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Effect of Ultrasonic Treatment on The As-Cast Grain Structure during The Solidification of An Al-2Cu Alloy

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The effect of the application of Ultrasonic Treatment (UST) over selected temperature ranges on the grain structure has been investigated for an Al-2Cu alloy melt. It was found that applying UST from 40°C above the liquidus to just above the liquidus brings the melt to a condition that is favourable for nucleation, survival of the nucleated grains and their subsequent transport throughout the melt. Continuing UST beyond the liquidus until about 25% solid fraction enhances both nucleation and convection ensuring the formation of a fine, uniform equiaxed grain size throughout the casting. The role of ultrasonication in grain refinement includes not only the nucleation and multiplication of grains close to the ultrasonic source at temperatures around liquidus but also acoustic streaming which generates a high degree of sustained convection that transports the nucleated grains and grain-seeds towards the solidification front, assuring their survival and distribution in the melt volume.

Keywords: *Solidification, Growth from melt, Alloys, Ultrasonic treatment, Grain refinement*

1. Introduction

Numerous works on the application of high intensity ultrasound in melt to achieve refinement of grains as well as suppression of columnar grain structure and formation of globular grain structure have been reported by researchers [1-3]. The influence of ultrasonication on the refinement of microstructure is based on the physical phenomena caused by high intensity ultrasound propagation in the melt, in particular acoustic cavitation and acoustic streaming [1-2]. While cavitation is the formation, oscillation and collapse of cavities in the liquid as a consequence of forces acting upon the liquid through alternating pressure at ultrasonic frequency [3-4], acoustic streaming represents the flow driven by absorption by the liquid of high amplitude acoustic oscillations and the momentum created by the oscillation of the cavitation region [5].

During Ultrasonic Treatment (UT) of the melt, cavitation bubbles nucleate and grow during the negative pressure portion of the sound field, and then collapse during intervals of increased pressure if the acoustic intensity is sufficiently high. The implosion of the bubbles results in intense local heating and high pressures with very short lifetimes, as well as violent liquid jets. Therefore, cavitation produces large instantaneous pressure and temperature fluctuations in the melt, and these pressure and temperature fluctuations may facilitate heterogeneous nucleation in the melt as reviewed elsewhere [5]. It is well understood that the cavitation zone is relative small, and it is the acoustic stream which also play a significant role in high intensity ultrasound applications to the grain refinement. It is proposed that acoustic streaming generates a high degree of sustained convection creating a favourable (i.e. more uniform) thermal environment throughout the melt which enhances survival of the new grains and provides time for their transport throughout the melt, and it is also the acoustic streaming induces a high degree of convection which then transport the new grain throughout a the casting during the nucleation stage therefore a large volume of the melt can be treated [6-7]. The level of both cavitation and acoustic streaming effects depend on the parameters of the ultrasonic field in the melt, and also properties of the treated materials. And these properties are highly influence by the melt temperature. Eskin [1] and Abramov [2] have both suggested that for ultrasonication the melt temperature plays an important role in controlling the melt properties particularly when it is close to the liquidus temperature. However, very few studies have been reported of the effect of the melt temperature on ultrasonic grain refinement of light alloys.

This work aims to understand the influence of melt temperature on the effectiveness of UT in promoting grain nucleation and formation of an Al-2 wt% Cu alloy. For this purpose, UT was applied at various starting and terminating temperatures by inserting and removing a molybdenum sonotrode without preheating into the alloy melt. The resulting microstructure were investigated in detail.

2. Experimental Procedures

The Al - 2 wt% Cu alloy was prepared from commercially pure aluminum (99.7%) and pure copper (99.9%) using an electric furnace in a 4 kg batch. The liquidus and solidus temperatures for this alloy were 655 and 620°C respectively calculated using ThermoCalc software.

