GRAIN REFINING OF PURE ALUMINUM

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Abstract

Grain refiners are currently used in casting aluminum alloys to reduce the grain size and to produce equiaxed grains during solidification of the alloys. Using inoculants to refine grains makes alloys castable but produces several disadvantages, including particle agglomerates, local defects, and impurities. In contrast, ultrasonic vibrations can be used in the place of grain refiners to refine grains without the disadvantages of using inoculants. In this study, high intensity ultrasonic vibrations were applied during solidification of pure aluminum. The grains of pure aluminum treated using ultrasonic vibrations were compared to the grains obtained using the TiB_2 grain refiner. It was found that the grain size in the pure aluminum ingots subjected to ultrasonic vibrations was comparable to those with the addition of grain refiners.

Introduction

Grain refining is a process by which the crystal size of the newly formed primary phase from the liquid is reduced by either chemical, physical, or mechanical means. The purpose of grain refining is to eliminate columnar grains and produce equiaxed grains [1]. It is an important process that makes aluminum metals and alloys castable using the direct chill (DC), or continuous casting (CC) process. Reducing grain size has the benefits of increasing the cast rate of ingots, improving resistance to hot tearing, minimizing elemental segregation, and enhancing mechanical properties, particularly ductility of aluminum wrought products [1-2]. The main method for grain refining is chemical grain refining. Chemical additions called grain refiners in molten metal reduce the grain size, which thus produces more equiaxed grains and decreases the size of the columnar grains. The addition of grain refiners such as TiB_2 to molten aluminum alloys decreases grain size through the means of heterogeneous nucleation by providing a surface onto which the aluminum solid phase can nucleate [3]. Inoculants in the melt also suppress the growth of columnar grains from the sides of the mold.

Grain refining through chemical means makes alloys castable but introduces several new problems to the melt. The current grain refiners have limited refining capabilities; the most powerful grain refiners commercially available can only achieve a grain size of 100 μm in aluminum alloys [3]. As the grain refiner is kept in the melt, it forms particle agglomerates or clusters of foreign particles in the melt. When the aluminum is cast, the particle agglomerates remain in the metal, which makes further processing steps difficult when working the aluminum [2]. The clusters also cause local defects in aluminum, such as "leakers" in beverage cans and "pin holes" in thin foil [4]. The use of grain refiners also introduces an excess amount of Ti in aluminum, causing a substantial decrease in electrical conductivity of pure aluminum. Pure aluminum is most often used for its property of high conductivity, so the addition of grain refiner reduces a favorable property of the aluminum.

A number of other grain refining methods, in addition to the chemical methods, have been explored in the past century. These methods include using physical fields, such as magnetic and electro-magnetic fields, and using mechanical vibrations [5]. High-intensity, low-amplitude ultrasonic vibration is one of the physical/mechanical means that has been demonstrated for grain refining of metals and alloys without using foreign particles [6-7]. The use of high intensity ultrasonic vibrations during solidification in place of grain refiners can produce smaller grains without the disadvantages of using grain refiners. As the waves go through the melt, cavitations create sites for heterogeneous nucleation to occur while acoustic streaming provides a current to distribute the nucleants around the melt. Acoustic streaming also provides energy to break off the dendrite arms of the solidifying grains, thus reducing grain size and providing new sites on which crystallization can occur. However, most of the experimental results using high intensity ultrasonic vibration has been obtained in aluminum alloys.

This work focused on grain refining of pure aluminum metal solidifying in a copper mold under high cooling rates. The purpose of this research was to evaluate grain refining of pure aluminum for electricity transition applications. Grain refining of pure aluminum solidifying at high cooling rates has proved to be difficult.

Experimental Procedure

High purity aluminum metal directly from a smelter (containing no residual grain refiners) was used. The use of such a pure metal was to isolate the effect of ultrasonic vibration from that of the residual grain refiners in the metal.

