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Grain Refinement of an Al-2 wt%Cu Alloy by Al₃Ti₁B Master Alloy and Ultrasonic Treatment

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Abstract. Both inoculation by AlTiB master alloys and Ultrasonic Treatment (UT) are effective methods of refining the grain size of aluminium alloys. The present study investigates the influence of UT on the grain refinement of an Al-2 wt% Cu alloy with a range of Al₃Ti₁B master alloy additions. When the alloy contains the smallest amount of added master alloy, UT caused significant additional grain refinement compared with that provided by the master alloy only. However, the influence of UT on grain size reduces with increasing addition of the master alloy. Plotting the grain size data versus the inverse of the growth restriction factor (Q) reveals that the application of UT causes both an increase in the number of potentially active nuclei and a decrease in the size of the nucleation free zone due to a reduction in the temperature gradient throughout the melt. Both these factors promote the formation of a fine equiaxed grain structure.

1. Introduction

Microstructural grain refinement of cast metallic alloys improves the mechanical properties of as-cast components and subsequent thermo-mechanical processing behaviour of semi-fabricated products such as billet and slab materials [1, 2]. The addition of master alloys containing inoculant particles is standard grain-refining practice for most commercial cast aluminium (Al) alloys where the most common grain refiners are based on Al-Ti-B and Al-Ti-C master alloys [3-6]. Alternatively, Ultrasonic Treatment (UT) has proved to be effective for the grain refinement of various Al alloys [7-12]. The effect of UT on microstructural evolution is derived from its physical influence on the molten melt, such as cavitation, acoustic streaming and radiation pressure [13]. Although much research has been undertaken over many decades, the mechanism of ultrasonic grain refinement is still a controversial issue. The current proposed mechanisms are mainly associated with the role of cavitation, including cavitation-induced dendrite fragmentation and cavitation-assisted heterogeneous nucleation [7-9, 13]. The present research investigates the combined effect of Al₃Ti₁B master alloy and UT on the grain refinement of an Al-2 wt% Cu alloy. The grain refining efficiency is evaluated for a range of master alloy additions with and without the application of UT.

2. Experimental procedure

The Al-2 wt% Cu alloy was prepared from commercial purity Al ingot (99.7%) and pure copper (Cu) (99.9%) using an electric furnace in a 4 kg batch. The equilibrium liquidus and solidus temperatures of



the alloy as calculated using ThermoCalc software are 655 and 620°C, respectively. The ultrasonic device consists of a 2-kW commercial ultrasound generator, an air cooled 20-kHz piezoelectric transducer and a sonotrode made of a molybdenum alloy with an 18-mm diameter tip. About 1 kg of the alloy was melted and preheated to 720±3°C inside a graphite-clay crucible with 90 mm top diameter, 60 mm bottom diameter and 120 mm in height. For samples without the addition of master alloy, the crucible containing the molten alloy was removed after reaching 720±3°C and transferred to the UT platform [14]. For samples with the addition of master alloy, the equivalent amounts of Al₃Ti₁B master alloy (in wt%) containing 50, 100, 500, 1000 and 2000 ppm Ti were introduced into the molten melt at 720±3°C, which then was mechanically stirred and held for 5 minutes before being transferred to the UT platform. The UT sonotrode was switched-on without preheating and then immersed 15 mm below the top surface of the melt. Two K-type thermocouples were inserted into the melt beside the sonotrode: one adjacent to the wall of the crucible and the other being placed 12.5 mm apart. Both thermocouples were positioned 45 mm above the bottom of the crucible. The temperature data was collected by a data-acquisition system with a sampling rate of four readings per second. The cooling rates from the measurement were 0.25±0.01 °C/sec without UT and 0.54±0.01 °C/sec with UT. The sonotrode was not inserted into the melt for the experiments without UT.

