# EFFECT OF SI AND Cu CONTENT ON THE SIZE OF INTERMETALLIC PHASE PARTICLES IN Al-Si-Cu-Mg-Fe ALLOYS

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## Abstract

Quasi-directionally solidified plates were sand cast using unmodified AI-xSi-yCu-O.1 Mg-0.5Fe alloy with two Si ( $x = 4.5$  or 9 mass%) and three Cu ( $y = 0$ , 1 or 4mass%) contents, and the size of the intermetallic phase particles ( $\beta$ -Al<sub>3</sub>FeSi and Al<sub>2</sub>Cu) assessed at constant secondary dendrite ann spacing (SDAS) using optical microscopy and back scattered electron images. Increasing the concentration of Si alone or in combination with  $Cu$  refined the  $B-AI<sub>5</sub>FeSi$ platelets, whereas increasing Cu at constant Si shows an SDAS and Si level dependent effect.

## Introduction

Recent work on Sr modified AI-Si-Cu-Mg-Fe-Mn alloys indicates that increasing Si content increases the ductility of high Fe containing alloys [I]. The enhanced ductility was explained with the help of the ternary AI-Si-Fe phase diagrams [2]: a higher level of Si shortens the solidification path and refines the  $\beta$ -Al<sub>5</sub>FeSi platelets. The effect was observed in the presence of Cu, Mg, Mn and Sr. further experiments aimed at verifying this hypothesis in ternary AI-Si-0.8Fe alloys contradicted the earlier conclusions, i.e., that increased Si led increased the size of the Fe-rich intermetallics [3]. It was thus suggested that the refining etfect of Si upon the Fe-rich intermetallics was related to the formation of low meting point Cu-based eutectics rather than just due to the sortening of the solidification path of the Al-Si base alloy. In the present work the effect of Cu (with levels of 0, 1 and 4 %) on the size of  $\beta$ -Al<sub>5</sub>FeSi platelets was studied. The Fe content (0.5%) was the same as those of reference [I]. Unmdified alloys were studied. The commercial software package, ThennoCalc™, was used in conjunction with the experiments to help understand the solidification sequence and the formation of intermetallics.

## Materials and experimental methods

Six alloy compositions, listed in Table I, were studied. The casting followed the description of Refs [4, 5]. Briefly, Al ingot (99.8% purity: main impurities 0.03% Si, 0.12%Fe) was molten in a 20kW induction furnace along with commercial purity Si. Cu, Fe as AlTab Fe 75% compacts, and Mg were subsequently added in the order; the liquid was degassed for about 20 minutes with Ar at a rate of 2 l/min  $(3.33 \times 10^{-5} \text{ m}^3/\text{s})$ , and poured into the mould after 2 minutes of holding.

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Samples for chemical analysis were taken prior to each pouring. Pouring was done at 1003 K (730'C) into a sand mould assembly, with heavy steel chills at the far ends, designed to produce three plates at a time, each  $160x120x15$  mm<sup>3</sup> in size. The mould contained the chills, had dimensions of  $32x40x140$  mm<sup>3</sup> and their surface was smooth. The mould assembly was inverted right after pouring to allow the plates to solidify in a quasi-directional manner upwards away from the chill, towards the risers. Furher details of the mould assembly can be found in Ref [4].

Longitudinal sections were cut from each cast plate and metallographically polished. Back scattered electron images were taken at predetermined locations selected using data from reference [4] to ensure constant SDAS in order to compare the size of Fe- or Cu-rich intermetallics. The SDAS selected for metallographic obseravations ( $\sim$ 50 and  $\sim$ 30  $\mu$ m), correspond to cooling rates of  $\leq$   $\degree$ C /s and  $\geq$   $\degree$ C /s, respectively. The cooling rates calculated between liquidus and solidus; CR1 cooling rate as mentioned in the Ref. [4]

#### Results

# THERMOCALC™ CALCULATIONS

The onsets of precipitation of each phase in the Cu-free alloys (whose composition matches the alloys of the prior studies of Refs. [3, 5]) are shown in Fig I as per the values listed in Tbale 2. The  $\beta$ -Al<sub>5</sub>FeSi phase precipitates in the pre-eutectic stage for all of the alloys except for the high Si ones where it forms in the post-eutectic stage. When present, Cu reduces the onset temperature of all reactions, namely,  $\beta$ -Al<sub>5</sub>FeSi phase and Al-Si eutectic temperatures, as well as a slight reduction in the values of  $T_{\beta-{\text{eut}}}$ , and  $T_{liq-{\text{eut}}}$ . The later two, however, were more than halved by the increase in Si content from 4.5 to 9.0%, i.e., increased Si level reduced the solidification range,  $(T_{liq} - T_{sol})$ .



Figure I: The temperatures at the onset of relevant reactions for the Cu-free alloys studied as per Table II. (Note that the Cu-free alloys of the work match those of the prior studies of Refs [3, 5]).

