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GRAIN SIZE CONTROL IN Al-4.8 wt.% Cu ALLOY BY COMPUTER-AIDED COOLING CURVE ANALYSIS

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Abstract

Generally Al–Ti and Al–Ti–B master alloys are added to the aluminium alloys for grain refinement. The cooling curve analysis (CCA) has been used extensively in metal casting industry to predict microstructure constituents, grain refinement and to calculate the latent heat of solidification. The aim of this study was to investigate the effect of grain refinement on the grain size of Al-4.8 wt.%Cu alloy by cooling curve analysis. To do this, alloy was grain refined by different amount of Al-5Ti-1B master alloy and all samples were solidified at constant cooling rate of 0.19 °C/s. The temperature of the samples was recorded using a K thermocouple and a data acquisition system connected to a PC. The results show that the segregating power of Ti is very high and it segregates to the nucleant–liquid interface which leads to constitutional supercooling within which other nucleant particles get activated for nucleation. Other results show that with considering the changes in the primary undercooling (ΔT_{RU}) as the main factor to determine the effectiveness of grain refinement process, it was found that by grain refinement, the value of undercooling decrease was approximately zero.

Keywords: Thermal analysis, grain size, grain refinement, cooling curve

Introduction

It is well known that metals and alloys usually solidify with coarse columnar grain structure under normal casting conditions unless the mode of solidification is carefully controlled. It is possible to develop fine equiaxed grains in the as cast structure either by increasing the number of nucleation sites or by grain multiplication [1,2]. One major advantage of generating a fine grain size is enhanced mechanical properties. Apart from the improved mechanical properties, grain size plays a significant role in controlling the soundness of the casting, preventing formation of serious casting defects such as porosity and hot tears that can severely impair product and process performance [2]. Grain refinement is generally understood to be achieved by providing numerous potent substrates for nucleation of crystals during solidification, combined with assuring

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a sufficient driving force for activation of a large number of nucleants present in the liquid. Grain refinement has attracted significant interest in the aluminum industry for several decades and grain size is generally well controlled in commercial operations by addition of master alloys which contain different combinations of Ti-B, Ti-C and Al-B [3-6]. The effect of solute on grain size is as important as that of nucleant particles since the segregation of solute during solidification plays a significant role in controlling the growth and survival of the nucleated crystals [7,8].

The thermal analysis is widely used in the evaluation of processing of aluminum alloys because it can provide several pieces of information regarding the alloy. Thermal analysis is based on the fact that the thermal events on a heating or cooling curve are directly related to phase transformation occurring in a sample. Many techniques are available to investigate the solidification of metals and alloys. Some of them were standardized such as DTA and DSC. Although these techniques are very accurate and well documented, they are inadequate for industries to investigating solidification of metals and alloys. The other way for investigating solidification of metals and alloys is the cooling curve analysis method. This technique is based on recording and analysis of the temperature versus time data collected during the solidification of the sample. In recent years computer-aided cooling curve analysis (CA-CCA) has been used to determine thermo-physical properties of alloys, latent heat and solid fraction. Being very simple to setup it can be widely used, especially in industries [9-11]. It just needs to place a thermocouple in the melt and allow the melt to solidify while the temperature is recorded as a function of time. The cooling curve does not always indicate all the reactions occurring during solidification of a casting clearly, due to the small amounts of heat evolved by certain phase transformations and more sensitive techniques should be developed. It has been found that the first derivative of the cooling curve can be employed to emphasize small heat effects not resolved on the cooling curve itself [12,13].

The aim of this study was to investigate the effect of grain refinement on the grain size of Al-4.8 wt.%Cu alloy by cooling curve analysis. To do this, alloy was grain refined by different amount of Al-5Ti-1B master alloy.

