UDK 661.862:544.57
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
MTAEC9, 58(2)185(2024)

EFFECT OF HIGH-INTENSITY ULTRASONIC TREATMENT ON REFINEMENT OF Al-5Cu ALLOY INGOTS

VPLIV VISOKOINTENZIVNEGA ULTRAZVOKA NA PREČIŠČEVANJE INGOTOV IZ ZLITINE Al-5Cu

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Prejem rokopisa – received: 2023-11-05; sprejem za objavo – accepted for publication: 2024-02-07

doi:10.17222/mit.2023.1038

The effect of high-intensity ultrasonic treatment combined with bottom cooling treatment on the refinement of Al-5Cu alloy ingots was studied. The results show that ultrasonic treatment combined with forced cooling at the bottom of ingots has a good refining effect, and the best refining effect can be obtained at 2000 W and after 120 s. The cross-section of an ingot exhibits almost 100 % refined equiaxed grains and no porosity. The method of combining ultrasonic treatment and forced cooling at the bottom of ingots not only influences the ingot refinement, but also decreases the porosity of the ingots with an increase in the ultrasonic duration. The ultrasonic degassing effect is due to the release of hydrogen in biofilms, which expand and grow gradually; finally, they burst on the melt surface, achieving the effect of degassing.

Keywords: high-intensity ultrasonic treatment, Al-5Cu alloy ingots, grain refinement, porosities of ingot

V članku je opisana študija vpliva visokointenzivne ultrazvočne obdelave v kombinaciji s specifičnim hlajenjem taline na rafinacijo ingotov iz zlitine Al-5Cu. Rezultati študije so pokazali, da ultrazvok v kombinaciji s prisilnim ohlajanjem dna ingotov dobro vpliva na rafinacijo oziroma udrobljenje (zmanjšanje velikosti) kristalnih zrn ingotov. Najboljši učinek obdelave ingotov so avtorji dosegli z močjo 2000 W za 120 s. Ingoti so imeli v preseku skoraj 100 %-no enakoosna drobna kristalna zrna in so bili praktično brez por. Izbrana metoda obdelave ingotov med ohlajanjem taline ni vplivala samo na udrobljenje kristalnih zrn temveč se je s podaljševanjem časa trajanja ultrazvoka zmanjšala tudi poroznost ingotov. Ugotovili so, da je razplinjevalni učinek ultrazvoka posledica sproščanja vodika, ki potuje v obliki mehurčkov (biofilma) navzgor, dokler se le-ti ne razpočijo na površini raztaljene kovine.

Ključne besede: visokointenzivna ultrazvočna obdelava, ingoti iz zlitine Al-5Cu, udrobljenje kristalnih zrn, poroznost ingotov

1 INTRODUCTION

Aluminum alloys are widely used in 3C (computer, communication and consumer electronics), aerospace and construction industries. Generally speaking, the manufacture of aluminum alloys always involves smelting.[1–3] However, if ultrasonic treatment is applied during smelting, the structure of an ingot can change from coarse columnar crystals to fine equiaxed crystals, and the porosity and segregation of the ingot are also restrained.[4,5] For aluminum alloys, refinement can be achieved by adding refining agents such as titanium, boron, manganese and other elements. Refinement can also be achieved through external field treatments such as ultrasonic field treatment, magnetic field treatment, pulse electric field treatment, and so on. Compared with the traditional chemical method, in grain refinement, the ultrasonic method is non-contaminative to the metal melt and non-harmful to the body, nor does it pollute the surroundings.[6–8] Ultrasonic treatment can refine a solidification structure steadily and rapidly. Some researchers found that the cavitation effect and sonic agitation caused by the high-frequency ultrasonic vibration have dual functions of refinement and degassing. The quality and mechanical properties of alloy ingots were also further improved, so high-intensity ultrasonic treatment has been widely favored by scholars around the world.[9–14] According to previous investigations, the factors affecting refinement comprise melt conditions, which include the chemistry component, pouring temperature, melt temperature at the beginning of ultrasonic vibration, etc.[15–20] This research, based on the work above, took an Al-5Cu alloy as an example, using forced cooling and ultrasonic treatment at the bottom of a clay-graphite crucible to achieve a combined refinement that was studied and investigated. Our investigation provides a new method for the industrial production of high-quality aluminum alloy ingots.

2 EXPERIMENTAL PART

2.1 Apparatus of the ultrasonic device

The experimental equipment includes an ultrasonic generator (its maximum output power is 2000 W and frequency is 20 kHz), a temperature control meter for the temperature control of the melt, a resistance furnace, an ultrasonic bracket (moving the ultrasonic probe up and down) and a clay-graphite crucible. The diameter of the