Table I: Target (left paired column) and actual (right, bold column) chemical compositions (mass %) of the alloys studied. The alloys nomenclature follows that of Ref. [4], i.e.: the superscripted/subscripted elements indicate higher/lower level)

Alloy	Chemical Composition <sup>†</sup>							
	Si		Сu		Mg		Fe	
Fe $\mathrm{Alg}_{\mathrm{IMg}}$	4.5	4.3	0.0	0.01	0.1	0.08	0.5	0.49
$\mathrm{Alg}_{\mathrm{ICuMg}}_{\mathrm{Fe}}$	4.5	4.4	1.0	0.96	0.1	0.08	0.5	0.48
Mg Fe	4.5	4.3	4.0	3.7	0.1	0.08	0.5	0.42
$\mathrm{Al}_{\mathrm{Si}}^{\mathrm{Si}}_{\mathrm{Mg}}^{\mathrm{Cu}}$	9.0	8.5	0.0	0.01	0.1	0.13	0.5	0.46
Fe $Al^{Si}$ CuMg	9.0	8.7	1.0	0.96	0.1	0.13	0.5	0.45
$AlSi$ Cu Έe Mg	9.0	8.5	4.0	3.79	0.1	0.12	0.5	0.43

Table II: ThermoCalc<sup>TM</sup> values for the onset of precipitation and the temperature range of key reactions. All values are in  $[°C]$ .



## BACKSCA TTERED ELECTRON IMAGES

Back scattered electron images were obtained at an SDAS of  $\sim$  30  $\mu$ m and  $\sim$  50  $\mu$ m are shown in (Fig. 2 (a-f)) and (Fig. 3 (a-f)) respectively. The Cu-free, Fig 2 a and d, and 0.1 Mg alloys showed a similar effect as in the alloys of Ref. [5] (the Fe content was  $\sim 0.8$  mass % in Ref. [5] against 0.5mass% in the present alloys): increasing the level of Si from 4.5 to 9 reduced the size of Fe intermetallics.

In low Si alloys at small SDAS (Fig. 2 a-c), the Cu-free alloy (Fig 2a) showed a large amount of long Fe-rich platelets, the size of which decreased when the Cu content increased to 4 mass% (Fig.2c). In the I mass% Cu alloy, the Fe-rich platelets exhbit a Igreater degree of interconnection than in the Cu-free alloy.

A high Si content (Fig. 2(d-f) and Fig.3(d-f)), refined the intermetallics at small SDAS regardless of the presense ofCu. At large SDAS, Cu does increased the size of the intermetallics (i.e., the effects were opposite to those of the low Si alloys).

 $^{\dagger}$  All other common impurities, such as Mn, Pb, Ti, Sn, Cr, Zn, Ni, etc., levels were well below 0.01 w.t.%



Figure 2: Back Scattered Electron images taken near chill end on the (left) low Si and (right) high Si alloys with (a & d) 0 Cu, (b & e) 1 Cu, and (c & f) 4 Cu. Average SDAS  $\sim$  30  $\mu$ m.



Figure 3: Back Scattered Electron images taken near riser end on the (left) low Si and (right) high Si alloys, with (a & d) 0 Cu, (b & e) 1 Cu, and (c & f) 4 Cu. SDAS  $\sim$  50  $\mu$ m.

## Discussion

All of the alloys with low Si  $(4.5 \text{ wt. } %)$  precipitate the  $\beta$ -Al<sub>3</sub>FeSi intermetallics in the preeutectic stage (Table 11 and Fig I). Introduction of Cu or increasing the Cu content reduces the  $T_{\beta$ -*eut* slightly (Table II) and possibly following the suggestion made in Ref. [3], the Cu rich pools of liquids isolate the  $\beta$ -AlsFeSi plates preventing them from forming long interconnected clusters. Alternatively, it may be speculated that if the  $A<sub>1</sub>$ Cu particles nucleate on the  $A<sub>1</sub>$ -AIsFeSi plates, then the lengthening process of the latter may somehow be disturbed by the newly nucleated particles; hence the overall size reduction of Fe-rich intermetallics. The multiple Cu pools lead to extensive comminution and this is the dominant effect, especially at small SDAS.

Prior work showed that increased Si increases the size of the  $\beta$ -Al<sub>5</sub>FeSi plates at 0.8 Fe level [3] whereas the more detailed study of ref. [5] made eveident the a cooling rate dependent evolution of  $\alpha$ -Al<sub>8</sub>Fe<sub>2</sub>Si and  $\beta$ -Al<sub>3</sub>FeSi phases at this high Fe level. In the present study, with only 0.5 Fe, the  $\beta$ - plates did not increase in size with increasing Si level (against Ref. [3]'s main conclusion); instead, it shows preferential formation of  $\alpha$ -Al<sub>8</sub>Fe<sub>2</sub>Si particles at small SDAS and of  $\beta$ -Al<sub>3</sub>FeSi plates at large SDAS. These observations can be easily rationalised with reference to different onset of precipitation temperature (Figs. 1 and 2, and Table II). The  $\beta$ -Al<sub>5</sub>FeSi plates evolve in the pre-eutectic stage in the low Si alloys with 0.5 Fe, and even in high Si alloys with 0.8 Fe. In the high Si alloy with  $0.5$  Fe, the  $\beta$ -Al<sub>5</sub>FeSi plates form in the post-eutectic stage, hence size refinement for increased Si occurs (Fig. 3). In the high-Cu low-Si alloys the  $\beta$ -Al<sub>5</sub>FeSi plates form in the pre-eutectic stage and no refining should be expected, still the Figs 2 and 3 showed a size refining effect compared to the low-Si (Cu-free) alloys. In fewer words, further growth of the pre-eutectically nucleated  $\beta$ -Al<sub>5</sub>FeSi plates in the post eutectic stage is prevented by the liquid Cu-rich pools.

