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Grain refinement of binary Al-Si, Al-Cu and Al-Ni alloys by ultrasonication

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ABSTRACT

A systematic experimental study has been made of the combined influence of solute content (1%, 2%, 3%, 5%) and ultrasonic intensity (0, 88, 350, 790, 1400 W/cm^2) on grain refinement of Al-Si, Al-Cu and Al-Ni binary alloys. The grain refining efficiency was analyzed using the growth-restriction-factor (GRF) model. Ultrasonication resulted in more activated nucleants in Al-Cu alloys than in Al-Si and Al-Ni alloys by the model. The combination of high solute content and high ultrasonic intensity produced significant grain refinement, including significant refinement of eutectic structures that formed in the Al-5%Si, Al-5%Cu and Al-5%Ni alloys. In particular, high solute content ensured achievement of homogeneous and consistent grain morphology in ultrasonic grain refinement. In addition, excellent grain refinement was achieved in both the Al-5%Cu alloy, which has an equilibrium freezing range of 100 °C, and the Al-5%Ni alloy, which has an equilibrium freezing range of just 5 °C, under the same applied ultrasonic intensity (1400 W/cm^2) conditions. The mechanisms for ultrasonic grain refinement of Al alloys are discussed based on experimental findings.

1. Introduction

In solidification processing, grain refinement can be achieved by increasing thermal undercooling through increasing cooling rates, constitutional undercooling through addition of selected solute elements, or by increasing the number of effective heterogeneous nuclei through inoculation, as summarized by Murty et al. (2002). Shaha et al. (2015) reported that a fine and uniform grain structure imparts good yield strength, high toughness, and improved machinability, and can result in a uniform dispersion of the second phase particles. During the last several decades, various methods have been employed during solidification for grain refinement of alloys, including inoculation, enhanced cooling, rheo-diecasting shown by Zhen et al. (2006), and physical methods such as mechanical stirring employed by Bhingole and Chaudhari (2012), electromagnetic vibrations shown by Yu et al. (2009) and ultrasound vibrations used by Patel et al. (2012). As shown by Radjai and Miwa (2002) and many other researchers, ultrasonic treatment (UST) of molten alloys can change the dendritic structure of the primary phase into equiaxed or non-dendritic structures.

A number of studies have focused on UST of Al alloy melts. Puga et al. (2011) observed that the grain morphology of cast AlSi9Cu3 alloy changed from dendritic to globular by UST and the grain size of α-Al decreased significantly to 41 μm. Feng et al. (2008) showed that the dendritic morphology of the primary α-Al in Al-23%Si alloy changed to the equiaxed one after the UST. Jian et al. (2005) obtained a globular and refined microstructure in A356 alloy by UST. Tuan et al. (2015) showed that applying UST at 720 °C resulted in a uniform grain structure in an Al-Mg-Sc alloy. Atamanenko et al. (2010) studied the effect of UST in both the liquid state and the semisolid state in Al-Cu, Al-Zr-Ti and Al-Zr-V alloys. Ultrasonic refinement of these alloys depended on the amplitude of the sonotrode, UST time, and volume of the melt. Das and Kotadia (2011) demonstrated that the α-Al phase in LM6 alloy can be modified into globular grains by UST. Li et al. (2006) observed that UST refined the microstructure of Al-1%Si alloy (herein all compositions are given in wt.%). Recently, Wang et al. (2014) showed that there is no effect of UST on grain refinement of Al-2%Cu alloy, when the UST is applied above the liquidus temperature. This finding suggests that ultrasonic grain refinement occurs within the liquidus-solidus range. Accordingly, the freezing range of an alloy can be an important factor that affects ultrasonic grain refinement.