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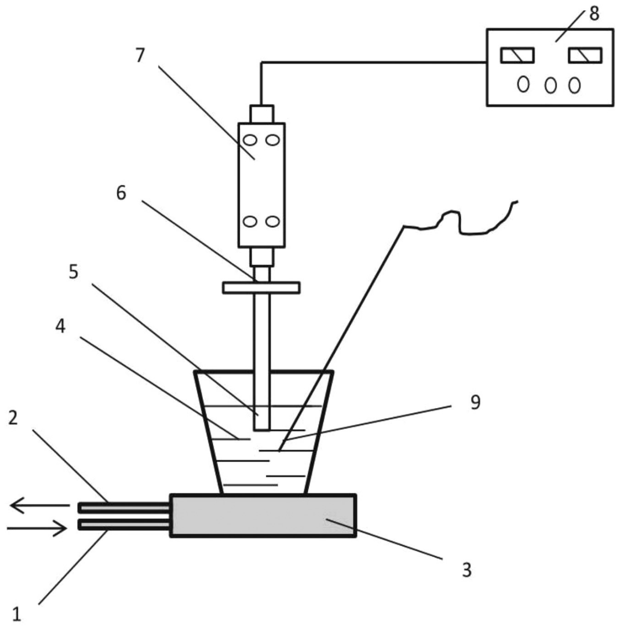

Figure 1: Schematic layout of the ultrasonic field device: 1 – water inlet; 2 – water outlet; 3 – copper plate; 4 – Al-5Cu alloy melt; 5 and 6 – ultrasonic probe; 7 – ultrasonic transducer; 8 – ultrasonic generator; 9 – thermocouple

crucible is 30 mm and the height is 40 mm. The apparatus is shown in **Figure 1**.

2.2 Experimental procedure

The Al-5Cu alloy used in the experiments was made of pure industrial aluminum (purity: 99.7 %) and electrolytic copper flakes in proportion to the melt. The specimens were divided into six groups, with the mass of each group being about 200 g, and put into the clay-graphite crucible for remelting. During the remelting process, when the temperature was stabilized at 800 °C, the clay-graphite crucible containing the melt was placed on a copper plate and ultrasonic treatment was performed using a preheated 500 °C ultrasonic probe inserted into the melt to a depth of 5 mm. When the melt temperature dropped to 750 °C, the copper plate was internally fed with circulating cooling tap water until the ingot solidified and cooled down to room temperature (about 25 °C). Experimental conditions were divided into six types: no ultrasonic treatment applied, bottom water cooling only, ultrasonic treatment only for 90 s without water cooling, and ultrasonic treatment combined with water-cooling treatment for (90, 120 and 150) s. Finally, the ingots were cut into two symmetrical pieces from the top downward for a microstructure observation. Sampling positions are shown in **Figure 2**. The composition and ratio of the macroscopic etching solution for the Al-5Cu alloy were HCl : HF : H_2O = 15 : 10 : 90 and the microstructure etching solution was a 5 % HF aqueous solution. The microstructures were observed using polarized light and the grain size was calculated. Image Pro was used to calculate the grain size.

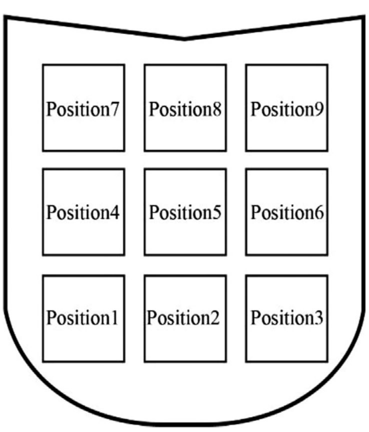

Figure 2: Sampling positions of Al-5Cu alloy ingots

3 RESULTS AND DISCUSSION

3.1 Macrostructures

Figure 3 shows the macrostructure change of Al-5Cu treated with ultrasonic treatment for different times and forced water cooling at the bottom of the ingot. It can be seen that the columnar crystals in the solidification structure of the naturally cooled ingot are relatively coarse, dendritic dendrites are developed and large pores are visible, as shown in **Figure 3a**. When the copper plate is placed in water without ultrasonic treatment, the coarse grains are obviously refined without ultrasonic treatment, the grain distribution is uniform and the grains of the unit area of the solidified structure of the ingot increase, as shown in **Figure 3b**. When ultrasonic treatment is combined with forced water cooling treatment at the bottom of an ingot for 90 s, the equiaxed crystal roundness

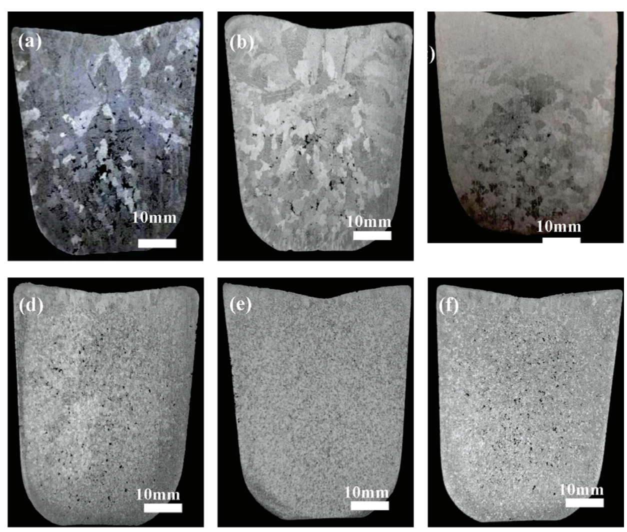

Figure 3: Macrostructures of Al-5Cu alloy ingots with or without ultrasonic treatment: a) normal cooling, b) forced water cooling, c) without forced water cooling but with ultrasonic treat. for 90 s, d) forced water cooling and ultrasonic treat. for 90 s, e) Forced water cooling and ultrasonic treat. for 120 s, f) Forced water cooling and ultrasonic treat for 150 s