An inverted cone Cu mold, shown in Figure 1, was used to cast the pure aluminum (solidification temperature: 660°C) in this study. The copper mold was chosen because pure aluminum has been cast in copper molds for making cables for transmitting electricity. The mold used in this study held up to 250 g molten aluminum and the cooling rates of the metal in the mold were higher than that of the wheel mold used for casting CC billets for making aluminum cables. The largest diameter of an ingot made using this mold was 50 mm (2 inches).

Grain refining using both chemical grain refiners and ultrasonic vibrations was evaluated. In the first experiment, control samples were cast at four different pouring temperatures (760°C, 730°C, 700°C, and 680°C) to establish the baseline for comparing the effect of grain refining.

In the second experiment, the Al-5Ti-1B master alloy (Tibor from KB Alloys) was added in the molten metal for grain refining. The effect of time on the fading of grain refiners was firstly investigated to establish the time for the inoculation of the pure metal. The fading test was done by adding 0.9wt% of grain refiners into the melt and pouring an ingot at 700°C at 20 minutes, 1 hour, 2 hours, 3.5 hours, 5.5 hours, and 20 hours, respectively. Grain refining using Tibor grain refiners was then carried out by adding 0.03%, 0.07%, 0.1%, 0.3%, 0.5%, 0.7%, 0.9%, 1.2%, or 1.5% of Tibor, followed by casting at a pouring temperature of 700°C.

In the third experiment, high intensity ultrasonic vibration was applied continuously on the metal being poured into the copper mold. Four pouring temperatures (760°C, 730°C, 700°C, and 680°C) were tested. The ultrasound system used in the study is illustrated in Figure 1. It consisted of an ultrasonic generator, a transducer, an ultrasonic horn, an ultrasonic radiator/wave guide to transmit ultrasonic vibration into aluminum melt. The transducer was capable of converting up to 1.5 kW of electric energy at a resonant frequency of 20 kHz. The power output of the ultrasonic system was adjustable. The ultrasonic radiator was placed at the metal ingot through a hole in the copper mold. Ultrasonic vibration was applied to the metal as soon as the metal was poured into the copper mold until the metal was solidified.

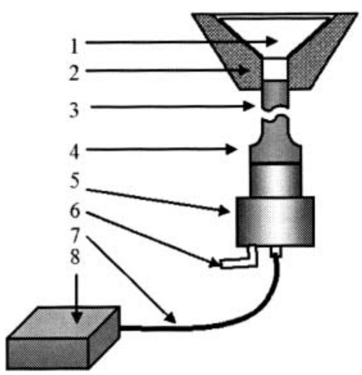


Figure 1: An Ultrasonic Vibrator. (1) Ingot, (2) Metal Mold, (3) Ultrasonic Radiator, (4) Ultrasonic Horn, (5) Transducer, (6) Cooling Air, (7) Cable, (8) Ultrasonic Generator

All the ingots were cut vertically and macro etched with three parts HCl to one part HNO_3 to reveal the grain size. Grain size of the columnar grains was calculated through (length of the longest grain + width of grain) divided by 2. Grain size of the equiaxed grains was measured under an optical microscope at a magnification of 100 times. More than 30 individual grains were measured at the center of each sample. The average grain size was reported.

Results and Discussion

Figure 2 shows the relationship between the grain size and the pouring temperature for pure aluminum and aluminum with an addition of 0.7 wt% Tibor. This amount of grain refiner addition was chosen based on experimental test to eliminate columnar grains in ingot that will be discussed later. Figure 3 illustrates the grain structure of two ingots with and without grain refiners.

Without the addition of grain refiners, the grain structure was columnar in pure aluminum ingots cast at all four temperatures, shown in Figure 3(a). The average grain size of the pure aluminum was around 16867.19 μm at 760°C and 15676.56 μm at 680°C. The grain size in ingot cast at lower temperature was slightly smaller than those cast at high temperatures. With the addition of 0.7 wt% Tibor master alloys, equiaxed grains were obtained in the ingot, shown in Figure 3(b). The average grain size of the ingot was reduced to an average of 397.76 μm at 760 °C and 466.45 μm at 680 °C.

