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Effect of Modification and Cooling Rate on Primary Grain in Al-Cu Alloy

M. Górny, G. Sikora *
AGH University of Science and Technology, Faculty of Foundry Engineering,
Department of Engineering of Cast Alloys and Composites, Reymonta 23, 30-059 Kraków, Poland
*Corresponding author. E-mail address:gsikora@agh.edu.pl

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Abstract

This paper presents a study of the effect of the modification and coolingrate on the grain countα(Al) in the Al-5Cu alloy. Research was performed on castings with walls thickness between 3 mm and 25 mm.Cooling curves were recorded to determine the cooling rate and the degree of undercooling at the beginning of solidification. It has been shown that cooling rate increases exponentially as the wall thickness of casting decreases. Moreover it has been demonstrated that the cooling rate of castings changes within a wide range (21ºC/s - 1ºC/s) when the wall thickness changes from 3 up to 25 mm. Metallographic examinations revealed primary grains (primary α(Al) grains).The paper show that the relationship between the grain count and the degree of undercooling (for non-modified and modified alloys) can be represented by the equation $N = N_v = n_p \cdot \exp(-b/\Delta T_\alpha)$, based on the Weibull's distribution of the size of nucleation sites.

Keywords: Modification, Cooling rate, Aluminium alloys, Grain refinement

1. Introduction

Number of primary grains is one of the most important factors for determining the quality of castings. The final grain density in the casting depends on the nucleation process at the beginning of solidification. Each nucleus gives rise to one single grain. Consequently, it represents the nucleation potential. The following factors significantly influences the number of primary grains in the casting [1,2]: chemical composition, modification, cooling rate,atmosphere, holding time and temperature of liquid metal. However, the measure used in practice to radically increase the number of grains is modification [3,4]. The number of primary grains depends on the type of nucleating agent, its usage and granulation, the overheating and modification temperatures as well as the time since the modification treatment. The benefits of modification are: improved fluidity, improved feeding, better distribution of porosity, better dispersion of secondary phases, improved surface finish, improved machinability, better mechanical properties, e.g. fatigue strength, and better pressure tightness [2]. The modification treatment increases the density of α(Al) phase nuclei as well as the amount of heat generated during solidification and, as a result, changes the degree of undercooling. The present knowledge of the overall relationship between the modification type, its amount and thecooling rate is extremely limited for specific alloys [2]. Literature contains only limited data about the relevant quantitative dependency of the cooling rate or the overcooling degree on the grain density of Al-5Cu alloys [5-7]. This is why the purpose of this study is to analyses the changes in the cooling rate of castings with walls of different thickness and making use of various physical-chemical states of the liquid metal and to demonstrate its significant effect on the grain density in castings.

2. Experimental Work

The experimental melts (Al-5Cu)were performed in a medium-frequency induction furnace. The furnace charge consisted of the following charge materials: aluminium (purity: 99.85%), AlCu50, AlMn75, and AlTi75. After the charge was melted, 1.2% by weight of a covering/refining flux was fed onto the metal surface. The bath having been heated to 750^{o}C, it was modified using the AlTi5B1 master alloy whose quantity was equal to 0.2% of the metal by weight. The base alloy A and the modified alloy B were cast into two sets of moulds made of a traditional moulding sand with bentonite, forming standard type Y ingots (according toASTM A536-84) with wall thicknesses, 3, 5, 13 and 25 mm,respectively.

The results of the chemical composition analysis of alloys A and B are shown in Table 1.

Table 1.
Results of chemical analyses of alloys A and B

Melt No.	% mass.						
	Si	Fe	Cu	Mn	Zn	Ti	Al
A (base alloy)	0.05	0.11	4.90	0.40	0.01	0.07	Rest
B (modified alloy)	0.02	0.04	5.00	0.39	0.01	0.11	Rest

In order to record cooling curves, 0.2 mm thick tips of Pt-PtRh10 thermocouples were placed in the geometric centres of the working part of the ingots. An Agilent 34970A multi-channel electronic module was used to record temperature as the function of time.

Samples for metallographic examinations were taken from the bottom working part of the ingots, and were then ground, polished and electrolytically etched using Barker's reagent. During the etching, electric current at 30 V was fed for the minimum time of 1 minute in accordance with the ASTM E407-07 standard. The metallographic examination was carried out with a Leica MZ6 optical microscope using polarized light.

The II option of the Jeffries method was used to determine the surface grain density N_A, and, after applying Saltykov's formula, this density can be noted as [8]:

$$N_A = \frac{N_i + 0.5N_c + 1}{A} , \tag{1}$$

where N_i is the number of grains contained in a rectangle with the surface area of A. N_c is the number of grains cut by the edges of the rectangle, excluding grains found in the corners.

