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## Optimization of grain refinement process in Al-Cu alloys

### ABSTRACT

The work presents an optimization of the grain refinement process of Al-Cu alloys with the use of selected commercially available master alloys. The optimization of the physicochemical state of the liquid metal was performed with the use of crystallization and cooling curves by selecting the holding time from the moment of introducing the master alloy as well as of the chemical composition of the melt by determining the initial content of the titanium and the main alloy element (copper) in the alloy that was intended for grain refinement (otherwise known as the base alloy). The optimization target was set to obtain the primary  $\alpha(\text{Al})$  grain with a size that is smaller than the optimum value of 220  $\mu\text{m}$ . Obtaining a high number of primary  $\alpha(\text{Al})$  grains ensures the minimization of the shrinkage porosity and minimizes the size of the interdendritic phases as well as the homogeneity of the microstructure, which leads to an improvement in the mechanical properties in the as-cast state. The study assumed a constant addition of grain-refining master alloys in a quantity equal to 0.2% of the metal by weight.

Light microscopy (LM), scanning microscopy (SEM), X-ray microanalysis (EDS), X-ray diffraction (XRD), a thermal analysis (AT), and a quantitative image analysis in the region of interest (ROI) were the methods that were used in the research.

The research concerns the characteristics of AlTi5, AlB3, and AlTi5B1 master alloys being used as grain-refining agents. The selection of the optimal AlTi5B1 master alloy for further research was made by examining the fading of the grain refinement effects for alloys that do not contain the initial titanium in the charge materials. After using the AlTi5B1 master alloy, the finest primary  $\alpha(\text{Al})$  grain was obtained, whose size reached the assumed optimum value of 220  $\mu\text{m}$ . In this case, only 100 ppm titanium and 20 ppm boron were introduced into the base alloy by the master alloy addition. In the same group of aluminum alloys after using the AlB3 master alloy (where three times more of the boron addition was introduced to the base alloy than in the case of the AlTi5B1 master alloy), the worst effects of the grain refinement of the primary  $\alpha(\text{Al})$  grain were observed. This result may be related to the fact that the microstructure of this master alloy contains borides (but mainly in the form of the  $\alpha(\text{AlB}_{12})$  phase). The optimal Al-B grain refiner should contain borides in the form of the AlB<sub>2</sub> phase.

The mentioned tendency also continued for the base alloys, which initially contained 0.16 wt.% of titanium. Based on these results, it was concluded that the boron is not a good grain-refinement agent for titanium-free Al-5Cu alloys when it is present in the grain-refining master alloy in the form of the  $\alpha(\text{AlB}_{12})$  phase.

During the research on the fading of the grain-refinement effects (mainly for the AlTi5B1 master alloy, but less for AlB3), it was observed that the higher the use of the initial titanium addition, the smaller the observed fading of the grain-refinement effects. Also, the obtaining of the super-

refined alloy was delayed over time. These changes indicate that, in the case of base alloys with higher levels of titanium content, the refining master alloys require more time to reach their maximum nucleation potential. In the case of the AlTi5B1 master alloy and the increased initial titanium content, the growth of crystals of the Al<sub>3</sub>Ti phase was observed along the primary  $\alpha$ (Al) grain with increased holding times of the liquid metal.

The effects of the grain refinement can be estimated based on the crystallization and cooling curves. This work presents the characteristics of the crystallization and cooling curves for base alloys and alloys with grains refined with various master alloys as well as with different initial titanium and copper contents. It has been shown that the equilibrium solidification temperature at the beginning of the solidification process should be related to the content of the main alloy component (copper), which can act in two ways in aluminum. Copper in aluminum alloys inhibits the growth of the primary  $\alpha$ (Al) grain through the segregation process (despite the addition of an AlTi5B1 master alloy) and change in the surface tension (reduces the surface tension at the nucleus-melt interface, and increases it at the melt-mold cavity interface).

The size of the primary  $\alpha$ (Al) grain is relevant, as it determines the final mechanical properties of the casting. In this study, grains of various shapes (from columnar through equiaxial shapes with differently developed secondary arms to globular ones) were observed. Therefore, the change in the shape of the primary  $\alpha$ (Al) grains may be one of the effects of the grain refinement of the primary structure. Along with the reduction of the size of the primary  $\alpha$ (Al) grains, the fragmentation and homogeneity of the distribution of the eutectic phases were observed by increasing their area fraction. As the research has shown, more-favorable mechanical properties are obtained when the average diameter of the primary  $\alpha$ (Al) grain is smaller which corresponds to a recalescence (that is close to 0°C in the considered cases) and the maximum degree of undercooling (which is less than 5°-7°C).

It has been shown that the process of the grain refinement of the primary structure is unstable and requires strict metallurgical control due to the holding time of the liquid metal. For a stabilized melting process, it is possible to obtain an optimal size of the primary  $\alpha$ (Al) grain for an alloy that does not contain the initial titanium content in the charge materials.

An optimization of the physicochemical state of the liquid metal combined with an increase in the cooling rate resulted in an increase in the tensile strength as well as elongation. The cooling rate increases exponentially as the wall thickness of the casting decreases. The research shows that the cooling rate of the castings varies within a broad range (23.7°-1.2°C/s) when the wall thickness varies from 3 to 25 mm (regardless of the physicochemical state of the liquid metal). The cooling rate represents the thermal conditions (of heat exchange) at the beginning of the crystallization process of the  $\alpha$ (Al) phase, which determine the final number of the  $\alpha$ (Al) phase grains for the given nucleation potential. The grain-refinement treatment significantly increases the  $\alpha$ (Al) phase nucleus density and the amount of heat generated during the  $\alpha$ (Al) phase formation, and it consequently reduces the maximum degree of undercooling.

Moreover, the experimental studies indicate that the relationship between the primary grain density and the maximum degree of undercooling (at variable cooling rates) for Al-Cu alloys can be described using the Fras or Oldfield models with high coefficients of determination. The percentage of the nucleation sites becoming active depending on the maximum degree of undercooling was estimated for the Fras and Log-normal models. The obtained results reach maximum values ranging from a few percentage points (2-5%) for the base alloy to over a dozen (13-23%) for the grain-refined alloys.

The size distribution of the nucleation sites can be approximated by using the Fras model based on the Weibull distribution with the good agreement of the theoretically determined volumetric grain densities with those obtained experimentally by using quantitative metallography. On the basis of this, it is possible to estimate the temperature uncertainty that is determined on the basis of the crystallization and cooling curves (which does not exceed 1°C).

The mechanism of the grain refinement of the primary  $\alpha(\text{Al})$  grain in Al-5Cu alloys was proposed with the use of an AlTi5B1 master alloy, which explains the shift the super-refined state in time with the increase of the initial titanium content in the charge materials.