The ultrasonic device consists of a 2 kW commercial ultrasound generator, an air cooled 20 kHz piezoelectric transducer and a sonotrode made of 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. The melt with crucible was then removed from the electric furnace and transferred to the experimental platform shown in Figure 1a, where the sonotrode was turned on 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 another placed 12.5 mm from the edge of the melt. Both thermocouples were placed 45 mm above the bottom of the crucible (Figure 1a). The temperature data was collected by a data-acquisition system at four readings per second.

UT experiments were conducted with fixed power input of 1 kW with an amplitude of 20 μm at the sonotrode tip applied over different temperature ranges, and two groups of experiments were

conducted. The first group of experiments were designed to study the effect of the UT starting temperature on the solidification structure, three UT temperature ranges (I, II, and III) as shown in Figure 1b have been investigated. For Ranges I to III, UT was applied from 694°C, 674°C and 655°C and was terminated after 4 minutes at 653.1°C, 651.3°C and 649.4°C, respectively. In order to find out how the UT stopping temperature influence grain structure, the 2nd set of experiments were conducted for 6 temperature ranges. In Range i, UT was applied from 714°C and terminated at 660°C which is 5°C above the liquidus temperature. For Ranges from ii to vi, UT was applied at 695°C and terminated after 0, 10, 20, 40 and 80 seconds after melt temperature reaching to the liquidus (as shown in Figure 1d) by removing the sonotrode then turning off the ultrasonic power to allow the melt to solidify. For all the experiments, the sonotrode was kept at the ambient temperature and turned on before immersion into the melt.

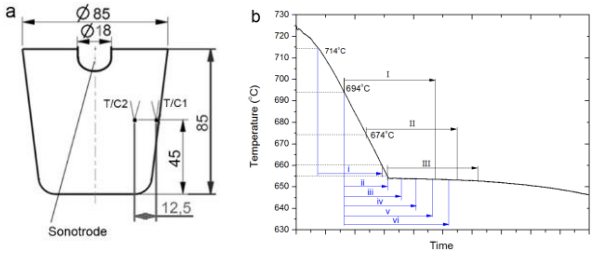

Figure 1. (a) Schematic of a cast sample showing the locations of the sonotrode and thermocouples (in mm), and (b) Schematic showing the temperature ranges over which UT is applied: (I) from 694°C (40°C above liquidus) for 4 min, (II) from 674°C (20°C above liquidus) for 4 min, and (III) from 655°C (liquidus) for 4 min; and (i) from 714°C (60°C above liquidus) to 660°C, from 694°C to (ii) 0 second, (iii) 10 seconds, (iv) 20 seconds, (v) 40 seconds, and (vi) 80 seconds after the liquidus temperature (655°C) is reached

Metallographic samples were sectioned along the central symmetrical axis, mechanically ground and polished using standard metallographic equipment for observation. Macroetching was done using a solution of hydrofluoric, nitric and hydrochloric acid. In order to measure the grain size, small samples were cut at a distance of 45 mm (the same height as the thermocouples) from the bottom of the sectioned piece. Micrographs were obtained using a Leica Polyvar microscope with polarized light after anodizing using a 0.5% HBF4 water solution for about 20 seconds at 30 VDC. The grain size was measured using the linear intercept method (ASTM E112-10).

3. Results and Discussion

The grain structure of the samples produced with UT starting from 40°C (Range I), 20°C (Range II), and 0°C (Range III) above the liquidus temperature for 4 minutes is shown in Figure 2. For Range I, UT applied 40°C above the liquidus temperature resulted in significant refinement throughout the sample as shown in Figures 2a with average grain size of 150 µm. When UT starting temperature was reduced to 20°C and 0°C above the liquidus for Ranges II and III, a well-developed columnar dendritic structure with no refinement is observed as shown in Figure 2b and 2c. It is clear that the melt temperature when UT is applied through an unpreheated sonotrode plays a critical role in refining the cast microstructure of the alloy and that a threshold temperature exists above which UT creates the conditions necessary for significant refinement of the α-Al grain structure.