Both the UST and the solute content affect the microstructural evolution during solidification in Al alloys. However, a systematic study of the combined effect of solute content and UST in Al alloys is lacking in literature (a study on Mg alloys was reported by Qian et al. in 2010b). In this research, the grain refinement in Al-Si, Al-Cu and Al-Ni alloys is studied by varying the respective solute content (1%, 2%, 3% and 5%) and the applied ultrasonic intensity (88, 350, 790 and 1400 W/cm^2). The resulting microstructure of each Al alloy is characterized and the combined effect of solute content and UST is discussed.

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Table 1
Basic data of the Al-Si, Al-Cu and Al-Ni systems (k: equilibrium solute partition coefficient; m: liquidus slope).

Alloy system	Eutectic point	Maximum solubility in α-Al	k	m	m(k-1)
Al-Si	577 °C, 12.6%Si	1.65%Si	0.130	−6.62	5.759
Al-Cu	548.2 °C, 33.2%Cu	5.65%Cu	0.17	−3.38	2.805
Al-Ni	639.9 °C, 5.7% Ni	0.24%Ni	0.042	−3.6	3.449

2. Experimental

A 20 kHz ultrasonic generator was used to generate vibrations in molten Al alloys using a 1.5 kW capacity ultrasonic generator unit (Model VCX 1500, Sonics and Materials, USA). The diameter and length of the niobium (Nb) acoustic radiator were 19 mm and 175 mm respectively. It should be noted that the frequency of the Nb radiator changes during UST with an increase in temperature due to being immersed in the melt and also with an increase solid fraction in the melt as solidification goes on. However, the variations are comparable for all the binary Al alloys processed in this work. The ultrasonic intensity is given by Eq. (1) according to Eskin (1998):

$$I = \frac{1}{2}\rho c(2\pi f A)^2 \tag{1}$$

where ρ is the density of molten metal, c is the speed of sound in the melt, f is the frequency and A is the amplitude. Four amplitudes of ultrasonic horn vibrations were selected, viz. 24 µm, 18 µm, 12 µm and 6 µm (measured using a contactless vibrometer at room temperature). They gave intensity values of about 1400 W/cm^2, 790 W/cm^2, 350 W/cm^2 and 88 W/cm^2 by Eq. (1), where $c \approx 1.3 \times 10^3$ ms^{-1} in molten Al according to Eskin (1998) and the density of molten Al was taken as $\rho = 2375$ kgm^{-3}. Eskin (1998) determined that the threshold ultrasonic intensity required to produce cavitation in molten Al is about 80 Wcm^{-2}. The lowest ultrasonic intensity of 88 W/cm^2 used in this research thus still exceeds this threshold intensity.

Binary alloys of Al-Si, Al-Cu and Al-Ni with solute contents of 1%, 2%, 3% and 5% were selected for ultrasonic grain refinement. Table 1 lists the basic data of the Al-Si, Al-Cu and Al-Ni systems as per the ASM Handbook Vol. 3 Alloy Phase Diagrams (ASM, 1992). In principle, all four selected Al-x% Cu (x = 1, 2, 3, 5) alloys are off-eutectic alloys (< 5.65%Cu, Table 1) while all four selected Al-x%Ni (x = 1, 2, 3, 5) alloys are hypoeutectic alloys (> 0.24%Ni, Table 1). As for the four selected Al-Si alloys, Al-1%Si is off-eutectic (< 1.65%Si, Table 1) while the rest three (Al-2%Si, Al-3%Si and Al-5%Si) are all hypoeutectic.

Commercial purity Al and respective master alloys were used to make Al–Si, Al-Cu and Al-Ni alloys. In each experiment, 300 g of the Al and master alloy were melted in a graphite crucible (inside diameter: 60 mm) in an electric resistance furnace at 700 ± 5 °C, which is at least 40 °C higher than the liquidus of each alloy. The melt was held for 30 min to allow for complete dissolution. A thermocouple was placed into the melt to record the cooling process during solidification. The Nb ultrasonic horn was preheated to ~700 °C prior to ultrasonic processing. The crucible was withdrawn from the furnace at 700 °C and was placed on refractory bricks. The Nb ultrasonic horn was then immersed into the melt and the system was switched on until solidification was nearly complete. A control sample was cast for each composition under identical conditions but without UST, which is referred to as “as-cast” alloy.

