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MICROSTRUCTURE AND THERMAL PARAMETERS DURING DIRECTIONAL SOLIDIFICATION OF A TERNARY AL-FE-MG ALLOY

Mateus Rovigati Gandolfi
Leonardo Fernandes Gomes
Bismarck Luiz Silva
José Eduardo Spinelli
Department of Materials Engineering, Federal University of São Carlos, UFSCar, 13565-905 São Carlos, SP, Brazil
mateus.gandolfi@gmail.com
gomeseng@gmail.com
bismarck_luiz@yahoo.com.br
spinelli@ufscar.br

Abstract. *Al-Fe alloys containing from 2.0 to 4.0wt%Mg characterizes chemical composition of some comercial alloys 5xxx series such as 5454, 5086 or 5083. These alloys are indicated for petrochemical platforms structures and shipbuilding industry due to their resistance to seawater and high humidity. The effects of magnesium addition in Al-Fe alloys are not properly established, mainly, considering the examination of aluminium α phase growth mode, the prevalence of eutetic phases and segregation. In contrast, Al-Fe alloys solidification is widely known from the point of view of microstructural analysis and thermal solidification parameters, namely growth rate (V_L) and cooling rate ($\dot{T}_L$). The iron addition near the eutetic composition allows to obtain higher mechanical strength, which is associated with the higher density and amount of hardening phases forming within the eutetic mixture.*
The present study aims to evaluate the addition of 1.2wt%Mg on the Al-1.5wt%Fe base alloy in order to obtain some resulting characteristics, such as the influences of thermal solidification parameters (growth rate - V_L and cooling rate - $\dot{T}_L$) on the microstructure of the aluminium alloy solidified under unsteady-state conditions. Samples were obtained by upward directional solidification (DS), followed by characterization through metallography, scanning electron microscopy (SEM) and X-Ray diffraction. The experimental microstructural evolution was determined for the ternary alloy so that it could be compared with that typically established for the directionally solidified Al-Fe alloy.

Keywords: *Metals and alloys, Microstructure, Solidification, Al-Fe alloys, Casting.*

1. INTRODUCTION

Casting alloys of the 5xxx series have a wide range of industrial applications because of their good mechanical properties, balance of the strength to weight ratio, weldability and high resistance to corrosion (Bazarnik, 2012; Alil, 2015). Parts manufactured with these alloys, containing magnesium, in marine applications, including structures, boats and ships have shown their good resistance and long life under conditions of partial, intermittent or total immersion (Davis, 1999). For example, 5083 is the usual aluminium alloy used in the manufacture of passenger, freight and naval vessels. Due to operating and environmental conditions, welded components and structures are subjected to a high level of loads. Therefore, microstructural degradation may be caused leading to failure (Katsas, 2006; Katsas, 2007).

Other industry sectors, such as automobile, are emphasized on greenhouse gas reductions and improving fuel efficiency in the transportation sector. Suppliers, assemblers, and component producers are investing significantly in lightweight materials research and development (Ghassemieh, 2011; Kabirian, 2014). One example is the replacement of steel with aluminium sheet that permits weight reduction of up to 47%, consequently, secondary weight savings can be performed because the lower loads will in turn permit lighter parts in the brakes, engine and suspension without loss of performance (Burger, 1995).

The solid solution formation is one of the main causes of the increase in mechanical resistance of the 5xxx series Al-Mg alloys (Bazarnik, 2012). Alloys in which the magnesium amounts remains in solid solution, or are partially precipitated as particles dispersed around the matrix, are generally more resistant to corrosion than commercially pure aluminium, resistant to salt water and some alkaline solutions (Korb, 1987).

Al-Fe binary system was investigated by Goulart, *et al.* 2010. Hypoeutectic alloys composition in the range of 0.5-1.5wt%Fe were characterized by transitory (growth rate, v>0,2mm/s) and stationary/Bridgman solidification conditions (50<v<1200 μm/s), the microstructures having been revealed for each of these conditions. Vickers microhardness profiles were determined in addition to further investigation of intermetallic nature, shape and distribution associated with the thermal solidification parameters (G, V e $\dot{T}$). Also, X-ray diffraction (XRD) and scanning electron microscopy (SEM) analyzes have been performed. The eutetic composition is about 1.8wt%Fe and the maximum solubility limit is 0.05wt%Fe. Under steady-state conditions the predominance of Al_3Fe (equilibrium phase) in the eutectic formation has been noted. On the other hand, with higher cooling rates ($\dot{T}$>1 °C/s), the equilibrium phase is replaced with the metastable phase (Al_6Fe).