of the solidification structure of the ingot increases, and the coarser columnar crystals become shorter. Compared with the ingot without the above treatment at the bottom of the ingot but with ultrasonic treatment, it can be seen from the solidification structure that the grain distribution is more uniform and the pores are reduced. When ultrasonic treatment is combined with forced water cooling treatment at the bottom of the ingot for 120 s, the effect of the combination is the best, and the grain refinement is the most obvious, as shown in **Figure 3d**. When ultrasonic treatment is combined with forced water cooling treatment at the bottom of the ingot for 150 s, the equiaxed grains begin to grow and the pores begin to become larger. If the ultrasonic treatment time is too long and the temperature of the melt is too low, cavitation bubbles in the melt are not discharged in time. Therefore, pores appear inside the ingot.

3.2 Microstructure

Figures 4 to **9** show a comparison of ingots' microstructures affected by different melt treatment methods as nine samples were taken from each ingot. When an ingot is cooled down normally, treated with forced water cooling or without forced water cooling but with ultrasonic treatment for 90 s, it can be found that the ingot has a large number of coarse dendrites, as shown in **Figures 4** to **6**. However, the grain size is at the millimeter level. When using forced water cooling and ultrasonic treatment of the melt for 90–150 s, the dendrites of the ingots are broken, and a large number of equiaxed crystals appear. The grain-size statistics are shown in **Figure 10**.

3.3 Mechanism of ultrasonic refinement and degassing by forced water cooling of the Al-5Cu alloy clay-graphite crucible bottom

When a melt is treated with ultrasonic treatment, the resulting cavitation bubbles grow and shrink in positive and negative cycles, as shown in **Figure 11**. There are two reasons for the grain refinement of the melt after ultrasonication: first, the growth of the cavitation bubbles can absorb the heat from the melt and then nucleate under the influence of cooling at the bubble interface. The subsequent rupture of the cavitation bubbles generates enormous energy to crush the initial nuclei and dendrites in the melt, increasing the number small grains. The generated acoustic flow is a liquid flow due to the acoustic pressure gradient, which also facilitates grain refinement. In addition, the macroscopic flow effect of the liquid phase generated by ultrasonication is manifested as a generalized reflux within the liquid, which can transport a large number of nuclei caused by nonuniform nucleation and dendrite fragmentation throughout the aluminum alloy melt. When forced cooling is applied to the

