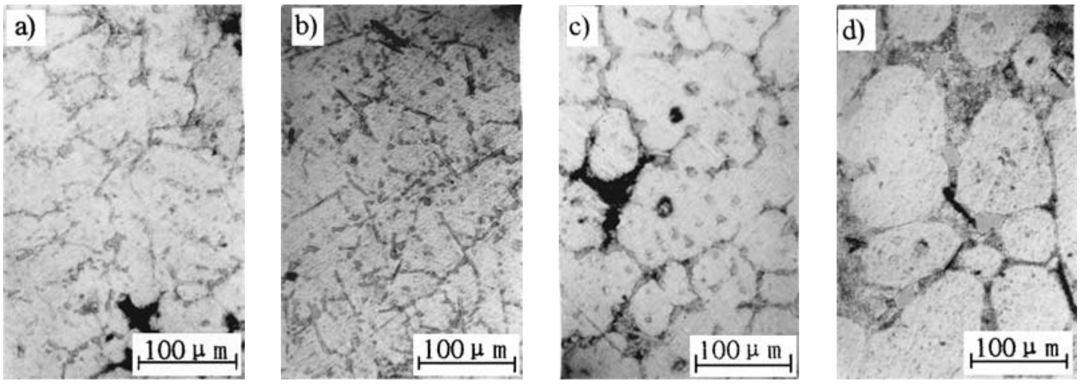

Figure 1 Micrographs showing transformation from solid to liquid remelt at 570°C for (a) 15 min, (b) 30 min, (c) 60 min, and (d) 180 min holding.

3. Results

3.1. Transformation procedure from solid to liquid

Fig. 1 shows the transformation from solid to liquid of the alloy when heated at 570°C. It was observed that the alloy starts to melt with eutectic fusion. Firstly, pin-shaped eutectic liquid regions appear at boundaries of grains in the specimen remelted for 15 minutes (Fig. 1a). Secondly, fiber-like eutectic liquid regions are present in the specimen remelted for 30 minutes (Fig. 1b). Thirdly, a liquid network is developed, some primary α crystals start to form individually and some liquid spots appear in the matrix of the crystal grains in the specimen when heated for 60 minutes (Fig. 1c). And finally, almost all eutectics melt and granular grains were isolated when heated for 180 minutes.

3.2. Structure transformation in the solid-liquid co-existing condition

Three stages of structure transformation were observed during evolution of α crystals in the specimens remelted at 573°C, 578°C and 574°C, respectively, and held for different durations.

1) The first stage is from the start of eutectic fusion to the phase constitution reaching the nominal liquid fraction. The main characteristics of this stage are the increase of liquid fraction, fusion of small α crystals, and the growth of α grains via coalescence of neighboring α grains.

2) The second stage starts from the moment when the specimen has approached the realization of equilibrium hypoeutectic composition of liquid. In this stage, the evolution procedure of collision-amalgamation-growth was observed because of a capability of the α crystals to shift and rotate freely.

3) The third stage takes place post realizing the nominal equilibrium composition of liquid. This stage is characterized by 1) the decrease of the growth rate of α crystals, 2) no amalgamation of spherical α crystals, and 3) fine α crystals adjoining to and surrounding to large α crystals in liquid. The later observation means that the liquid composition near the surface of the large α crystals has reached hypoeutectic.

The structure evolution of the specimens remelted at 573°C is shown in Fig. 2. Fig.2a and b show structure features in specimens remelted for 15 and 45 minutes holding during the first stage of the evolution. As the figures indicate, smaller α crystals disappeared via melting while some α crystals coalesce to form large crystals, and the quantity of liquid increased. Fig. 2c and d show the structure features in specimens for 75 and 120 minutes holding during the second stage. The phenomena of amalgamation-coalescence-growth and