Provided that Al-Fe alloys with composition around the eutetic point, the occurrence of a gradually intermetallic particles transition of Al_3Fe and Al_6Fe can be typically observed with an $Al_{13}Fe_4$ intermetallic interaction (Goulart, 2009). This phase formation is affected not only by thermal solidification parameters but also by Fe content. Thus, experimental systems allowing transient flow of heat extraction may promote the increase on the spectrum of experimental values improving the experimental data interpretation. Studies considering transient solidification conditions of ternary alloys related with the microestructural evolution, solute segregation and porosity formation are rare in literature (Atwood, 1999; Voller, 2004). The main difficulty is related to the resolution of ternary alloy solidification mode and intermediate phases, reactions and connections between them. This is quite noticed in the case of developing solidification mathematical models for these alloys.

Al-Fe hypoeutectic alloys with Fe adding in the range from 0.5 to 1.5wt%Fe show fully cellular microstructure according to the results presented by Goulart, *et al.*, 2009. The intercellular spacing (λ_c) increases with decreasing of solidification thermal variables as tip growth rate (V_L) and cooling rate ($\dot{T}$). The λ_c variation range obtained was from 5μm to 20 μm during transient unidirectional solidification. Other important experimental evidences were recently obtained in present work by Goulart, *et al.*, 2010; concerning the nature, distribution and Al-Fe intermetallic morphology in these alloys, its variations as a function of the solute content and cooling rate, and contributions as reinforcement particles of cell growth during ingot submission to hardness and tensile strength tests. The presence of several intermetallic phases (Al_3Fe, $Al_{13}Fe_4$ e Al_6Fe) proved to have minor contribution in relation with Vickers microhardness variation. This indicates clearly that the intercellular spacing controls the mechanical properties.

There are few experimental works related to Al-Mg alloys in the literature, especially considering results under unsteady-state conditions. Pereira, *et al.*, 2007 performed one of the few systematic studies with upward vertical unidirectional solidification of hypoeutectic Al-5wt%Mg, Al-10wt%Mg and Al-15wt%Mg alloys. The dendritic growth in all three cases was characterized by the presence of a much more branched structure in Al-15wt%Mg alloy. However, the effect of Mg content in λ_1 and λ_2 was not determined. The authors maintain that primary and secondary dendrite spacing are strongly influenced by the thermal conditions, decreasing with increases of tip growth rate (V_L) and cooling rate ($\dot{T}$).

Based on the importance of solidification structure evaluation in aluminium alloys, its influence on materials development with optimized mechanical properties, a need for a better knowledge of the mechanisms involved in cellular/dendritic growth, and the absence in literature of experimental investigations considering transient solidification for multicomponent alloys, the present work focuses on the transient solidification of Al-Fe-Mg alloys, fixing the Fe content in 1.5wt%Fe with adding of 1.2wt%Mg. The comprehensive microstructural evolution of the ternary Al-1.5wt%Fe-1.2wt%Mg alloys with respect to solidification parameters such as growth rate (V_L) and cooling rate ($\dot{T}$). The effects of Mg addition are clarified.

2. EXPERIMENTAL PROCEDURE

To permit a transient heat flow condition to be achieved a directional solidification system was used in this experiment as detailed in Figure 1. Heat is directionally extracted only through a water-cooled low carbon steel bottom, promoting vertical upward directional solidification (Silva, 2012).

