Short Communication

Investigation on microstructural and microhardness evolution in as-cast and T6/heat-treated samples of a horizontally solidified AlSiCu alloy

Fabrício A. Souza b, Marlo O. Costa b, Igor A. Magno b, Jacson M. Nascimento b, Adriana P. Silva b, Thiago S. Costa a, Otávio L. Rocha a,b,*

a Federal Institute of Education, Science and Technology of Pará — IFPA, 66093-020, Belém-Pará, Brazil
b Faculty of Mechanical Engineering, Federal University of Pará — UFPA, 66075-110, Belém-Pará, Brazil

ARTICLE INFO

Article history:
Received 16 January 2019
Accepted 28 June 2019
Available online 18 July 2019

Keywords:
Unsteady-state horizontal solidification
T6-heat treatment
Microstructural evolution
Microhardness
AlSiCu alloys

ABSTRACT

In this investigation, in order to evaluate the influence of microstructures on the microhardness of an aluminum-based multicomponent alloy, samples of the horizontally solidified Al-7 wt.%Si-3 wt.%Cu alloy were subjected to the T6 heat treatment. Microhardness (HV) measurements were carried out in specific zones, like in the center of the dendrites (α/Al-rich) and interdendritic regions. As expected, in as-cast samples, it has been observed high HV values in the interdendritic regions, due to the presence of the harder phases in the eutectic mixture (Si particles + Al2Cu (θ) + Fe intermetallic phases). As a highlight, hardening has been observed in both analyzed regions (α/Al-rich and interdendritic), and a similar behavior in the HV evolution, between the two investigated regions, has also been evidenced. In addition, mathematical equations have characterized the HV dependence as a function of the aging time (tageing).

© 2019 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

1. Introduction

It is known that aluminum alloys present combinations of unique properties, with emphasis on high strength/weight ratio, achieving economic notoriety in many industrial applications, such as in the automotive and aerospace industries, civil construction, pressure vessels for cryogenic applications, and in several other areas [1–4]. We highlight the casting alloys of the 4xx.x group, based on binary Al-Si alloys containing 5–12% Si (mass fraction), which find many applications where combinations of moderate strength and high ductility and impact resistance are required [4–9]. The addition of alloying elements increases the strength, through solids or precipitation hardening processes [10–18].

The 3XX.X Series alloys, containing Cu and Mg or both, are most commonly used in the military, aeronautics and
automotive industries [12–15]. They are also the most important casting alloys because combine the benefits of silicon with those of heat treatment alloys. In general, these alloys are found in the following systems: Al–Si–Mg, Al–Si–Cu or Al–Si–Cu–Mg. In Al–Si–Cu alloys, the Si and Cu contents range from 5 to 22 wt% and 0 wt% to 4.5 wt%, respectively. Most of these alloys have nominal magnesium contents, ranging from 0.3 to 0.6 wt% [7–13,19].

The effects of the conditions of the solidification and heat treatment processes and their interrelationships on the microstructure and mechanical properties of aluminum-based ternary alloys have been recently investigated by Costa et al. [2] and Magno et al. [3,17] for AlSiCu alloys, Chen et al. [13], Sekar et al. [14] and Pramoda et al. [15] for AlSiMg alloys, respectively. Costa et al. [2], Magno et al. [3] and Chen et al. [13], have reported in their investigations on the favorable influence of $\lambda_2$ high and low $T_R$ and $\lambda_2$ values, respectively, on the T6 heat treatment, especially in the step of the solution treatment. Sekar et al. [14] have elaborated studies on the influence of as-cast (gravity casting, vacuum casting and squeeze casting methods) and T6 heat treatment conditions on mechanical properties (hardness, bending and double shear) and the results found for all casting methods showed that hardness and bending strength increased after heat treatment; on the other hand, the double shear strength of all these three castings decreased after the assumed heat treatment. However, these investigations did not explore the operational processing effects in specific regions of the cast and heat treated microstructures, such as the hardening evolution in the aluminum-rich primary phase and the interdendritic regions.

