Effect of Titanium and Boron on the Stability of Grain Refinement of Al-Cu Alloy

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Abstract

The present research was conducted on thin-walled castings with 5 mm wall thicknesses. This study addresses the effect of the influence of different master alloys, namely: (1) Al-5%Ti-1%B, (2) Al-5%Ti and (3) Al-3%B, respectively on the structure and the degree of undercooling ($\Delta T_\alpha = T_\alpha - T_{min}$, where T_α - the equilibrium solidification temperature, T_{min} - the minimum temperature at the beginning of α(Al) solidification) of an Al-Cu alloy. The process of fading has been investigated at different times spent on the refinement treatment ie. from 3, 20, 45 and 90 minutes respectively, from the dissolution of master alloys. A thermal analysis was performed (using a type-S thermocouple) to determine cooling curves. The degree of undercooling and recalescence were determined from cooling and solidification curves, whereas macrostructure characteristics were conducted based on a metallographic examination. The fading effect of the refinement of the primary structure is accompanied by a significant change in the number (dimension) of primary grains, which is strongly correlated to solidification parameters, determined by thermal analysis. In addition to that, the analysis of grain refinement stability has been shown with relation to different grain refinements and initial titanium concentration in Al-Cu base alloy. Finally, it has been shown that the refinement process of the primary structure is unstable and requires strict metallurgical control.

Keywords: Quality management, Metallography, Al-5Cu alloy, Fading effect, Primary grains of α(Al)

1. Introduction

Aluminum-copper alloys are important engineering materials used in applications requiring high tensile properties. Al-Cu alloys are extensively in use for high quality castings in industrial applications, such as aviation or automotive. The main factors that influence the structure of Al-Cu alloys are chemical composition [1-2], cooling rate [3-5], liquid treatment (melt refining, impurity control and grain refining) [6], and heat treatment [2, 6-9]. A refined dendritic structure, small secondary dendrite arm spacing (SDAS) and less dendrite branching leads to a homogeneous structure [10]. Grain refinement is a function of the time which elapses from the introduction of the modifier (grain refiner) to liquid metal. After dissolving the modifier, this liquid metal is in

the so-called super-modified state. This condition is maintained for a certain period of time, depending on the type and usage of a modifier, melt temperature and slag used, furnace atmosphere, mixing of the melt and the chemical composition of liquid metal. This characteristic feature of grain refinement and inoculation processes are called fading effects. That is why the so-called grain refiner and inoculants differ from microalloying, the effects of which are independent of time [11-12]. Literature provides limited data [11-14] on the primary structure - maximum undercooling relationship in the Al-5Cu system as a function of time (fading effects) lapsed from the grain refinement treatment. The purpose of this study was to investigate the effect of stability in modifying the primary structure - dendrites of α(Al) phase in the aluminum-copper type of alloys, using selected master alloys (1) Al-5%Ti-1%B, (2) Al-5%Ti and (3) Al-3%B as well as examining

the effect of the initial titanium content in the base alloy on the stability of these effects.

2. Experimental Procedure

Within the framework of this study, four Al-5Cu alloys were melted. The experimental melts were prepared in a medium-frequency induction furnace. The furnace charge consisted of the following charge materials: aluminum (purity: 99.85%), AlCu50, AlMn75, and AlTi75. After the charge was melted, the covering and refining flux (1.2 wt.%) was added onto the metal surface. The liquid metal has been heated to 750°C and then grain refiner (master alloy) was introduced to the melt. This process was conducted using the following master alloys: AlTi5B1 (alloy I and IV), ATi5 (alloy II) and AlB3 (alloy III) the quantity of was equal to 0.2% by weight of the metal. The base Al-5Cu alloy has the following chemical composition: Mn: 0.88-0.98; Ti*: 0.008-0.012; Si: 0.022-0.075; Fe: 0.078-0.082; Al: Balance. The liquid metal was cast into dried molds made of a traditional molding sand with bentonite, of standard Y-shaped type ingots (according to ASTM A536-84) with a wall thickness of 5 mm in the bottom part. The pouring temperature was 750°C. The base alloy was poured and then grain-refined alloys after the time of 3, 20, 45 and 90 minutes respectively, were obtained from the addition of the master alloy.