Increasing the Cu content to the low Si alloys reduces the size of the  $\beta$ -Al<sub>3</sub>FeSi plates in small SDAS, but the effect requires a critical level of Cu which is a function of the cooling rate and the Fe and Si content. The critical level of Cu seems determined to the solubility in AI, which is around I mass%. That is, an excess of Cu above this concentration is required for the retining of Fe-platelets to occur (ie. Formation of  $Al<sub>2</sub>Cu$  is required).

At large SDAS in the low Si alloys, increased Cu, actually increased the size of  $\beta$ -Al<sub>5</sub>FeSi plates. Since the Fe-intermetallic particles nucleate in the pre-eutectic stage and due to the slow cooling rate there would be much time and space for the independent growth of these plates to grow larger before the nucleation of the detrimental  $A1<sub>2</sub>Cu$  particle which could prevent the lengthening of these plates. In the slow cooling condition, i.e. SDAS  $>50 \mu m$ , Cu forms both blocky and fine eutectic  $A_2Cu$  in the low Si alloys. In high Si alloys it only forms very fine eutectic Al<sub>2</sub>Cu. Increasing Si increases the size of the  $\beta$ -Al<sub>5</sub>FeSi plates in Cu free alloys with 0.8 mass% Fe [3, 5]. Increasing Si with Cu reduces this effect. Hence, addition of Cu to high Si alloys not only improve its strength but also be beneficial by improving ductility especially for alloys made using slow cooling processes such as sand castings. Again Cu addition to low Si alloys (which form with SDAS of  $\leq 25 \mu m$ ) is also beneficial for refining the  $\beta$ -Al<sub>5</sub>FeSi plates. The reason for the refining effect of high level of Si on the  $Al_2Cu$  is, increasing Si in low cooling condition provides more nucleation sites, post eutectically formed  $\beta$ -AI,FeSi plates[6], for the eutectic Al<sub>2</sub>Cu and the formation of this fine eutectic precipitation prevent the lengthening of B-Al<sub>5</sub>FeSi plates in the later part of solidification.

### **Conclusions**

- High level of Si and/(or) Cu decreased the amount and size of the  $\beta$ -Al<sub>3</sub>FeSi platelets in AI-Si-Cu-Mg-Fe casting alloys, especially at small SDAS if the Fe level is below the Si dependent critical level for the formation of the pre-eutectic platelets (in the post eutectic stage).
- The Fe containing intermetallics become script-like, alpha-Al $_8$ Fe<sub>2</sub>Si, at small SDAS while at large SDAS they take a plate-like,  $\beta$ -Al<sub>5</sub>FeSi, shape.
- At small SDAS, in the low Si  $(4.5\%)$  alloys, Cu actually leads to decreased size of the interemetallics whereas **in** the Cu-free, high-Si alloy, the plates are replaced by mixture of the irregular alpha- $A_{1x}Fe_{2x}Si$  and  $B-A_{1x}FeSi$  platelets. ie. in the Cu containing alloys, plates are formed even at small SDAS.

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#### **References**

- [1]C.H. Caceres, I.L. Svensson, and J.A. Taylor: *International Journal of Cast Metals Research* 2003, vol. 15, pp. 531-543,
- [2]C.H. Caceres and *1.A.* Taylor: *Shaping Casting: The John Campbell Symposium* 2005, vo!. I, pp. 245-254,
- [3]C.H. Caceres, B. Johannesson, J.A. Taylor, A. Canales-Nunez, M. Cardoso, and J. Talamantes: *Shape Casting: 2nd Jnternational Symposium* 2007, vol., pp. 135-141,
- [4]T. Sivarupan, c.H. Caceres, and lA. Taylor: *Metallurgical and Materials Transactions A*  September 2013, vol. 44 A pp. 4071-4081, DOI: 10.1007/s11661-013-1768-x.
- [5]T. Sivarupan, c.H. Caceres, and .LA. Taylor: *Materials Science Forum* 2013, vol. Light Metals Technology 2013, pp. 107,
- [6]M.C. Eva Tillova, Lenka Hurtalova, Peter Palcek: *Acta Metallurgica Slovaca Coriference, Vol.* 3, *2013, p.* 196-2012013, vol., pp. 10. I 2776/amsc.v3iO. 127.