Notably, the literature [1–4,10–13,16,17] is unanimous about the strong influence of the solidification thermal and microstructural parameters on the mechanical properties of metals and their alloys. Study of solidification conditions on the morphology of the intermetallic phases in aluminum alloys have been elaborated in the last two decades. Samuel et al. [7], Djurdjevic et al. [8] and Li et al. [9] have reported that cast AlSiCu alloys the Al$_2$Cu intermetallic phase can be seen in the block and eutectic morphologies or as a mixture of both types. According to the authors, higher cooling rates favor the formation of eutectic type, and the block phase is more difficult to dissolve during the solution heat treatment. However, there is still a large gap in the literature that needs to be explored on the effects of the size and morphology of intermetallic phases on mechanical properties during the solidification path of AlSiCu alloys.

It is well known that in casting Al–Si alloys the presence of iron as an impurity is unavoidable and, due to its extremely low solubility in aluminum, Fe forms intermetallic compounds during the solidification of these alloys [1–4,11–13,17]. Generally in Al–Si alloy, Fe-rich phases can be grouped into three kinds of morphologies: polyhedral or star-like, Chinese script and platelet [10,11], and the most observed among them is $\beta$–Al$_3$SiFe with platelet-like morphology [11].

It is important to emphasize that there are still doubts in the literature about the influence of the solution treatment on the as-cast microstructures, especially on the dendritic spacings. It is very well known that the Al-rich as-cast primary phase (matrix), generally dendritic, hardens after the T6 heat treatment. On the other hand, there are no studies in the literature regarding the effects of the phases that do not dissolve on the mechanical properties within the interdendritic regions, as well as the effects of the microstructural solidification parameters (as $\lambda_2$) on the size, morphology and distribution of Si, Al$_2$Cu and Fe particles after T6 treatment. In this sense, the present contribution aims to investigate experimentally the effects of solidification and precipitation hardening processes parameters on the microstructure, intermetallic phases and microhardness in both matrix and interdendritic regions. Mathematical expressions have been proposed to predict the HV evolution with the aging time ($t_{\text{aging}}$) in both the matrix and the interdendritic regions.

2. Experimental procedure

Experiments on the investigated alloy, involving the preparation, quantitative and qualitative chemical analysis, transient horizontal solidification and the obtained thermal date, the resulting ingot as well as its characterizations in macrostructural and microstructural scales, are detailed in our recent publication [10]. The solidification set-up was designed in such a way that heat was extracted only through the water-cooled mould, promoting horizontal directional solidification.

Samples with the dimensions of $10 \times 10 \times 60 \text{ mm}$ were selected from the horizontally solidified ingot and submitted to the T6 heat treatment. The maximum length of 60 mm was defined as a function of the variation of the cooling rate ($T_R$) along the length of the ingot. It is emphasized that the $T_R$
values range from 21 °C/s to 0.53 °C/s along the length of the ingot, from the cooled base to the position in as-cast ingot equal to 60 mm. Obviously, a high range of $T_R$ values allows a large variation of the as-cast microstructure, since the cooling rate exerts a strong influence on dendritic spacing.

Fig. 1 shows the drawing scheme of samples that were selected from the as-cast horizontal ingot and submitted to the T6 heat treatment, followed recommendations of ASTM B-597 standards, that is: solution treating performed during 3 h at 495 ± 2 °C, followed by quenching in warm water (70 ± 2 °C), aging for 1, 2, 3, 4, 6, 8, 16 and 24 h at 155 ± 2 °C and air cooling. Immediately after the heat treatment, both samples were subjected to microstructural characterization. The microstructural parameter analyzed was the secondary dendrite arm spacing ($\lambda_2$). More details on the T6 heat treatment have been reported in the references [1,17].

In order to evaluate the effect of the precipitation hardening treatment on the as-cast samples, microhardness test was performed on both as-cast and heat treated samples. The HV measurements were obtained at the center of the dendrites and within the interdendritic regions. $\lambda_2$ and HV measurements were carried out at the following positions of the as-cast and heat-treated samples 5, 10, 15, 30 and 60 mm from the cooled mold. The methodology used for the $\lambda_2$ and HV measurement has been detailed in our recently published work [1]. The HV measurements followed the ASTM E384 standard and were conducted using a test load of 50 g and a dwell time of 10 s. A Shimadzu HMV model was used and the adopted Vickers microhardness has been the average of at least 20 measurements on each as-cast and heat treated samples. In addition, the scanning electron microscope (SEM TESCAM, VEGA LMU) coupled to an energy dispersion spectrum (EDS X-MAX 20, Oxford) was used to investigate the morphology and distribution of the phases in both as-cast and heat treatment samples.

