The role of ultrasonic treatment in refining the as-cast grain structure during the solidification of an Al-2Cu alloy

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Abstract. The effect of Ultrasonic Treatment (UT) over selected temperature ranges during cooling and solidification of an Al-2Cu alloy melt on the grain structure and cooling behaviour of the alloy has been investigated using a molybdenum sonotrode introduced without preheating. UT was applied over various temperature ranges before, during and after the nucleation of primary aluminium grains. It was found that ultrasonic grain refinement was achieved only when UT was applied from more than 20°C above the liquidus temperature until below the liquidus temperature after nucleation has occurred. Applying UT from 40°C or 60°C above the liquidus to just above the liquidus brings the melt to a condition that favours nucleation, survival of the nucleated grains and their subsequent transport throughout the melt. Continuing to apply UT beyond the liquidus for a short time enhances both nucleation and convection thereby ensuring the formation of a fine, uniform equiaxed grain size throughout the casting. The lack of grain refinement when UT was applied from 20°C above the liquidus temperature or from temperatures below the liquidus temperature is attributed to the formation of a strong solidified layer on the sonotrode which hinders the effective transmission of ultrasonic irradiation into the liquid metal. The application of a preheated sonotrode showed that formation of a solid layer can be prevented by preheating the sonotrode to 285°C. Thus, an appropriate amount of superheat of the liquid metal or sufficient preheating of the sonotrode is necessary for ultrasonic grain refinement when a sonotrode is introduced into the melt.

1. Introduction

Using grain refinement techniques to promote the formation of a fine, uniform and equiaxed grain structure during casting processes is common industrial practice in order to improve the downstream processing for as-cast intermediate products [1,2], or structural uniformity for consistent performance, as well as mechanical properties for as-cast finished products [3]. The method widely used to refine the grain structure of a casting is by the addition of a grain refiner master alloy containing potent nucleant particles [4]. Research has also been undertaken to refine the microstructure by the application of an external field [5-10], including electric current pulse (ECP) [5], pulsed magnetic field (PMF) [6], pulse magneto-oscillation (PMO) [7, 8], and ultrasonic treatments (UT) [9-11].

Literature has shown that the introduction of high intensity UT to a metal or alloy melt can result in a noticeable reduction in grain size, often accompanied by changes in grain morphology [9-14]. The influence of UT on microstructural refinement is based on the physical phenomena that occur during the propagation of high intensity ultrasound in the melt, including ultrasonically-induced cavitation and acoustic streaming. A number of hypotheses have been proposed on this basis. The major ones may be summarised as cavitation-enhanced heterogeneous nucleation, cavitation-induced dendrite fragmentation, and vibration-stimulated separation of wall crystals from the sonotrode surfaces [12, 13, 15, 16]. Each of these mechanisms has been reviewed in Ref. [16]. Ultrasonication can be introduced in a variety of approaches [12, 16]. For a light metal or alloy melt UT is typically realized by inserting a sonotrode into the melt. When the sonotrode is immersed into the melt without preheating (1) heat is extracted from the melt by the relatively cold sonotrode, (2) the temperature distribution in the melt may become more uniform when acoustic streaming-induced convection predominates in the ultrasonicated region, and (3) the melt temperature may increase due to ultrasonically-induced cavitation being converted into thermal energy [15]. The melt temperature and its distribution are clearly two important factors affecting the effect of UT. Eskin [12] and Abramov [13] 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, no systematic study has 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 temperatures by inserting a molybdenum sonotrode without preheating into the alloy melt. The resulting microstructure and cooling curves were investigated in detail.

2. Materials and 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 were calculated using ThermoCalc software and the chemical composition of the Al-2Cu alloy is presented in Table 1.

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 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. The melt was exposed to air during melting and during the application of UT while casting. 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). 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, I to V, as shown in Figure 1b. For Range 1, UT was applied from 714°C and terminated at 660°C which is 5°C above the liquidus temperature. For Ranges II to V, UT was applied from 714°C, 694°C, 674°C and 655°C and was terminated after 4 minutes at 653°C, 653.1°C, 651.3°C and 649.4, respectively. For all the experiments, the sonotrode was kept at the ambient temperature and turned on before immersion into the melt. The temperature data was collected by a data-acquisition system at four readings per second.

The solidified samples were sectioned along the centre symmetrical axis and prepared for microscopic observation using standard metallographic procedures. Optical macrographs and micrographs of the grain structures were obtained by Kaiser 5450 and Leica Polyvar microscopes, respectively.