The UT experiments were conducted with a fixed power input of 1 kW at an amplitude of 20 μm from 695 °C which is 40 °C above the liquidus temperature, for 4 minutes during subsequent solidification and ceasing at 653°C. A previous investigation [14] has revealed that an appropriate amount of superheat of the liquid metal and continued application of UT for a period of time below the liquidus temperature for ultrasonic grain refinement are essential when an unpreheated sonotrode is introduced into the melt. Superheat of 40°C above the liquidus temperature can insure that there is no formation of a strong solidified layer on the sonotrode. This solidified layer could dampen the UT effect in the surrounding liquid which may result in a significant reduction in cavitation and acoustic streaming thus preventing enhanced nucleation and rapid grain transport while continued application of UT for a period of time below the liquidus temperature promotes nucleation therefore refinement can be achieved.

Metallographic samples were sectioned along the centre symmetrical axis and mechanically ground and polished for observation. In order to measure the grain size, small samples were cut at 45 mm (the same height as the thermocouples and 25 mm below the sonotrode) from the bottom of the sectioned piece. Micrographs were obtained by a Leica Polyvar microscope with polarized light after anodizing using a 0.5% HBF₄ solution for about 20 seconds at 30 V. The grain sizes were measured using the linear intercept method (ASTM E112-10). The value of the growth restriction factor, Q , was calculated using the Al-Cu-Ti ternary phase diagram generated by ThermoCalc.

3. Results

3.1 Ultrasonic refinement of Al-2Cu alloy

Solidification of the Al-2 wt% Cu alloy without UT and without the addition of master alloy produces a coarse dendritic macrostructure (figure 1a). Figure 2a shows the typical equiaxed dendritic grain structure obtained from the centre of the ingot. The average dendritic grain size is about 1500 μm.

With UT applied from 40°C above the liquidus temperature of the Al-2 wt% Cu alloy for 4 minutes during subsequent cooling, the macrostructure presents a uniform fine grained structure throughout the whole ingot (figure 1b). The anodized microstructure of samples from the central part is shown in figure 2b, and the average grain size is about 150 μm, indicating that significant grain refinement results from the application of UT.

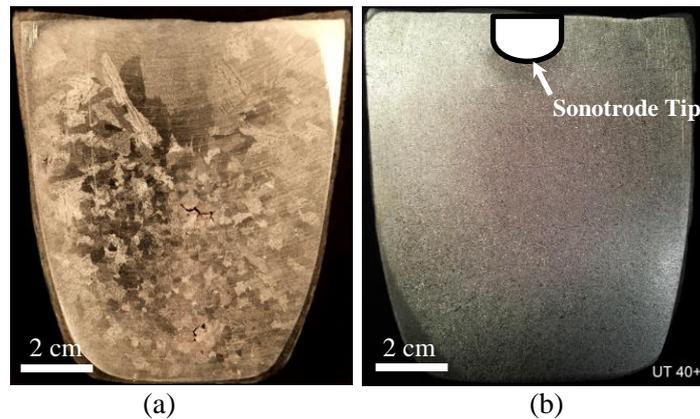


Figure 1. The macrostructure of Al-2% Cu alloy ingot samples (a) without UT and (b) with UT applied at 40 °C above the liquidus temperature for 4 minutes.

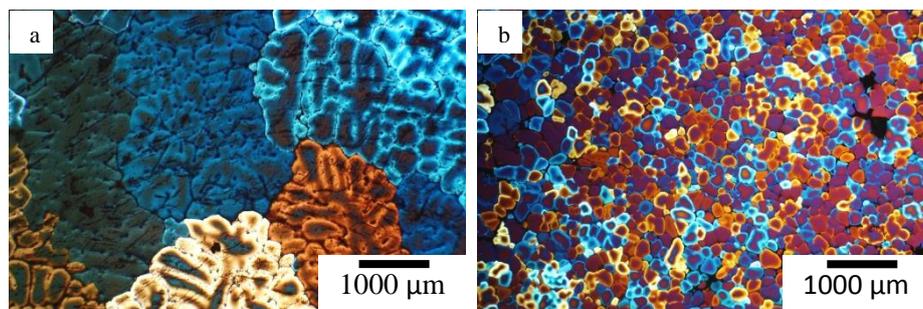


Figure 2. The microstructure of anodized samples from the central part of the Al 2Cu alloy ingot samples: (a) without UT and (b) with UT.

