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## TAILORING OF TRANSIENT THERMAL PARAMETERS AND MICROSTRUCTURE VIA UPWARD SOLIDIFICATION OF THE EUTECTIC Al12.6Si ALLOY (wt.%)

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**Abstract.** Casting Al-Si alloys are the most widely used as cast products in the automotive industries. In this sense, to perform a thermal and microstructural analysis with the eutectic Al12.6Si alloy (wt.%), an as-cast ingot of the aforementioned alloy was obtained via transient upward solidification, using a water-cooled directional solidification device. Temperature data have been generated from the solidification process, and this has enabled a thermal analysis of solidification conditions by means of growth and cooling rates ( $V_L$  and  $T_R$ , respectively) and by secondary dendrite arm spacing ( $\lambda_2$ ). Optical microscopy has been used to characterize the microstructure and an image processing system as well as a software have been used for measurement of  $\lambda_2$ . An association between  $\lambda_2$ ,  $V_L$  and  $T_R$  has been elaborated, and mathematical expressions given by general formulas  $\lambda_2=A.(V_L \text{ and } T_R)^{-n}$  have been proposed to characterize the variation of secondary dendritic spacing as a function of solidification thermal parameters. A comparison with the literature for hypoeutectic Al-Si alloys has been conducted.

**Keywords:** Unsteady-state horizontal solidification; Transient heat flow parameters, eutectic AlSi alloy.

## 1. INTRODUCTION

The phenomenon of solidification can be considered fundamentally as an unsteady-state mass and heat transfer process that contemplates the phase change from the state liquid to the solid state. The rate at which latent heat is extracted and transferred through the metal/mold system, has a direct relationship with the solidification velocity and, in turn, with the thermal and structural parameters involved in the liquid/solid phase change which directly interfere in the mechanical properties of the casting product. In this sense, the analysis of the heat transfer process that occurs during solidification presents a very significant importance in the design and control of metal /mold systems (Barros et al. 2016; Carvalho et al. 2013, 2014, 2018; Peres et al. 2003, Santos et al. 2001; Ferreira et al. 2008; Rocha et al. 2020).

Figure 1a shows a representative schematic of a reference element from the metal/mold system that best illustrates the analysis of heat transfer during upward directional solidification, respectively. It is observed all the modes of heat transfer that can occur along the solidification, such as thermal conduction in metal and mold, Newtonian transfer at the metal/mold interface, convection in the liquid metal and the mold/environment interface and radiation of the mold to the environment. Figure 1b and 1c show an illustrative scheme of imperfect contact and heat flux at the metal/mold interface.

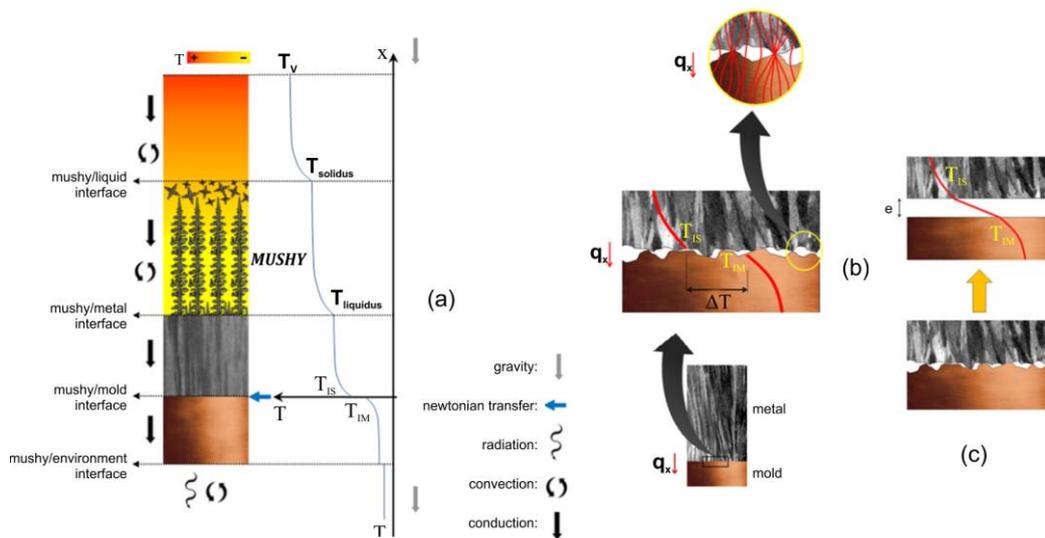


Figure 1. Schematic representation of a reference element of an upward solidification: (a) temperature gradients and heat-transfer modes from the metal/environment interface; (b) contact at the metal/mold interface; (c) Newton's equivalence model for the metal/mold interface. Adapted from the literature (Santos et al. 2001; Ferreira et al. 2008).

