Influence of thermal parameters on the dendritic arm spacing and the microhardness of Al-5.5wt.%Sn alloy directionally solidified

Influência dos parâmetros térmicos sobre os espaçamentos dendríticos primários e na microdureza da liga Al-5,5%Sn solidificada direcionalmente

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Abstract

Al-Sn alloys are widely used in tribological applications. In this study, thermal, microstructural and microhardness (HV) analysis were carried out with an Al-5.5wt.%Sn alloy ingot produced by horizontal directional transient solidification. The main parameters analyzed include the growth rate (V_L) and cooling rate (T_R). These thermal parameters play a key role in the microstructural formation. The dendritic microstructure has been characterized by primary dendritic arm spacing (λ_1) which was experimentally determined and correlated with V_L, and T_R. The behavior presented by the Al-5.5wt.%Sn alloy during solidification was similar to that of other aluminum alloys, i.e., the dendritic network became coarser with decreasing cooling rates, indicating that the immiscibility between aluminum and tin does not have a significant effect on the relationship between primary dendritic arm spacing and the cooling rate. The dependence of the microhardness on V_L, T_R and λ_1 was also analyzed. It was found that for increasing values of T_R, the values of HV decrease. On the other hand, the values of HV increase with increasing values of λ_1.

Keywords: *Horizontal directional solidification, unsteady-state heat flow, primary dendrite arm spacing, microhardness, Al-Sn Alloys.*

Resumo

As ligas Al-Sn são amplamente utilizados em aplicações tribológicas. Nesse estudo, análises térmica, microestrutural e dureza (HV) foram realizadas ao longo de um lingote da liga Al-5,5%Sn, obtido por solidificação direcional horizontal transitória. Os principais parâmetros analisados incluem a velocidade de deslocamento da isoterma liquidus (V_L) e a taxa de resfriamento (T_R). Esses parâmetros térmicos desempenham um papel fundamental na formação da microestrutura. A microestrutura dendrítica foi caracterizada através dos espaçamentos dentríticos primários (λ_1), os quais foram determinados, experimentalmente, e correlacionados com V_L, e T_R. O comportamento apresentado pela liga Al- 5,5% Sn, durante a solidificação,é semelhante ao de outras ligas de alumínio, isto é, observa-se rede dendrítica mais grosseira com a diminuição da taxa de resfriamento, indicando que a imiscibilidade entre o alumínio e estanho não tem um efeito significativo sobre o relação entre o espaçamento dendrítico primário e taxa de resfriamento. A dependência da microdureza em V_L, T_R e no λ_1 foi também analisada. Verificaram-se menores valores de HV para maiores T_R. Por outro lado, os valores HV aumentam com valores crescentes de λ_1.

Palavras chave: Solidificação, regime transitório de extração de calor, espaçamentos dendríticos primários, ligas Al-Sn.

1. Introduction

Alloys of the Al-Sn system are known to have good mechanical and tribological properties, making the alloys of this system suitable for applications requiring satisfactory wear resistance, for example automotive bearings (Yuan *et al*., 2000). The metallographic structure is characterized by a heterogeneous aluminum matrix with tin particles dispersed throughout the matrix. This type of structure determines the tribological behavior of the alloy, with the unyielding matrix being responsible for mechanical strength while the particles of Sn act as a solid lubricant (Perrone *et al*., 2002). Due to the limited miscibility of Sn in Al, it is expected that the rapid solidification conditions significantly affect the tribological properties by the modification of the microstructure (Cruz, 2008; Cruz *et al*., 2010).

