Evolution of Eutectic Spacing During Unidirectional Solidification of Al-Ni Alloys

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Hypoeutectic Al-Ni alloys show a ductile phase \( \alpha \) distributed with a \( \beta \) phase Al\(_{3}\)Ni fragile where \( \beta \) serves as reinforcement of the structure of the material. The eutectic composition alloys obey the relationship: \( \lambda^{2} \cdot v = C \), where \( \lambda \) is the eutectic spacing, \( v \) is a tip growth rate and \( C \) is a constant. The aim of this study is to establish correlations between \( \lambda \) and \( v \) for hypoeutectic Al-1%, 3% and 5% Ni alloys. Unsteady-state upward directional solidification experiments were performed, as well as metallography, dissolution of the aluminum matrix and scanning electron microscopy (SEM). The interphase spacing of the three Al-Ni alloys decreased with increasing tip growth rate, with a predominance of a rod-like morphology on intermetallic. It was observed that parameters such as tip growth rate, cooling rate and temperature gradient decreases as the solidification front advances. It was further observed that a single experimental law \( \lambda = 1.2 \cdot v^{-0.5} \) illustrates the evolution of the interphase spacing for any examined alloy.

Keywords: Al-Ni alloys, eutectics, unidirectional solidification, interphase spacing

1. Introduction

In order to optimize the properties of aluminum alloys, such as lightness, mechanical strength and corrosion, it is necessary to study characteristics such as microstructural aspect, dendrite arm spacing and porosity, and their evolution during the solidification process\(^1\). In the particular case of Al-Ni alloys, within the range of hypoeutectic compositions, the solidification microstructure is composed of a dendritic matrix rich in aluminum (\( \alpha \)) surrounded by a eutectic mixture \( \alpha + \beta \), where \( \alpha \) is rich in aluminum and \( \beta \) is enriched by the intermetallic Al\(_{3}\)Ni, with a predominance of a rod-like morphology\(^2,3\). Such eutectic mixture nucleate in a cooperative and alternative way during growth and remains located between the dendritic arms. The measurement of the interphase spacing is commonly used to quantify the eutectic arrangement along foundry components. Both dendritic arrangement and eutectic mixture configuration can affect the mechanical properties of the mentioned alloys.

The variation of interphase spacing (\( \lambda \)) is commonly expressed by the classic relationship for growth of eutectic systems proposed by Jackson and Hunt\(^4\), as shown in Equation 1, where \( v \) is tip growth rate and \( C \) is a constant.

\[ \lambda^{2} \cdot v = C \] (1)

The unsteady-state regime of solidification in casting alloys has been the object of extensive studies\(^5,7\), but there is a lack of studies that emphasize the solidification transient heat extraction, which is close to the industrial reality. Most of the solidification studies use a Bridgman device which promotes steady-state regime\(^2,3\).

The evolution of the interphase spacing of Al-Fe hypoeutectic alloys under both steady-state and unsteady-state conditions was investigated in a recent study. Two experimental relations have been proposed\(^7\), as shown in Equation 2. In this case, the eutectic mixture was examined within intercellular regions once cellular structures prevailed in all examined Al-Fe alloys.

\[ \lambda = 1.6 \cdot (v^{-1/2}) \text{ and } \lambda = 3.8 \cdot G^{-1/2} \cdot v^{-1/4} \] (2)

This study aims to determine the solidification thermal parameters (tip growth rate – \( V_{t} \), temperature gradient – \( G_{s} \) and cooling rate - \( T_{c} \), interphase spacing and the resulted interrelations between these experimental data. The solute concentration was varied (1.0, 3.0 and 5.0 wt. (% Ni)) to evaluate the effect of the solute content in the interphase spacing. The applicability of the expression proposed by Jackson and Hunt\(^5\) has been checked for a wide range of solidification conditions.

2. Experimental Procedure

Directional solidification experiments with hypoeutectic Al–Ni alloys (1.0, 3.0 and 5.0 wt. (% Ni)) were carried out in a water-cooled solidification apparatus, which is characterized by unsteady-state heat flow conditions, as described in a previous study\(^6\).

In the unsteady-state solidification system, heat is directionally extracted only through a water-cooled bottom made of low carbon steel (SAE 1020), promoting vertical upward directional solidification. A stainless steel split mold was used having an internal diameter of 60 mm, a height of 157 mm and a wall thickness of 5 mm.

The Al-Ni partial phase diagram was also computed by Thermo-Calc(Thermo-Calc software is an exclusive copyright property of the STT Foundation (Foundation of Computational Thermodynamics, Stockholm, Sweden)) and it is shown in Figure 1.

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Continuous temperature measurements in the casting were monitored during solidification via the output of a bank of fine type-K thermocouples (made from 0.2 mm diameter wire), sheathed in 1.6 mm diameter stainless steel tubes and positioned at 4, 8, 12, 17, 22, 37, 52 and 88 mm from the heat-extracting surface at the bottom. The thermocouples were connected by coaxial cables to a data logger interfaced with a computer, and the temperature data, read at intervals of 0.1 second, were acquired automatically. From these data thermal solidification parameters were inferred ($V_L$ - tip growth rate, $\dot{T}$ - tip cooling rate and $G_L$ - temperature gradient).