Before pouring, the thermocouples of type S (Pt-PtRh10) were placed in the geometric centers of the bottom part of the ingots. An Agilent 34970A multi-channel electronic module was used to record temperature with a time step of 0.02 s.

Samples for metallographic examinations were taken from the bottom part of the ingots. They were grinded, polished, and electrolytically etched, using Barker's reagent (0.5 % HBF4). During the etching, an electric current at 30 V was fed for a period of 1-2 min in accordance with the ASTM E407-07 Standard. The metallographic examination was carried out with an optical stereomicroscope using polarized light at a magnification of 20x. The average diameter of the primary grain α(Al) and (by using the planimetric method) the surface grain density N_A was determined.

3. Results and Discussion

3.1. Thermal Analysis

Figure 1 shows the cooling curves recorded for both base alloys (Alloy no. I and IV) and also grain-refined alloys ie. after 3 minutes from the introduction of these master alloys.

The addition of the grain refiner reduced the maximum degree of undercooling ($\Delta T_\alpha = T_\alpha - T_{min}$, where T_α - the equilibrium solidification temperature, T_{min} - the minimum temperature at the beginning of α(Al) solidification) as compared to the base alloy.

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*Not modified alloy IV differed in the amount of AlTi75 alloy additions in charge materials in order to obtain different titanium contents. In this alloys amount of titanium was set to 0.24 % mass.

AlTiB master alloy (Alloy I) decreased the degree of undercooling by 7.9 °C, and significantly reduced recalescence by 1.3 °C as compared to the base alloy. In the case of the master alloy Al5Ti (Alloy II) the degree of undercooling decreased by 5.6 °C, and recalescence had not changed significantly. In the case of the master alloy AlB3 (alloy III) the smallest change in the maximum degree of undercooling was observed, which amounts to 4.1 °C while recalescence decreased by only 0.3 °C.
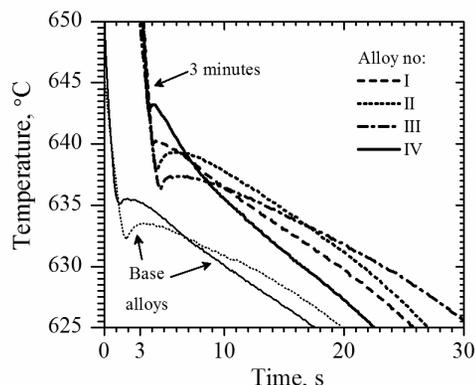
Fig. 1. Cooling curves recorded for base alloys (no. I and IV) and after 3 minutes from the introduction of master alloys (I to IV)

The addition of titanium to the base alloy at 0.24% (alloy IV) resulted in a reduction in the degree of undercooling of this base alloy by 2.9 °C, and recalescence by 1 °C. Modification of this alloy (IV) using AlTi5B1 master alloy, significantly reduced the degree of undercooling by 7.5 °C and led to a slight increase in recalescence by 0.2 °C.

3.2. Metallographic Examinations

Figure 2 shows selected macrostructures of analyzed alloys, (a) alloy I after 3 minutes of grain refinement (b) alloy III after 3 minutes of grain refinement, (c) alloy II in its initial state, (d) melt II 3 minutes after grain refinement (e) alloy IV in its initial state, and (f) alloy IV after 3 minutes of grain refinement.
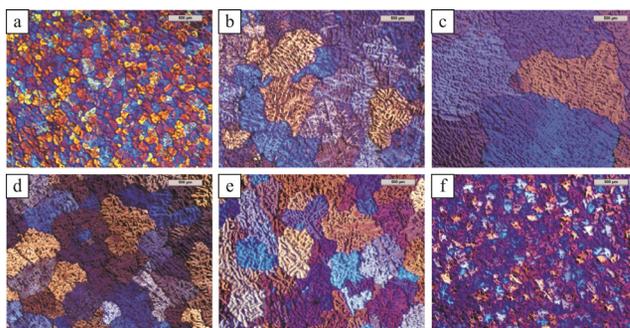
Fig. 2. Macrostructures: (a) alloy I after 3 minutes of grain refinement (b) alloy III after 3 minutes of grain refinement, (c) alloy II in its initial state, (d) melt II 3 minutes after grain refinement (e) alloy IV in its initial state, and (f) alloy IV after 3 minutes of grain refinement