It is known that the as-cast microstructure and its properties depend strongly on the solidification thermal parameter (Costa et al. 2015; Dias et al. 2020; Gündüz et al. 2004; Kaya et al. 2003; Kaya, Çadirli, and Gündüz 2007), and the most studied in the literature are the interfacial heat-transfer coefficient ( $h_i$ ) and growth and cooling rates ( $V_L$  e  $T_R$ , respectively) and solidification local time ( $t_{SL}$ ) (Kurz, Fisher, and Trivedi 2019). Studies on thermal analysis during directional solidification that correlate  $h_i$ ,  $V_L$  and  $T_R$  with the length of the dendritic scale, characterized by primary, secondary and tertiary dendritic arm spacing ( $\lambda_1$ ,  $\lambda_2$ , and  $\lambda_3$ ), have been carried out in recent decades (Barros et al. 2016; Carvalho et al. 2013, 2014; Costa et al. 2015; Dias et al. 2020; Gündüz et al. 2004; Kaya et al. 2003, 2007; Kurz et al. 2019; Peres et al. 2004; Rocha et al. 2020).

The literature presents theoretical work focusing on the microstructural evolution of aluminum-based binary alloys. Theoretical mathematical laws on dendritic growth, given by the general formula  $\lambda_{1,2,3} = \text{Constant} \cdot (V_L, T_R)^{-n}$ , have been proposed and properly validated (Bouchard and Kirkaldy 1997; Kirkwood 1985; Kurz et al. 2019; Mortensen 1991; Rappaz and Boettinger 1999; Rappaz and Thévoz 1987; Tiryakioğlu 2019), which have been represented by experimental expressions (Barros et al. 2016; Carvalho et al. 2013, 2014; Costa et al. 2015; Ferreira et al. 2018; Gündüz et al. 2004; Kaya et al. 2003, 2007; Peres et al. 2004; Quaresma, Carlos, and Garcia 2000; Rocha, et a. 2003).

Casting Al-Si alloys represent 85% to 90% of all aluminum alloys, since Si element increases the mechanical and corrosion resistance of these alloys (Costa et al. 2015; Dias et al. 2020; Peres et al. 2004). Investigations on dendritic growth with hypoeutectic Al-Si alloys ( $\text{Si} < 12.5\text{wt.}\%$ ) are the majority in the literature (Carvalho et al. 2013, 2014; Dias et al. 2020; Gündüz et al. 2004; Kaya et al. 2003, 2007; Peres et al. 2004). In the case of secondary dendritic spacings ( $\lambda_2$ ), the  $n$  exponent has assumed values equal to  $2/3$  and  $1/3$  for  $V_L$  and  $T_R$ , respectively (Barros et al. 2016; Bouchard and Kirkaldy 1997; Carvalho et al. 2013, 2014; Costa et al. 2015; Ferreira et al. 2018; Peres et al. 2004; Quaresma et al. 2000; Rocha et al. 2003). In eutectic Al-Si alloy ( $\text{Si} \cong 12.6\text{wt.}\%$ ) as well as hypereutectic Al-Si alloys (Reyes et al. 2017), the morphology of primary phase has been observed as halos dendrite. Studies on correlation of secondary arms ( $\lambda_2$ ) of halo dendrites with  $V_L$  and  $T_R$  are scarce or almost unknown, being more frequent studies on primary arms. Thus, the present work has as main goal to elaborate a thermal analysis ion secondary dendritic growth via transient upward solidification of the Al12.6Si alloy (wt.%).

## 2. EXPERIMENTAL PROCEDURE

The investigated alloy was prepared from its individual elements Al and Si ( $\text{Al} > 99.6\%$  and  $\text{Si} > 99.7\%$ ). The resulting ingot of the Al-12.6wt.% Si alloy was obtained by means of a water-cooled directional solidification device, as shown in Figure 2a. It is also possible to observe the heat transfer modes active in the solidification process as well as the direction of gravity (Figure 2b). A data acquisition system connected to eight thermocouples, inserted into the metal in positions from the cooled base (heat transfer surface), has been used, as seen in Figure 2c. The complete details of the device as well as the operational principle of the upward directional solidification procedure have been described in our recent work (Rocha et al. 2020).

