


# Effect of Squeeze Casting on Microhardness and Microstructure of Al-3Cu-xSi Alloy

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Aluminum alloys of the ANSI series 319 present Si and Cu as the main alloying elements and the mechanical strength of these alloys can be improved by the precipitation of the metastable  $Al_2Cu$  phase. Squeeze Casting is a casting process in which the cast metal is deposited in a metal mold and solidified under certain pressure exerted by a punch. This process provides changes in the microstructure which lead to changes in the mechanical properties of the material such as the increase in the microhardness of the processed alloy. alloys Al-5,5% Si-3% Cu, Al-7,5% Si-3% and Al-9% Si-3% Cu alloys were solidified by the squeeze casting process to analyze the microstructural and microhardness variations. Pressures of 0 MPa, 50 MPa, 100 MPa and 150 MPa were used. In general, with increasing pressure and composition, the  $\lambda_2$  values decrease from the metal/mold interface, especially at pressures of 150 MPa and Composition of 9%Si, on the other hand the increase of these variables leads to the formation of more phases  $Al_2Cu$  metastable that have a decrease in the Microhardness of the material, in addition to the formation of other phases due to the proximity to the eutetic.

**Keywords:** Squeeze Casting, Microstructure, Microhardness, Dendritic Spacing, Al-Si-Cu alloys.

## 1. Introduction

The main technological issue in the development of new pressure assisted casting processes is related to the increase of the holding pressure during the alloy solidification. In fact, it is evident that, by increasing the pressure, the overall quality of the casting generally increases in terms of a smoother surface, lower porosity and higher mechanical properties<sup>1</sup>. In die casting, higher holding pressures lead to higher production runs too. However, it is not yet clear what is the main role of the pressure during casting as many other process parameters are present and their effect cannot be easily separated from the effect of the pressure. Moreover, different results may be obtained depending on the alloy to cast and on its sensibility to the casting process. For aluminum alloys, the application of a holding pressure during cooling plays a very important role in defining casting properties. A significant microstructural refinement was already observed during the solidification of an Al-Cu alloy under high pressure (Han et al., 1994). In this case the average dendrite cell size changes from 30  $\mu m$ , without applied pressure, to 5  $\mu m$  less than 1.7 GPa. For an Al-Si alloy (designated as B390) a great microstructure refinement was also observed as a consequence of the pressure application (up to 100 MPa) and higher mechanical properties were measured in terms of hardness and tensile strength but no material model has never been proposed<sup>2</sup>. Other studies showed that in direct and indirect squeeze casting, apart from the pressure, other process parameters strongly affects microstructure and mechanical properties such as melt and die temperature, die geometry<sup>3,4</sup> and melt flow.

In such conditions, it is very difficult to understand what process parameter is the main one in determining the final mechanical properties of the cast alloy. As pressure increases, together with microstructure refinement, other structural modifications can occur such as shrinkage, porosity change<sup>5</sup> and macrosegregation<sup>6,7,8</sup>. As a result, a complete process map can be obtained with the definition of the process window where no micro and macro-defects occur<sup>5,6</sup>.

Aluminum alloys have properties of great industrial interest, such as: low density, good corrosion resistance, high thermal and electrical conductivity, good combinations of mechanical properties, good workability in machining processes and mechanical forming. These alloys can be divided into two groups. One contains those alloys for which the mechanical properties are controlled by work hardening and annealing. Commercial purity aluminum and alloys of the Al-Mg and Al-Mn systems are common examples. The second group comprises alloys such as Al-Cu-Mg, Al-Mg-Si, Al-Cu-Si and Al-Zn-Mg-Cu that respond to age or precipitation hardening<sup>9,10</sup>.

Particularly Al-Cu-Si alloys of the ANSI series 319.1 and 333.1 (in which compositions lie mostly within the ranges 5.5–10 Si and 3.0–4.0% Cu) are extremely important considering their applicability in the automotive and aeronautics industries, in the manufacture of components such as cylinders, pistons, engine blocks, etc. Their main characteristics are good castability and strength at relatively high temperatures, low coefficient of thermal expansion and good wear resistance<sup>11–15</sup>. In general, the Al-Si-Cu alloys are used for many of the applications listed for the binary alloys but where higher strength is needed.

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Increasing demands on such properties have pointed to the need for close microstructural control through tighter specification of composition, casting practice, and subsequent heat treatment<sup>9, 11, 13</sup>.

In unmodified Al-Si alloys, the Si particles have a plate-like morphology, which can act as crack initiators and have negative influence on ductility<sup>[11]</sup>. The ductility can be improved by changing the morphology of the Si particles towards a more fibrous form. This can be done using high cooling rates during casting, by addition of a chemical modifier, by exposing the casting to high temperature for long periods (heat treatments), or by a combination of these processes<sup>9, 11-14, 16, 17</sup>. For instance, the magnitude of cooling rate during solidification of Al-(Si, Cu) alloys, determines the fineness of the microstructure and other microstructural features such as fraction, size and distribution of intermetallic phases and segregation profiles of solute in the interdendritic regions<sup>[18]</sup>. Large and brittle intermetallic phases can be formed during slow solidification, with deleterious effects to the mechanical properties. Increased cooling rates result in finer microstructures and improved mechanical properties<sup>15, 18, 19, 20</sup>.

