Aluminum Alloys: Fabrication, Characterization and Applications II
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SOLIDIFICATION AND PROCESSING OF ALUMINUM BASED IMMISCIBLE ALLOYS

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Abstract

The Al-Sn and Al-Pb based immiscible alloys have significant potential for bearing applications. However, the mixing and understanding of solidification process for immiscible alloys have been long standing challenges for their development. This paper presents solidification and microstructural evolution of the Al-Sn-Cu alloys and also describes the mechanism of effective mixing by the intensive shearing. The experimental work was also focused on analyzing the effects of shear rate, temperature and time on Sn droplets size and their distribution. Results have been compared with earlier study on Al-Si-Pb alloys. Experimental results suggest that the intensive shearing process produces homogeneous and finely dispersed Sn and Pb droplets.

Introduction

The solidification studies of immiscible alloy systems such as Al-Bi, Al-Sn, Al-Pb, Al-Si-Pb, Al-Pb-Si etc. is important from scientific and technical point of view [1-3]. The Al-Sn and Al-Pb based alloys have been commonly accepted for having excellent tribological and mechanical properties. These kinds of alloy system are suitable for engineering applications, particularly self lubrication bearing materials [1, 2]. Owing to the lower solubility, the parent liquid is decomposed into two distinct immiscible liquid phases when it passes through the immiscibility gap [1-3], and then followed by severe segregation due to the large density difference between two different density liquid phases [1,2]. In Al-Sn and Al-Pb alloy systems, phase separation occur when the Sn and Pb content are higher than 0.09 wt.% and 0.2 wt.%, respectively. To overcome segregation problem in immiscible alloy many methods have been proposed, such as stir casting, ultrasonic, rheocasting and rapid solidification. Recently, Fan *et. al* [4] developed a melt conditioning advanced shearing technology (MCAST) device to create a fine and homogeneous liquid dispersion within the miscibility gap and then the viscous force offered by semi-solid slurry to counterbalance the gravity force and the Marangoni effect [4,6].

In the present study, the immiscible Al-Sn-Cu alloys were successfully synthesized within the semi-solid region using the well developed MCAST device and results are compared with earlier study on Al-Si-Pb alloys system. It is observed that the final microstructures of alloys are strongly influenced by the viscosity of the system, shear forces, turbulence and cooling rate.

Experimental procedure

The (90-x)Al-xSn-10Cu immiscible alloys for x = 20, 30 and 45 were prepared from commercial pure aluminium with appropriate addition of 99.99 wt.% pure Sn and Cu and Al-Si-xPb alloys for x = 3.8, 7.2 and 17.2, were prepared from A357 alloy with appropriate addition of 99.97 wt% pure Pb [6]. All compositions in this paper are given in wt%. The melt was prepared in a graphite clay crucible in electric resistance furnace. The furnace temperature was gradually increased and held 200 °C above critical temperature (T_c) for 2 hours to homogenize the melt.

The MCAST device used in this work for intensive shearing is combined with high pressure die casting (HPDC) machine (DCC280, LK® Machinery, Hong Kong). The combination of MCAST and HPDC is called MC-HPDC. The detailed explanation about MCAST has been described elsewhere [7, 8]. The Al-Sn-Cu and Al-Si-Pb alloys melt were poured into the MCAST device at 650 °C and 620 °C, respectively. The pouring temperature was well above the T_c to avoid phase separation of L' and L'' (L' is Al-rich liquid and L'' is Sn-rich or Pb-rich liquid) before the shearing commenced and then multi-phase mixture was sheared at desired speed, time and processing temperature (T_p). For microstructural comparison purpose melt was directly transferred to the HPDC machine without shearing, which is referred to as conventional HPDC process.

To investigate microstructural features with optical microscope (OM) the samples were mounted and ground using standard metallographic polishing techniques. In the process of microstructural characterization, the equivalent diameter (d) and shape factor (F) were calculated by $d = \sqrt{4A/\pi}$ and $F = 4\pi A / P^2$; where, A is the total area and P is the peripheral length of the particles. When F is equal to 1, it represents a perfect spherical particle.

Results

HPDC

Figure 2 shows the OM image of Al-45Sn-10Cu alloy produced by conventional HPDC process. Segregation of the Sn droplets (dark grey in contrast) can be seen at the centre of the tensile specimen. Due to presence of temperature gradient during solidification Sn droplets migrate from low temperature region to the high temperature region [9].

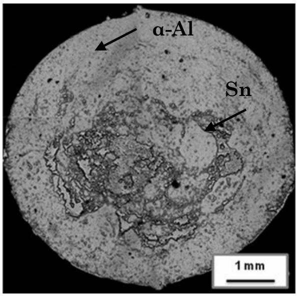

Figure 2. The cross section view of Al-45Sn-10Cu alloy tensile specimen produced with conventional HPDC.