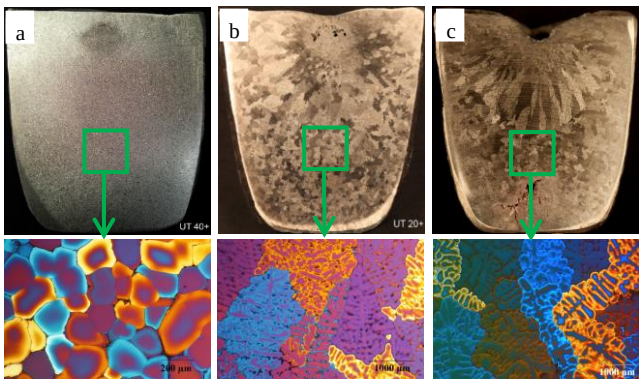

Figure 2. The macro and micro grain structure of the samples: UT applied for 4 minutes from (a) 0°C, (b) 20°C, and (c) 40°C above the liquidus during cooling as indicated by Ranges I, II, IIII in Figure 1b.

Figure 3 shows both macro- and micro-structure for the 2nd group of experiments on the variety UT termination points. When UT was applied from 714 to 660°C where 660°C is 5°C above the liquidus (Range i in Figure 1b) the grain structure of the sample consists of coarse equiaxied dendrites in the central region and lower part of the sample and columnar dendritic grains at the top (Figure 3a).

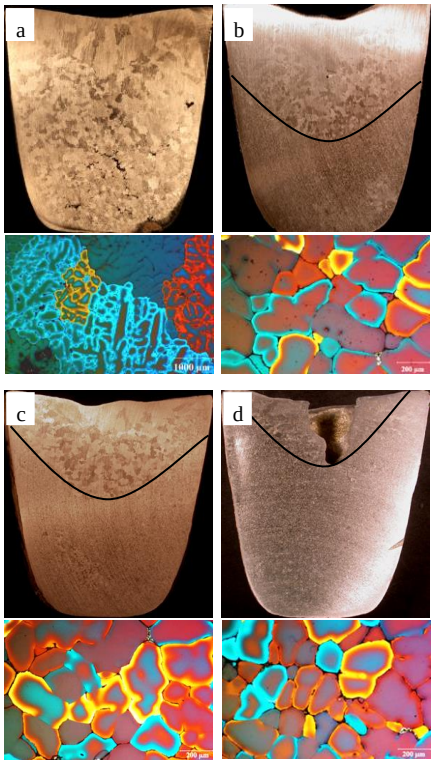

Figure 3. Grain structure of the samples (a) range i with UT until just before solidification begins in the temperature range 714-660°C, and with UT started at 694°C and stopped at (b) 0 s for range ii, (c) 20 s, the range iv, (d) 80 s range vi after the liquidus (655°C) is reached (see Figure 1b for reference).

When application of UT from 694°C and terminating at different duration time from 0 to 80 seconds after reaching the liquidus temperature (655°C), the vertical cross section of the casting is clearly divided into two regions, i.e. a coarse grain region at the top and a fine grain region at the bottom as indicated by the superimposed lines. Terminating UT at the liquidus temperature resulted in grain refinement of about 50% of the volume as shown in Figure 3b, while about 63% and 90% of volume has been refined through increasing UT duration up to 20 and 80 seconds after melt passed the liquidus temperature.

Figure 4a plots the fraction of the refined volume in percentage vs UT duration time after 655°C, and with increasing UT duration after 655°C from 0 to 10 second the proportion of the coarse and fine grained regions does not change much, while after that the fine equiaxed grained region progressively increases at the expense of the coarse grained region. This revealed that UT duration is important for generating a larger fine equiaxed grain.