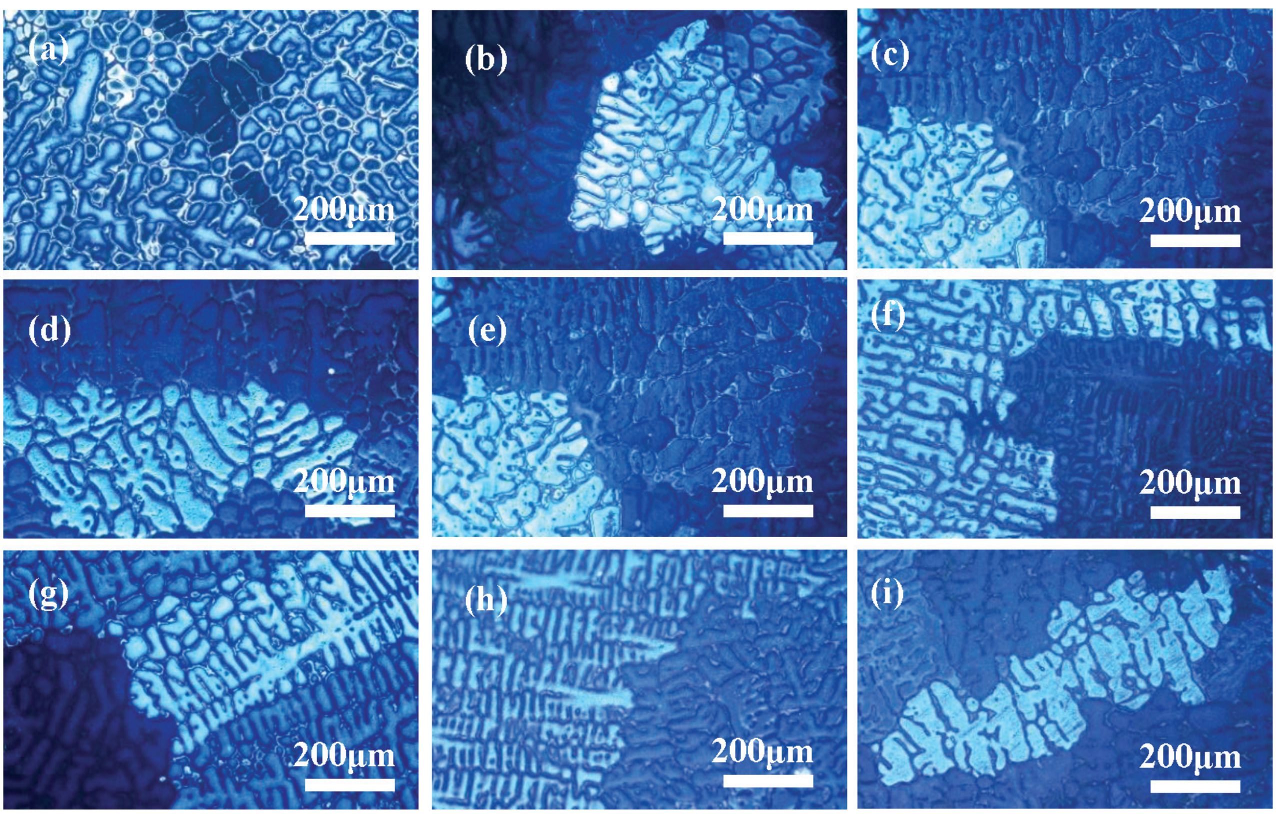

Figure 4: Microstructure of the Al-5Cu alloy ingot after normal cooling: a) position 1, b) position 2, c) position 3, d) position 4, e) position 5, f) position 6, g) position 7, h) position 8, i) position 9

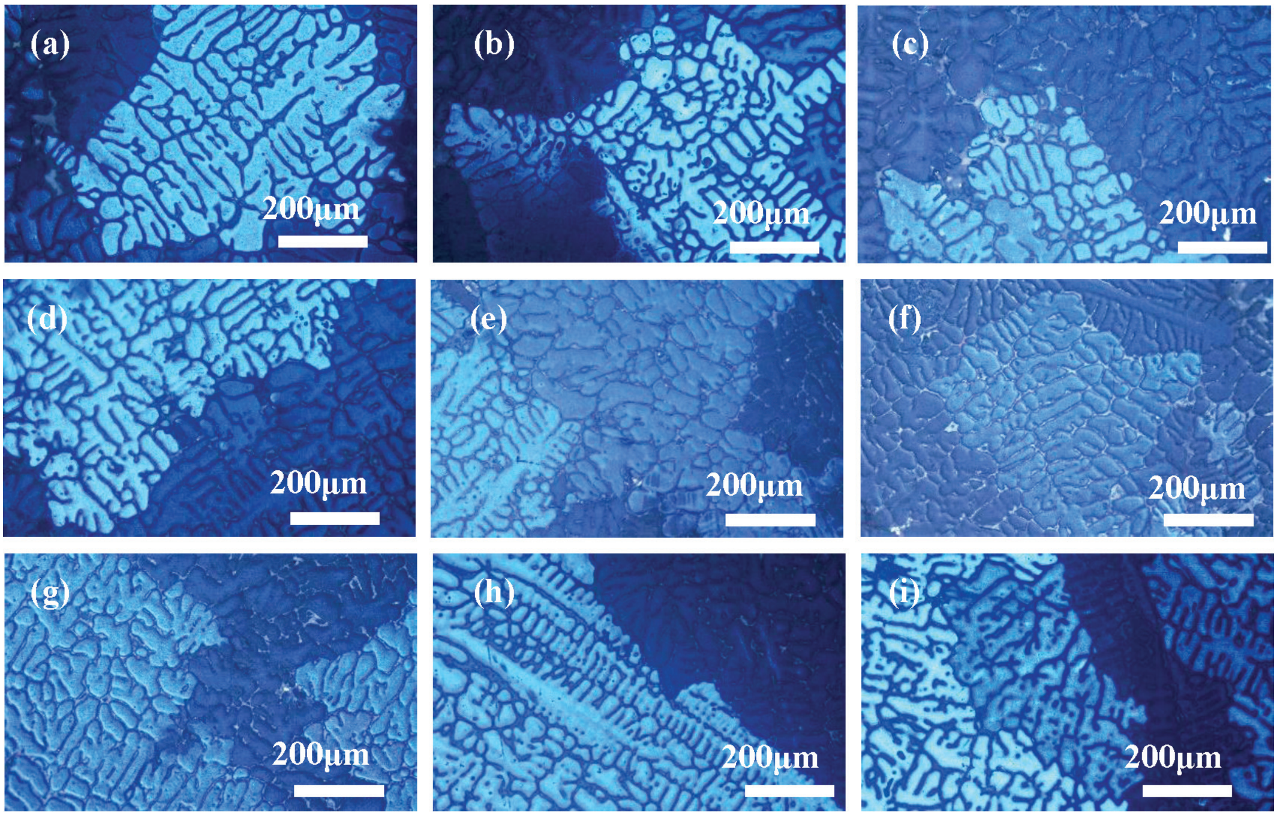

Figure 5: Microstructure of the Al-5Cu alloy ingot after forced water cooling: position 1, b) position 2, c) position 3, d) position 4, e) position 5, f) position 6, g) position 7, h) position 8, i) position 9