Figure 3 shows the change in the average grain diameter (Fig. 3a), the surface density of the grain (Fig. 3b), and the relative effectiveness of grain refinement ** (c) as a function of time from the addition of the master alloy.
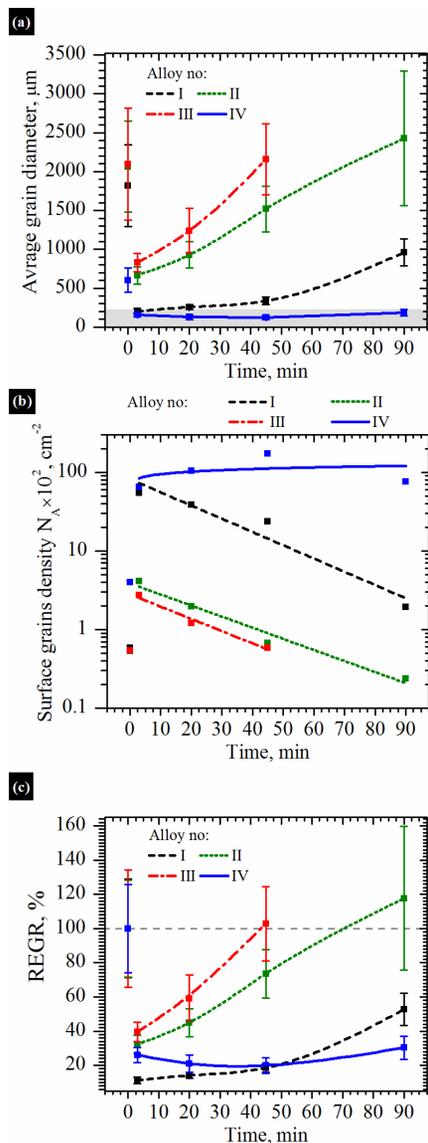
Fig. 3. Variation in the average grain diameter (a), the surface density of α(Al) grains (b) and the relative effectiveness of grain refinement (c) as a function of time, from the addition of the master alloy

This study was aimed at determining the effectiveness of the grain refiners in the Al-5Cu alloys. Research was performed for two variants of its initial state. In the first variant, the effectiveness of grain refiners containing no titanium in the base

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** Relative effectiveness of grain refinement (REGR) was determined using the following equation: $REGR = d_i/d_w \times 100\%$, where: d_i – given grain diameter, d_w – grain diameter determined in the base alloy.

alloy was investigated, whereas in the second variant, the studies deal with the effectiveness of grain refiners of Al-5Cu alloys when titanium was introduced to the base alloy at the level of 0.24% Ti. From Fig. 3a it follows that, in the case of the first variant, most effective (in terms of grain refinement) is the grain refiner AlTi5B1 (used in Alloy I). As a result of its actions, the smallest mean grain diameter was attained as compared to alloy II (in which AlTi5 was used as a grain refiner) and alloy III (in which AlB3 was used as a grain refiner). The effect of grain refiner is manifested by a decrease in the size of primary α(Al) grains in this alloy. This effect holds without significant changes in up to 45 minutes from the addition of the master alloy. At this time, a significant increase in grain size (almost double) with respect to its state just after the addition of the grain refiner (the so-called super-modified state) can be observed. The second in terms of effectiveness of grain refinement is the master alloy AlTi5 (used in alloy II). In this case, the effect is not as stable as in the case of master alloy AlTi5B1.