When a metallic alloy is solidified, the most frequently observed solid morphology is the dendritic microstructure (Kaya *et al*., 2013), which is characterized by its microstructure parameters. Numerous solidification studies (Okamoto and Kishitake, 1975; Rocha *et al*, 2003; Peres, *et al*., 2005; Carvalho *et al*., 2013), all for Al-based alloys, have been reported with a view to characterizing the microstructure parameters, such as primary dendrite arm spacing (λ_1) and secondary dendrite arm spacing (λ_2) as a function of V_L and T_R; all destined for vertical upward directional solidification. In this case the influence of the convection is minimized when the solute is rejected for to the interdendritic regions, providing the formation of an interdendritic liquid denser than the global volume of liquid metal. When the process is carried out vertically downward, the system provides the melt convection that arises during the process. In the horizontal unidirectional solidification, when the chill is placed on the side of the mold, convection in function of the composition gradients in the liquid always occurs. An interesting feature of the horizontal configuration is the gradient of solute concentration and density in a vertical direction because solute-rich liquid precipitates, whereas free solvent-crystals rise due to buoyancy force. Moreover, there will also be a vertical temperature gradient in the sample as soon as a thermosolutal convection roll emerges. In spite of these particular physical characteristics, only a few studies (Nogueira *et al*., 2012) have reported these important effects of melt convection and direction of growth on dendrite arm spacing for this particular case.

Realizing the influence of structural parameters on the mechanical properties of the produced material, some researchers began studies that correlate the microstructure and properties, such as in the 50s, when Hall and Petch (1951; 1953) proposed a relationship that relates the grain diameter versus the yield stress or hardness of the material,

$$H=H_0+k\times d^{(-1/2)} \text{ and } \sigma_e=\sigma_i+k\times d^{(-1/2)} \quad (1)$$

where H is the hardness of the material; σ_e is the yield stress, "d" is the average grain size; H_0, σ_i and k are particular constants obtained experimentally for the material. However, for some metallic systems, the dendrite arm spacing may have a more significant effect on the resulting mechanical properties of the material than the actual grain size.

The microhardness analysis is reported by Fan et al. (2010) as a relatively simple way to accomplish the complex task of predicting mechanical properties of alloys. In the case of Ti–Al alloys, Vickers microhardness (HV) is already used for quality control in the production of turbochargers, automotive valves and compressor blades. Kaya et al. (2004) reported the effect of the lamellar spacing of Pb–Cd, Sn–Zn and Bi–Cd eutectic alloys on microhardness. In some recent studies with binary Al–Fe hypoeutectic alloys, Hall–Petch type relationships between hardness and the scale of the microstructure have been proposed for alloys solidified under transient heat flow conditions (Silva *et al*., 2012). It was shown that the hardness increases with the increase in alloy solute content and with the decrease in cell/dendritic spacing for any alloy examined. In recently published studies, Kaya *et al*. (2008; 2013) correlated the microstructural parameters (λ_1 and λ_2) of aluminum based alloys with hardness values, measured on the transverse and longitudinal sections of the cast product under steady-state solidification conditions. The results showed that the increasing of the dendritic arm spacing promoted reduction of the tested hardness, i.e., the studies indicate that the structure refinement degree yields materials with higher hardness and therefore with greater wear resistance. Thus, the aim of the present work was to experimentally investigate the influence of thermal parameters on the dendritic arm spacing and the microhardness of the Al-5.5wt.%Sn alloy directionally solidified in a horizontal solidification device.

2. Experimental procedure

2.1 - Sample Preparation and Solidification

Horizontal unidirectional solidification experiments were carried out with the Al-5.5wt.%Sn alloy in unsteady-state heat flow conditions in order to simulate solidification conditions typical of industrial foundry processes. The casting assembly used in solidification experiments was recently published (Nogueira *et al*., 2012, Carvalho *et al*., 2013). It was designed in such a way that the heat was extracted only through the water-cooled system placed in the lateral mold wall, promoting horizontal directional solidification. The carbon steel mold used had a wall thickness of 3 mm, a length of 110 mm, a height of 60 mm and a width of 80 mm. The lateral inner mold surfaces were covered with a layer of insulating alumina and the upper part of the mold was closed with refractory material to prevent heat losses. The thermal contact condition at the metal/mold interface was also standardized with the heat extracting surface being polished.