The volumetric graindensityN_v was calculated using the Voronoi relationship [9]:

$$N_v = 0.568(N_A)^{3/2} . \tag{2}$$

3. Research results and discussion

3.1. Thermal analysis

Figure 1 shows cooling curves recorded in castings with walls 3, 5, 13 and 25 mm thick, respectively, for the basealloy A.

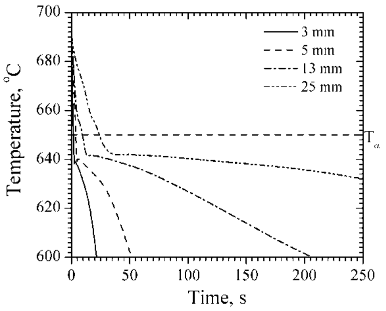


Fig. 1. Cooling curves of castings with walls thickness 3, 5, 13 and 25 mm.Alloy A (base alloy)

The cooling curves obtained were used to determine the Q cooling rate and the maximum degree of undercooling ΔT_α at the beginning of the solidification. The cooling rate Q of castings was determined in the vicinity ($T_\alpha \div T_\alpha + 5^{o}C$) of the equilibrium alloy solidification temperature of phase α(Al). The undercooling value ΔT_α was determined from the following relationship:

$$\Delta T_\alpha = T_\alpha - T_m , \tag{3}$$

where: T_α is the equilibrium temperature of the beginning of the solidification of the α(Al) primary phase read from the Al-Cu equilibrium system for the researched copper content;
T_m is the minimum temperature at the beginning of the α(Al) phase solidification.

Table 2 shows the experimentally determined values of the cooling rate Q and the maximum undercooling degree ΔT_α for the castings obtained.

Table 2.
Results of thermal analysis

Wall thickness g [mm]	Alloy A (base alloy)		Alloy B (modified alloy)	
	Maximum degree of undercooling ΔT_α, [^{o}C]	Cooling rate Q [^{o}C/s]	Maximum degree of undercooling ΔT_α, [^{o}C]	Cooling rate Q [^{o}C/s]
3	11.80	21.62	8.35	21.53
5	10.93	13.67	3.90	9.62
13	8.71	2.30	3.23	2.12
25	8.26	1.17	0.20	1.61

The research show that the cooling rate has a very similar course for the base alloy and the modified alloys (Table 2). The recorded cooling rate varies within a broad range (21.62ºC/s - 1.17ºC/s) when the wall thickness changes from 3 to 25 mm for the base alloy A and the modified alloy B. The cooling rate impacts the maximum degree of undercooling at the beginning of the α(Al) phase solidification and, as a result, also influences the structure of the examined alloy. If the cooling rate Q rises, this increases the maximum degree of undercooling ΔT_m at the beginning of solidification, contributing to increasing the driving force of the solidification process**.** This is of particular importance for the modified alloy B, in which the degree of undercooling values are much lower than in the base alloy. Increase of the degree of undercooling along with the growing cooling rate is greater in the modified alloy than in the base alloy.

The dependency of the cooling rate as a function of wall thickness can be approximated using an equation of the following form:

$$Q = 1.44 + 55.52 \cdot \exp(-0.338 \cdot g) \qquad (4)$$

with a high correlation coefficient of R= 0.99.

Cooling rate of the modified alloy and the unmodified alloy is the same. Liquid metal in the modified and unmodified alloy has various physical-chemical states. This state can be represented by a different number of nucleation sites of the α(Al) phase. Modification causes millions of new nucleation sites to appear. The size (d) distribution of sites in the modified alloy, which can be represented using the $n_i(d)$ curve, ensures a much greater number of nuclei at a lower level of undercooling. An equation identifying the number of α(Al) phase nuclei created at the maximum undercooling ΔT_αcan be presented based on the Weibull distribution of nucleation sites given by the equation of the following form [10]:

$$N_v = n_p \cdot \exp\left(-b/\Delta T_\alpha\right) \qquad (5)$$

where:

n_p- the number of all particles - substrates found in the liquid metal,

b – the nucleation coefficient.

Eq. (5) describes the relationship between the maximum degree of undercooling and the nucleus count, and thus the grain count as well (because each nucleus produces one single grain).Equation 5accounts for the number of all particles (α(Al) phase nucleation sites) which depends on factors influencing the physical-chemical state, such as: the type and quantity of the modifier added, the time and liquid metal temperature, slag and the furnace atmosphere, molten metal mixing or the chemical composition of the alloy. Not all sites found in liquid metal are involved in the nucleation process. It is estimated [11,12] that only a negligible part of them take part in the process of α(Al) phase nucleation. More sites found in the liquid metal can be activated by raising the undercooling degree. For a given physical-chemical state of liquid metal, the undercooling degree can be raised by accelerating the cooling rate, which is strictly connected with the thickness of casting walls as well as the type of mould material and the casting temperature. The cooling rate represents the thermal conditions (of heat exchange) at the beginning of solidification, which in turn determine the final number of α(Al) phase grains for the given physical-chemical state.