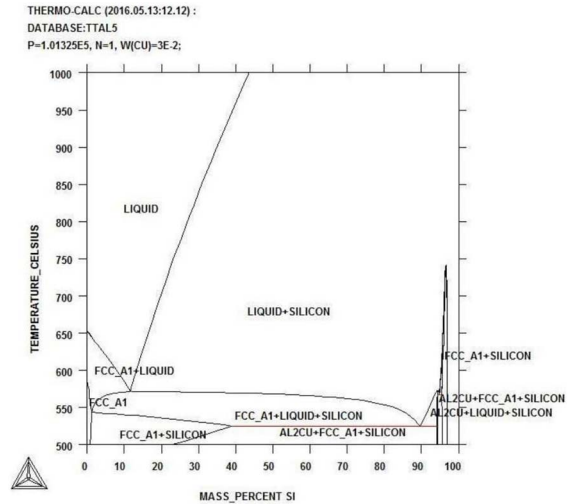
Depending on the purity of the base material, some of the alloying elements and impurities will be part of the Al- $\alpha$  solid solution and the remainder will precipitate in the form of intermetallic compounds in the grain contours and between the dendritic arms. However, the Fe element is one of the most important elements to be controlled due to its tendency to form intermetallic compounds of high hardness, high brittleness and difficult to dissolve by homogenization treatments, type Al<sub>15</sub>(Mn, Fe)<sub>3</sub>Si<sub>2</sub> and  $\beta$ -Fe<sub>3</sub>AlSi. In addition, they can act as stress concentrators because they have low ductility and low fracture toughness at room temperature<sup>5</sup>. Such characteristics are detrimental to the mechanical properties of the alloys. However, rapid solidification involving high cooling rates has been observed as a process that may reduce or even inhibit the growth of the intermetallic phases especially of the  $\beta$ -Al<sub>3</sub>FeSi phase<sup>[6]</sup>.

The aim of the present study is to experimentally examine the influence of melt thermosolutal convection, as well as, the variation of pressure on secondary dendrite arm spacing and microhardness during the squeeze casting process of Al-5.5wt.%Si-3.0wt.%Cu, Al-7.5wt.%Si-3.0wt.%Cu, Al-9wt.%Si-3.0wt.%Cu alloys.

## 2. Experimental Procedure

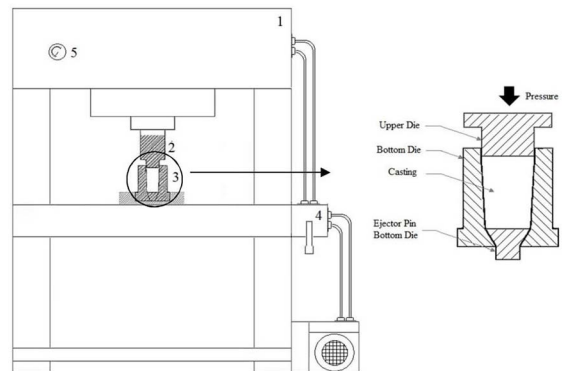
The alloys was prepared in an electric muffle furnace and poured into a cylindrical die of inner diameter of 50mm and height of 100mm at temperatures of 20% above the melting temperature, thus Al-5,5% Si-3% Cu ( 617 ° C), Al-7,5% Si-3% Cu ( 603 ° C) and Al- Cu ( 592 ° C) were cast with temperatures of of 740 ° C, 723 ° C and 710 ° C respectively. Pressure of 0, 50, 100 and 150 MPa was applied by means of a 100 tons hydraulic press and held until the end of solidification.

The experiments were performed with Al-Si-Cu (Al-5.5wt.%Si-3.0wt.%Cu, Al-7.5wt.%Si-3.0wt.%Cu and Al-9wt.%Si-3.0wt.%Cu alloys). The phase diagram of the Al-Si-Cu system, shown in Figure 1, permits a clear visualization of equilibrium phases in the hypoeutectic range of compositions.



**Figure 1.** Partial phase diagram of the Al-Si-Cu system.

In this work, 12 ingots were produced for the three compositions and with the respective pressures. The squeeze casting solidification (SCS) device used in this study is schematized in Figure 2.