The alloy was melted in situ and heated until a superheat of 10% above the liquidus temperature (T_L) using an electrical furnace. Approaching the superheat temperature, the mold was taken from the heater and set immediately on a water cooled carbon steel chill. Water was circulated through this cooling jacket keeping the carbon steel plate during the solidification at a constant temperature of about 25^{0}C and thus inducing a longitudinal heat transfer from the mold. During the solidification process, temperatures at different positions in the alloy samples were measured and the data were acquired automatically. For the measurements, a set of five fine type K thermocouples, arranged as shown in Figure 1, was used. The thermocouples were sheathed in 1.6 mm diameter steel tubes, and positioned at 5, 10, 15, 30, and 50 mm from the heat-extracting surface. The thermocouples were calibrated at the melting point of Al, exhibiting fluctuations of about 0.4^{0}C and 1^{0}C respectively, and connected by coaxial cables to a data logger interfaced with a computer. Previous measurements of the temperature field were carried out confirming that the described experimental set-up fulfills the requirement of a unidirectional heat flow in horizontal direction.

2.2 - Measurements of Growth Rate and Cooling Rate

Experimental cooling curves for the five thermocouples inserted into the casting during solidification of the alloy investigated in this study are shown in Figure 1. It is well known that the primary dendritic arm spacing are dependent on solidification thermal variables such as V_L and T_R, all of which vary with time and position during solidification. In order to determine more accurate values of these parameters, the results of experimental thermal analysis have been used to determine the displacement of the liquidus isotherm, i.e., the thermocouples readings have also been used to generate a plot of position from the metal/ mold interface as a function of time corresponding to the liquidus front passing by each thermocouple. A curve fitting technique on such experimental points has generated power functions of position as a function of time. The derivative of this function with respect to time has yielded values for V_L. The T_R profile was calculated by considering the thermal data recorded immediately after the passing of the *liquidus* front by each thermocouple. The method for measuring the tip cooling rate was used recently by Rocha et al. (2003).

2.3 - Metallographic Analysis

Selected transverse (perpendicular to the growth direction) sections of the directionally solidified specimens at 10, 15, 20, 30, 40, 50, 60, 70, 80, 90 and 100 mm from the metal-mold interface were polished and etched with a solution of 5%NaOH in water for micrograph examination. Image processing system Olympus BX51 and Image Tool (IT) software were used to measure primary arm spacing (about 20 independent readings for each selected position, with the average taken to be the local spacing) and their distribution range. The method used for measuring the primary arm spacing on the transverse section was the triangle method utilized by Rocha *et al*.(2003).

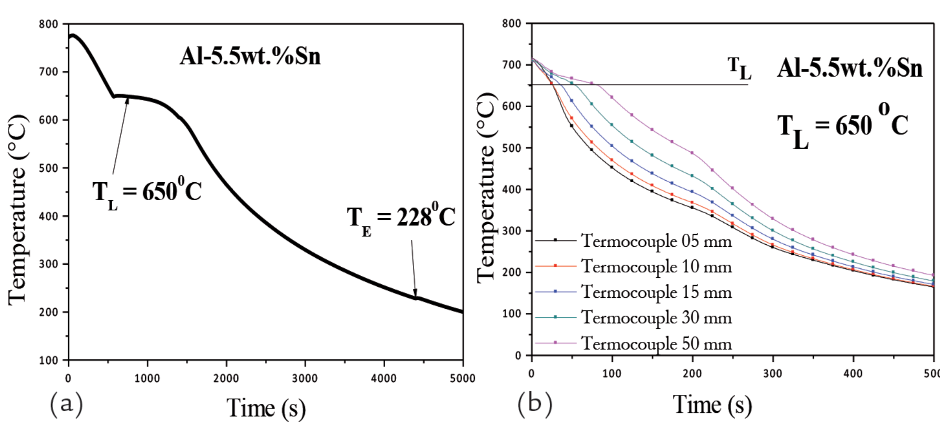

Figure 1
(a) Cooling curves showing the *liquidus* (T_L) and solidus (T_E) temperatures of the alloy studied and (b) Cooling curves recorded by five thermocouples inside the casting.