3. Results and discussion

As mentioned, the thermal data resulting from the horizontal solidification can be observed in our article [10]. From these
Fig. 3 – HV variation with the position in the as-cast ingot and secondary dendrite arm spacing: (a) matrix versus interdendritic region and (b) interdendritic region.

Experimental mathematical expressions given by $T_R = 123(P)^{-1.25}$ and $\lambda_2 = 50.6(T_R)^{-1/3}$ have characterized the $T_R$ and $\lambda_2$ variation as a function of ingot cooling rate, (Fig. 2a and b, respectively). It is observed that the cooling water exerts a strong influence on the $T_R$ and $\lambda_2$ values, since higher and lower $T_R$ and $\lambda_2$ values, respectively, finer microstructures are achieved at positions in ingot close to the cooled base.

Fig. 3 shows the results of HV measurements in the regions of the as-cast samples established for studies, that is, it has been carried out on the center at the dendrites ($\alpha$-Al-rich phase) and within the interdendritic regions. Obviously, higher HV values have been observed within the interdendritic regions due to the presence of harder phases that constitute the eutectic mixture (Si particles + Al$_2$Cu and Fe intermetallic compounds). An average HV value equal to 60 kg/mm$^2$ has been obtained on the region of the Al-rich phase (Matrix) to the length of the as-solidified ingot; on the other hand, within the interdendritic regions (IR), a progressive increase of HV values has been observed, as can be seen in Fig. 3a and b represents the HV behavior with the variation of $\lambda_2$ in IR. It can be seen

<table>
<thead>
<tr>
<th>Point</th>
<th>Weight (%)</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Al</td>
<td>Si</td>
<td>Cu</td>
<td>Fe</td>
<td>Phase</td>
</tr>
<tr>
<td>1</td>
<td>95.33</td>
<td>1.35</td>
<td>3.28</td>
<td>0.04</td>
<td>$\alpha$-Al</td>
</tr>
<tr>
<td>2</td>
<td>27.08</td>
<td>71.07</td>
<td>1.74</td>
<td>0.07</td>
<td>$\alpha$-Al + Si</td>
</tr>
<tr>
<td>3</td>
<td>20.42</td>
<td>12.87</td>
<td>66.38</td>
<td>0.34</td>
<td>$\alpha$-Al + Al$_2$Cu(O) + Si</td>
</tr>
<tr>
<td>4</td>
<td>22.74</td>
<td>28.26</td>
<td>48.92</td>
<td>0.08</td>
<td>$\alpha$-Al + Al$_2$Cu(O) + Si</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Point</th>
<th>Weight (%)</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Al</td>
<td>Si</td>
<td>Cu</td>
<td>Fe</td>
<td>Phase</td>
</tr>
<tr>
<td>1</td>
<td>96.46</td>
<td>1.21</td>
<td>2.27</td>
<td>0.06</td>
<td>$\alpha$-Al</td>
</tr>
<tr>
<td>2</td>
<td>2.46</td>
<td>97.54</td>
<td>0.3</td>
<td>-</td>
<td>$\alpha$-Al + Si</td>
</tr>
<tr>
<td>3</td>
<td>14.32</td>
<td>28.25</td>
<td>57.2</td>
<td>0.23</td>
<td>$\alpha$-Al + Al$_2$Cu(O) + Si + Fe</td>
</tr>
<tr>
<td>4</td>
<td>46.01</td>
<td>6.66</td>
<td>27.64</td>
<td>19.69</td>
<td>$\alpha$-Al + Al$_2$Cu(O) + Si + Fe-rich phase</td>
</tr>
</tbody>
</table>

Fig. 4 – SEM microstructures for two positions in the ingot from the cooled base, obtained by: (a) $P = 5$ mm and (b) $P = 70$ mm, both by BSSs with mapping and microanalysis by EDS.
that the Hall–Petch expression given by $HV = 106 – 163(\lambda^2)^{-0.5}$ characterizes the HV dependence as a function of $\lambda^2$. This is due to the harder particles located within the interdendritic regions that increase in size with the increase of $\lambda^2$, specifically the Si particles that present with coarser and elongated lamellar morphologies.