The least effective is master alloy AlB3 (used in alloy III). Note, the high efficiency of master alloy AlTi5B1 in the second variant (Fig. 3). In this case, the addition of titanium to the base alloy (alloy IV) resulted in the highest degree of refinement and also the longest in terms of duration time, which is continuous during the entire test time (90 minutes).

Variation in the surface density of the grains as a function of time which has elapsed since the grain refinement treatment, is shown in Figure 3b. These results are consistent with changes in the size of primary grains (Fig. 3a).

Figure 3c shows the relative effectiveness of grain refinement (REGR) in comparison to its initial state. An analysis of this parameter (REGR) indicates that the maximum efficiency was attained for the grain refiner in the first variant, ie. when titanium is not present in the base alloy (alloy I). In this case, the percentage reduction amounted to 11.5 % of the original grain size, while in the case of the second variant (alloy IV) 26%, although in alloy IV, the smallest primary α(Al) grain was attained. These results indicate the importance of the initial concentration of titanium in the base alloy as well as the final Ti/B ratio, taking into account their content in master alloys. The results of these studies demonstrate that analysis of the effectiveness of grain refiners should take into account the initial content of both titanium and boron in the initial state (ie. in the base alloy).

The next important aspect, after the degree of grain refinement, is the stability in time of the modification (ie. grain refinement) process.

From Fig. 3a and 3b it is clear that the initial content of titanium plays an important role in the stability of modification effects. In the case of the second variant (alloy IV) this stability is, as already mentioned, much higher than in the case of the first variant. Taking these two variants into account, the difference in primary grain size after 90 minutes, amounts to 776 μm (Fig. 3a).

Figure 4 shows the relationship between surface density and average grain diameter as a function of the maximum degree of undercooling for the initial state (ie. for this base alloy) and for the alloy received after 3 minutes after the grain refinement treatment (using aforementioned master alloys ie. AlTi5B1, AlTi5, AlB3). From Fig. 4 it follows that the smaller the maximum degree of undercooling, the smaller the resulting primary α(Al) grains, which correspond to a higher surface grain

density. This trend is characteristic for castings with walls of the same thickness, but of various physicochemical states of liquid metal, which depend on the type and quantity of the grain refiner added, and the time and liquid metal temperature, slag and furnace atmosphere, molten metal mixing, or even the chemical composition of the alloy [4-5, 14-16].
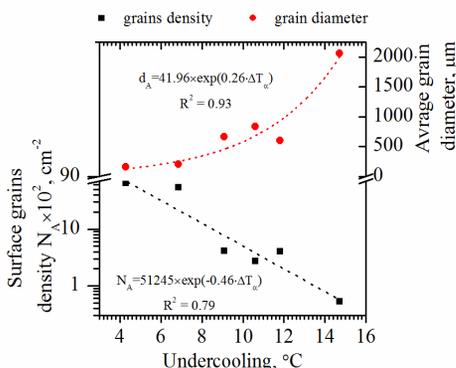
Fig. 4. Primary grain diameter and the surface grain density vs the maximum degree of undercooling

From the above analysis, it is shown that the nucleation process of Al-Cu alloy is strongly related to the type of substrates for heterogeneous nucleation. This study suggest, that the most effective nucleation effect can be achieved by the simultaneous presence of titanium and boron, which contribute to the high efficiency of grain refinement, together with long-term stability of the modification (grain refinement) process.

4. Conclusions

The following conclusions can be drawn from the present experimental investigation:
1) The most effective modifying results, exhibiting high efficiency of grain refinement and long-term stability, was obtained by the simultaneous presence of titanium and boron in Al-Cu alloys.
2) This paper shows the relative effectiveness of grain refinement (REGR) of primary grains, for selected master alloys and two different initial states of liquid metal (related to different initial contents of Ti in base alloys).
3) Stability effects of grain refinement (number of grains, ΔT_m, recalescence) depends on the initial content of titanium and the type of modifier used. It can be concluded that the fading effects of grain refinement of the primary structure is accompanied by a change in the number of $\alpha(Al)$ grains, ΔT_m and recalescence. The refining process is thus highly unstable and requires strict metallurgical control.

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