2.4 - Measurement of Microhardness

The mechanical properties of any solidified materials are usually determined with hardness tests, tensile strength tests, ductility tests, etc. Since true tensile strength testing of the solidified alloys gave inconsistent results with wide scattering, due to strong dependence on the solidified sample surface quality, the mechanical properties were monitored by hardness testing, which is one of the easiest and most straightforward techniques (Kaya *et al.*, 2013).Thus, microhardness measurements in this work were carried out using a Shimadzu HMV-2 hardness measuring test device using a 50 g load and a dwell time of 10 s, according to the ASTM: E 384-11e1 and Kaya *et al.* (2008, 2013). The adopted Vickers microhardness was the average of at least 20 measurements on each sample, according method applied by Dias Filho (2013).

3. Results and discussion

Figure 2 presents microstructures of a cross section of the samples at 10, and 60 mm from metal/mold interface, showing the primary dendrite arms. Figure 3(a) shows the average experimental values of primary dendritic spacing as a function of distance from the metal/mold interface obtained in this work. It is observed that dendrite arm spacing increases with the distance from the heat-extracting surface of the alloy investigated. It is noted that this behavior is reflected in correlation with the growth rate and cooling rate, i.e., primary spacing increases with decreasing thermal parameters, as shown in Figures 3 (b) and (c).

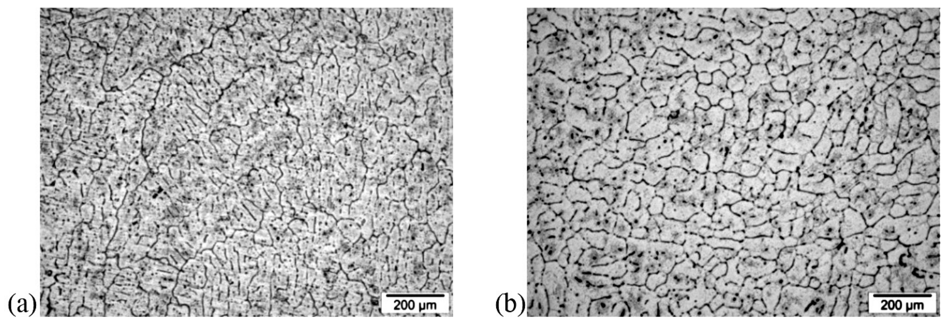

Figure 2
Solidification microstructure: (a) P = 5 mm, V_L = 0.75 mm/s, T_R = 6.6 ^{0}C/s and λ_1 = 65 µm; (b) P = 60 mm, V_L = 0.60 mm/s, T_R = 1.30 ^{0}C/s and λ_1 = 119 µm;