Fig. 4 shows longitudinal SEM micrographs obtained for two positions in the as-solidified ingot (5 and 70 mm from the cooled interface). The SEM images of Fig. 4a and b have been obtained by means of secondary electrons (SE) and scattered retro electrons (BSSs), respectively. The images by BSSs are extremely useful when it is desired to observe impurities.
or intermetallic phases in which there is considerable difference of density between the elements. The density difference between the matrix (Al) and the Si particles is small, therefore, both appear in gray color, but clearly distinguishable. On the other hand, the density difference between Cu and Fe with the matrix is higher, with both appearing white in the SEM image, but with a marked contrast.

Besides that, in order to better identify the elements and their influences in the formation of phases that constitute the investigated alloy, in this work compositional maps and point microanalyses by EDS on each constituent phase have been carried out and the results are presented in both Fig. 4a and b. The EDS mapping has allowed to separate the Al, Si, Cu and Fe elements by specific colors and the point microanalysis by EDS aims to elaborate a qualitative and quantitative chemical analysis of these elements. The use of these analyzes combined with observations obtained from the literature [7–11,13] on the morphology of the second phases that form during the solidification path of AlSiCu alloys, it has contributed in the characterization of the Fe/AlCu intermetallic compounds and Si particles.

Thus, by examination of the as-cast SEM microstructures shown in Fig. 4, it is clearly evidenced the effect of high cooling rates on the formation fibers/spheroidized-like Si particles and finer AlCu intermetallic compounds of block and eutectic types, as observed in Fig. 4a. On the other hand, as the cooling rate decreases with the advance of solidification, the dendritic spacings (KS) increase and the Si particles are able to grow and exhibit lamellar morphology, as seen in Fig. 4b. Certainly, the presence of coarser Si particles and AlCu/Fe intermetallic phases justify the highest HV values observed at positions with higher KS.

Fig. 5a shows for both investigated regions (α-Al and IR) the HV evolution with the aging time (taging) for all the analyzed positions. As expected, by analyzing the HV values on the matrix, a gradual hardening is observed with increasing of taging, reaching a maximum value equal to 16 h, and from this time a slight decrease in the HV value has been evidenced, probably due to the overaging, whose phenomenon is very well known in the literature due to the increase in the size of metastable precipitates (θ → Al2Cu) and returning to stable phase (θ → Al2Cu), now dispersed in the matrix (α-Al) and not within the interdendritic regions.

Fig. 5c shows the results of the average HV values for both investigated regions, calculated from the measurements of each position which have been reported in Fig. 5a and b. As one of the highlights of this work, mathematical expressions given by HV = 84.75 + 1.72(taging) – 0.04(taging)² and HV = 74.5 + 0.98(taging) – 0.03(taging)² have been proposed, which characterize the HV variation with aging in both matrix and interdendritic regions, respectively. As cab be observed, SEM images by EDS/scan mapping of microindentations have been obtained within the interdendritic regions and are shown in the upper parts of Fig. 5c and d. It is noted the presence in the eutectic mixture of Si particles and block-type Al2Cu intermetallic compounds, which did not dissolve during solution treatment. In addition, it is observed more spheroidized-like Si particles for the heat treated sample at position equal to 5 mm, due to higher cooling rates at this position.

Fig. 5d shows also qualitative and quantitative elemental microanalysis by EDS. It is observed from the result of point 2 that the Cu and Fe amounts present depict the presence of ω-Al2Cu2Fe phase, which does not dissolve during the step of the solution treatment. According to Dons [5], the β-AlFeSi phase becomes the ω phase, presenting a morphology similar to that of Al2Cu. This author has reported that the β-AlFeSi + ω-Al2Cu2 phases can coexist in as-cast structures and the Si + ω-Al2Cu2Fe phases have been observed in heat treated structures. Cerri et al. [6] have also reported on the presence of an intermediate phase between the β and ω phases, that is, the AlFeSiCu phase has been observed after the solution treatment. In general, it is known that both Si and Fe particles do not dissolve during solution treatment.