Experimental Procedure

Binary Al-4.8 wt.% Cu alloy was used in this study. About 300 g of alloy was melted in a graphite crucible in an electric furnace. After some preliminary experiments superheat temperature was selected as 740°C to reduce its harmful effects. Commercially available Al-5Ti-1B was used as grain refiner which was added to the melt at the last stage of melting. The amounts of grain refiner added to the melt was 0.05, 0.1 and 0.12 wt.%. For homogenous distribution of the grain refiner in the melt, it was stirred with and alumina rod for 20 s and held in 720°C for 5 min. The melt was then cast in a 100-200 °C preheated sand mold and solidified at cooling rate of 0.19 °C/s [14,15]. The sand mold had a cavity 40 mm in diameter and 50 mm in depth, and the wall thickness was 10 mm. To check the effectiveness of the grain refinement, the undercooling criteria as shown in Figure. 1 was used. The temperature of the sample was recorded by a K thermocouple with the diameter of 0.15 mm placed at the center of the mold and an Advantech 4718 data acquisition system connected to a PC.

Samples for microstructural examination were taken from a location close to the thermocouple tip. Samples were ground, polished and etched with Keller's (10ml HF, 20ml HNO3, 20ml HCl, and 50 ml distilled water) reagent. The microstructure was examined by an Olympus BX60 optical microscope.

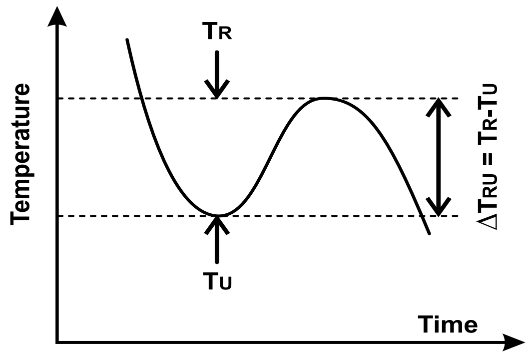

Figure 1. Undercooling criteria to examine the effectiveness of the grain refinement process.

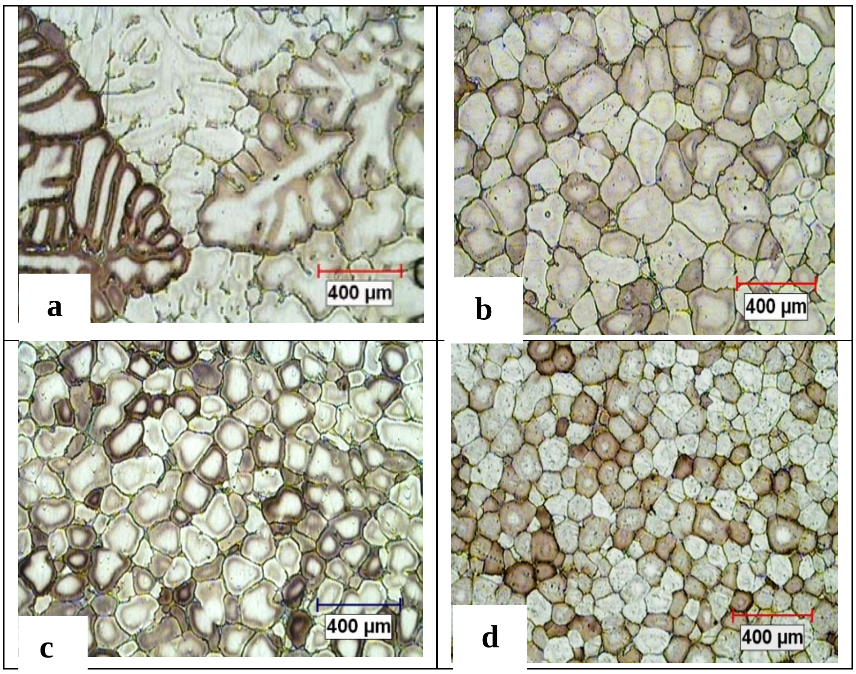

Figure 2. Micrographs of samples with different grain refinment level, a) Base alloy, b) 0.05 wt%, c) 0.1 wt%, d) 0.12 wt%.