3. Results

3.1. Grain structures

3.1.1 Ultrasonic treatment before or after the nucleation stage (Ranges I and V).

Figure 2a shows the grain structure of the samples solidified without UT which consists of coarse equiaxied dendrites in the central region and lower part of the sample and columnar dendritic grains at the top. When UT was applied from 714 to 660°C where 660°C is a few degrees above the liquidus (Range I in Figure 1b) a similar macrostructure to that shown in Figure 2a is produced. In addition, when UT was applied from the liquidus temperature (Range V) there was no obvious change in the macrostructure, Figure 2c, except the top columnar region became larger. Figure 2 reveals that when UT is terminated just before the liquidus temperature or applied below the liquidus temperature no refinement to the grain structure occurs.

3.1.2 Ultrasonic treatment applied from the liquid phase region through the solidification period (Ranges II, III and IV).

The grain structure of the samples produced with UT starting from 20°C (Range IV), 40°C (III) and 60°C (II) above the liquidus temperature for 4 minutes is shown in Figure 3. For Range IV a well-developed columnar dendritic structure with no refinement, Figure 3a, is observed. However, an increase in the UT starting temperature from 20°C to 40°C and 60°C above the liquidus resulted in significant refinement throughout the sample as shown in Figures 3b and 3c with a grain size in both conditions being reduced to the range 150-200 µm.

It is clear from Figure 3 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

3.2. Effect of UT on cooling curves

Figure 4a presents the cooling curves obtained without UT and with UT applied from 40°C above the liquidus for 4 minutes. Without UT the melt cooled at a constant rate of 0.25±0.01°C/sec down to the liquidus temperature, and the temperatures recorded by the two thermocouples located near the crucible wall and 12.5 mm distance towards the centre are fairly close with a difference of a few degrees (Figure 4a). This can be attributed to a low temperature gradient caused by slow cooling through the hot crucible walls. It can also be observed in Figure 4a that when the unpreheated sonotrode was immersed into the melt an immediate drop in melt temperature of about 13°C occurs followed by a decrease in the cooling rate to a stable but still faster rate of about 0.54±0.01°C/sec. The unpreheated sonotrode acted as a significant pathway for heat extraction from the melt. In this case the two cooling curves overlapped, indicating that a negligible temperature gradient exists which may be due to acoustic streaming and convection promoting a uniform temperature distribution within the melt.

Terminating UT and removing the sonotrode before nucleation commenced (Range I) resulted in a cooling rate approaching the liquidus temperature that is similar to that obtained without UT. Applying UT at 674°C exhibits the highest value of the cooling rate, 0.6±0.01^{o}C/sec, when it approaches the liquidus temperature, while applying UT from 694°C and 714°C the cooling rate is lower, 0.5±0.01^{o}C/sec. All cooling rates obtained with UT are higher than 0.25±0.01°C/sec for the condition without UT.

Figure 4b is a detailed view of cooling curves from thermocouple T/C 2 over a narrow temperature range from 653°C to 658°C showing the cooling behaviour during the nucleation stage. The timescale is arbitrary as all curves are displaced so that each curve can be clearly observed. The point at which nucleation begins is when the cooling curves first exhibit deflection. The dashed line superimposed on Fig. 4b indicates the approximate location where deflection is first observed showing that the nucleation temperatures are similar except for the cooling curve for Range IV. However, the undercooling at which recalescence begins varies. A lower recalescence temperature and a lower plateau temperature after recalescence can both be caused by a lower rate of nucleation (i.e. a lower rate of latent heat release) and/or a higher rate of heat extraction by the mould walls. When UT is applied at 40°C (III) and 60°C (II) above the liquidus temperature the recalescence temperatures are close to 655°C, the liquidus temperature. This suggests that there has been a significant release of latent heat. Given the resulting fine equiaxed grain size, it can be concluded that this arises from the growth of a large population of nucleated crystals compared to the case without UT. Additionally, the high degree of acoustic streaming heats the mould walls more rapidly resulting in a lower rate of heat extraction as indicated by the higher plateau temperatures. The curves for Ranges I and IV are similar but have lower recalescence and plateau temperatures compared with Ranges II and III indicating a lower nucleation rate and possibly cooler mould walls. In both cases acoustic streaming is not active when nucleation occurs because UT was terminated above the liquidus in Range I and was impeded by a solid chill layer forming on the sonotrode in Range IV. Range IV has the fastest cooling rate because UT was applied at a much lower temperature and heat would still be extracted by a colder sonotrode. Also of note is that the nucleation temperature is higher which has probably been caused by the rapid generation of latent heat by the formation of the chill layer on the colder sonotrode not far from the thermocouple. Thus, both Ranges I and IV would not heat the mould as effectively as in II and III. The curve when UT was not applied has the lowest recalescence and plateau temperatures because, despite a lower cooling rate, the nucleation rate is very low.