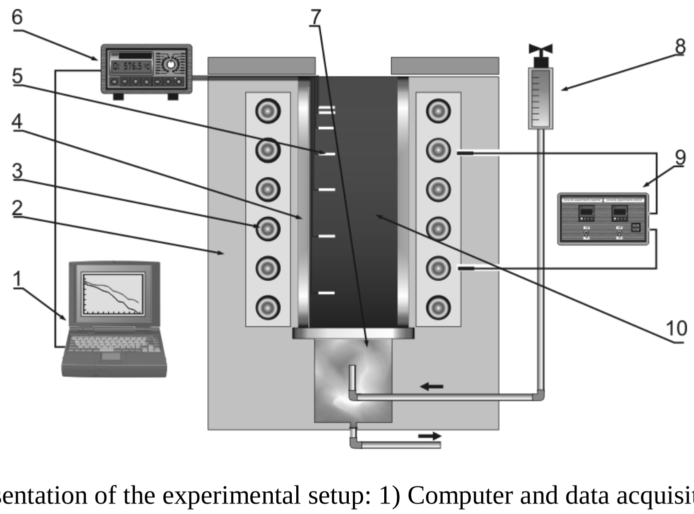


Figure 1. Schematic representation of the experimental setup: 1) Computer and data acquisition software; 2) Insulating ceramic shielding; 3) Electric heaters; 4) Mold; 5) Thermocouples; 6) Data logger; 7) Refrigeration chamber; 8) Rotameter; 9) Temperature controller, 10) Casting.

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. To minimize radial heat losses, the lateral inner mold surface was covered with a layer of insulating alumina. A thin (3 mm) 1020 carbon steel was used to close the bottom part of the mold (Silva, 2012).

Al-Fe-Mg alloy was prepared by melting weighed quantities of aluminium, iron and magnesium of high purity elements (99.99%) in a silicon carbide crucible inserted into the induction furnace. The alloy was poured in the mold and the electric heaters were disabled so that directional solidification could be initiated, and at the same time, the controlled water flow was opened. During solidification, continuous temperature measurements in the casting were performed by fine type K thermocouples placed along the casting length.

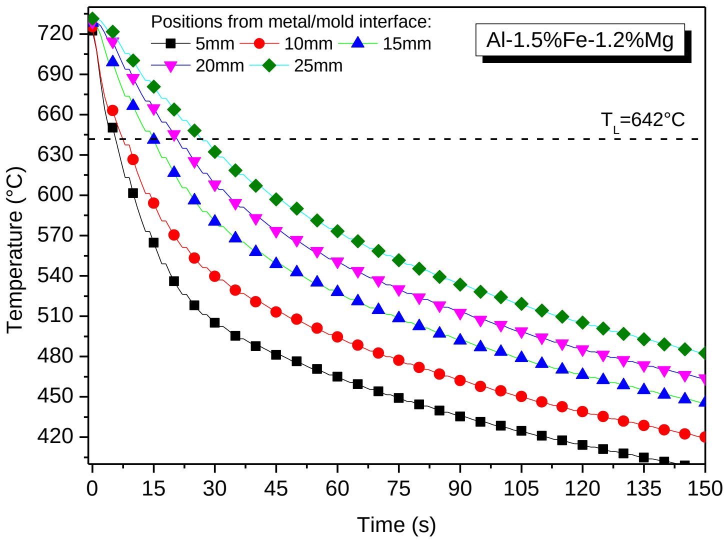
The cylindrical ingot was sectioned along its vertical axis, sanded with a 1200 mesh sandpaper and etched with an acid solution (Poulton's reagent: 5 mL H_2O, 30 mL HNO_3, 60 mL HC*l* and 5 mL HF) to reveal the macrostructure. A solution of NaOH in water was used to reveal the microstructures for metallography. The primary and secondary dendrite arm spacings (λ_1 and λ_2) were measured using an optical microscopy by examining transverse and longitudinal sections of the casting. Furthermore, representative micrographs were obtained with an optical image processing system (Brito, 2015). The triangle method was employed to quantify the primary dendrite arm spacing (λ_1), on transverse sections of the casting. Further, the secondary dendrite arm spacing (λ_2), was determined adopting the intercept method on longitudinal samples. This method is typically employed in the examination of longitudinal sections in the direction of heat flow, measuring the length between the center of a particular secondary branch to another center aligned with the first (Gündüz, 2002).

Beyond optical microscopy, microestructural characterization was complemented by the use of a Scanning Electron Microscopy (SEM), equipped with an Energy Dispersive Spectrometer (EDS). These analyzes were performed to the overall rating of phases present in the Al-1.5wt%Fe-1.2wt%Mg alloy casting. Samples of different positions along the length of the DS casting were selected and subjected to X-ray diffraction (XRD) analyses. Therefore, the present phases and the peak intensities were related with the cooling rate evolution along the length of the casting.