Figure 3 shows the effect of grain refiner additions on average grain size of Al-4.8 wt.%Cu alloy. Grain size decreased rapidly with increasing grain refiner content up to 0.05

wt.%, while further increases in grain refiner content, up to 0.12 wt%, resulted in a more gradual reduction in grain size. However, by further addition of grain refiner (>0.1 wt.%) to the alloy, the average grain size almost remains constant and the excess addition of the grain refiner does not have a considerable effect on the macrostructure of the alloy.

The grain refinement ability of Al-5Ti-1B in Al-4.8 wt.%Cu is demonstrated if Figure 4. It is observed that there was a reduction of the sample grain size with the addition of the grain refiner. Two mechanisms have been proposed for the grain refining process [16]. 1: Nucleant effects in grain refinement, 2: Solute effect.

The presence of some particles like $TiAl_3$, TiB_2 and AlB_2 are known to be effective for grain refinement while $TiAl_3$ is known to be a potent nucleating site for aluminum [16-18]. Jones and Pearson [19] have demonstrated that a ternary Al–Ti–B grain refiner is 4–5 times more efficient than a binary Al–Ti grain refiner having approximately the same titanium content, in terms of the decrease in grain size.

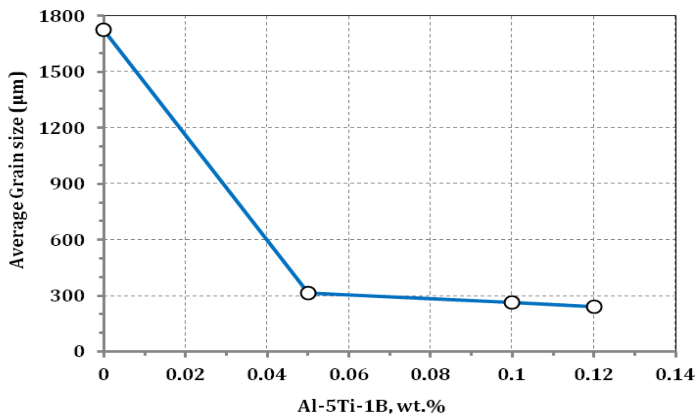

Figure 3. Grain size behavior as a function of grain refiner content in Al-4.8 wt.%Cu.

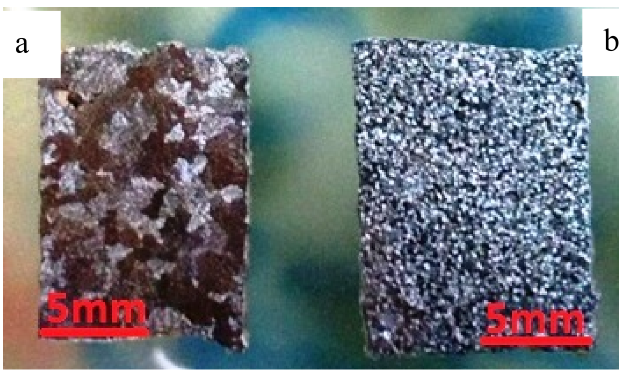

Figure 4. Macrostructure of Al-4.8 wt.%Cu alloy, a: base alloy, b: after addition of Al-5Ti-1B master alloy.

2-3 Solute effect

Addition of solute generates constitutional undercooling in a small diffusion layer ahead of the advancing solid/liquid interface which restricts growth of the crystal since the diffusion of the solute occurs slowly, thus limiting the rate of crystal growth. In addition, further nucleation occurs in front of the interface (in the diffusion layer) because nucleants in the melt are more likely to be activated in the constitutionally undercooled zone where there is an additional driving force, i.e. undercooling, for nucleation. The effect of solute has been investigated experimentally in various aluminium alloy systems [20-22]. This effect is usually explained in terms of the growth restriction factor (GRF). The growth restriction factor can be calculated using binary phase diagrams. GRF equals:

$$\sum_i m_i C_{0,i}(k_i - 1) \tag{1}$$

where m_i is the slope of the liquidus line, k_i is the distribution coefficient and $C_{0,i}$ is the initial concentration of element *i* when the liquidus and solidus are assumed to be straight lines. The growth restriction factor is thus a measure of the segregating power of all elements ahead of the solidification front [20,23]. It should be noted that this equation is only applicable for solute present in dilute concentration levels. The GRF values for different solute elements in aluminium alloys have been determined using binary phase diagrams and are shown in Table 1. The $mC_0(k - 1)$ value of the solute element has been used to determine how important a particular solute addition is on grain size**.** For titanium, $mC_0(k - 1)= 245.6$ while, for copper, it is only 2.8; hence, small amounts of titanium will produce a large decrease in grain size, while much larger amounts of copper are required for a similar effect [24].