3.2. Macroscopic examinations

Figure 2 shows examples of macrostructures portraying primary α(Al) phase grains in the base alloy A and the modified alloy B.

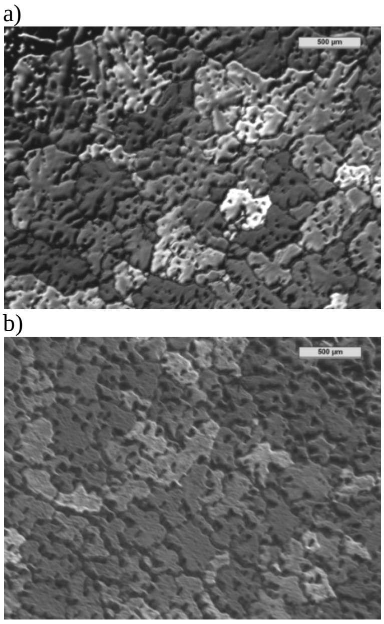
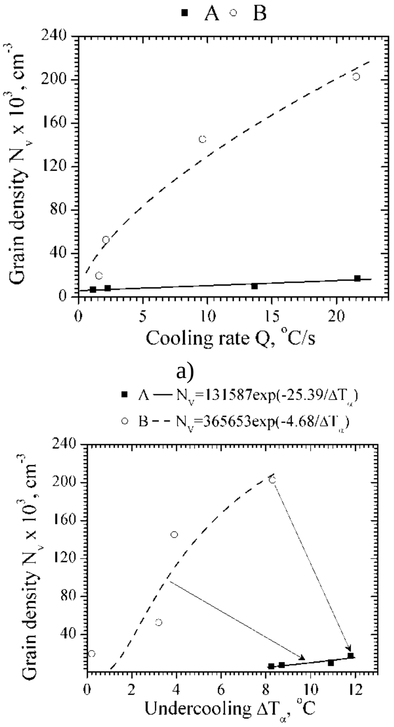


Fig. 2. Macrostructures of electrolytically etched samples cut from castings with wall thickness of 13 mm (a)base alloy, (b) modified alloy

Figure 3 presents the relationship between the density of grains N_v as a function of the casting cooling rate Q and the density of primary grains as a function of the maximum degree of undercooling ΔT_α for the original alloy A and modified alloy B.

Figure 3 suggests that, for a given physical-chemical state, increasing the cooling rate boosts the primary grain count. It has already been mentioned that the cooling rate influences the maximum degree of undercooling at the beginning of the α(Al) phase solidification, which, in turn, determines the number of nuclei, and thus of the grains (dendrites) of the α(Al) phase. This is shown in Fig. 3b.

Figure 3 also suggests that for a given physical-chemical state, if the maximum degree of undercooling increases, so does the number of grains. In the case of castings with walls of the same thickness, but of various physical-chemical state, if the maximum undercooling degree goes up, the number of grains goes down (arrows in Fig. 3b).



a)

b)

Fig. 3.Primary grain density as a function of the cooling rate (a), the primary grain density as a function of the maximum degree of undercooling (b) (original alloy – A, modified alloy – B)

The transition from the first physical-chemical state (unmodified alloy) to the second one (modified alloy) brings about a change in the density of sites n_p(from $n_p = 131587/cm^3$ to $n_p= 365653/cm^3$) and in the nucleation coefficient b (from b = 25.39°C to b = 4.68°C). The rise in the number of nuclei (as a result of treatment of an alloys with the AlTi5B1 modifier) and thus also of the primary grains in Al-Cu alloys results from an increase in the total number of heterogenous nucleation sites caused by the formation of new particles in the liquid metal. The latter number drives the solidification kinetics. In temperature terms, it is reflected in the metal cooling curve. It should also be noted that if the physical-chemical state changed (as a result of modification), the degree of undercooling ΔT_α fell, which was connected with an increase in the quantity of the solidification heat generated by a greater number of primary grains of the α(Al) phase forming.

4. Conclusions

1.Experimental research indicates that the connection between the density of primary grains and the degree of undercooling (for an unmodified and a modified alloy) can be noted in the following form with a high correlation coefficient:

a) for the basealloy state:

$N_v = 131587\exp(-25.39/\Delta T_\alpha)$and $R^2 = 0.82$

b) for the modified alloy state:

$N_v = 365653\exp(-4.68/\Delta T_\alpha)$and $R^2 = 0.87$

2. The cooling rate varies within a wide range (21ºC/s-1ºC/s) when the wall thickness is changed from 3 to 25 mm and this is accompanied by a significant variation in the maximum undercooling at the beginning of the α(Al) phase solidification.

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