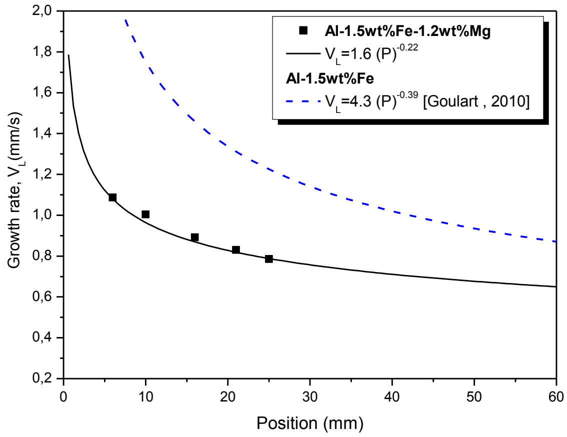
3. RESULTS AND DISCUSSION

Figure 2a shows the cooling curves corresponding to the thermal responses of thermocouples inserted along the length of the Al-1.5wt%Fe-1.2wt%Mg alloy casting. The thermal readings have been used to provide plots of position from the metal/mold interface and the corresponding time of the *liquidus* front passing by each thermocouple. The derivatives of these functions with respect to time gave values for the growth rate (V_L), as shown in Figure 2b. The cooling rate was determined along the castings lengths, by considering the thermal data recorded immediately after the passage of the *liquidus* front by each thermocouple. The resulting experimental $\dot{T}$ values are shown in Fig. 2c.

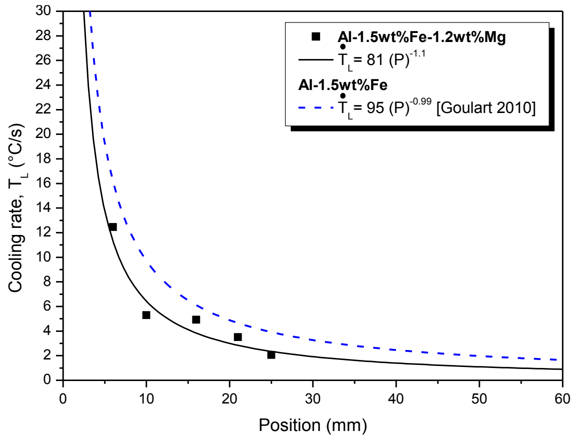
The experimental tendencies proposed by Goulart, *et al.*, 2010 have been inserted in the graphs for comparison purposes (blue dash lines). It is clearly observed that the Mg containing alloy is associated with lower growth rates and cooling rates profiles, especially for positions very close to the cooled surface of the casting. These solidification thermal parameters may be directly connected to the metal/mold thermal conductance, which is represented by an interfacial heat transfer coefficient, h_i (Cheung, 2009). Such coefficient determines the heat ability to flow across the metal/mold interface during solidification. One of the main factors affecting h_i and, as a consequence, growth rate and cooling rate is the wettability. The wettability is related to the fluidity of the molten alloy, which is a function of solute content considering a certain metallic system. According to Kim, *et al.*, 2013 Mg addition may accelerate the formation of Mg-based oxides and hard inclusions during alloying and casting due to its high oxygen affinity, causing the drop in the melt fluidity. Thus, the decrease on V_L and $\dot{T}$ values is essentially caused the lower fluidity induced by Mg alloying.



(a)



(b)



(c)

Figure 2. (a) Experimental cooling curves for the Al-1.5wt%Fe-1.2wt%Mg alloy sample in the water-cooled directional solidification system; (b) Growth rate against position and (c) cooling rate against position. T_L is the *liquidus* temperature.

Representative optical microstructures referring to cross and longitudinal sections of the Al-Fe-Mg alloy examined can be seen in Figure 3. The dendritic arrays located at different positions from the cooled surface of the casting reveal the coarsening of the dendritic arrangement. This is due to the differences in cooling rate associated with positions close to cooled surface of the castings (higher cooling rates) compared with those representing positions farther from the bottom of the castings (lower cooling rates).