MC-HPDC

Al-Sn-10Cu alloys. Figure 3 shows the microstructures of Al-Sn-Cu samples produced with MC-HPDC. The Sn droplets (dark grey in contrast) in all samples are dispersed uniformly in Al matrix. A good distribution and fine size of Sn droplets achieved at optimum processing parameters. As the wt.% of Sn increases the average Sn droplets size increases from 4 μm to 22 μm with almost constant shape factor (Figure 4(a)). No significant segregation has been observed throughout cross section of the tensile specimen as shown in Figure 3. Figure 4(b) reveals that α-Al particles are also spherical in shape. The size of the α-Al particles vary between 40 μm to 50 μm with different Sn concentrations. The microstructures produced after shearing with varied shearing time and intensity have been also characterised for their respective volume fraction of α-Al particles and Sn droplets [9].

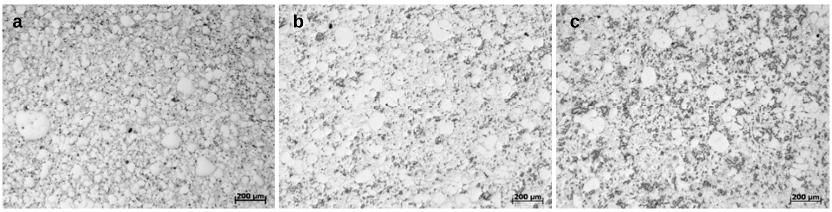

Figure 3. Optical micrographs of various (90-x)Al–xSn–10Cu alloys produced by the MC-HPDC process under optimal processing parameters (a) x = 20; T_p = 580 °C; shearing speed 800 rpm for 60 s (b) x = 30; T_p = 580 °C; shearing speed 800 rpm for 60 s(c) x = 45; T_p = 535 °C; shearing speed 800 rpm for 180 s.

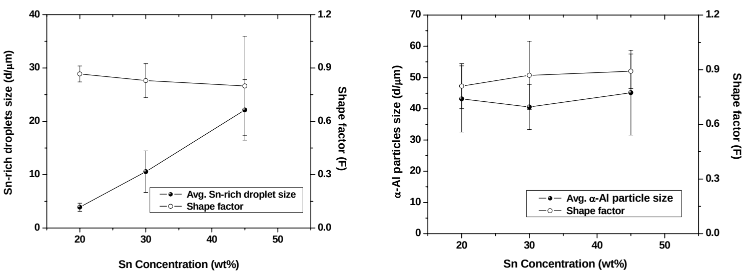

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Figure 4. Effect of intensive shearing on (a) Sn droplets size and shape (b) α-Al particles size and shape as a function of Sn concentration.

Al-Si-Pb alloys. The resultant microstructures of Al-Si-Pb alloys are shown in Figure 5. The average size of the Pb droplets (black color in contrast) in Al-Si-3.8Pb alloy is 2.6 μm and the shape factor of the Pb droplets is 0.89. In Figure 6(a) by increasing Pb concentration from 3.8 wt.% to 17.2 wt.% the droplet size increases from 2.6 μm to 14 μm, while shape factor has decreased from 0.89 to 0.82. Similar to the Al-Sn-Cu alloys system, there has been no significant segregation found. In addition, primary α-Al particle are observed to distribute homogeneously and finely throughout the sample along with the uniform and well distributed Pb droplets. The size and shape of the α-Al particles have not changed much with increasing the wt.% of Pb in these alloys (Figure 6(b)).

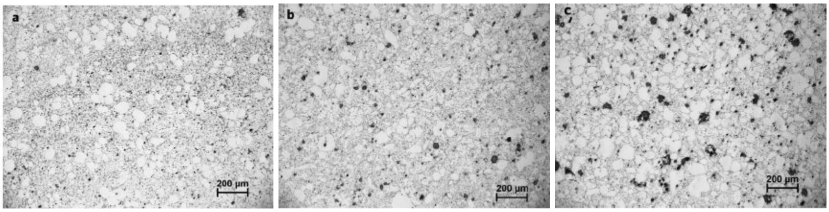

Figure 5. Optical micrographs of various Al–Si–*x*Pb alloys produced by the MC-HPDC process under optimal processing parameters (a) $x = 3.8$; $T_p = 605$ °C; shearing speed 500 rpm for 120 s (b) $x = 7.2$; $T_p = 605$ °C; shearing speed 500 rpm for 120 s (c) $x = 17.2$; $T_p = 595$ °C; shearing speed 500 rpm for 120 s [6].

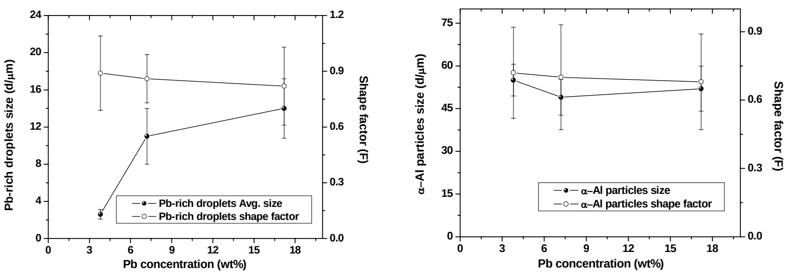

Figure 6. Effect of intensive shearing on (a) Pb droplets size and shape (b) α-Al particles size and shape as function of Pb concentration.