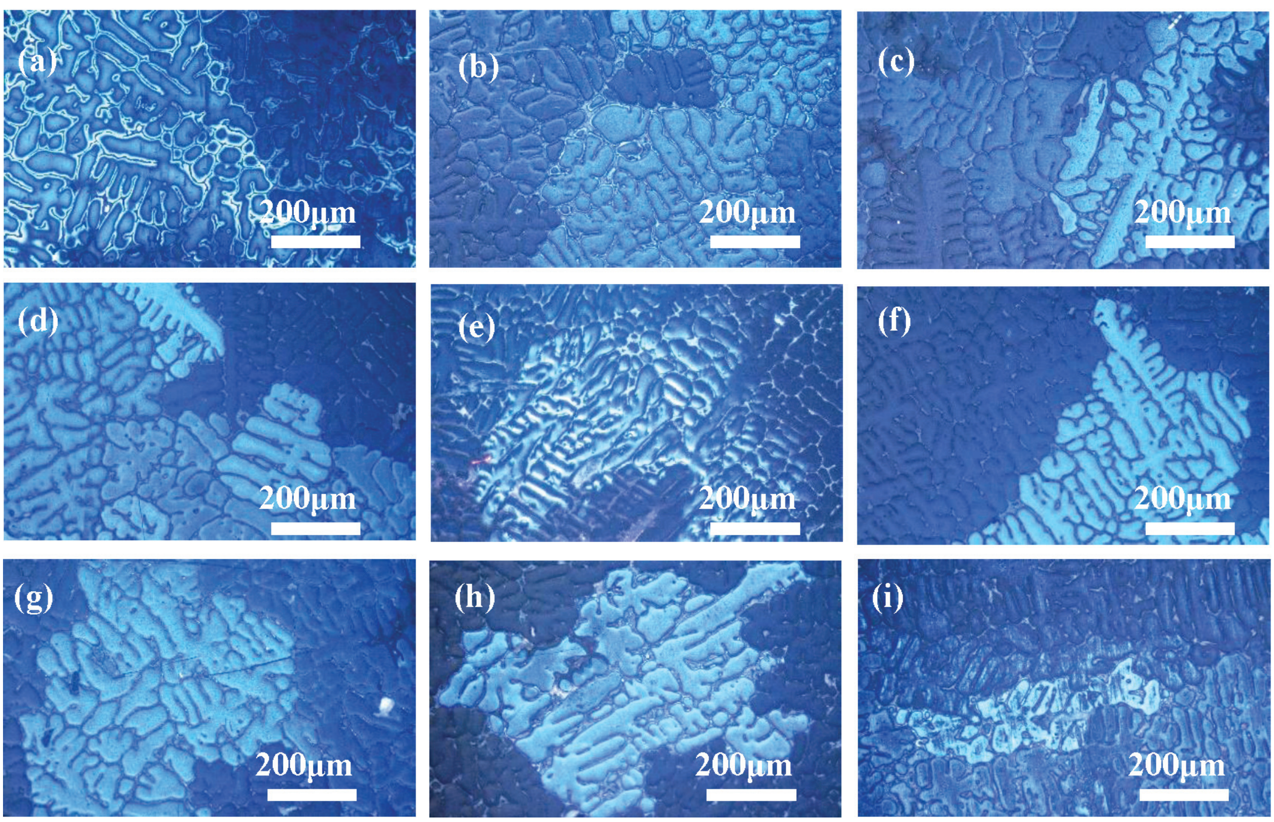

Figure 6: Microstructure of the Al-5Cu alloy ingot without forced water cooling but with ultrasonic treat. for 90 s: position 1, b) position 2, c) position 3, d) position 4, e) position 5, f) position 6, g) position 7, h) position 8, i) position 9