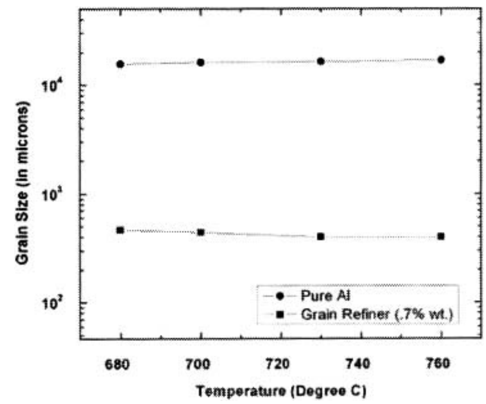
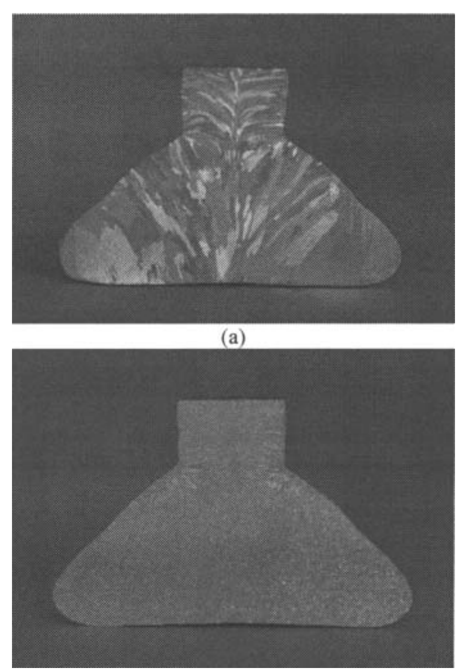


Figure 2: Effect of pouring temperature on the grain size of pure aluminum with and without using Tibor for grain refining.



(b)
Figure 3: Microstructure of pure aluminum poured at 700°C. (a) Without grain refiner, and (b) with an addition of 0.7 wt% of grain refiners.

Experiments on the fading of grain refiners were then carried out. After the Tibor alloy was stirred into the melt, the first ingot was poured immediately. Subsequent ingots were then poured at various times. The results, plotted in Figure 4, indicated that the Tibor master alloy did not show significant fading in the 20 hour time period. The size of the grains was around 275 μm for all times tested. The grain size had a standard deviation of 46 μm in the ingot poured at 0.33 hours after the grain refiner was added and a standard deviation of 36 μm in the ingot poured at 20 hours after the master alloy had been added. The mean grain size was 300.50 μm and 256.20 μm, respectively.

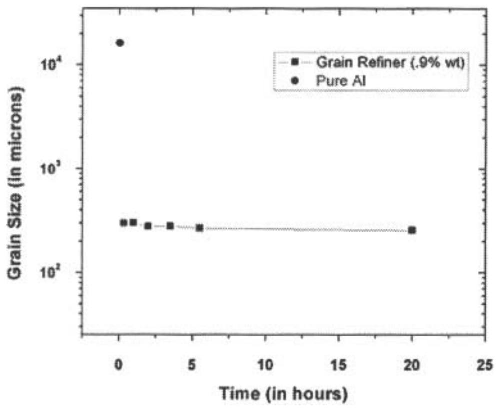


Figure 4: Effect of time on the fading of grain refiners (with an addition of 0.9 wt% of Tibor) in aluminum metal poured at 700°C.

Since fading was not an issue for the Tibor master alloy, chemical grain refining was carried out after being added into the melt for 0.5 hours. Figure 5 shows the curves of grain size vs. the percentage of grain refiner addition and Figure 6 shows the microstructure of selected ingots containing various amount of grain refiners.