4. Discussion

The two main UT refinement mechanisms that have been proposed are cavitation-induced dendrite fragmentation and cavitation-enhanced heterogeneous nucleation [12, 13]. This study has shown that the temperature at which UT is applied plays a critical role in refining the cast grain structure. The initial chill effect from immersion of the sonotrode into the melt resulted in a rapid decrease of 13°C. When UT was applied from 20°C above 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 [17, 18], 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 [19], and (2) improved wetting of the insoluble inclusions by the melt [11]. 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 [20-23]. 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 [20]. 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 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 and 60°C above the liquidus temperature. However, when the sonotrode is immersed at 20°C above the liquidus temperature (Range IV) or applied from the liquidus temperature (Range V), 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. Evidence is provided by Figures 2c and 3a which show a separate solidified region of fine grain size under the depression left by the sonotrode which is not observed in Figures 3b or c.

In this study UT was introduced by an unpreheated sonotrode. To determine whether preheating the sonotrode has an effect, Range IV was repeated with the sonotrode preheated to 285°C. Whereas Range IV produced a coarse macrostructure (Fig. 3a), Range IV with a preheated sonotrode produced a refined microstructure (Fig. 5). It is likely that preheating allowed the sonotrode to reach a higher temperature sooner which was sufficiently high to prevent a strong solidified layer forming on the sonotrode. Thus, acoustic streaming is able to occur unimpeded delivering a refined structure as occurs when Ranges II and III were applied. This result implies that preheating simply extends the period when acoustic streaming is active. When UT is applied from a higher temperature, for example Range III, preheating may have little effect as the temperature gradient in the melt is already sufficiently reduced to ensure a good thermal environment for significant nucleation without the application of preheating. This result indicates that there is a stage before a fully solidified layer around the sonotrode forms where partial solidification occurs but in this case the crystals are readily removed by acoustic streaming and/or cavitation. This would support the detachment mechanism described above but whether this occurs in all situations needs to be verified by further work.

In contrast, it was found that the wall crystal mechanism from a titanium sonotrode has a negligible effect on the grain refinement of a Mg-3Al-1Zn (wt.%) alloy under a range of experimental conditions [16] indicating that detachment may or may not occur depending on other casting conditions. For example, another factor that may affect ultrasonic grain refinement is the influence of the sonotrode material and this issue has not as yet received serious attention. Unlike the molybdenum alloy sonotrode material used in this study, a titanium sonotrode is often used for UT of molten magnesium and magnesium alloys. Other variations include a niobium sonotrode [25] and in some cases titanium sonotrodes are coated with a boron nitride layer to prevent attack by aluminium alloys. In addition, recent work on ultrasonication of Mg-Al and Mg-Zn alloys [26, 27] and of aluminium alloys [25] has shown that ultrasonic grain refinement depends significantly on alloy chemistry (i.e. solute type and solute content). These observations add to the complexity of understanding the mechanisms of ultrasonic grain refinement of metals and alloys. A generic theory for ultrasonic grain refinement remains elusive despite the large amount of experimental data produced to date.

5. Concluding remarks

With an unpreheated sonotrode, UT was applied over selected temperature ranges during the solidification of an Al-2Cu alloy to determine the critical range that produces a refined equiaxed macrostructure. It was determined that UT leads to significant refinement of the Al-2Cu alloy when applied from a melt temperature greater than 20^{o}C above the liquidus temperature. 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.

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 or both 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 above the liquidus temperature and terminated just before the liquidus temperature, no refinement occurred suggesting that UT does not contribute to the wetting of inclusions or they were already well wetted, or the conditions for their activation were not met for the experimental conditions used in this study.

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. However, when the sonotrode was preheated to 285°C a refined structure was produced indicating the sonotrode reached a higher temperature sooner resulting in less solidification before acoustic streaming was activated. Thus, an appropriate amount of superheat of the liquid metal or sufficient preheating of the sonotrode is necessary for ultrasonic grain refinement when a sonotrode is introduced into the melt.