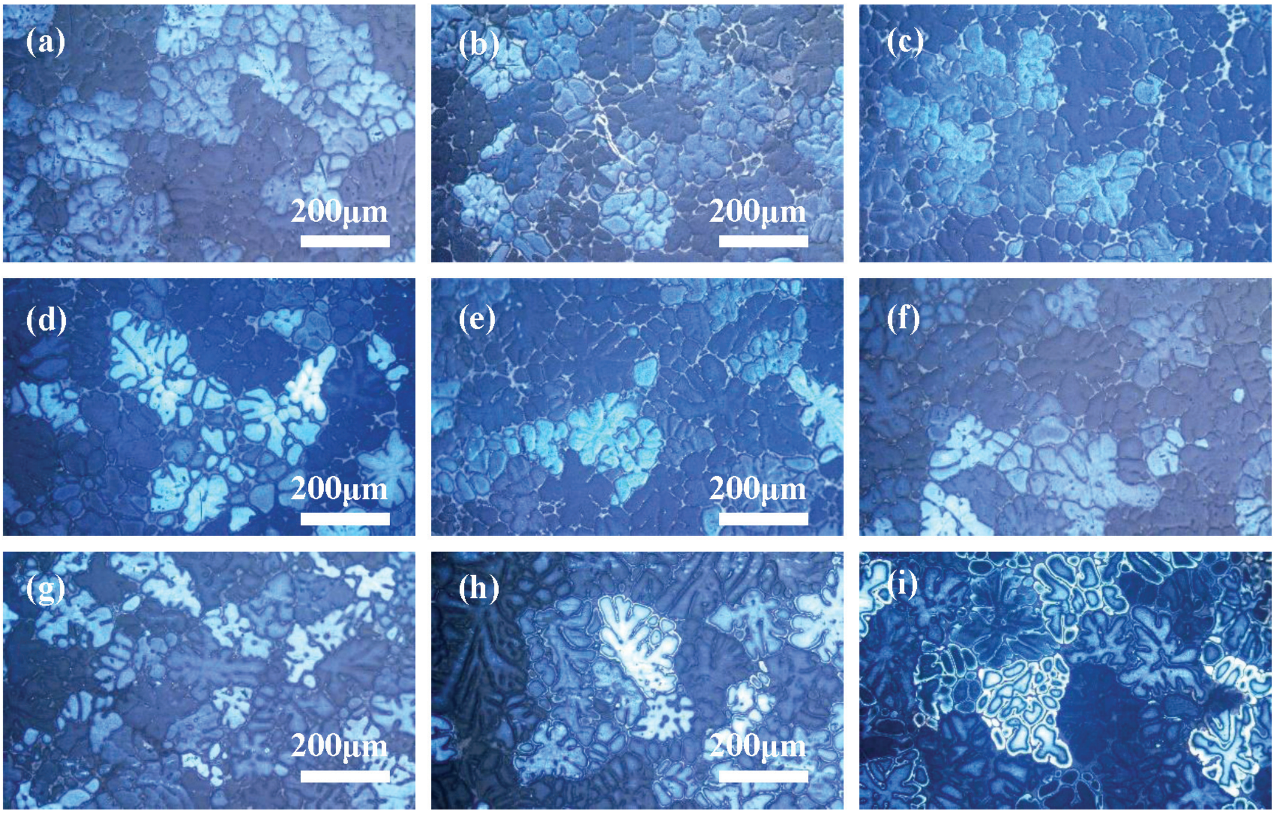

Figure 7: Microstructure of the Al-5Cu alloy ingot after forced water cooling and ultrasonic treat. for 90 s: position 1, b) position 2, c) position 3, d) position 4, e) position 5, f) position 6, g) position 7, h) position 8, i) position 9

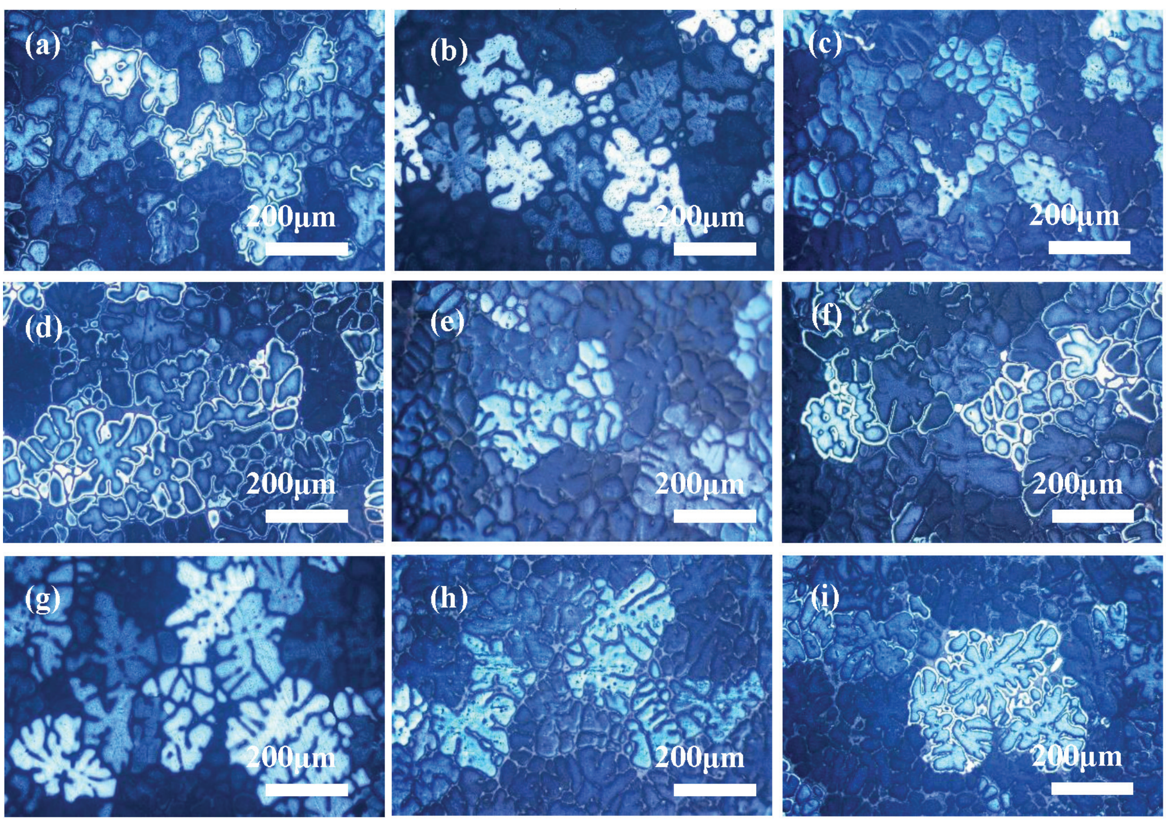

Figure 8: Microstructure of the Al-5Cu alloy ingot after forced water cooling and ultrasonic treat. for 120 s: position 1, b) position 2, c) position 3, d) position 4, e) position 5, f) position 6, g) position 7, h) position 8, i) position 9