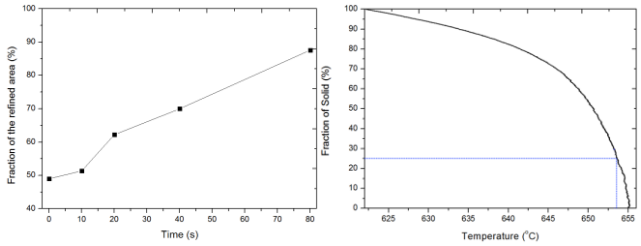

Figure 4. (a) The fraction of the refined region and the grain size versus UT duration after the liquidus temperature is reached (see Figure 1d for reference) and (b) a temperature–solid fraction plot with the coherency fraction and temperature marked by blue dashed lines.

The relationship between melt temperature and solid fraction has been measured by the TGA-TG as plotted in Figure 4b. It shows that the with the temperature decreased the solid fraction increased rapidly in the early stage of the solidification, when temperature reached to 653.1°C, the recorded temperature for Range vi, the solid fraction is about 25%.

It is clear that under the UT even the refined spherical grain can be formed in the nucleation stage, no matter of how powerful of the cavitation on enhancing the nucleation; there is still no guaranty to achieve a fully refined structure throughout the sample. A fully refined equiaxed structure can only be achieved when UT generates lots of grains during the nucleation and UT energy keep these grains suspend and move before melt lost its flow ability.

The two main UT refinement mechanisms that have been proposed are cavitation-induced dendrite fragmentation and cavitation-enhanced heterogeneous nucleation [1-2]. This study has shown that the temperature at which UT is applied plays a critical role in refining the cast grain structure. Initial chill effect from immersion of the sonotrode into the melt resulted in a rapid decrease of 13°C [7]. When UT was applied from 20°C above or at the liquidus temperature this chill effect would be greater adjacent to the sonotrode leading to significant solidification of the surrounding liquid while the bulk melt temperature also decreased. This hinders or prevents effective transmission of the ultrasounds from the sonotrode into the surrounding liquid metal and therefore makes melt cavitation difficult to occur. Accordingly, no grain refinement is observed. According to Ohno [8], once a solid shell of columnar grains form on a cold contact with high cooling capacity, it is difficult for the columnar crystals to separate even if movement of molten metal exists. By increasing the temperature at which UT is applied, less chill solidification occurs and the solidified shell will remelt possibly releasing solidified grains into the melt, allowing cavitation and acoustic streaming to occur resulting in significant refinement of the grain structure. Convection from enhanced acoustic streaming creates a uniform temperature field which provides favorable thermal conditions for the survival and transport of the nucleated grains in the melt and their continued growth.

Cavitation-enhanced nucleation has been explained as (1) increased pressure caused by the collapse of bubbles leading to an increase in the melting point of the surrounding liquid, which is equivalent to generating increased undercooling and, therefore, enhanced nucleation can be expected [9], and (2) improved wetting of the insoluble inclusions by the melt [6]. UT applied in the liquid and terminated before the liquidus temperature is reached results in no refinement indicating that the ultrasound-induced pressure rise is not maintained during the nucleation stage. On the other hand, if cavitation wets the insoluble inclusions making them more potent nucleation sites, then it would be expected that UT before nucleation commences should refine the structure to some degree. The fact that the grain structure was not refined when UT was applied only in the liquid state (Range i) suggests that the cavitation-enhanced nucleation mechanism due to the increased pressure and melting point caused by the collapse of the cavitation-induced bubbles may only act at and below the liquidus temperature of the alloy.