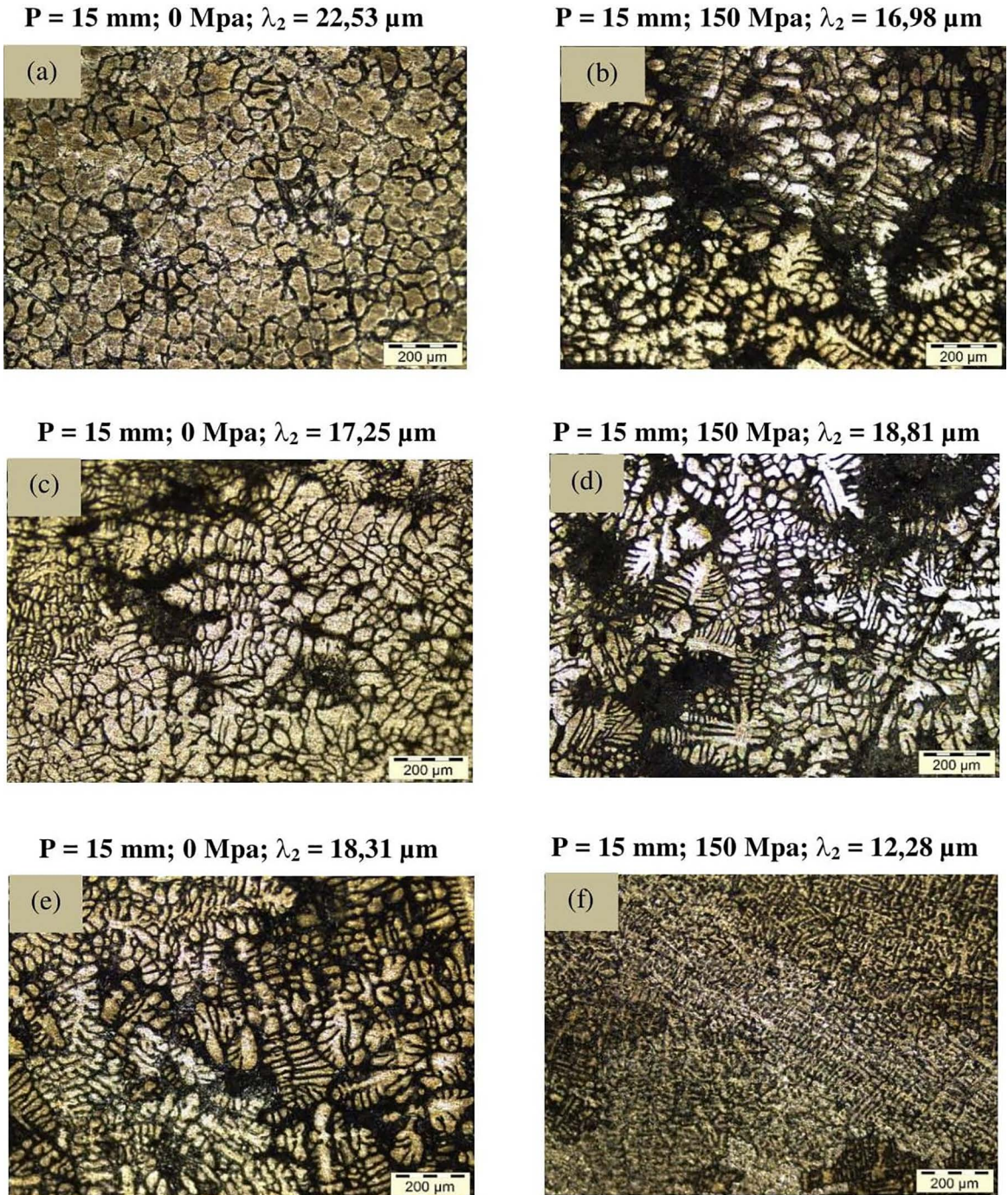


**Figure 2.** Schematic representation of the squeeze casting experimental solidification setup: (1) hydraulic press; (2) upper die; (3) cylindrical mold; (4) Hydraulic pressure control.

Microstructural investigations were carried out following standard metallography procedures and using image analysis techniques on a transverse section cut 30 mm from the bottom of the samples as shown in Figure 3. In order to characterize and quantify the dendritic structure, selected longitudinal sections of the as-cast part (in position 35 mm from the base of the ingot) were polished and etched with an acid solution (2%HF, 10%HNO<sub>3</sub> e 88%H<sub>2</sub>O) for microstructural analysis.







**Figure 4.** Revealed micrographs for the investigated alloy, showing the dendritic microstructures of the samples: (a) Al-5.5wt.%Si-3.0wt.%Cu (b) Al-7.5wt.%Si-3.0wt.%Cu and c) Al-9wt.%Si-3.0wt.%Cu.

According to Runxia Li in his study for alloys Al-xSi ( $x = 15, 17.5$  and  $22 \text{ wt}\%$ ) solidified under pressure of  $600 \text{ MPa}$  the increase of the silicon concentration in the alloy, as well as the use of high pressure, caused a greater refinement of the structure and consequently an increase in microhardness of  $15.91\%$ ,  $12.23\%$  and  $17.48\%$  with respect to gravity casting<sup>22</sup>.

Chelladurai in his study for LM6 aluminium alloy with  $2.5\text{--}10 \text{ wt}\%$  of copper coated short steel fiber reinforced composites were prepared using squeeze casting process, found that the increase of Cu in the coating of the  $2.5$  and  $10\%$  steel fibers caused an increase in the Brinell hardness from  $48$  to  $72 \text{ BHN}$ <sup>23</sup>.