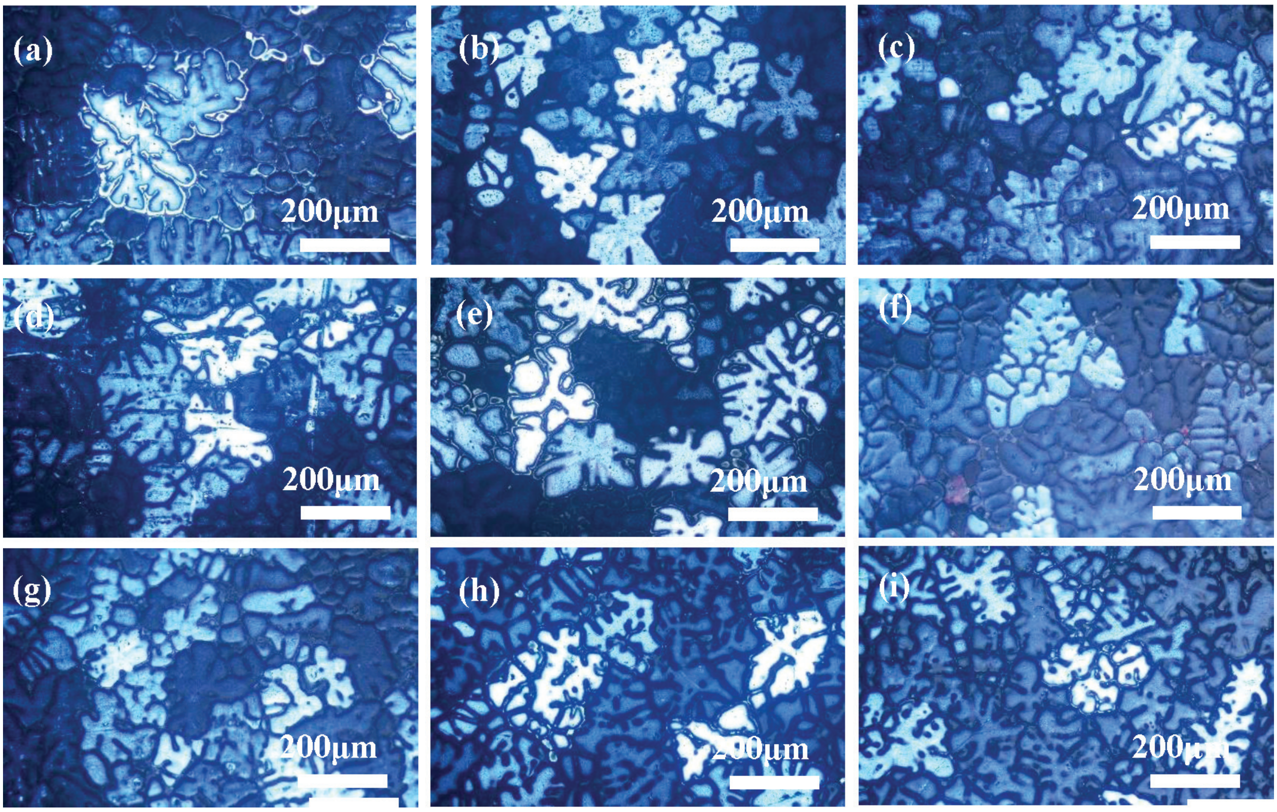

Figure 9: Microstructure of the Al-5Cu alloy ingot under forced water cooling and ultrasonic treat. for 150 s: position 1, b) position 2, c) position 3, d) position 4, e) position 5, f) position 6, g) position 7, h) position 8, i) position 9

bottom of the clay-graphite crucible, the fine equiaxed crystals produced by ultrasonication fall downward and solidify into refined equiaxed grains due to the acoustic flow. The pores below are gradually discharged, so a dense structure starts to form from the bottommost part of the melt. The best results are obtained when this stage is achieved in 120 s. After further extending the ultrasonic treatment time to 150 s, many pores appear due to a decrease in the aluminum liquid temperature, causing the melt viscosity to be too large to hinder the upward floating of the gas.

Figure 12 shows the ultrasonic treatment and forced water cooling mechanism at the bottom of the clay-graphite crucible for this experimental process. When a copper plate is placed at the bottom of the clay-graphite crucible for water cooling, the fine isometric crystals formed by ultrasonic vibration keep falling and solidify quickly. The metal liquid above fills in the gaps between them, thus forming a dense structure. In contrast, in the case of non-forced cooling at the bottom of the clay-graphite crucible, the melt solidifies everywhere although it also has a degassing effect under the ultrasonic action. The cooling rates are comparable in all directions, making most of the melt's gas obstructed by the dendrites formed by the melt before the solidification; it remains in the melt to form pores before it can escape completely.