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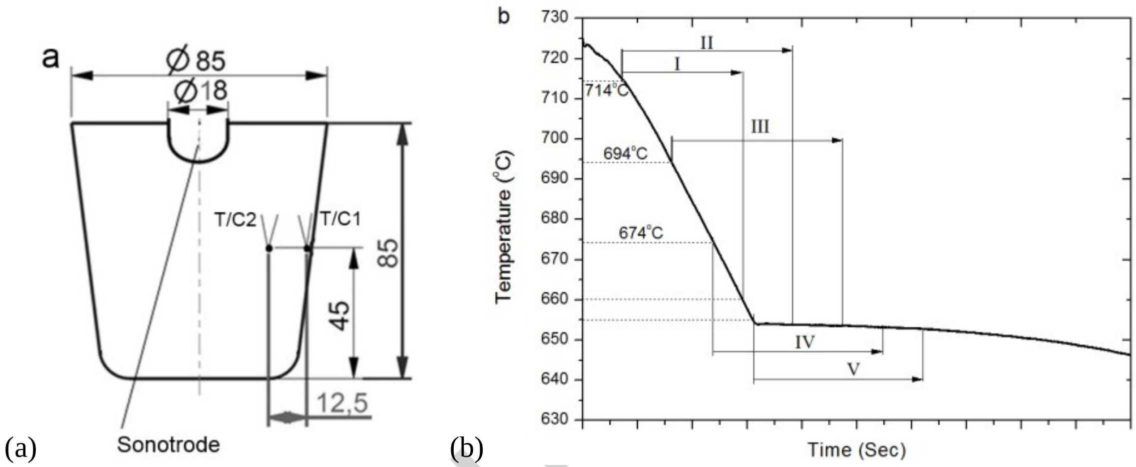
Table 1. Liquidus and solidus temperatures, and the chemical composition of the Al-2Cu alloy.

Liquidus (°C)	Solidus (°C)	Chemical composition (wt%)				
		Al	Si	Cu	Fe	Ti
655	620	Bal	0.03	1.97	0.08	<0.01



(a) (b)

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 714°C (60°C above liquidus) to 660°C, (II) from 714°C for 4 min to 653.0°C during cooling, (III) from 694°C (40°C above liquidus) for 4 min to 653.1°C during cooling, (IV) from 674°C (20°C above liquidus) for 4 min to 651.3°C during cooling, and (V) from 655°C (liquidus) for 4 min during cooling to 649.4°C.

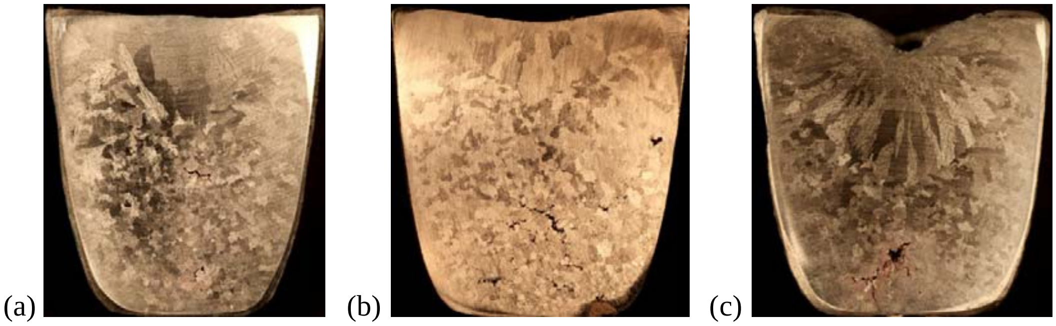


Figure 2. The macroscopic grain structure of the samples: (a) without UT, (b) range I with UT until just before solidification begins in the temperature range 714-660°C, and (c) range V with UT applied for 4 minutes starting at 655°C.