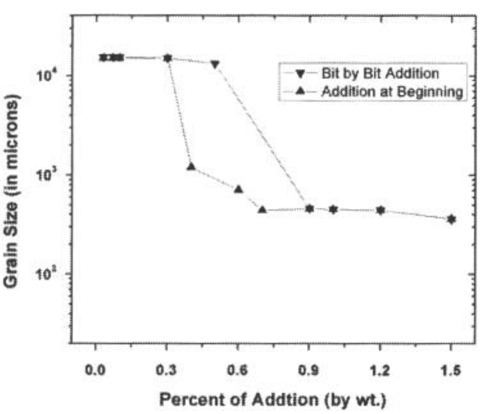
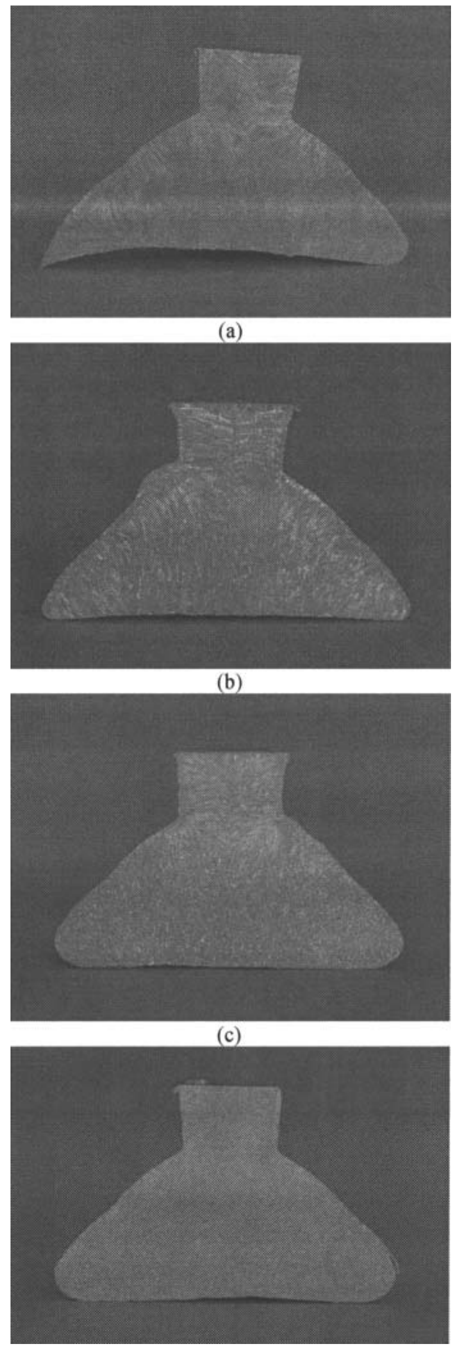


Figure 5: The effect of the addition of grain refiners on the grain size in aluminum Ingots.

Initially, the grain refiners were added into the melt bit by bit. 6.82 kg of pure aluminum was melted. 0.03% Tibor was then added into the melt for pouring an ingot. More Tibor was added into the remaining melt to make ingots containing the next level of grain refiner content until the grain refiner content in the melt reached 1.5%. The results of "bit by bit addition" of grain refiners are shown in the top curve in Figure 5. Surprisingly, no much grain refinement was achieved when the grain refiner addition was about 0.5 wt%, which was 5 times of the recommended amount of grain refiner addition used by the aluminum industry for grain refining. Equiaxed grains were obtained in ingots containing more than 0.9% of Tibor.



(a)

(b)

(c)

(d)

Figure 6: Microstructure of pure aluminum poured at 700°C. (a) 0.3 wt% Tibor, (b) 0.4 wt% Tibor, (c) 0.6 wt% Tibor, and (d) 1.5 wt% Tibor.

The addition method for grain refiners was then changed to "addition at the beginning" by adding the whole amount of grain refiners at once after the melt was prepared. The melt was then held for 0.5 hours before pouring. The bottom curve in Figure 5 shows the experimental results.