An alternative mechanism is crystal nucleation on the sonotrode surface followed by detachment. Detachment has been proposed as the major refinement mechanism for magnetic field treatments including pulsed magnetic field (PMF), magnetic stirring in the weld pool and solidification during casting [10]. For example, PMF refinement has been attributed to the detachment of heterogeneous nuclei from the mould wall due to melt vibration and subsequent separation of nuclei in the melt caused by melt convection [11]. As mentioned above, when a cold sonotrode is immersed into the melt the significant temperature drop initiates the formation of solidified crystals on the sonotrode surface. Depending on the condition of the sonotrode surface and the surrounding melt, the solidified grains will either remelt, continue to grow or be detached and swept into the bulk of the melt. The newly nucleated grains on the radiating face of the sonotrode will be subjected to alternating tensile or compressional loads due to vibration at a rate of 20 kHz. Further, large instantaneous pressure and temperature fluctuations associated with cavitation will have a significant impact on grain stability on the sonotrode surface. These fluctuations may lead to a continuous cycle of nucleation, growth and detachment of crystals from the radiating surface of the sonotrode, and these grains or fragments would then be distributed by acoustic streaming within the bulk of the melt, increasing the number of grains and, therefore, a refined structure may be achieved.

This detachment mechanism may explain the good refinement achieved when immersing the sonotrode at 40°C above the liquidus temperature. However, when the sonotrode is immersed at 20°C above the liquidus temperature (Range II) or applied from the liquidus temperature (Range III), columnar dendrites form on the surface of the sonotrode and radially grow into the bulk of the melt

forming a solidified shell on the sonotrode surface. It is expected that the solidified shell will significantly reduce cavitation in the melt below the chilled zone along with weakening of acoustic streaming, in particular the solidified shell will prevent detachment, thereby favouring continued coarse columnar growth.

When UT is applied from a higher temperature, for example Range I, the temperature gradient is sufficiently reduced to ensure a good thermal environment for significant nucleation. This result indicates that there is a stage before a fully solidified layer around the sonotrode forms where partial solidification occurs but the crystals are readily removed by acoustic streaming. This would support the detachment mechanism described above but whether this occurs is all situations needs to be verified by further work.

In the current study, terminating UT at the liquidus (655°C) resulted in grain refinement of about 50% of the volume, and the grains were refined to about 220 µm as shown in Figure 3b. Increasing the UT duration time results in increase of the refined volume. The combination of the refined nondendritic or less-dendritic grains in the bottom region and coarse developed dendrites in the upper region of the sample (Figures 3b–d) can be explained as follows (1) refined grains are generated in the cavitation zone under UT during the nucleation stage of solidification; (2) as long as UT continues, the nucleated grains are distributed into the bulk of the melt by acoustic streaming with keeping the solidified grains in suspension and moving, therefore a uniform refined grain structure can be achieved in the lower part of the crucible where the chances of grain survival is maximum due to the bulk of the melt being below the liquidus temperature; (3) terminating UT after establishment of the coherent solidified network lead to a fully refined grain structure within the coherent domain of the casting.

4. Conclusion

UT leads to significant refinement of the Al-2Cu alloy when applied from a melt temperature 40°C above the liquidus temperature and continuing UT beyond the liquidus until about 25% solid fraction throughout the casting. It is proposed that acoustic streaming generates a high degree of sustained convection creating a more uniform thermal environment throughout the melt which enhances survival of the new grains and provides time for their transport throughout the melt. In addition to creating the above conditions, continued application of UT for a period of time below the liquidus temperature promotes nucleation. UT enhanced nucleation may be due to an increase in the liquidus temperature caused by an increase in pressure due to cavitation. It is also possible that the nucleated grains are generated by a continuous process of nucleation, growth and detachment of crystals from the radiating surface of the sonotrode. Once nucleation occurs by either of the above mechanisms on or near the sonotrode, the new grains are then swept into the melt by the high degree of convection caused by acoustic streaming.

When UT was applied from 20°C or from just after the nucleation stage no refinement was observed. It is proposed that this lack of refinement is due to the formation of a strong solidified layer on the sonotrode that dampens the UT effect in the surrounding liquid resulting in a significant reduction in cavitation and acoustic streaming thus preventing enhanced nucleation and rapid grain transport.

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