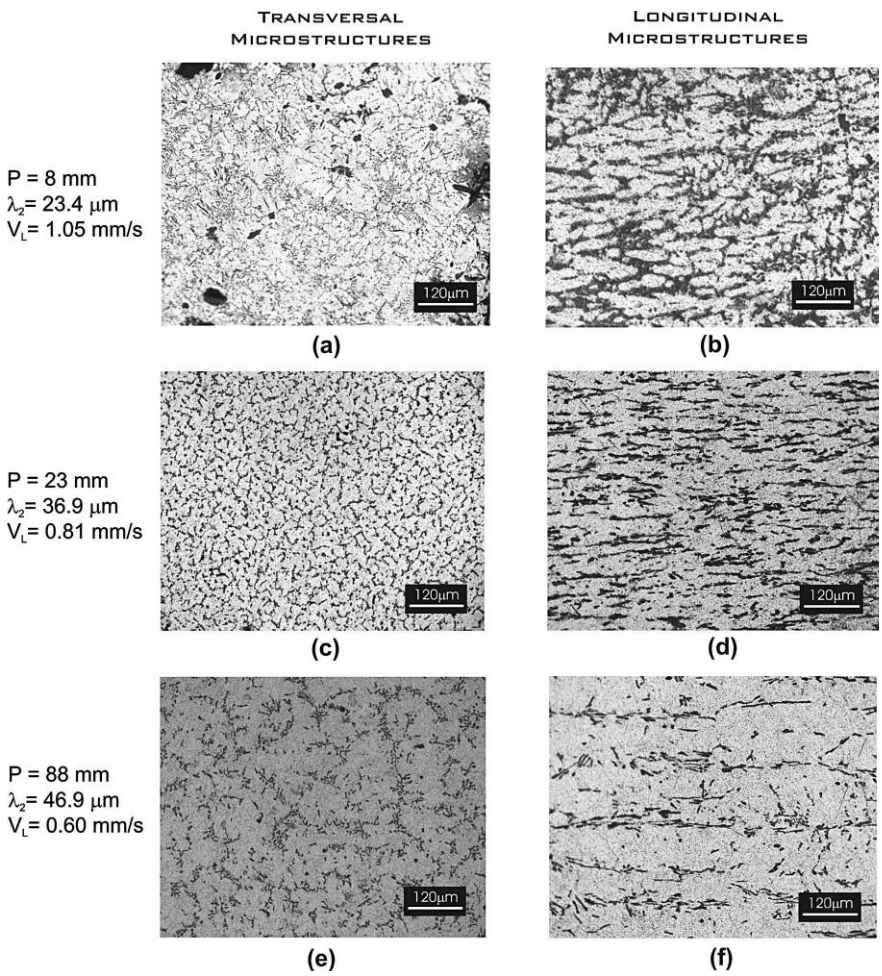
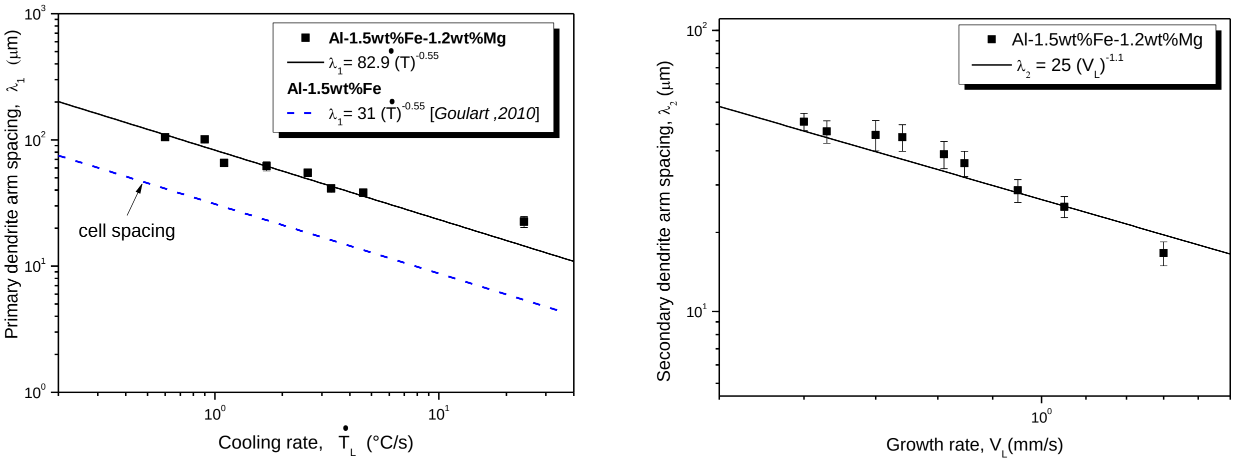


Figure 3. Typical optical microstructures of transverse (left) and longitudinal (right) sections of the Al-1.5wt%Fe-1.2wt%Mg. P is the position from the cooled surface of the DS casting.