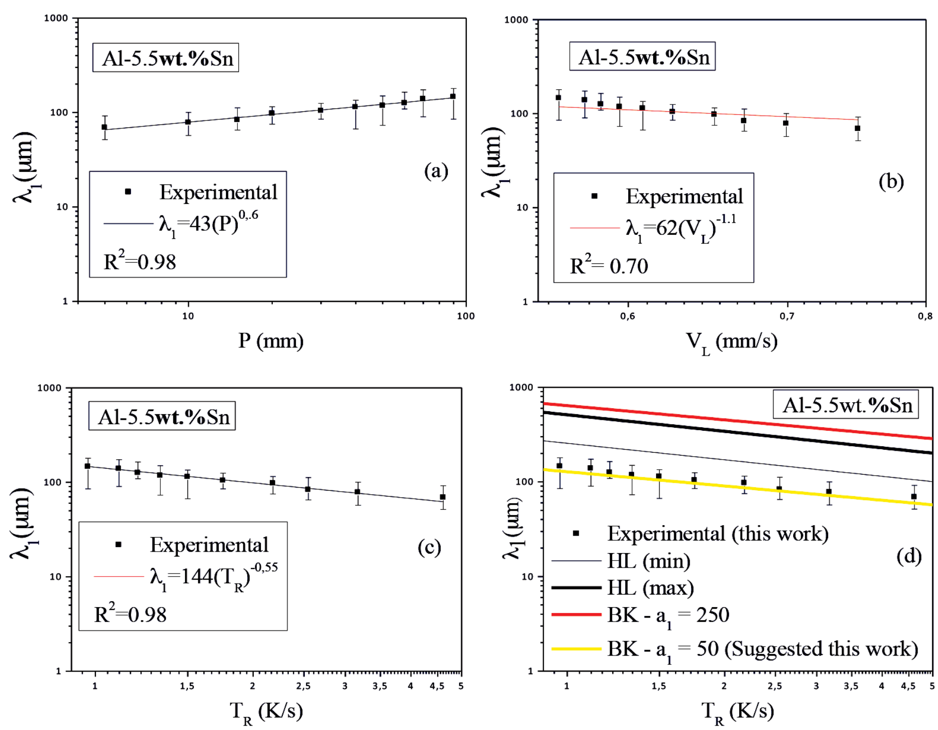

Figure 3
(a) Primary dendrite arm spacing as a function of: (a) distance from metal-mold interface; (b) tip growth rate; (c) tip cooling rate and (d) Comparison of experimental and theoretical primary dendrite arm spacing as a function of cooling rate. The R^2 is the least-squares correlation coefficient.

Results from literature for Aluminum based binary alloys: Al-Cu (Rocha et al,, 2003), Al-Si (Peres *et al.*, 2005; Carvalho *et al.*, 2013) and Al-Sn (Cruz *et al.*, 2010) alloys also indicate an increase in spacing with decreasing V_L and T_R. Observed by Cruz *et al.* (2010), the Al-Sn is an immiscible binary alloy system, with an Al-rich dendritic matrix involving an Sn-rich eutectic mixture distributed along the interdendritic regions. For positions closer to the cooled casting, i.e., at high values of the cooling rate, a more extensive distribution of the eutectic mixture is attained, which is consequently associated with the fineness of the interdendritic spacing. Notice that Figure 3(c) shows that power laws equal to -1.1 and −0.55 characterize the experimental variation of primary spacing with growth rate and cooling rate, respectively, *i.e*: $\lambda_1 = 62(V_L)^{-1.1}$ and $\lambda_1 = 144(T_R)^{0.55}$. This is in agreement with observations reported by Rocha *et al* (2003A),Peres et al(2005), Carvalho et al (2013) and Cruz *et al.*(2012) who stated that exponential relationships λ_1 = constant $(T_R)^{-0.55}$ best generate the experimental variation of primary dendritic arms with cooling rate along the unsteady-state solidification of Al–Cu, Al–Si and Al-Sn alloys, respectively.

Figure 3(d) shows the comparisons between the present experimental results of primary spacing with theoretical predictions furnished by unsteady-state predictive models: Hunt–Lu's model (HL) (1966) and Bouchard–Kirkaldy's model (BK) (1997), with a calibration factor a_1 = 250 for Al–Sn alloys, as suggested by these authors and a_1 = 50, suggested this work. It is observed that the theoretical values calculated from both models overestimate the experimental values. On the other hand, for a_1 = 50, a good approximation is observed between the experimental results and those calculated by BK. Similarly, Cruz (2008) also noted that the experimental data did not fit the model of Bouchard-Kirkaldy for primary growth. The author attributes this behavior to the extensive range of solidification of the Al-Sn system, and the large difference in density between the two components (virtually immiscible), which significantly increases under heat extraction conditions in the vertical direction, which phenomenon can be attributed to results observed in this work. The predicted dendritic spacing calculated for unsteady solidification by Hunt-Lu's and Bouchard- Kirkaldy's models are subjected to deviations caused mainly by the unaccounted diffusion relaxations and coring reductions for primary spacing. The thermophysical properties of the Al-5.5wt.%Sn alloy, used to calculate the theoretical values of λ_1 were obtained from Themo-Calc software and are summarized in Table 1.Comparison between the results obtained in this work with those of Cruz (2008) and Okamoto-Kishitake (1975) is performed, as shown in Figure 4. The results of Cruz underestimate Okamoto-Kishitake's values. On the other hand, noted is an excellent approximation of obtained experimental results with those of Okamoto-Kishitake. This can be attributed to the compositions of the alloys being very close.