3.2 The influence of Al3Ti1B master alloy on grain refinement

Figures 3 a, b and c show the effect of adding (a) 50 ppm Ti, (b) 100 ppm Ti, (c) 1000 ppm Ti on the microstructure near the centre of the ingot. Figure 4a plots the measured grain sizes versus Ti content up to 2000 ppm Ti where the grain size decreases from about 230 microns to 125 microns. The diminishing effect of further high levels of master alloy is typically observed as there is a cube root relationship between the number of particles nucleating grains and the grain size [15].

3.3 The influence of UT on the refinement of Al-2 wt% Cu alloy with additions of master alloy

Figures 3 d, e and f show the microstructures of the Al-2Cu alloy ingot samples with (d) 50 ppm Ti, (e) 100 ppm Ti and (f) 1000 ppm Ti after the application of UT. Figure 4a shows that the grain size is consistently smaller than when UT is not applied and ranges from about 130 microns to just under 100 microns.

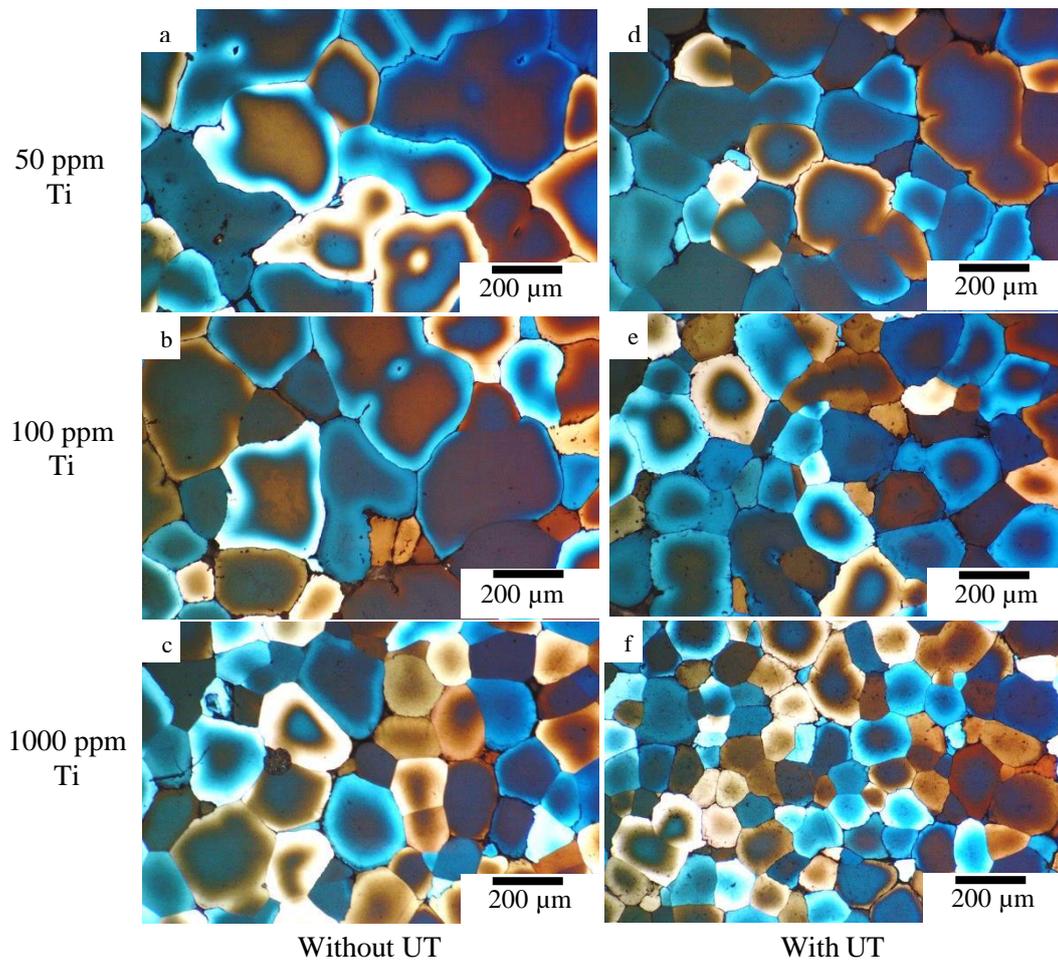


Figure 3. The microstructure of Al-2Cu alloy ingot samples with equivalent amounts of 50 ppm Ti, 100 ppm Ti and 1000 ppm Ti where (a), (b) and (c) are without UT and (d), (e) and (f) are with UT.

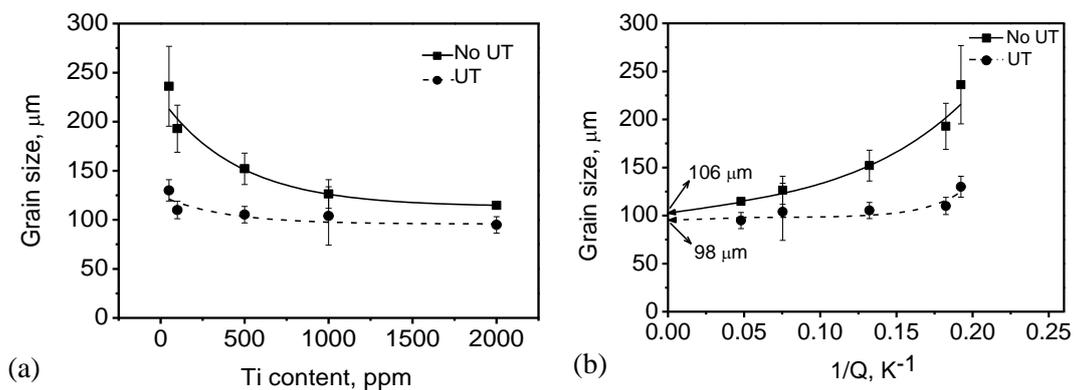