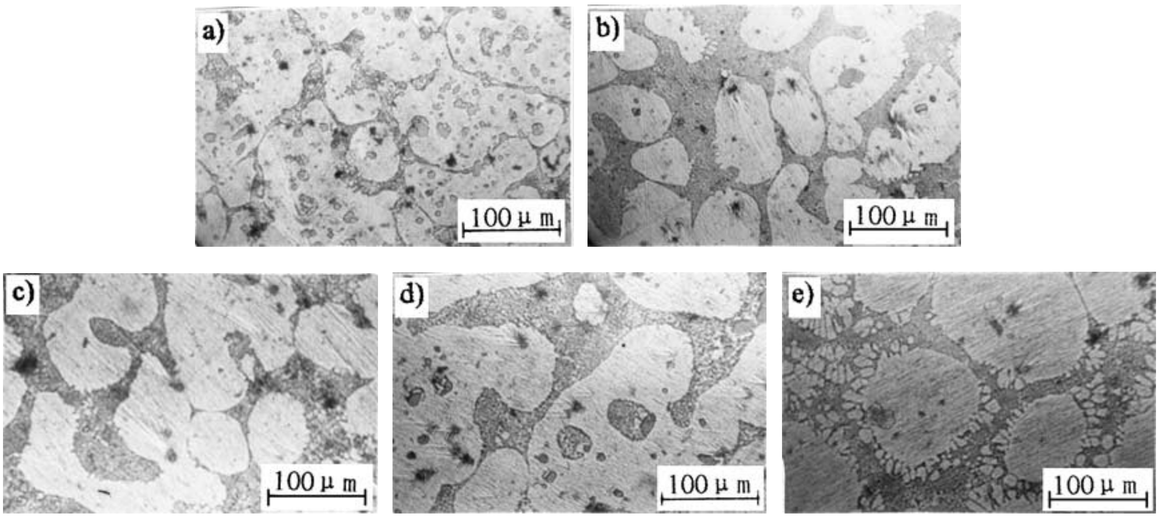

Figure 2 Structure evolution of the specimen held for (a) 15 min, (b) 45 min, (c) 75 min, (d) 120 min, and (e) 240 min at 573°C.

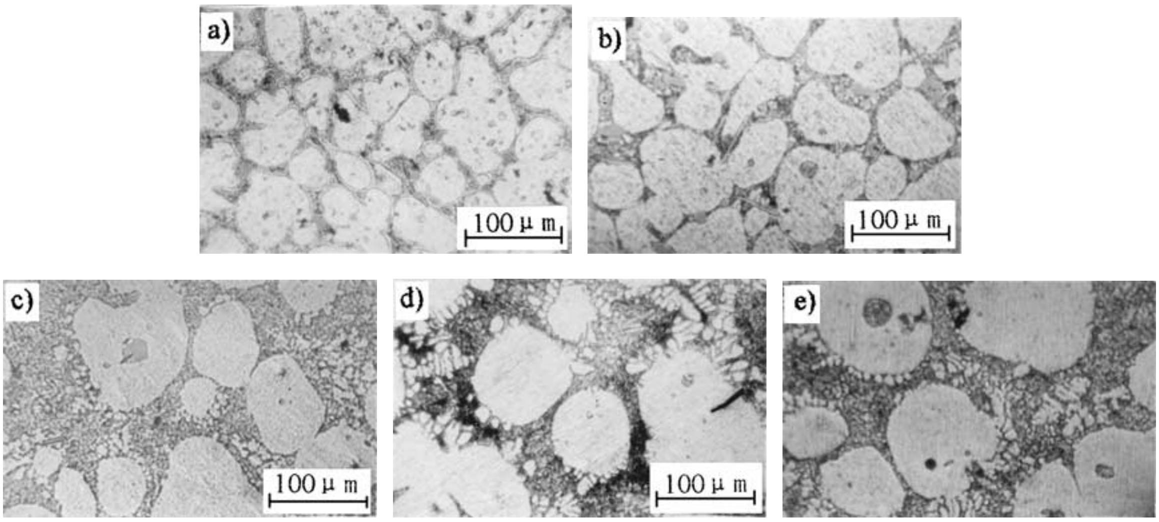

Figure 3 Structure evolution in specimens subjected to (a) 15 min, (b) 30 min, (c) 60 min, (d) 120 min, and (e) 180 min at 578°C.

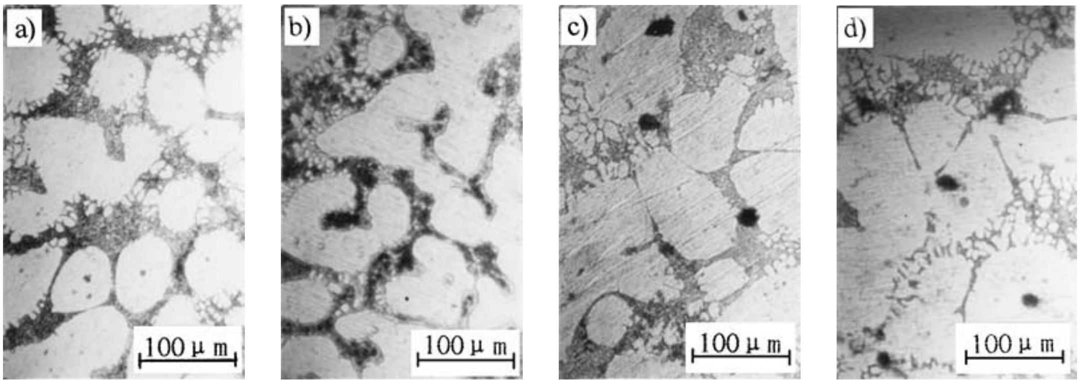

Figure 4 Structure evolution of the specimens remelt at 584°C for (a) 15 min, (b) 45 min, (c) 75 min, and (d) 120 min holding.

approaching the equilibrium quantity of liquid are obvious. Fig. 2e shows the appearance of spherical α crystals in the specimen for 240 minutes holding during the third stage, in which very fine α crystals exist in liquid and adjoin to the surface of the large α crystals and growth rate of the large α crystals is very low.