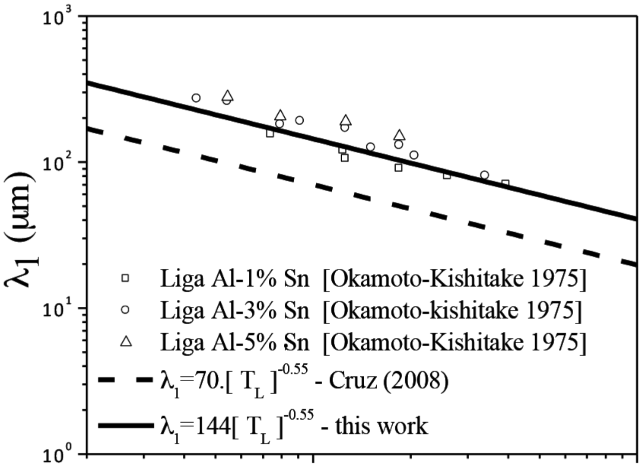

Figure 4
Comparison between the results obtained in this work with those of Cruz (2008) and Okamoto-Kishitake.

The dependence of the microhardness on the growth rate, cooling rate and the primary dendrite arm spacing for the investigated alloy is shown in Figures 5 (a), (b), (c) and (d), respectively. It can be seen from Figures 5 (a) e (b) that an increase in the growth rate and cooling rate leads to a decrease in the microhardness. The variation of the microhardness as a function of the dendritic spacing can be noted in Figures 5 (c) and (d), respectively. It can be observed that an increase in the λ_1 values also increases the HV values. This result is in accordance with the study of Cruz (2010), which observed that for the Al-Sn alloys, the wear volume decreases with increasing λ_1. According to the author the lower wear volume observed for coarser dendritic structures for the Al-Sn alloys seems to be associated with the larger Sn-rich interdendritic regions. He also assumed that the lubrication effect of the soft Sn-rich areas seems to be improved for coarser microstructures. Variation of micro-indentation hardness with solidification and microstructure parameters in the Al based alloys was investigated recently by Kaya *et al.* (2008, 2013). The exponent value of λ_1 obtained in this work (0.30) is between to the values of 0.28, 0.40, and 0.43, 0.35obtained by Kaya *et al.* (2008, 2013) for Al-based alloys (Al-Si, Al-Cu and Al-Ti alloys), solidified in an Bridgman system.

Properties	**Density**	**Latent heat of fusion**	**Solute diffusivity**	**Gibbs–Thomson coefficient**	**Solidus tempera-ture**	**Liquidus temperature**	**Partition coefficient**	**Liquidus slope**
Symbol/units	ρ_S [kg/m³] (solid) ρ_L [kg/m³] (liquid)	L [J/kg]	D [m²/s]	Γ [m.K]	T_E [°C]	T_L [°C]	k_O	m_L(K/wt pct)
Al-5.5wt.%Sn	3011.6 2819.8	39.700.0	3 x 10^{-9}	9 x 10^{-8}	227	650	0.041	7.2

Table 1
Thermophysical properties of the Al 5.5wt.%Sn alloy used in the HL and BK models (Thermo-Calc).