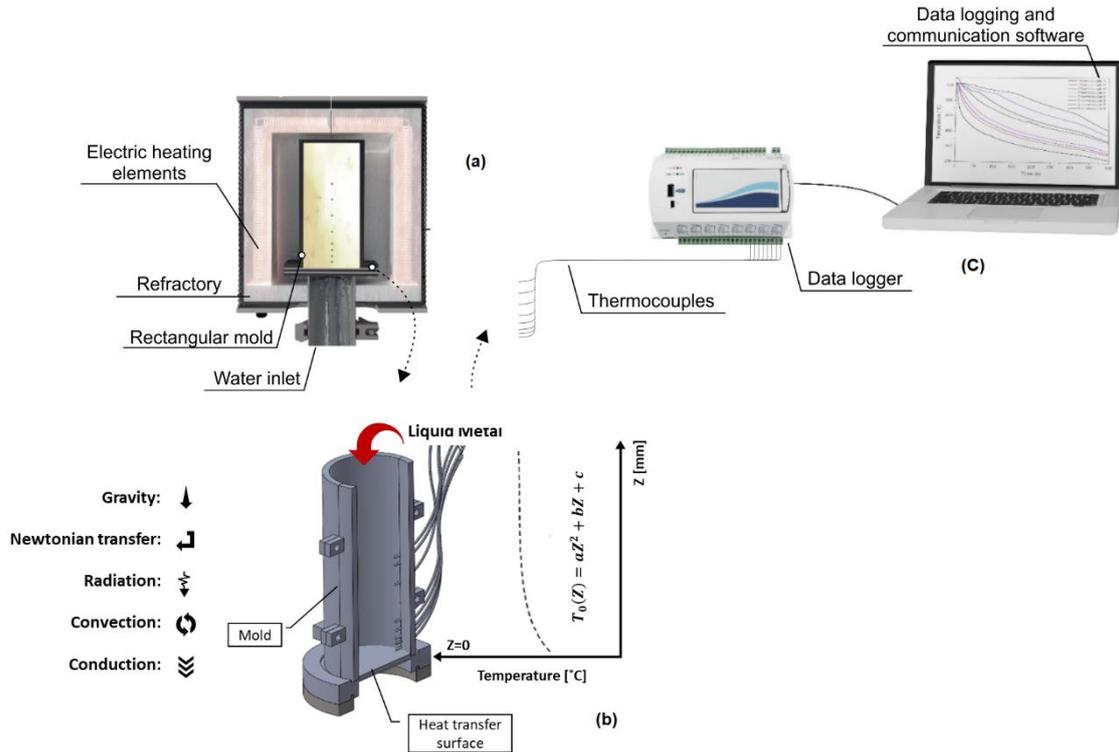


Figure 2. Schematic representation of the solidification furnace: (a) water-cooled upward solidification device, (b) detail of the ingot mold with positioning of the thermocouple and the heat transfer modes and (c) data acquisition system (Rocha et al. 2020).

Once the ingot was obtained, longitudinal samples were sectioned and the microstructure has been characterized as well as the secondary dendritic spacings have been measured following methodology from the literature (Costa et al. 2015; Ferreira et al. 2018; Peres et al. 2004; Quaresma et al. 2000; Rocha et al. 2003b), as can be seen in Figure 3. Image processing system MOTIC and the Image J software were used for measurement of  $\lambda_2$  independent readings (~20) for each selected position, with the average taken to be the local spacing and their distribution range.