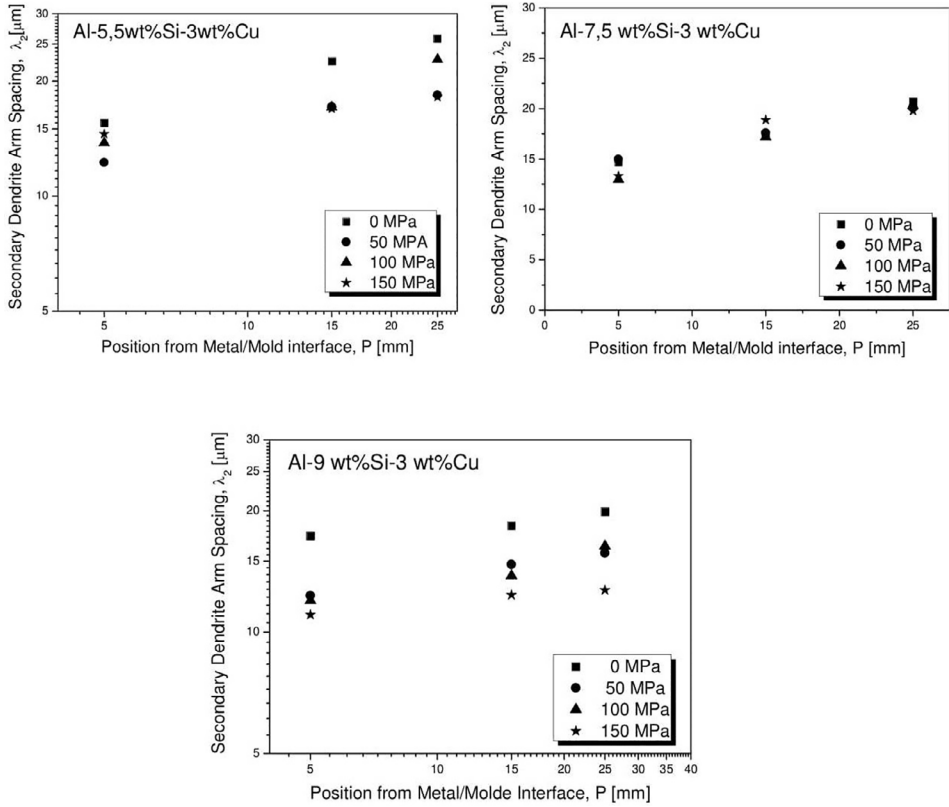


Figure 5. Correlation between the secondary dendrite arm spacing with position from metal/mold interface.