No significant refining was observed when the addition of grain refiners was lower than 0.3 wt%, shown in Figure 6(a). The grain size decreased with increasing addition of grain refiners. When the addition of grain refiners was 0.4 wt%, the grains were still columnar but the diameter and the length of the columnar grains shown in Figure 6(b) were much smaller comparing with the control sample shown in Figure 3(a) and the ingot containing 0.3 wt% of grain refiners shown in Figure 6(a). Trace amount of columnar grain can also be seen in the ingot containing 0.6wt% grain refiners shown in Figure 6(c). Equiaxed grains about 400 μm were obtained when the amount of grain refiners added into the melt reached 0.7 wt%, as shown in Figure 3(b). Further increase in the addition of grain refiners decreased the size of the equiaxed grains only slightly. Figure 6(d) shows the equiaxed grain in an ingot containing 1.5wt% of Tibor. Grain size around 300 μm could be achieved in pure aluminum solidified in the copper mold using Tibor as the grain refiner.

It was concluded that the "bit by bit addition" of grain refiners in the melt resulted in an increase in the use of grain refiner for a small amount of melt. Some of the TiB_2 particles could be removed by a layer of dross on the melt surface when a small amount of grain refiners was added at the top of the melt.

The use of continuous ultrasonic vibrations during the solidification process of pure aluminum produced equiaxed grains which are comparable to that with the addition of 0.7wt% grain refiners. Figure 7 shows the difference in the grain sizes in the ingots treated with ultrasonic vibrations and those treated with grain refiners at different temperatures. Figure 8 shows the microstructure of the ingot cast at 760 °C with either a 0.7 wt% Tibor or subjected to ultrasonic vibration.

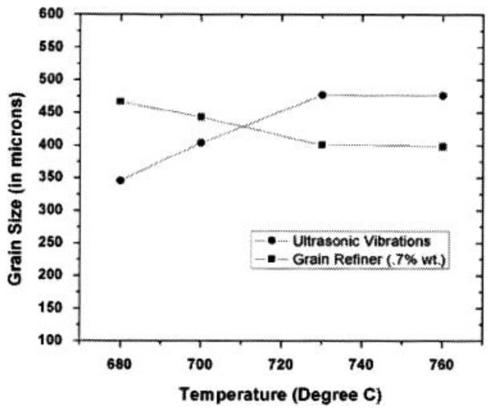
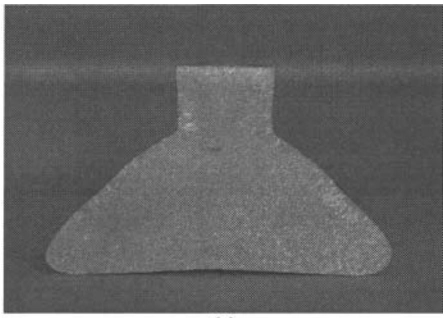
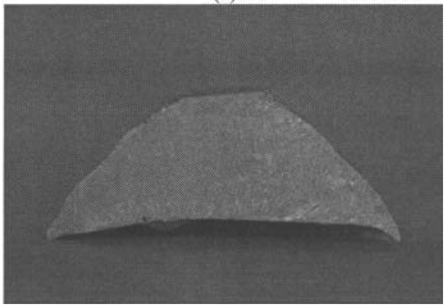


Figure 7: Comparison of the grain size of ingots treated with ultrasonic vibrations (20 kHz) and with the addition of 0.7 wt% Tibor at different temperatures.

At higher temperatures, the use of grain refiners resulted in a smaller grain size. The average grain size of the grain refined ingot at 760°C was 397.76 μm, shown in Figure 8(a), while the average grain size of the ultrasonic vibrations treated ingot was 475.82 μm, shown in Figure 8(b). The standard deviation of the grain sizes was around 169 μm and 95 μm, respectively, which shows that the ultrasonic vibrations produced more uniform grains than did the Al-Ti-B grain refiner. At lower temperatures, the ultrasonic vibration treatment is more effective than the adding of grain refiners. Figure 7, at 680°C, shows that the grain size of the ultrasonic vibration treatment was around 350 μm while the average grain size of the grain refiners was around 460 μm. The standard deviation was 60 μm for the ultrasonic vibration treated ingot and 95 μm for the grain refined ingot, which again shows that the ultrasonic vibrations treated ingots have a smaller and more uniform grain structure.



(a)



(b)

Figure 8: Microstructure of pure aluminum poured at 760°C. (a) 0.7 wt% Tibor, and (b) With ultrasonic vibration.