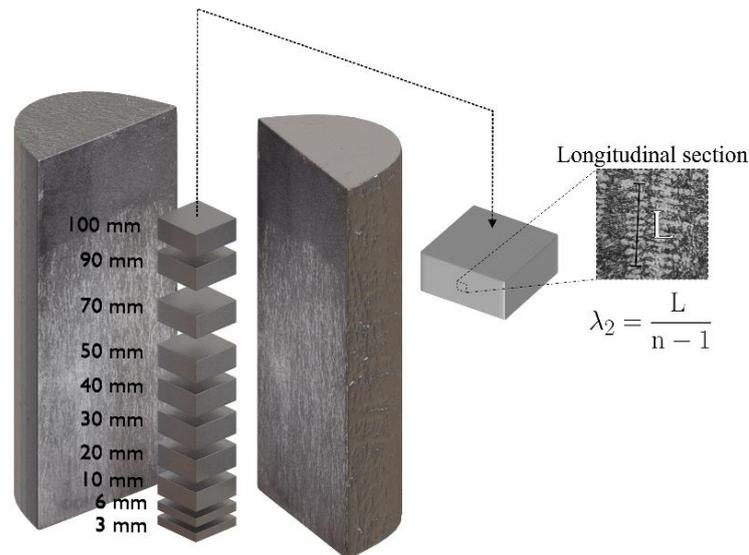


Figure 3. Detailing of the resulting as-cast ingot from the upward solidification, showing the longitudinal samples and the technique of  $\lambda_2$  measurement (Adapted of Rocha et al. 2020).

### 3. RESULTS AND DISCUSSION

Figure 4 shows the results of thermal processing, and experimental curves for each thermocouple, inserted in the metal, which have been published in our recent work (Rocha et al. 2020). It was allowed to determine experimentally the growth and cooling rates ( $V_L$  e  $T_R$ ), as shown in Figure 3b. The intersection of the *liquidus* temperature ( $T_L$ ) of the investigated alloy with the respective curves has allowed to obtain the solidification kinetics, represented by the mathematical expression given by  $P=1.65(t)^{0.77}$ .  $V_L$  and  $T_R$  have been determined by the derivatives  $d[1.65(t)^{0.77}]/dt$  and  $dT/dt$  at the point of intersection of  $T_L$  with the respective experimental curves, respectively (Barros et al. 2016; Carvalho et al. 2013, 2018; Costa et al. 2015; Ferreira et al. 2018; Peres et al. 2004; Quaresma et al. 2000; Reyes et al. 2017; Rocha et al. 2003; Rocha et al. 2020). It is observed that the cooling water imposes high  $V_L$  and  $T_R$  values close to the heat-transfer surface (Figure 1b), which gradually decrease with the progress of vertical solidification. It has been due to the progressive formation of the solidified layer, which acts during the solidification as resistance to the heat extraction by conduction, as observed in Figure 1a.

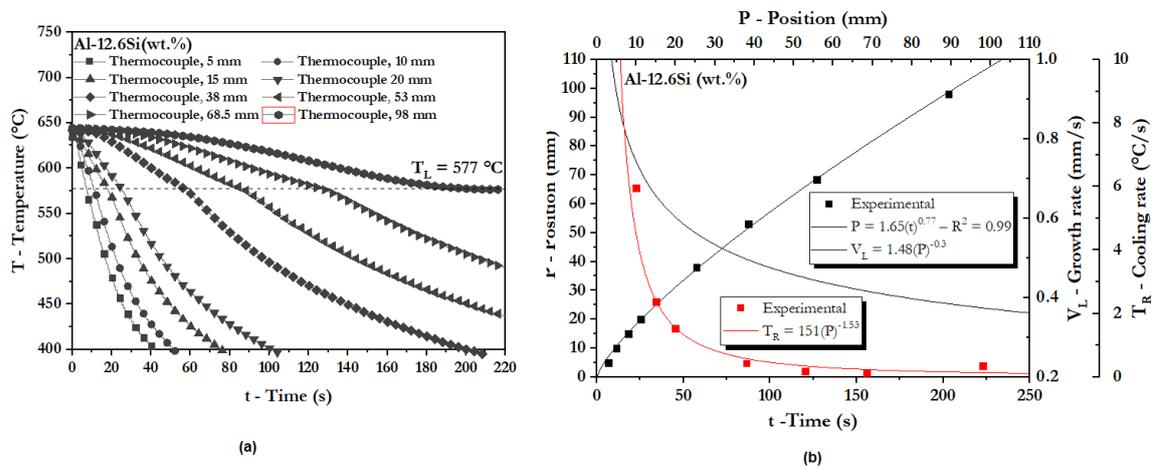


Figure 4. Thermal analysis resulting from the upward solidification of the Al12.6Si alloy (wt.%): (a) experimental curves, (b) thermal solidification parameters.