Table 1. Segregating power of some elements in aluminium [24].

Solute Element	K_i	m_i	$(K_i-1)m_i$	Maximum Concentration (wt.%)	Type of Reaction
Ti	9	30.7	245.6	15	Peretectic
Ta	2.5	70	105	0.1	Peretectic
V	4	10	30	0.1	Peretectic
Hf	2.4	8	11.2	0.5	Peretectic
Mo	2.5	5	7.5	0.1	Peretectic
Zr	2.5	4.5	6.5	0.11	Peretectic
Nb	1.5	13.3	6.6	0.15	Peretectic
Si	0.11	-6.6	5.9	12.6	Peretectic
Cr	2	3.5	3.5	0.4	Peretectic
Ni	0.007	-3.3	3.3	6	Peretectic
Mg	0.51	-6.2	3	3.4	Peretectic
Fe	0.02	-3	2.9	1.8	Peretectic
Cu	0.17	-3.4	2.8	33.2	Peretectic
Mn	0.94	-1.6	0.1	1.9	Peretectic

3-3 Thermal Analysis

The cooling curve and its first derivate curve for the no-refined and refined samples are presented in Figure 5 a-b. According to cooling an derivative curves, it can be seen that the solidification consisted of two steps: at first step the solidification starts by development of a dendritic network. First inflection point on the curves results from the evolution of the heat of solidification of the α_{Al}. At the second step at low temperatures, however, solidification is followed by a eutectic reaction [9]. During solidification two phenomena occur simultaneously, *i.e.* nucleation and growth. As the temperature decreases (increasing the level of undercooling) the nucleation rate rises and, at the same time, growth of the formed nucleus will release the latent heat of solidification which reduces the cooling rate until it reaches to a minimum temperature; recalescence temperature. Thereafter the temperature rises again to its normal growth temperature due to release of more latent heat. After the end of recalescence, the nucleation process is completed and no more new α_{Al} particles will form.