The structure evolution in specimens remelted at 578°C is shown in Fig. 3. Evolution process at this temperature is as same as that at 573°C with an only difference of an acceleration of every stage. Fig. 3a and b show the structure evolution in the first stage in the specimens subjected to 15 min and 30 min holding. Fig. 3c presents the structure evolution in the second stage in the specimen subjected to 60 min holding and Fig. 3d and e present the evolution in the third stage after 120 min and 180 min holding.

A structure evolution in the specimens remelted at 584°C is shown in Fig 4. Fig. 4a presents the evolution in the first stage for 15 min holding, Fig 4b presents the second stage for 45 min holding and Figure 4c and d do the third stage for 75 min and 120 min holding. More attention should be paid to the fact that the liquid composition of the alloy reaches hypoeutectic so early because of high diffusion rate of atoms of Al, Cu, Mg at the elevated temperature such that during only 15 min holding, many fine α crystals in the liquid adjoin large α crystals. All three stages proceeded faster at this heating temperature. The structure evolution stages of the specimens at different remelting temperatures for different holding times are listed in Table I. As the table indicates, every stage is obviously accelerated with raised heating temperature.

TABLE I Structure evolution in the specimens subjected to variable holding at different temperatures

Holding time (minute)	15	30	45	60	75	120	240
573°C melting	I		I		II	II	III
578°C melting	I	I		II		III	III
584°C melting	I		II		III	III	

3.3. Variations of diameters of α crystals

Relationships between mean diameter (d) of α crystals and holding time (t) of specimens remelted at 573°C, 578°C and 584°C are presented in Fig. 5. As the figure shows, the variation curves of diameters are parabolic, which means higher growth rates of α crystals in the early stage of remelting and lower growth rates with increased holding time, and the growth rate increases sharply when remelting temperature was raised. The relationships between α crystal size and holding time at different temperatures are obtained by non-linear regression analysis of the experimental data as follow [8]:

$$d^{4.004} - d_0^{4.004} = 6.583t \quad \text{(for 573°C)} \tag{1}$$

$$d^{2.38} - d_0^{2.38} = 1.480t \quad \text{(for 578°C)} \tag{2}$$

$$d^{1.84} - d_0^{1.84} = 1.640t \quad \text{(for 584°C)} \tag{3}$$

where, d_0 is the mean diameter when $t = 0$.

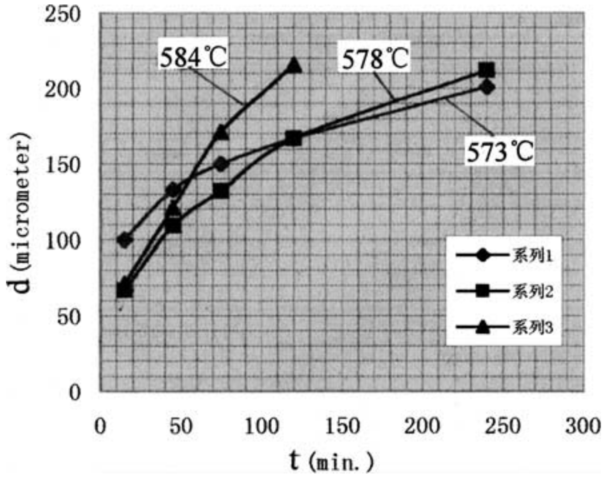

Figure 5 Relationship between mean diameter d and holding time t at 573°C, 578°C and 584°C temperatures.

The correlation coefficient (r) takes the values of 0.9853, 0.9906 and 0.9939 for Equations 1, 2, and 3 at the temperatures of 573°C, 578°C and 584°C, respectively.

4. Discussion

4.1. Structure evolution

In the first stage of the evolution, the quantity of liquid increases because of a high diffusion rate of Si, Cu and Mg atoms from liquid to solid caused by the difference of the concentrations in liquid and α crystal. With the uniformity of the alloying elements by the diffusion, the melting point of the boundary of an α crystal will decrease and a fusion of its surface will occur. Meanwhile, the fine α grains melt more quickly than large ones because their melting point is lower than that of the coarse α crystals owing to their high curvatures. On the other hand, adjoined α grains that have matched lattice orientation will coalesce to coarsen under the driving force of system energy decrease, specially the decrease of surface energy during coarsening.