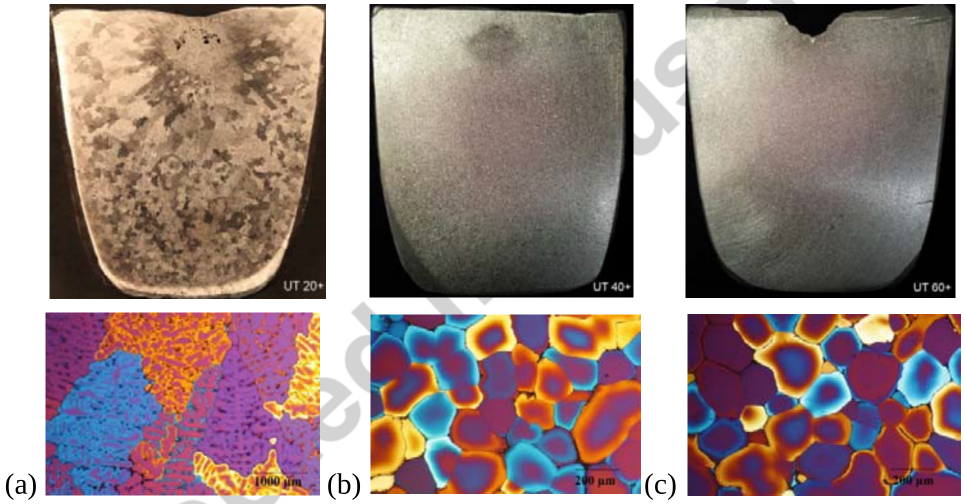
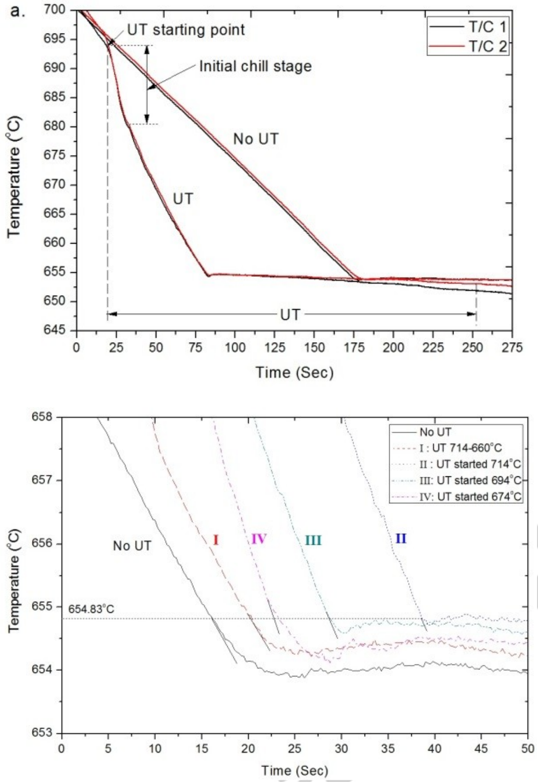


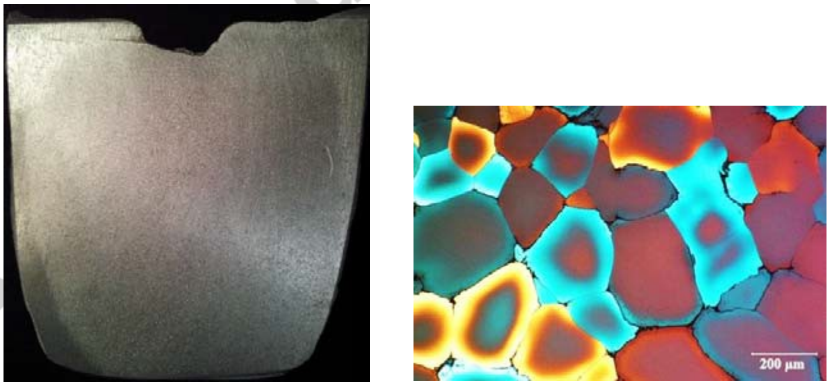
Figure 3. The macro and micro grain structure of the samples: UT applied for 4 minutes from (a) 20°C, (b) 40°C, and (C) 60°C above the liquidus during cooling as indicated by Ranges IV, III, and II in Figure 1b.



(a)

(b)

Figure 4. Cooling curves: (a) with and without UT and (b) cooling curves focused on the nucleation stage from thermocouple 2 (T/C2) indicated in Figure 1a.



(a)

(b)

Figure 5. (a) The macro and (b) microstructure for the sample where the sonotrode was preheated to 285^{o}C and subjected to UT over Range IV.

Highlights

This paper has highlighted that ultrasonic treatment of alloy melts has two important functions.

1. 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.
2. In addition to creating the above conditions, continued application of UT for a period of time below the liquidus temperature promotes nucleation and the acoustic streaming-induced convection provides rapid transport of the new grains into the bulk of the melt.

The equal importance of these factors in achieving a fine equiaxed grain size, in particular the first factor, has not been clearly elucidated before.

Further, we have highlighted the role of solidification is preventing the full potential of UT being realised. For example, when UT was applied from 20^{o}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.

This work also provides evidence that when UT is applied from above the liquidus temperature and terminated just before the liquidus temperature, no refinement occurred suggesting that UT does not contribute to the wetting of inclusions.