After completing the conventional metallography procedures, the microstructures were revealed along the transverse sections of the castings by both optical microscopy (1000× magnification) and scanning electron microscopy (magnification 5000×). The acid solution used to reveal the microstructures was 0.5% HF diluted in $\text{H}_2\text{O}$, with sample exposure varying from 1 to 10 minutes.

According to literature [10], the interphase spacings ($\lambda$) can be measured based on two techniques of quantification: intercept method and triangle method. Böyük and Marasli [11] claim that more reliable measurements can be obtained in transverse samples. Then, the interphase eutectic spacing was measured by the linear intercept method, with at least 50 measurements for each position.

**Figure 1.** Al-Ni partial phase diagram furnished by the software ThermoCalc AB, version N.

**Figure 2.** Tip growth rate evolution for hypoeutectic alloys: a) Al-1 wt. (%) Ni; b) Al-3 wt. (%) Ni and c) Al-5 wt. (%) Ni.
Figure 3. Cooling rate evolution for hypoeutectic alloys: a) Al-1 wt. (%) Ni; b) Al-3 wt. (%) Ni; and c) Al-5 wt. (%) Ni. The arrows indicate the position in which the microstructure was observed.
Figure 4. Typical microstructures of eutectic Al-Ni structures obtained in MEV: a) Al-1 wt. % Ni – P = 5 mm; b) Al-1 wt. (%) Ni – P = 20 mm; c) Al-3 wt. (%) Ni – P = 10 mm; d) Al-3 wt. (%) Ni – P = 60 mm; e) Al-5 wt. (%) Ni – P = 5 mm; and f) Al-5 wt. (%) Ni – P = 30 mm. P is the position from the metal/mold interface.
3. Results and Discussion

The experimental tip growth rates ($V_L$) were determined for all alloys analyzed by derivative functions of position versus time of liquidus isotherm passage. The values of the liquidus temperatures of the studied alloys can be found in previous study performed by Canté\textsuperscript{2}.

The functions established for position as a function of time were obtained from the intersections of the lines of each liquidus temperature ($T_L$) with the cooling curves for each position of the thermocouples. The cooling rate ($\dot{T}$) was determined by considering the thermal data recorded immediately after the passing of the liquidus front by each thermocouple. The temperature gradient ($G_L$) was determined according to the ratio of $\dot{T}$ and $V_L$.

The data acquisition system, in which temperature readings are collected at a frequency of 0.5 seconds, permits accurate determination of both the slope of the experimental cooling curves, cooling rate evolution and growth rate.

Figures 2 and 3 show the evolution of $V_L$ and $\dot{T}$ for Al-Ni alloys. As the solidification front advances, the values of both parameters decrease. This effect is reversely translated resulting in increasing of the as-cast microstructure (primary and secondary dendritic spacing) for positions far from metal/mold interface. Typical optical microstructures (transverse sections) can be seen in Figure 3 in order to emphasize the mentioned effect.

Figure 5 shows typical SEM images of the interdendritic regions, i.e., emphasizing the eutectic mixture configuration: distribution, size and morphology. It can be noted that the rod-like Al$_3$Ni has prevailed. When comparing the microstructures of Figures 4a and b (Al-1 wt. (%) Ni) and the Figures 4e and f (Al-5 wt. (%) Ni), it can be clearly seen that a higher fiber density was found for the highest amount of nickel, due to the increase of eutectic fraction into interdendritic regions with increasing Ni content.

Figures 4c and d (Al-3 wt. (%) Ni) show clear distinction with regard to the interphase spacing with larger $\lambda$ values being observed in the case where the value of $V_L$ is lower (Figure 4d).

Based on images obtained by SEM and optical microscopy several measurements were performed in order to determine the interphase spacings along the Al-Ni castings. The average, maximum and minimum $\lambda$ values are shown in Figure 5. A single experimental law, Equation 3, is able to represent the evolution of the interphase spacing for all examined Al-Ni alloys with $V_L$, as shown in Figure 6.

$$\lambda = 1.2 \times (V_L)^{-1/2}$$

This means that the increase in the solute content does not affect the interphase spacing for hypoeutectic Al-Ni alloys. The exponent $-1/2$ proposed by Böyük and Marasli\textsuperscript{11} is appropriate to state the unsteady-state growth of the eutectic mixture despite the slight overestimation.
The intermetallic Al-Ni prevails with rod-like morphology, and a higher fiber density in the alloys with higher solute content was observed. The spacing $\lambda$ increases as the tip growth rate $V_L$ decreases.

A single experimental law: $\lambda = 1.2 \left( V_L \right)^{1/2}$ can be applied to encompass the evolution of the interphase spacing of the three hypoeutectic Al-Ni alloys with tip growth rate $V_L$. It is observed that increase in nickel content does not affect the interphase spacings for these hypoeutectic alloys.

The experimental law: $\lambda = 2.5G_L^{-1/2}V_L^{-1/4}$ seems to be more appropriate than the first law obtained: $\lambda = 1.2 \left( V_L \right)^{1/2}$. The interrelation between $\lambda$ and more than one solidification thermal parameter seems to be more representative of the eutectic growth in hypoeutectic Al-Ni alloys.

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