Figure 5 shows the  $\lambda_2$  variation with the position in the as-cast ingot as well as with the transient solidification thermal parameters,  $V_L$  and  $T_R$ . Mathematical expressions given by  $\lambda_2 = 8.7(V_L)^{-0.66}$  and  $\lambda_2 = 13.7(T_R)^{-0.24}$  have characterized the  $\lambda_2$  dependence as a function of  $V_L$  and  $T_R$ , respectively. It can be observed that finer microstructures, i.e., lower  $\lambda_2$  values have been obtained for as-caste samples close to the heat transfer surface, where  $V_L$  and  $T_R$  became higher. The microstructure observed in this work has been of halo dendrite, consisting of a branched network of primary and secondary arms, as seen in Figure 6. Reyes et al. (2017) and Rocha et al. (2020) have also observed it.

The exponent equal to the 0.66 (2/3) found in the mathematical correlation  $\lambda_2$ - $V_L$  is in agreement with the theoretical (Bouchard and Kirkaldy 1997) and experimental (Carvalho et al. 2018; Peres et al. 2004; Spinelli, Peres, and Garcia 2005) predictions for secondary dendritic growth proposed in the literature for hypoeutectic Al-Si alloys, as well as for other binary and multicomponent Al-based alloys (Barros et al. 2016; Costa et al. 2015; Quaresma et al. 2000; Rocha et al. 2003). On the other hand, the  $n$  exponent ( $\cong 0.24$ ) proposed in this work to characterize the  $\lambda_2$  growth law with  $T_R$ , for the eutectic Al-Si alloy, is different from that suggested for hypoeutectic Al-Si alloys ( $0.66 \cong 2/3$ ) (Carvalho et al. 2018; Peres et al. 2004; Spinelli et al. 2005). Peres et al. (Peres et al. 2004) have depicted for upward solidified hypoeutectic Al-xSi alloys (x=3, 5, 7 and 9wt.%) the following experimental equations  $\lambda_2=A.(V_L)^{-2/3}$  ( $A=32, 32, 26$  and  $22$ , respectively). Spinelli et al. (Spinelli et al. 2005) have reported for downward solidified hypoeutectic Al-xSi alloys (x=5, 7 and 9wt.%) the following experimental expressions  $\lambda_2=A.(V_L)^{-2/3}$  ( $A=31, 25, \text{ and } 22$ , respectively).

More recently, Carvalho et al. (2018) have obtained for the horizontally solidified Al-9wt.%Si alloy the following mathematical expression  $\lambda_2=18.48(V_L)^{-2/3}$ , whose exponent (2/3) has been exactly equal to the found in the present work, as well as that reported for upward solidified Al-xSi alloys (Peres et al. 2004; Spinelli et al. 2005). It can be observed that the  $A$  constants values found in the literature (Carvalho et al. 2018; Peres et al. 2004; Spinelli et al. 2005) have higher than those obtained in the present work ( $A=8.7$ ). It has allowed to deduce that the secondary arm spacing in halo-like dendrites, observed as a primary phase in the eutectic Al-Si alloy, are smaller than the secondary dendrite arm spacing often seen in hypoeutectic Al-Si alloys.

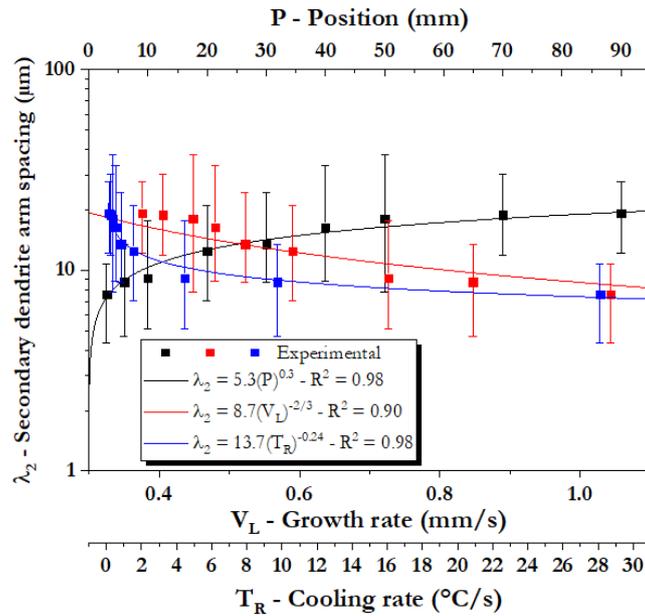


Figure 5.  $\lambda_2$  variation with the position as-cast ingot, growth rate and cooling rate