Figure 4. (a) The relationship between grain size and Ti content with and without UT application. (b) The relationship between grain size and inverse growth restriction factor Q .

4. Discussion

The average grain size of ingots with and without the application of UT is plotted against the equivalent content of Ti in figure 4a. There is a significant decrease in grain size from 50 to 500 ppm Ti content in both master alloy addition alone and the combined master alloy and UT cases, and the grain size decreases slowly with further increases in the Ti content. However, while plotting grain size against composition shows a diminishing effect, the number density of active particles may in fact be increasing at the same rate in both cases. Figure 5 converts grain size into grain density [16] which is directly related to the number of active nucleant particles. It should be noted that the rate of increase in grain density diminishes as the Ti content increases. This curvature may be due to the fact that the increasing amount of Ti solute added by the master alloy is increasing the growth restriction factor, Q , which has a greater effect on grain size reduction at low levels of addition [15].

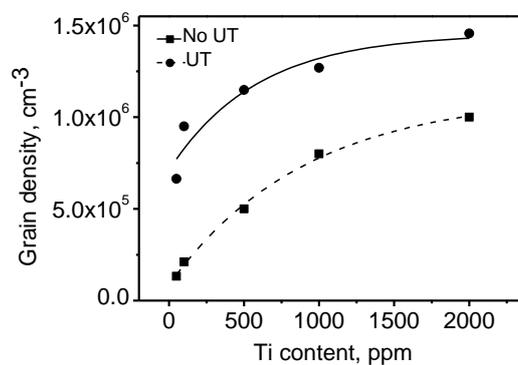


Figure 5. The relationship between grain density and Ti content with and without the application of UT.