The pouring temperature affects the grain size in ingots subjected to ultrasonic vibration. The grain size decreased with decreasing pouring temperature. This trend is in good agreement with experimental results obtained in aluminum alloys subjected to ultrasonic vibration [6]. Optimal grain refining effect using ultrasonic vibration can be achieved when the melt is poured into a mold at temperatures within 10°C above the liquidus temperature of the alloy. The minimum pouring temperature in this study was slightly higher than the optimal pouring temperature for pure aluminum. Still equiaxed grains have been obtained in the ingots subjected to ultrasonic vibrations.

The grain size in pure ingots subjected to ultrasonic vibration during its solidification stage is still much larger compared to that in A356 alloy. Jian et al [8] tested grain refining in A356 alloy using an identical ultrasonic system. They poured A356 alloy melt at 634°C, 20°C above its liquidus, into a similar copper mold as used in this study, and obtained spherical grains. The size of the spherical grains was in the range of 40 μm to 70 μm. This was almost one order of magnitude smaller than the size of equiaxed grains obtained in this study on pure aluminum ingots. The combination of using both grain refiners and ultrasonic vibration was then tested in the present study. The results indicated that no much further grain refining can be achieved when the grain size was about 300 μm.

Ultrasonic grain refining is closely related to cavitation and acoustic streaming in the melt. Qian *et al* [9] summarized that two types of mechanism have been proposed for ultrasonic grain refining in metals and alloys. The first type of mechanism is based on the cavitation enhanced nucleation and the second on the cavitation induced fragmentation of dendrites [6-7, 10-11]. Jian et al [7] vibrated an A356 alloy starting at various temperatures and found that grain refining was more effective in forming spherical grains when the vibration was applied near the liquidus temperature of the alloy. They suggest that cavitation enhanced nucleation may play a more important role in reducing the grain size using high intensity ultrasonic vibration. Han et al [7,12] suggested that a rapid expansion of cavitation bubbles during the rarefaction phase of the ultrasonic vibration undercools the liquid at the bubble-liquid interfaces, resulting in nucleation on the bubble surfaces. The collapse of these bubbles during the compressive phase of the vibrations distributes the newly formed grains throughout the melt for grain refining. Another mechanism based on cavitation enhanced nucleation considerations assumes that cavities and cracks on the substrate surfaces that pre-exist in the melt can be wetted by the shock waves and acoustic streaming, enabling these substrates to act as effective nucleation sites [6].

It is unclear which mechanisms operate for ultrasonic grain refining of pure aluminum. The addition of 0.3 wt% Tibor seems not enough to eliminate columnar grains. It is difficult to believe that ultrasonic cleaning of the surfaces of the solid substrate can play a more effective role in grain refining than the addition of 0.3 wt% Tibor. Also the grain size in pure aluminum subjected to ultrasonic vibration is much larger than that in A356 alloy. It is unclear why ultrasound induced nucleation is not that effective in pure aluminum compared with that in A356 alloy. The growth restricting effect of solute elements in A356 alloy must have played an important role in retarding the growth rates of the equiaxed grains that leads to the formation of small and spherical grains in the A356 alloy.

Conclusion

It is difficult to eliminate columnar grains in a pure aluminum ingot solidified in a copper mold. It requires an addition of 0.7 wt% Tibor to totally eliminate columnar grains in ingots cast under the experimental conditions of this study. This amount is much higher than the amount of grain refiners needed to eliminate columnar grains in commercial aluminum alloys, typically 0.1wt%.

High intensity ultrasonic vibration can effectively eliminate columnar grains in pure aluminum ingots over a wide range of pouring temperature between 680°C to 760°C tested in this study. The size of the equiaxed grains is comparable to that with an addition of 0.7 wt% of Tibor grain refiners under the experimental conditions of this study. The grain size in ingots subjected to ultrasonic vibration decreases with decreasing pouring temperatures. This is an indication that the mechanism by which grains are refined is related to the nucleation and the survival of the small nuclei, or embryos of grains formed under the influence of high intensity ultrasonic vibration.

Acknowledgements

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