Figure 6 shows the typical solidification microstructures revealed for two longitudinal positions from the cooled surface (see Figure 1a). It is observed significant changes in the length of the microstructure scale in the as-cast sample further away from the cooled base ( $P=30\text{mm}$ ), since the greater thermal resistance to heat extraction in the metal/mold system has prevailed through the heat conduction mode, due to the increasing formation of the solid layer, as observed in Figure, and in these conditions, it results lower  $V_L$  and  $T_R$  values and, as a consequence, coarser microstructure has been found.

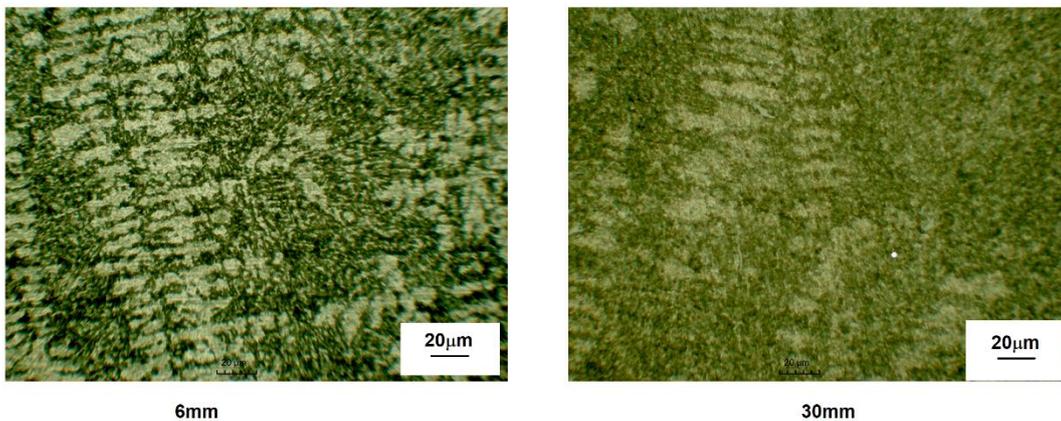


Figure 6. Typical solidification microstructure for two samples in the ingot from the cooled base.

#### 4. CONCLUSIONS

The main conclusions reached in this work were:

1. The water-cooled solidification device imposes high growth and cooling rates values close to the heat-transfer surface, providing an inverse mathematical relation of  $V_L$  and  $T_R$  with the position in the as-cast ingot, given by the general formula  $(V_L, T_R) = \text{Constant} \cdot P^{-n}$ .
2. The as-cast microstructure of the analyzed primary phase, for the Al-12.6Si alloy (wt.%), under the assumed solidification conditions in this work, has been characterized by a network of halos dendrite, according to a recently published work (Rocha et al. 2020).
3. As expected, the solidification conditions ( $V_L$  and  $T_R$ ) have influenced the length of the microstructural scale, where finer microstructures have been found in samples close to the heat-transfer surface, i.e., lower  $\lambda_2$  values for higher  $V_L$  and  $T_R$  values.

- Power mathematical expressions given by  $\lambda_2=8.7(V_L)^{-0.66}$  and  $\lambda_2=13.7(T_R)^{-0.24}$  have characterized the  $\lambda_2$  variation as functions of  $V_L$  and  $T_R$ . Despite the primary phase has been observed as halo-type dendrite, it has been observed that the exponent equal to 2/3 found for the eutectic Al-Si alloy, investigated in this work, is perfectly in agreement with the literature for hypoeutectic Al-Si alloys. On the other hand, the exponent equal to 0.24, found for secondary arms of halo dendrite, does not represent the growth laws of  $\lambda_2$  ( $2/3 \cong 0.66$ ) proposed in the literature for Al-based hypoeutectic alloys.
- Finally, through comparative analysis with the literature for other Si compositions, it has been possible to verify that the  $\lambda_2$  values decrease with the increase of Si content. It is in agreement with the theoretical prediction for  $\lambda_2$  growth, proposed by Bouchard-Kirkaldy (Bouchard and Kirkaldy 1997).

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