Other effects of UT could be that cavitation and acoustic streaming promote a finer distribution of TiB_2 particles and/or deagglomeration of TiB_2 clusters creating more sites for nucleation [16]. In addition, the collapse of cavitation bubbles may enhance the wetting of particles which may improve their efficiency for grain refinement [7].

By calculating Q for the alloy composition (2 wt% Cu plus the amount of Ti solute), the relationship between grain size and Q with and without UT is presented in figure 4b. Note that the curves of best fit are not straight lines as observed when the particle number density is kept constant [15]. Curves are observed when both the particle number density and Q increase simultaneously [17]. Figure 4b clearly shows that UT decreases the gradient of the curve of best fit which may indicate that the nucleant potency of the refining particles has increased. Atamanenko et al. [9] found that UT alters the potency of the nucleant particles in Al-Zr-Ti alloy. However, in this case the effect of UT is likely to be related to the refinement of the particles. In the present study the nucleant particles are only TiB_2 , and though the number of active particle may change with UT due to the deagglomeration and better dispersion the change in slope may be due to other factors as well. The gradient of the curves in figure 4b can be significantly influenced by change in the size of the Nucleation-Free Zone (x_{NFZ}) surrounding each nucleated grain with change in the value of Q [17] according to [5]

$$x_{NFZ} = 5.6 \left(\frac{D \cdot z \Delta T_n}{v Q} \right) \quad (1)$$

where D is the diffusion coefficient, v is the rate of growth of the solid-liquid (S-L) interface, and $z \Delta T_n$ is the incremental amount of undercooling required to activate the next nucleation event as the temperature gradient moves towards the thermal centre of the casting. x_{NFZ} contributes to the final grain size. The other factor contributing to the grain size is the distance between the most potent particles in the melt which is denoted x_{Sd} [18]. The sum of x_{NFZ} and x_{Sd} equals the grain size.

The non-linear relationship shown in figure 4b derives mainly from the non-linear dependency of x_{sd} on $1/Q$ in this study.

Therefore, a change in any of the parameters in equation (1) will change the gradient. The parameter z in equation (1) represents the effect of temperature gradient on the size of x_{NFZ} . Because the temperature gradient decreases due to UT treatment [14] the value of z decreases thus reducing x_{NFZ} and, therefore, the gradient of the UT curve in figure 4b. The other point to note is that the intercept with the y-axis in figure 4b which defines x_{sd} for an infinite number of active particles, decreases when UT is applied. By extrapolating the curves to the y-axis a value of 106 is obtained without UT and with UT the intercept is 98. Although this appears to be a small difference, it represents an increase in grain density of about 20% implying the number of activatable particles has increased by a similar amount with the application of UT. In addition, for a given composition a reduction in x_{NFZ} also enables more TiB_2 particles to nucleate grains [18].

5. Summary

The application of UT to an Al-2 wt% Cu alloy without master alloy additions resulted in significant grain refinement when applied from 40 °C above the liquidus temperature for 4 minutes during subsequent cooling. The separate application of UT and the addition of Al₃Ti₁B master alloy to an Al-2 wt% Cu alloy both individually produced considerable grain refinement. Further refinement was achieved by applying both methods together. The relationship between grain size and the inverse growth restriction factor indicates that UT changes the nucleation efficiency of the master alloy by lowering the temperature gradient before and during the nucleation stage in the melt which decreases the size of the nucleation-free zone, and by increasing the proportion of activatable TiB_2 particles along with the addition of the UT induced nucleation by about 20% through the nucleation stage. Both of these factors lead to a reduction in grain size.

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