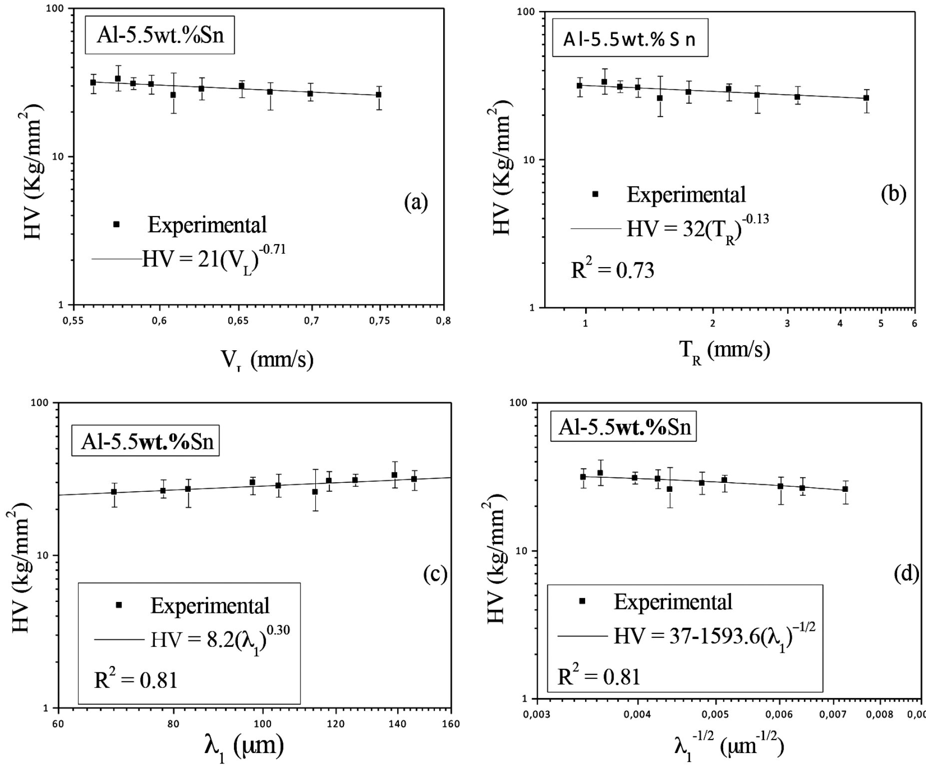

Figure 5
Variation of microhardness as a function: (a) growth rate; (b) cooling rate; (c) primary dendrite arm spacing (λ_1) and (c) $\lambda_1^{-1/2}$.The R^2 is the least-squares correlation coefficient

Okamoto-Kishitake (1975).

4. Conclusions

The principal results of the present work can be summarized as follows:

(1) Experimental observations show that the values of λ_1 decrease as the V_L and T_R increase. Relations between λ_1 with the growth rate and cooling rate were obtained by the following experimental laws: $\lambda_1 = 62(V_L)^{-1.1}$ and $\lambda_1=144(T_R)^{-0.55}$.

(2) The values of HV for directionally solidified Al-5.5wt%Sn alloy were measured. It was found that the values of microhardness increase with the decreasing of the values of V_L as well as with the increasing of the values of the λ_1. Microhardness has shown to be significantly affected by the thermal parameters and by the scale of the primary dendrite arm spacing. Experimental laws have been determined correlating HV with V_L, T_R and λ_1, giving as results: $HV = 21(V_L)^{-0.71}$, $HV = 32(T_R)^{-0.13}$, $HV = 8.2(\lambda_1)^{0.30}$ and $HV = 37 - 1593(\lambda_1)^{-1/2}$.

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6. References

BOUCHARD, D., KIRKALDY, J. S. Prediction of dendrite arm spacings in unsteady and steady-state heat flow of unidirectionally solidified binary alloys. ***Metallurgical and Materials Transactions*** B, v. 28B, p. 651-663, 1997.

CARVALHO, D. B., GUIMARÃES, E. C., MOREIRA. A. L., MOUTINHO, D. J., DIAS FILHO, J. M., ROCHA, O.L. Characterization of the Al-3wt.%Si alloy in unsteady-state horizontal directional solidification. ***Materials Research***. v. 16.n.4, p. 874-883, 2013.

CRUZ, K.A.S. Microestrutura de solidificação, resistências mecânica e ao desgaste de ligas Al-Sn e Al-Si. Campinas, SP, 2008. 211 p. Tese de Doutorado em Engenharia Mecânica.