In this work a comparative study with results from the literature has been carried out for directionally solidified AlSiCu alloys, as shown in Table 1. It is important to inform that the average HV values compared have been considered the closest to the assumed conditions in the present investigation (3 h at 155 ± 2 °C of aging). In addition, the solidification thermal and microstructural parameters have been very close. This is seen that precipitation hardening has occurred for the three analyzed cases. The average HV values obtained equal 87, 80 and 84.5 kg/mm² are among the minimum and maximum HV values of the 20 measurements (75–98 kg/mm², respectively) achieved in the present work. Therefore, it is possible to predict that the hardening levels have been the same. It is important to note that the maximum hardening reached in this work has been obtained for HV = 92 kg/mm² (average between the HV values of both regions) at the aging time equal to 16 h, as seen in Fig. 5c.

### Table 1 – Comparison with the literature.

<table>
<thead>
<tr>
<th>References</th>
<th>Alloy (wt.%)</th>
<th>Solidification growth direction</th>
<th>Solution treatment conditions</th>
<th>Aging treatment conditions</th>
<th>Average of HV</th>
</tr>
</thead>
<tbody>
<tr>
<td>[2]</td>
<td>Al3Cu5.5Si</td>
<td>Upward</td>
<td>5 and 8 h at 490 ± 2 °C</td>
<td>3h at 155 ± 2 °C</td>
<td>76 kg/mm²</td>
</tr>
<tr>
<td>[17]</td>
<td></td>
<td>Horizontal</td>
<td>5 and 8 h at 490 ± 2 °C</td>
<td>3h at 155 ± 2 °C</td>
<td>62 kg/mm²</td>
</tr>
<tr>
<td>This work</td>
<td>Al3Cu7Si</td>
<td>Horizontal</td>
<td>3h at 495 ± 2 °C</td>
<td>1, 2, 3, 4, 6, 8, 16 and 24h at 155 ± 2 °C</td>
<td>72 kg/mm²</td>
</tr>
</tbody>
</table>

4. Conclusions

Considering the results obtained from this investigation, the following conclusions were drawn:

1. It has been evidenced that both solidification thermal and microstructural parameters, that is, high and low TR and
\(\lambda_2\) values, respectively, positively influenced the solution step during the T6 heat treatment, since spheroidized-like Si particles have been observed on the as-cast and heat treated samples at positions in the ingot closest to the cooled base.

(2) As expected, hardening of the matrix has been observed with increasing aging time.

(3) As a highlight, the progressive increase of HV with the ingot position has been noted within the interdendritic regions of both as-cast and heat treated samples. This has been due to the presence of harder particles contained in the eutectic mixture (Si particles + Al\(_2\)Cu/Fe intermetallic compounds) which, with the increase of \(\lambda_2\), they have found equilibrium conditions more favorable to increase in size and, as a consequence, hardening the interdendritic region.

(4) Hall–Petch mathematical expressions given by

\[
\text{HV} = 84.75 + 1.72(t_{\text{asgng}}) - 0.04(t_{\text{gng}})^2
\]

and

\[
\text{HV} = 74.5 + 0.98(t_{\text{asgng}}) - 0.03(t_{\text{gng}})^2
\]

have been proposed to characterize the HV dependence as a function of aging time for both matrix and interdendritic regions.

(5) The hardening levels achieved in this work have been similar to those reported in the literature for as-cast samples of the same investigated alloys system (AlSiCu) under similar T6 heat treatment conditions.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgments

The authors acknowledge the financial support provided by IFPA — Federal Institute of Education, Science and Technology of Pará, UFPA — Federal University of Pará, and CNPq — The Brazilian Research Council (grants 302846/2017-4 and 400634/2016-3), FAPESP — Amazon Foundation of Support to Study and Research (grants ICAAF 064/2016) and CAPES — Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brazil - Finance Code 001.

REFERENCES