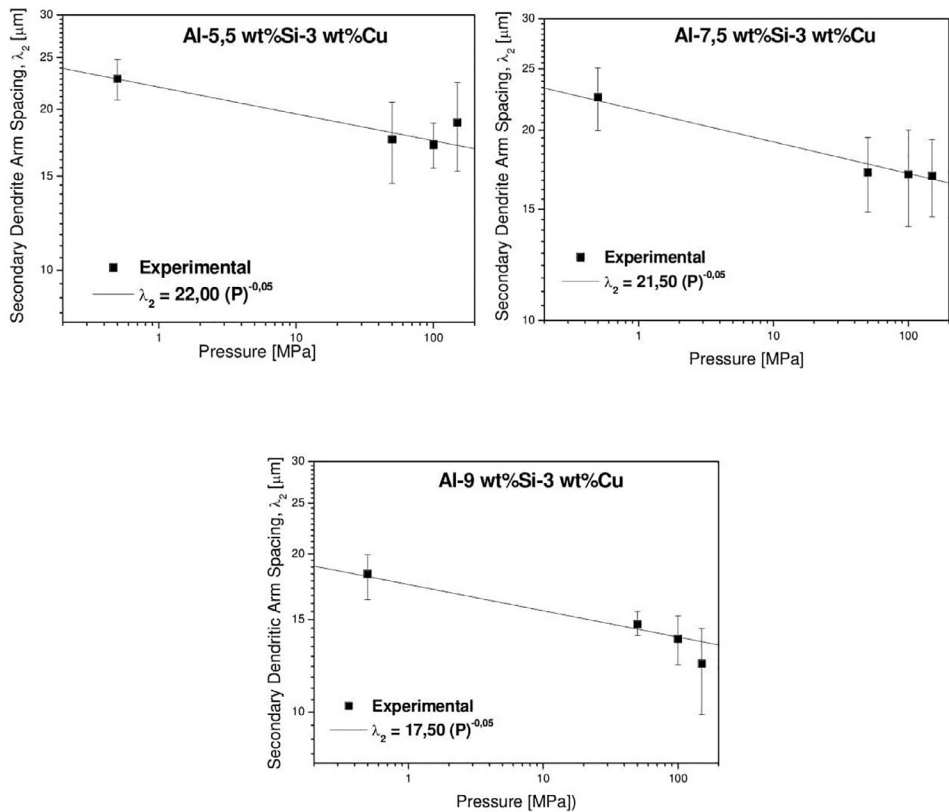
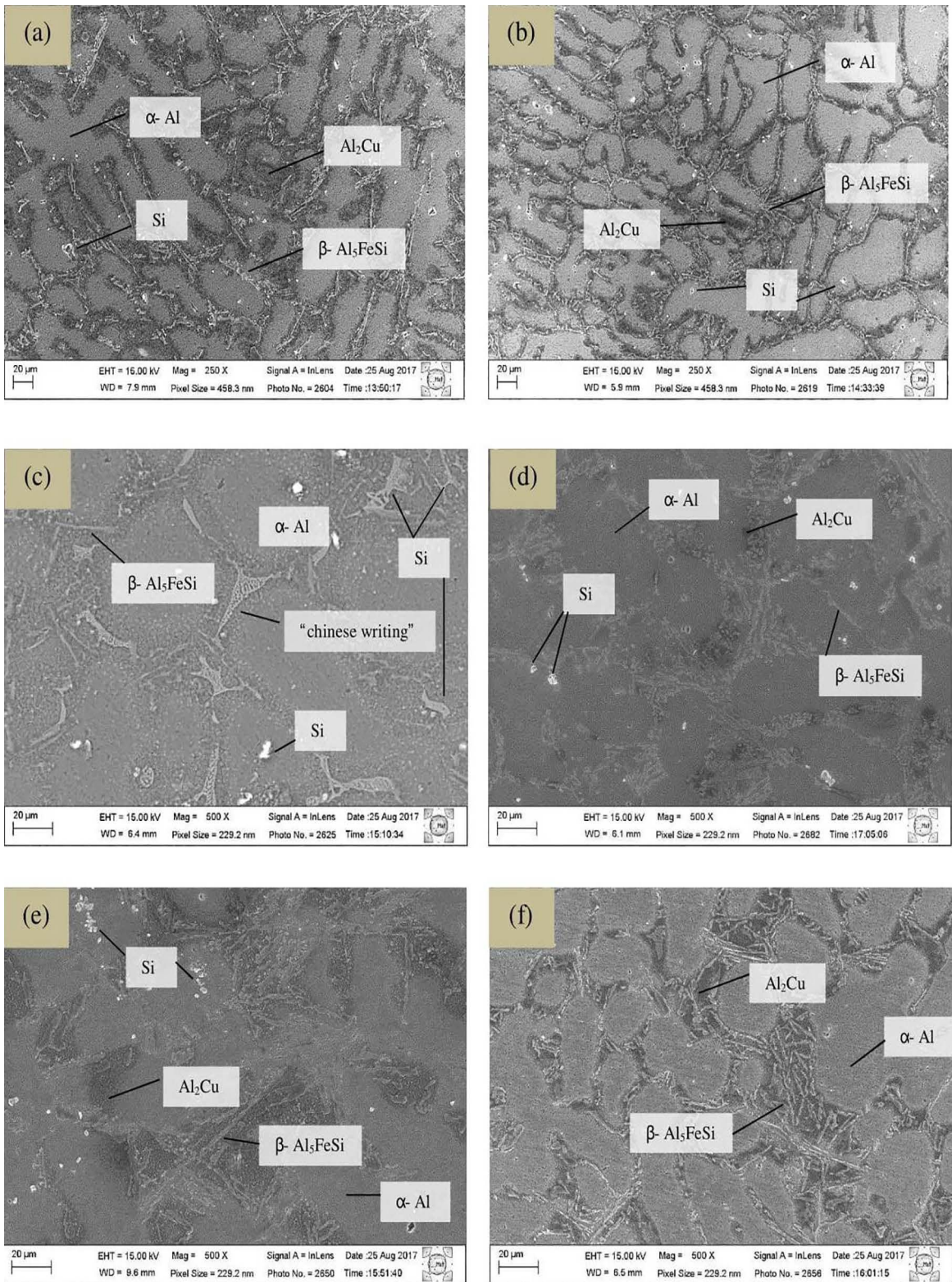
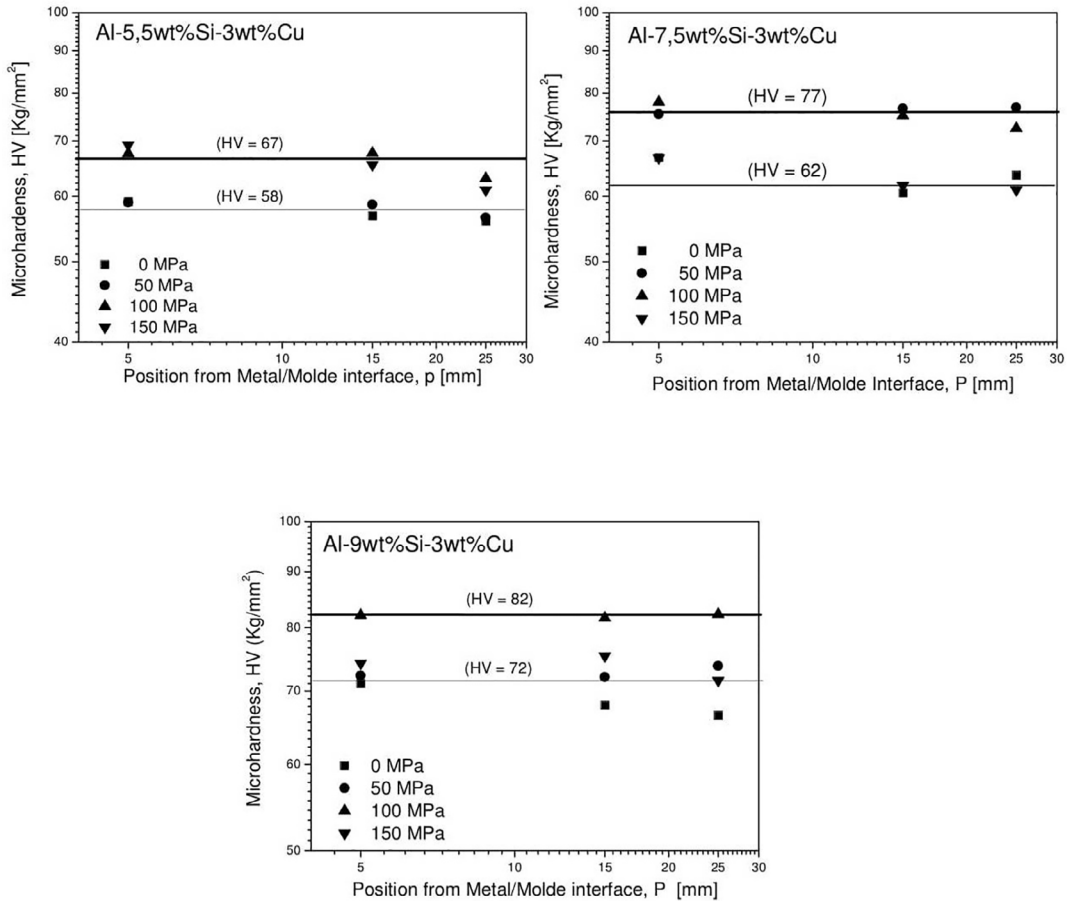