CRUZ, K.S, MEZA, E.S, FERNANDES, F.A.P, QUARESMA, J.M.V, CASTELETTI, L. C, GARCIA, A. Dendritic arm spacing affecting mechanical properties and wear behavior of Al-Sn and Al-Si alloys directionally solidified under unsteady-state conditions. ***Metallurgical and Materials Transactions A***, v. 41, p. 972-984, 2010.

DIAS FILHO, J. M. Análises térmica e microestrutural na solidificação de ligas monofásica e peritéticas do sistema Zn-Ag. Campinas, SP, 2013, 109 p. (Dissertação

de Mestrado).
FAN, J., LI, X., SU, Y., GUO, J., FU, H. Dependency of microhardness on solidification processing parameters and microstructure characteristics in the directionally solidified Ti–46Al–0.5W–0.5Si alloy. ***Journal of Alloys and Compounds***, v. 504, p. 60-64, 2010.
HALL, H.O. The deformation and ageing of mild steel: III **DISCUSSION OF RESULTS. PROCEEDINGS OF THE PHYSICAL SOCIETY**, 53.v. 64B, p. 747–753, 1951.
HUNT,J.D, LU,S.Z. Numerical modeling of cellular/dendritic array growth: spacing and structure predictions. ***Metallurgical Materials Transactions*** A, v. 27A. p. 611-23, 1966.
KAYA, H., GÜNDÜZ, M., ÇADIRLI, E., UZUN, O. Effect of growth rate and lamellar spacing on microhardness in the directionally solidifiedPb–Cd, Sn–Zn and Bi–Cd eutectic alloys. ***Journal of Materials Science***, v. 39, p. 6571–6576, 2004.
KAYA, H., ÇADIRLI, E., BÖYÜK, U., MARASLI, N. Variation of microindentation hardness with solidification and microstructure parameters in the Al based alloys. ***Applied Surface Science***, v. 255, p. 3071–3078, 2008.
KAYA, H., ÇADIRLI, E., BÖYÜK, U., MARASLI, N. Influence of growth rate on microstructure, microhardness, and electrical resistivity of directionally solidified Al-7 wt% Ni hypo-eutectic alloy. ***Metals and Materials International***, v. 19, p. 39-44, 2013.
NOGUEIRA, M.R., CARVALHO, D.B., MOREIRA, A.L., DIAS FILHO, J.M., ROCHA, O.L. Espaçamentos dendríticos primários da liga Sn-5%Pb solidificada direcionalmente em um sistema horizontal. ***Revista Matéria***, v. 17, n. 2, p. 1009-1023, 2012.
OKAMOTO, T., KISHITAKE, K. Dendritic structure in unidirectionally solidified aluminum, tin, and zinc base binary alloys.Journal of Crystal Growth, v. 29, p. 137-146, 1975.
PERRONE, A., ZOCCO, A., de ROSE, H., ZIMMERMANN, R., BERSANI, M. Al-Sn thin films deposited by pulsed laser ablation. ***Materials Science and Engineering*** C, v. 22, p. 465-468, 2002.
PETCH, N.J. The cleavage strength of polycrystals. ***Journal of the Iron and Steel Institute***, v. 174, p. 25–28, 1953.
ROCHA, O.L., SIQUEIRA, C.A., GARCIA, A. Heat flow parameters affecting dendrite spacings during unsteady-state solidification of Sn-Pb and Al-Cu alloys. ***Metallurgical and Materials Transactions A***, v.34A, p. 995-1006, 2003.
SILVA, B.L., GARCIA A., SPINELLI, J.E. The effects of microstructure and intermetallic phases of directionally solidified Al-Fe alloys on microhardness. ***Materials Letters***, v. 89, p. 291–295, 2012.
YUAN, G.C., LI, Z.J., LOU, Y.X., ZHANG, X.M. Study on crystallization and microstructure for New Series of Al-Sn-Si Alloys. ***Materials Science and Engineering*** A, v. 280, p. 108-115, 2000.

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