Specimens for microstructural examination were cut along the longitudinal section of each casting. They were ground and polished following standard metallographic procedures, and then etched with Keller’s reagent. A Leica DMI 5000 M microscope and Zeiss scanning

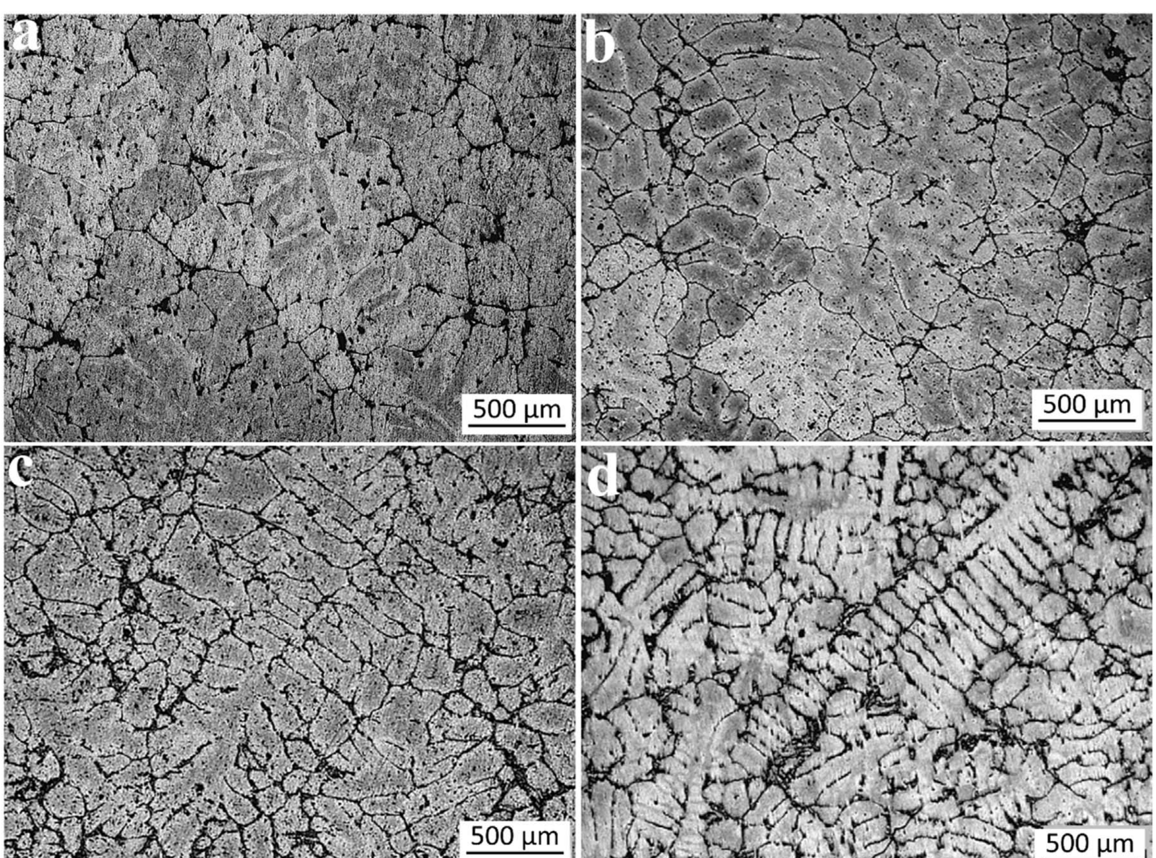

Fig. 1. Optical micrographs of Al-Si alloys cast without UST containing (a) 1%, (b) 2%, (c) 3%, and (d) 5% Si.