Figure 6. Espaçamento dendrítico secundário em função da pressão. (a) Al-5,5wtSi-3wtCu, (b) Al-7,5wtSi-3wtCu e (c) Al-9wtSi-3wtCu.





**Figure 7.** Micrographies of alloys cast by the squeeze casting process. (a) Al-5,5%wtSi-3%wtCu, 0MPa; (b) Al-5,5%wtSi-3%wtCu, 150MPa; (c) Al-7,5%wtSi-3%wtCu, 0MPa; (d) Al-7,5%wtSi-3%wtCu, 150MPa; (e) Al-9%wtSi-3%wtCu, 0MPa and (f) Al-9%wtSi-3%wtCu, 150MPa.



**Figure 8.** Microhardness dependence as a function of position from metal/mold Interface. (a) Al-5,5wtSi-3wtCu, (b) Al-7,5wtSi-3wtCu e (c) Al-9wtSi-3wtCu.

#### 4. Conclusion

The aim of present research was to evaluate the effects of squeeze casting on microstructures and microhardness of the Al-Si-Cu alloy solidified under pressure, allowed we to have the following conclusions.

In general, secondary dendritic spacing increases as we move away from the metal/mold interface towards the center of the ingot, behavior generated by the solidification front that advances from the metal/mold interface to the center of the ingot. This seems to indicate that, during the primary ramify, the higher solute lateral rejection in the higher Si content alloy, provokes an even greater degree of instability in the interdendritic region, which favors the appearance of a greater amount of secondary dendritic protuberances, decrease so the secondary spacing.

With increasing pressure and composition the secondary dendritic spacing tend to decrease, being more perceptible for the concentration of 9% Si and pressure de 150 MPa. According to Figure 6, the variable concentration seems to be more significant than the pressure variation, as evidenced in the studies of Gomes<sup>24</sup> and Runxia<sup>22</sup>.

A complete dendritic arrangement was shown to characterize the Al-rich matrix of the microstructures of the ternary Al-Si-Cu alloys. The secondary phases within the interdendritic regions of the ternary alloy included Al<sub>2</sub>Cu intermetallic particles, microstructure was composed of phases;  $\alpha$ -Al,  $\beta$ -Fe<sub>3</sub>AlSi, Al<sub>2</sub>Cu,  $\alpha$ -Al<sub>15</sub>(Mn, Fe)<sub>3</sub>Si<sub>2</sub>, Si and complex structures. According to research<sup>21</sup> on the Al-Si-Cu alloy, increased cooling rates may inhibit or reduce the growth of intermetallic phases, especially phase  $\beta$ -Fe<sub>3</sub>AlSi, which can act as stress concentrators and decrease ductility and fracture.

The microhardness values of the ternary Al-Si-Cu alloy shown to be quite higher as the Si content in the alloy increases. This is attributed to the more complex arrangement of phases developed in the ternary alloy. This increase is more significant for the Al-9wt.%Si-3.0wt.%Cu composition due to the proximity to the eutectic point of Si and larger formations of the intermetallic phase  $\beta$ -Fe<sub>3</sub>AlSi which increases the concentration of the alloy tension and consequently increases the microhardness.

According to the obtained results we can establish the pressure of 150 MPa, seems to be the most indicated when it is desired higher microhardness and more refined spacing values, on the other hand the increase in the composition decreases the ductility of the material due to the emergence of metastable phases.