In the second stage, the diffusion controlled fusion and the melting of small α grains terminated because of the decrease of the concentration difference of solutes between liquid and α crystals, and the composition of liquid reaches equilibrium. Therefore, α crystal coarsening occurs via the amalgamations-coalescences of α crystals under the effects of thermal convection of the melt moving and of mutual attraction of α crystals. Figs 2c and d, 3c and 4b demonstrate this amalgamation and coalescence evolution. The α crystal coarsening occurs spontaneously because system energy decreases with the decrease of interface area between liquid and solid during the coarsening process.

The coarsening rate in the first stage is greater than that in the second stage because the probability of amalgamation of α crystals in the first stage is larger than that in the second stage. In the third stage, Ostwald ripening growth plays a dominant role in α crystal coarsening instead of the amalgamation-coalescence because equilibrium composition has been realized and thermal convection has disappeared. The coarsening rate in this stage is very low as diffusion rate of solutes in liquid is lower in Ostwald ripening model [9–11].

4.2. Power exponent relationship between mean diameters, (d), and holding time, (t).

As shown by Equations 1, 2 and 3, the power exponents of the parabolic relationship between d and t vary with the remelting temperature. The higher the temperature, the smaller the power exponents are. As pointed out by G. Wan and P. R. Sahm, the power exponent is affected by convection intensity, the higher the intensity is, the smaller the power exponent is [9]. Their viewpoints are very consistent with the results obtained in this investigation. The transforming from solid to liquid and the diffusing of solute atoms during heating will result in convection. The higher the remelting temperature, the more intensive convection is, the higher the probabilities of the amalgamations and coalescences are, and the smaller the power exponents are.

Furthermore, according to the results obtained, another mechanism of coalescence growth is found at the beginning of remelting at a very low remelting temperature in this investigation different from the mechanisms of the amalgamations-coalescence and Ostwald ripening in the structure evolution in the alloy. This coalescence growth occurs within two neighbored grains via a thin film of liquid between them. This thin film acts as medium to get a perfect matching between their crystal lattices. The growth rate in this mechanism is low because the liquid film is thin and the viscosity of the liquid is very high.

Some researchers [9–11] pointed out that the exponent in the parabolic growth equation is not 3 in a semisolid melt with convection in the growth mechanisms of the amalgamations-coalescence and Ostwald ripening. In the experiment with convection of Wan and Sahm, the exponent is 7/3 instead of 3[9], and in the work of Akaiwa, Hardy and Voorhees, it is 2.7 at intermediate volume fractions of solid and is a function of the magnitude of the convection [11].

In this investigation, the exponent of the parabolic growth equation is 4.004 in the alloy remelted at 573°C which corresponds to a low volume fraction of liquid and the growth of α crystals is dominated by the mechanism of the coalescence of lattice matched neighboring crystals. It is expected that, when the remelting temperature is lower, the exponent becomes greater. However, the exponent decreases from 2.38 to 1.84 with raised temperatures. In this situation, liquid fractions are higher and convection in the melt is intensified, and crystal growth follows the amalgamations-coalescence and Ostwald ripening mechanisms. It is expected that the limiting minimum value of the exponent is 1 when convection intensity reaches it's maximum at a very high remelting temperature close to liquidus. The reason for the limiting minimum value of the exponent of 1 is that Ostwald ripening growth, occurring following the amalgamations-coalescence growth, totally depends on the atom diffusion, which is a time-dependent factor, and will lead to a decline of growth rate of crystals with the holding time, and if the exponent is smaller than 1, the curve of an equation showing the relation of diameters of α crystals to remelting time will rise steeply, which corresponds to a rapid increasing of growth rate that shouldn't occur because of the growth of Ostwald ripening.

5. Conclusions

1) The eutectic starts its fusion in a form of spot-like eutectic liquid, then forms fiber-like and net-like liquid at boundaries when AlSi6.5Cu2.8Mg alloy is remelted.

2) The structure evolution procedure of the alloy can be divided into three stages. In the first stage, the quantity of liquid increases in company with the fusion of eutectics and small α grains and α crystal coarsening occurs in the form of adjoined crystals coalesce. Thermal convection is a main factor to cause the amalgamation and coalescence in the second stage. In the third stage, α crystal coarsening follows Ostwald ripening model caused by volume diffusion instead of coalescence model because of a minor convection in the melt.

3) Power exponents of parabolic relationship between α crystal diameter and holding time vary with remelting temperatures. The exponents are 4.004, 2.38 and 1.84 corresponding to the remelting temperatures of 573°C, 578°C and 584°C, respectively. The values of the exponents vary with different mechanisms of α crystal growth. At low temperature near solidus in the semi-solid temperature range, α crystals grow in a coalescence growth mechanism and the exponent is over 3. At high temperature, the mechanisms of the amalgamation-coalescence and Ostwald ripening dominate the growth and the exponent is less than 3 because of great convection in the melt and greater than 1 because of very small diffusion rate of atoms in Ostwald ripening growth.

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