By comparing both experimental growth laws depicted in Fig. 4a, the addition of Mg to the Al-1.5wt%Fe alloy resulted in increasing λ_1 value if considered a certain cooling rate. Despite the comparison regarding $\lambda_{1/c}$ values, as a matter of fact, the binary alloys are characterized by cellular structures and the Mg-modified alloy by dendritic pattern. The following experimental growth laws $\lambda_1=82.9(\dot{T})^{-0.55}$ and $\lambda_c=31(\dot{T})^{-0.55}$ have been derived, where the experimental λ_1 average values are plotted together with the error bars corresponding to the range of minimum and maximum values. This means that relatively large dendritic spacings may be obtained due to the Mg addition, achieving a coarser microstructural scale as that reported for Al-Fe alloys during upward vertical solidification. The lateral instabilities and arms typically found during dendritic growth seems to be able to enlarge the α-Al structures which may increase the spacing between them. Anyway, dendritic growth allows a better distribution and alternation of the reinforced phase to be obtained if compared with cells. Normally, more complex morphologies of the α-phase dendrites are associated with higher mechanical resistance. A -1.1 exponent seems to be appropriate in power function equations relating λ_2 to the growth rate, as shown in Figure 4b. The exponent -0.55 has also been adopted for representing the evolution of λ_1 of other multicomponent Al-based alloys such as Al-Cu-Si and Al-Fe-Ni (Ferreira, 2010; Canté, 2013).



(a) (b)

Figure 4. Experimental correlations between dendrite arm spacing and solidification thermal parameters for Al-1.5wt%Fe-1.2wt%Mg alloy: (a) λ_1 evolution as a function of cooling rate and (b) λ_2 evolution as a function of growth rate.

Under equilibrium or slow cooling conditions if the magnesium content in the Al-Fe-Mg alloy is lower than 14.1wt% at 723K there is a possibility of a single phase to be formed (Mondolfo, 1976). The level of cooling must be examined in combination with nominal composition and segregation so that the tendency to form second phases can be established. Mondolfo (1976) affirmed that in nonequilibrium conditions or moderate to rapid cooling conditions, the Mg_5Al_8 compounds tend to appear, even at magnesium contents of 2-3wt%Mg and at low iron content concentrations. However, as can be seen in both Figs 5 and 6 Al-Fe intermetallic compounds were formed together with Al-rich α phase in the Al-1.5wt%Fe-1.2wt%Mg alloy. A negligible presence of Al-Mg intermetallic can be seen in Fig. 6, with most of the correspondent 2θ values being coincident with those for the α-Al phase. Most of Mg content of the ternary alloy was probably consumed in solid solution with the α-Al phase. XRD results confirm that secondary phases based on AlFe have been massively formed at the α-Al dendrite boundaries.

In the case of the directionally solidified Al-Fe-Mg specimens of the present study, the SEM images obtained (see Figure 5) suggest that the level of cooling rate can affect the nature of the formed Al-Fe intermetallic. Rapid solidification conditions (high cooling rates) can induce the replacement of the stable Al–Al_3Fe eutectic (plate-type in Figure 5c) with the metastable eutectic Al–Al_6Fe (fiber-type in Figure 5a), as indicated by black arrows. The presence of this kind of transition indicates that further investigation is needed so that the factors affecting the nature of the intermetallic could be determined such as cooling rate and chemical composition.