## 5. Acknowledgements

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## 6. References

- Han YS, Kim DH, Lee HI, Kim YG. Effect of applied pressure during solidification on the microstructural refinement in an Al-Cu alloy. *Scripta Metallurgica et Materialia*. 1994;31(12):1623-1628. DOI: [http://dx.doi.org/10.1016/0956-716X\(94\)90453-7](http://dx.doi.org/10.1016/0956-716X(94)90453-7)
- Maeng DY, Lee JH, Won CW, Cho SS, Chun BS. The effects of processing parameters on the microstructure and mechanical properties of modified B390 alloy in direct squeeze casting. *Journal of Materials Processing Technology*. 2000;105(1-2):196-203. DOI: [http://dx.doi.org/10.1016/S0924-0136\(00\)00527-6](http://dx.doi.org/10.1016/S0924-0136(00)00527-6)
- Lee JH, Won CW, Cho SS, Chun BS, Kim SW. Effects of melt flow and temperature on the macro and microstructure of scroll compressor in direct squeeze casting. *Materials Science and Engineering: A*. 2000;281(1-2):8-16. DOI: [http://dx.doi.org/10.1016/S0921-5093\(99\)00746-7](http://dx.doi.org/10.1016/S0921-5093(99)00746-7)
- Kim SW, Durrant G, Lee JH, Cantor B. The effect of die geometry on the microstructure of in direct squeeze cast and gravity die cast 7050 (Al-6.2Zn-2.3Cu-2.3Mg) wrought Al alloy. *Journal of Materials Science*. 1999;34(8):1873-1883. DOI: <http://dx.doi.org/10.1023/A:1004531830333>
- Hong CP, Lee SM, Shen HF. Prevention of macrodefects in squeeze casting of an Al-7 wt pct Si alloy. *Metallurgical and Materials Transactions B*. 1998;31(2):297-305. DOI: <https://doi.org/10.1007/s11663-000-0048-5>
- Hong CP, Shen HF, Cho IS. Prevention of macrosegregation in squeeze casting of an Al-4.5 wt pct Cu alloy. *Metallurgical and Materials Transactions A*. 1998;29(1):339-349. DOI: <https://doi.org/10.1007/s11661-998-0185-z>
- Gallerneault M, Durrant G, Cantor B. Eutectic channelling in a squeeze cast Al-4.5wt%Cu alloy. *Scripta Metallurgica et Materialia*. 1995;32(10):1553-1557. DOI: [http://dx.doi.org/10.1016/0956-716X\(94\)00030-L](http://dx.doi.org/10.1016/0956-716X(94)00030-L)
- Gallerneault M, Durrant G, Cantor B. The squeeze casting of hypoeutectic binary Al-Cu. *Metallurgical and Materials Transactions A*. 1996;27(12):4121-4132. DOI: <http://dx.doi.org/10.1007/BF02595660>
- Polmear I. *Light Alloys. From Traditional Alloys to Nanocrystals*. 4<sup>th</sup> ed. Oxford: Butterworth-Heinemann; 2006.
- Rout PK, Gosh MM, Gosh KS. Influence of Aging Treatments on Alterations of Microstructural Features and Stress Corrosion Cracking Behavior of an Al-Zn-Mg Alloy. *Journal of Materials Engineering and Performance*. 2015;24(7):2792-2805. DOI: <https://doi.org/10.1007/s11665-015-1559-1>
- Sjölander E. *Heat treatment of Al-Si-Cu-Mg Casting Alloys*. [PhD Thesis]. Jönköping: Jönköping University, School of Engineering; 2011.
- Mohamed AMA, Samuel FH, Al kahtani S. Influence of Mg and solution heat treatment on the occurrence of incipient melting in Al-Si-Cu-Mg cast alloys. *Materials Science and Engineering: A*. 2012;543:22-34. DOI: <https://doi.org/10.1016/j.msea.2012.02.032>
- Samuel AM, Doty HW, Valtierra S, Samuel FH. Relationship between Tensile and Impact Properties in Al-Si-Cu-Mg Cast Alloys and their Fracture Mechanisms. *Materials & Design*. 2014;53:938-946. DOI: <https://doi.org/10.1016/j.matdes.2013.07.021>
- Shabestari SG, Shahri F. Influence of modification, solidification conditions and heat treatment on the microstructure and mechanical properties of a356 aluminum alloy. *Journal of Materials Science*. 2004;39(6):2023-2032. DOI: <https://doi.org/10.1023/B:JMSE.0000017764.20609.0d>
- Costa TA, Moreira AL, Moutinho DJ, Dias M, Ferreira IL, Spinelli JE, et al. Growth direction and Si alloying affecting directionally solidified structures of Al-Cu-Si alloys. *Materials Science and Technology*. 2015;31(9):1103-1112. DOI: <https://doi.org/10.1179/1743284714Y.0000000678>
- Nikseresht Z, Karimzadeh F, Golozar MA, Heidarbeigy M. Effect of heat treatment on microstructure and corrosion behavior of Al6061 alloy weldment. *Materials & Design (1980-2015)*. 2010;31(5):2643-2648. DOI: <https://doi.org/10.1016/j.matdes.2009.12.001>
- Ferreira IL, Moutinho DJ, Gomes LG, Rocha OL, Garcia A. Modeling and experimental analysis of macrosegregation during transient solidification of a ternary Al-6wt%Cu-1wt%Si alloy. *Philosophical Magazine Letters*. 2009;89(12):769-777. DOI: <http://dx.doi.org/10.1080/09500830903292320>
- Carvalho DB, Guimarães EC, Moreira AL, Moutinho DJ, Dias Filho JM, da Rocha OL. Characterization of the Al-3wt.%Si alloy in unsteady-state horizontal directional solidification. *Materials Research*. 2013;16(4):874-883. DOI: <http://dx.doi.org/10.1590/S1516-14392013005000072>
- Kaya H, Çadirli E, Büyüç U, Maraşlı N. Variation of microindentation hardness with solidification and microstructure parameters in the Al-based alloys. *Applied Surface Science*. 2008;255(5 Pt 2):3071-3078. DOI: <https://doi.org/10.1016/j.apsusc.2008.08.080>
- Kaya H, Çadirli E, Büyüç U, Maraşlı N. Measurements of the microhardness, electrical and thermal properties of the Al-Ni eutectic alloy. *Materials & Design*. 2012;34:707-712. DOI: <http://dx.doi.org/10.1016/j.matdes.2011.05.030>
- Triveño Rios C, Penna RS, Botta Filho WJ, Kiminami CS, Bolfarini C. Solidificação Rápida de Ligas do Sistema Al-Si-Cu. In: *CBECIMAT 2002*; 2002 Nov 9-13; Natal, RN, Brazil. p. 3599-3606.
- Li R, Liu L, Zhang L, Sun J, Shi Y, Yu B. Effect of Squeeze Casting on Microstructure and Mechanical Properties of Hypereutectic Al-xSi Alloys. *Journal of Materials Science & Technology*. 2017;33(4):404-410. DOI: <https://doi.org/10.1016/j.jmst.2017.02.004>