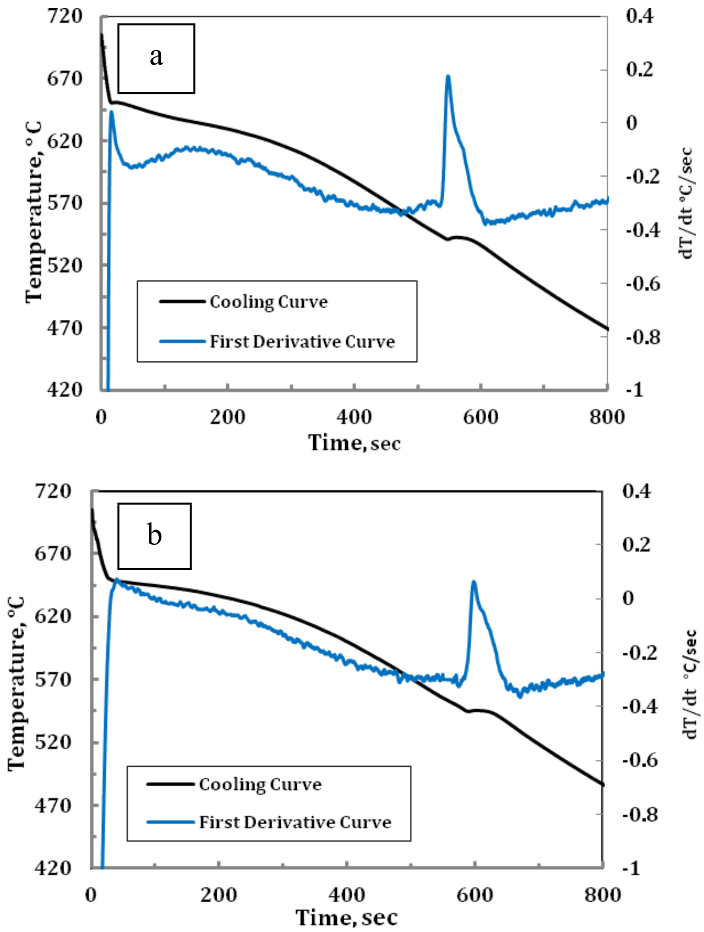

Figure 5. Cooling curves and first derivative for samples, a: no-refined, b: refined .

The occurrence of the recalescence depends on the density of the nucleation sites. If the melt is no-refined, a greater driving force in the form of undercooling will be necessary to start solidification of the primary grains. Once nucleation is completed, evolution of latent heat tends to decrease the undercooling and recalescence occurs. The apparent undercooling then is defined as the difference between the minimum temperature relating to the beginning of solidification and the maximum temperature reached by the alloy during solidification. This apparent undercooling, ΔT_{RU}, is shown in Figure 1. The beginning of cooling curve for the no-refined and refined samples is shown in Figure 6. It can be seen that the grain refinement affects the shape of the cooling curve at the beginning of solidification. In refined sample, for example, with a sufficient number of effective nuclei, nucleation will occur in a shorter time. This indicates that there is almost no energy barrier for nucleation and that the grain size of the casting will be fine. The shape of the cooling curve at the beginning of the solidification process gives a good indication of the number of nuclei present in the melt. When there are a great number of nuclei, the curve exhibits low undercooling, (as illustrated in the Figure. 6 by dotted line). When there are few nuclei, a higher extent of undercooling may be expected as can be seen for no-refined sample (Figure. 6, solid line). This Figure clearly shows that the melt needs to be undercooled before nucleation of new α_{Al} particles occurs. It should be noted that ΔT_{RU} does not represent the undercooling ΔT, required for the nucleation of solid. ΔT_{RU} is only the point at which heat loss from the sample becomes less than the rate of heat generation from latent heat evolution [25].

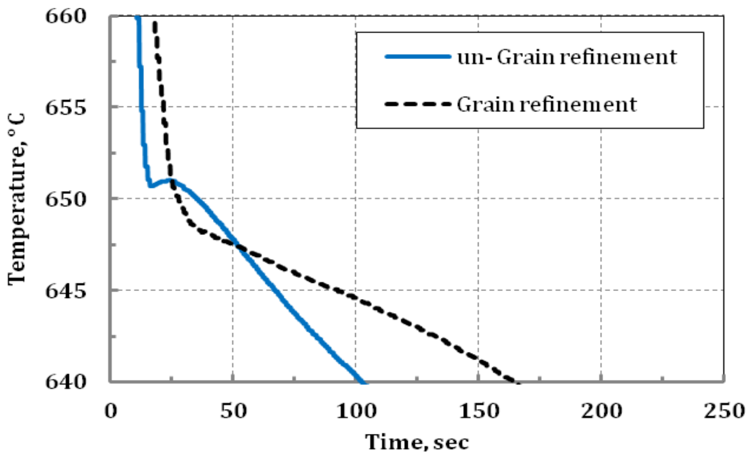

Figure 6. Comparison between the cooling curves of no-refined and refined samples at the beginning of the solidification.

Conclusion

Undercooling parameter from the cooling curve has been routinely used for the quality process control giving a prompt information about efficiency of grain refining additions into aluminium melt. Smaller ΔT_{RU} parameter, means higher potency of master alloy leading to smaller casting grains in as-cast structure. In a well refined alloy, for example, with a sufficient number of effective nuclei, nucleation will occur in a shorter time, and a undercooling approximately decreases to zero. This indicates that there is almost no energy barrier for nucleation and that the grain size of the casting will be fine. If the melt is not grain refined or partially refined, a greater driving force in the form of undercooling will be necessary to start solidification of the primary grains.

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