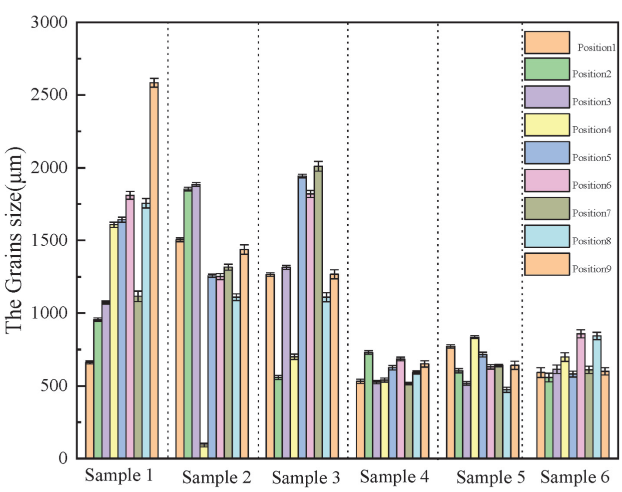

Figure 10: Grain-size distribution of Al-5Cu alloy ingots after ultrasonic treatment

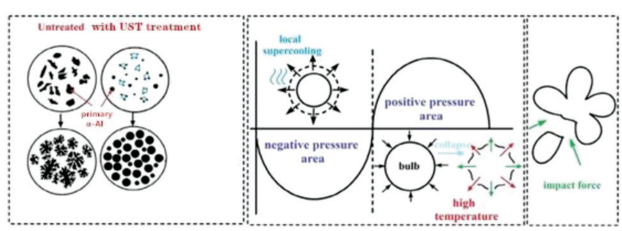

Figure 11: Schematic of grain refinement

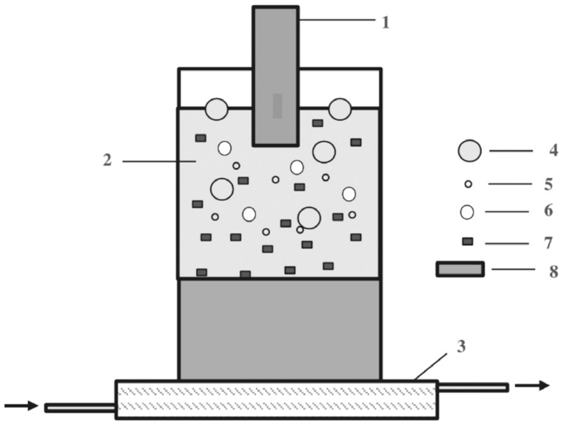

Figure 12: Schematic diagram of aluminum alloy with ultrasonic degassing: 1– ultrasonic probe, 2 – aluminum, 3 – copper plate in water, 4 – cavitation bubbles formed by ultrasonic treatment, 5 – gas in the melt, 6 – hydrogen diffuses into cavitation bubbles, 7 – refined equiaxed grains, 8 – forced cooling zone

Recently, it has been reported that the porosities in the ingots are due to the formation of bifilms in the liquid metal. Campbell[21] indicates that there is a layer of compact oxide films on the melt surface. When a metal melt is poured stably, these oxide films have limited effects on the quality of ingots but they prevent the oxidization of the melt. However, there is turbulence when the melt is poured. It makes the oxide films on the melt surface fold collapse and finally enter into the interior of the metal melt. During this process, air is entrained in the melt, forming bubbles, which is a phenomenon of bubble entrainment.

Firstly, bifilms are very thin, usually a few nanometers, thus, oxide films are broken by the ultrasonic cavitation during the compression and expansion stages of vibration. Hydrogen atoms in the melt enter biofilms; being wrapped and continually diffused or separated in bifilms, they cause an increase in the oxide film volume. Otherwise, these bifilms continually expand and absorb hydrogen from the melt by streaming, and when the pressure in these bifilms reaches a certain level, the bifilms burst and escape from the melt surface. Secondly, these oxide films contain less gas; they continually affect each other and bond with each other due to long-term stirring of acoustic streaming, which also can remove the gas from the melt. Therefore, the bond of oxide films substantially reduces the hydrogen content and, finally, the possibility of porosity formation. It is reported that the partial/local temperature and pressure caused by the cavitation effect are nearly 104 K and 103 MPa, respectively. Therefore, the fragments of the oxide film are the result of ultrasonic cavitation.

4 CONCLUSIONS

The method of combining ultrasonic treatment and forced cooling at the bottom of ingots cannot influence the ultrasonic ingot refinement. Conversely, with increases in the ultrasonic duration, the equiaxed grain occupancy in ingots improves rapidly, even reaching 100 %. In this experiment, we obtained fine and uniform equiaxed crystals in the ingot sections treated with 2000 W for 90–150 s. The USD degassing effect is due to a release of hydrogen in the bifilms, which expand and rise gradually. Finally, they burst on the melt surface, resulting in the effect of degassing.

Acknowledgements

The authors acknowledge the financial support from the Key Project of the Education Department of Liaoning Province [LJKZ1171], China Postdoctoral Fund [2020M680978], Liaoning Province Doctoral Start-Up Fund Project [2020-BS-260], Natural Science Foundation of Fujian Province [2021J05232] and Science and Technology Planning Project of Longyan [2021LYF9012].

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