23. Chelladurai SJS, Arthanari R, Nithyanandam N, Rajendran K, Radhakrishnan KK. Investigation of Mechanical Properties and Dry Sliding Wear Behaviour of Squeeze Cast LM6 Aluminium Alloy Reinforced with Copper Coated Short Steel Fibers. *Transactions of the Indian Institute of Metals*. 2018;71(4):813-822. DOI: <https://doi.org/10.1007/s12666-017-1258-8>
24. Gomes LG. *Dendritic microstructure, macrosegregation and microporosity in the solidification of ternary Al-Si-Cu alloys*. [Thesis]. Campinas: Faculty of Mechanical Engineering, State University of Campinas; 2012. 178 p.
25. Chelladurai SJS, Arthanari R. Investigation on mechanical and wear properties of zinc-coated steel wires reinforced LM6 aluminium alloy composites by squeeze casting. *Surface Review and Letters*. 2019;26(1):1850125. DOI: <https://doi.org/10.1142/S0218625X18501251>
26. Chelladurai SJS, Arthanari R, Krishnamoorthy K, Selvaraj KS, Govindan P. Investigation on mechanical properties of squeeze cast LM6 aluminium alloy reinforced with zinc coated steel-wire mesh. *Materiali in Tehnologije/Materials and Technology*. 2018;52(2):125-131. DOI: <https://doi.org/10.17222/mit.2017.019>
27. Chelladurai SJS, Arthanari R, Krishnamoorthy K, Selvaraj KS, Govindan P. Effect of Copper Coating and Reinforcement Orientation on Mechanical Properties of LM6 Aluminium Alloy Composites Reinforced with Steel Mesh by Squeeze Casting. *Transactions of the Indian Institute of Metals*. 2018;71(5):1041-1048. DOI: <https://doi.org/10.1007/s12666-017-1235-2>
28. Chelladurai SJS, Arthanari R, Thangaraj AT, Sekar H. Dry sliding wear characterization of squeeze cast LM13/FeCu composite using response surface methodology. *China Foundry*. 2017;14(6):525-533. DOI: <https://doi.org/10.1007/s41230-017-7101-3>
29. Chelladurai SJS, Arthanari R, Selvarajan R, Athanarsamy S, Arumugam S, Veerakumar G. Investigation on mechanical properties and wear behaviour of squeeze cast LM13 aluminium alloy reinforced with copper coated steel wires. *Zeitschrift für Physikalische Chemie*. 2018;232(12):1787-1806. DOI: <https://doi.org/10.1515/zpch-2017-1093>
30. Chelladurai SJS, Arthanari R, Selvarajan R, Ravichandran TP, Ravi SK, Petchimuthu SRC. Optimisation of dry Sliding Wear Parameters of Squeeze Cast AA336 Aluminium Alloy: Copper Coated Steel Wire-Reinforced Composites by Response Surface Methodology. *Journal of Metalcasting*. 2019;13(2):354-366. DOI: <https://doi.org/10.1007/s40962-018-0258-8>