Discussion

In HPDC produced alloys, the higher density Sn droplets were accumulated at the central area of tensile specimen. The volume fraction of droplets and their size was increased from the mould wall to centre position of mould. This occurs because nucleation starts at the surface of mould and proceeds inward, but due to the migrating nature of the droplets from a low temperature region to high temperature region, the segregation occurs at the centre of the specimen at place where liquid solidifies last [1,2,10,11], which is described by the well known Marangoni motion. The coalescence mechanism mainly depends on the size and volume fraction of the L″ droplets [1, 11]. The coalescence takes place by the transfer of matter in which larger droplets grow by absorbing smaller ones and some droplets collide with each other to from a single one by mutual loss of surface energy due to joining [11].

In the MC-HPDC process, when the liquid alloy is fed into the MCAST (above the T_c) and the melt cools quickly to the barrel temperature set by the control system, which is usually just below the monotectic temperature (T_m), where primary α-Al already start to precipitate. At the same time, the melt separates rapidly into two immiscible liquids through nucleation and growth of liquid droplets in miscibility gap. Under the intensive shear mixing action created by the twin screws, the liquid droplets attain fine particle size, as a result of the dynamic equilibrium between two opposite processes, coagulation and breakup of liquid droplets. The stages of the process from the homogeneous liquid to fine and uniformly distributed Sn and Pb droplets in Al matrix is shown schematically in Figure 7.

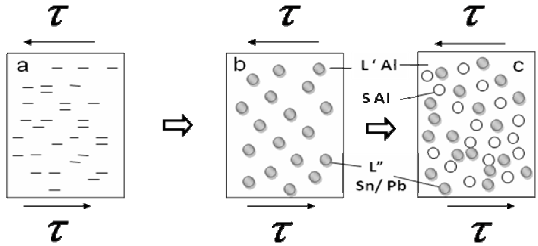

Figure 7. Schematic illustration of the rheomixing process for achiving a uniform distribution of soft phase in Al alloy matrix (a) homogenious liquid (above the T_c); (b) creation of the L'' droplets in L'; (c) rheomixing: formation of a primary α-Al solid phase (S) in L' through a monotectic reaction.

The final size of liquid droplets will be dictated by the intensity of shear mixing action and the thermo-physical properties of the system, such as viscosity, interfacial tension, etc. When the melt reaches a temperature below the T_m, a solid phase will form from one of the liquid phases through the monotectic reaction [5]. It is well known that the viscosity of the semi-solid slurry increase exponentially with the volume fraction of the solid phase and decrease dramatically with increasing shearing rate and shearing time. By careful selection of the processing temperature, the viscous force was kept high enough to counterbalance the gravity force. Consequently, the alloy system is stabilized for the final solidification of the remaining liquid, normally by a eutectic reaction at a lower temperature. Therefore, viscosity helps to inhibit agglomeration or to slow down diffusion of the Sn and Pb droplets. The effect of viscosity of the semi-solid slurry on Stokes motion (U_s) and Marangoni motion (U_m) is given by:

$$U_s = \frac{2g\Delta\rho(\eta + \eta^{'})}{3\eta(2\eta + 3\eta^{'})} r^2, \tag{1}$$

$$U_m = \frac{2\left|\frac{dT}{dx}\right|\left|\frac{d\sigma}{dT}\right|\kappa}{(2\eta + 3\eta^{'})(2\kappa + \kappa^{'})} r, \tag{2}$$

Where, $\Delta\rho$ is the density difference between the two liquids, g is gravitational acceleration, r is the size of the liquid droplet, κ and $\kappa^{'}$ are conductivity of liquid matrix and droplets respectively; η and $\eta^{'}$ are viscosities of the liquid matrix and droplets respectively. dT/dx is the temperature gradient and $d\sigma/dT$ is the variation of the interfacial energy between the two liquid phases with change in temperature. During intensive shearing, the melt temperature is extremely uniform throughout the entire volume of the liquid mixture. According to equation (2), $U_m = 0$, therefore segregation of L'' droplets are negligible during solidification of intensively sheared melt.

The initial size distribution of droplets is inhomogeneous in the melt conditioner. Refinement and dispersion of droplets occur at later stages when increased the time of shearing in Al-Sn-Cu alloy [9]. The observed decrease in droplet size with shear rate is related not only to the breakup process but also to the shear-induced coalescence. The coalescence can be accelerated by the same factors that favor the drop breakup, i.e. high shear rate and reduced viscosity ratio. Therefore, the minimum droplet size under given shear mixing conditions is a dynamic balance between two opposite processes, droplet breakup and coalescence.

Summary

1. The MC-HPDC process produces a uniform dispersion of Sn and Pb droplets in Al alloy matrix. The size of the α-Al primary phase is approximately 50 μm and the average size of the Sn and Pb droplets increases with Sn and Pb concentration.

2. The Sn and Pb metallic droplets can be broken up more easily in the viscous fluid under high shear rate conditions and can achieve more spherical shape in thick viscous turbulent flow. Increasing shear rate speed up the droplets breakup process and will also lead to the spherical and fine droplet formation.

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