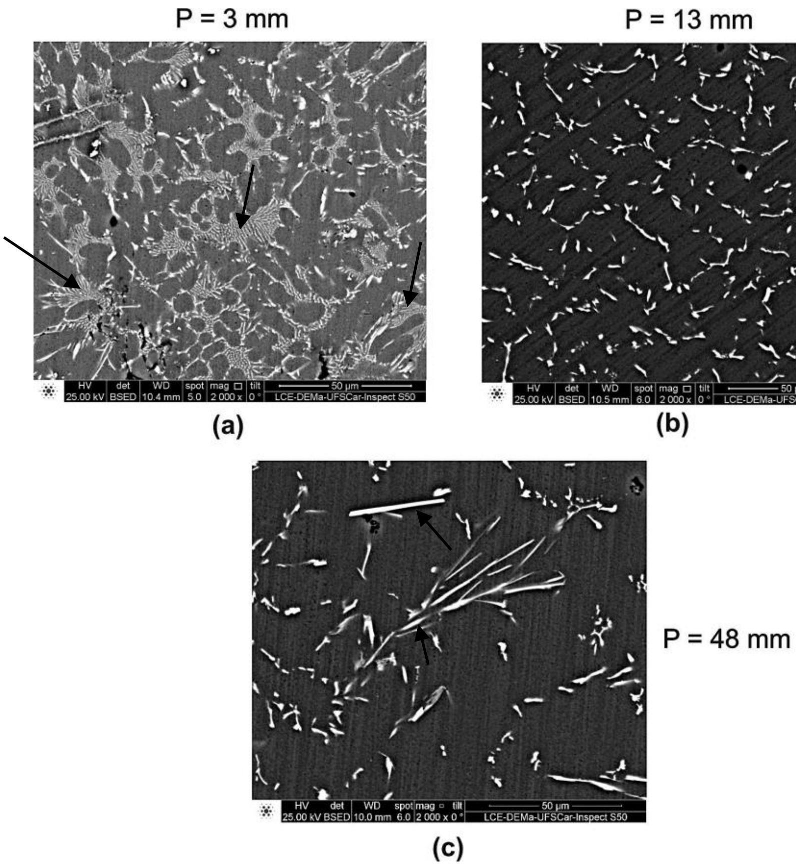


Figure 5. SEM images detailing the phases formed during the directional solidification of the Al-1.5wt%Fe-1.2wt%Mg alloy casting. P is the position from the cooled surface of the casting.

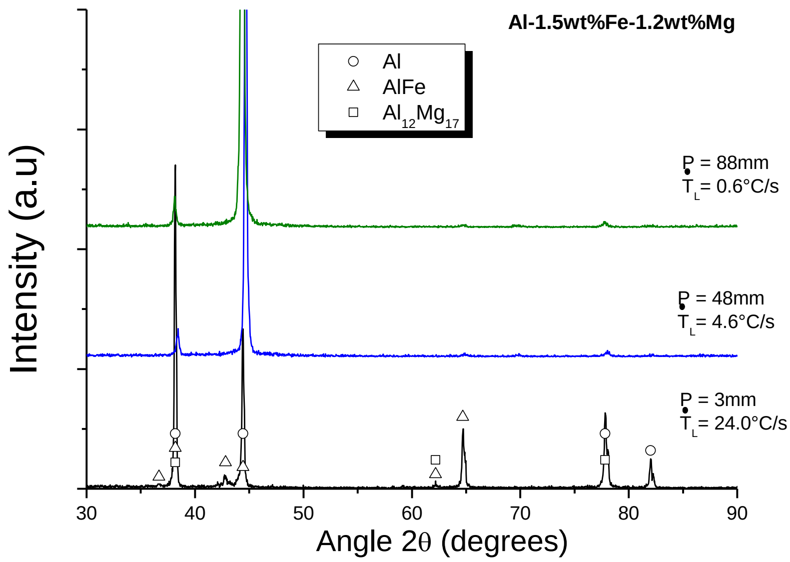


Figure 6. X-ray diffractograms obtained for the directionally solidified Al-1.5wt%Fe-1.2wt%Mg alloy solidified under unsteady-state condition.

4. CONCLUSIONS

The effect of cooling rate on the dendritic evolution of the ternary Al-1.5wt%Fe-1.2wt%Mg has been examined along the length of a directionally solidified casting. The experimental set up allowed cooling rates in the range 24.0< $\dot{T}$ <1.0 K/s to be associated with the corresponding microstructures along the casting length. As observed in the literature for the binary Al-1.5wt%Fe alloy, a -0.55 exponent was able to represent the λ_1 variation against cooling rate. However, the dendritic microstructural scale of the ternary alloy is coarser than the cellular microstructure observed for the binary Al-Fe alloy during upward vertical solidification. It was shown that the lower growth rates and cooling rates characterize the ternary alloy. The minor content of Mg seems to provoke a decrease on the alloy fluidity and as consequence a decrease on the heat transfer efficiency.

5. ACKNOWLEDGEMENTS

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