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# Crystallization of the Structural Components of Multiple Remelted AlSi9Cu3 Alloy

**M. Matejka \***, **D. Bolibruchová**, **M. Kuriš**University of Zilina, Faculty of Mechanical Engineering, Department of Technological Engineering,  
Univerzitna 1, 010 26 Zilina, Slovak Republic

\* Corresponding author. E-mail address: marek.matejka@fstroj.uniza.sk

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## Abstract

At present, Al-Si-Cu based alloys (with a typical representative AlSi9Cu3 alloy) represent more than half of the castings used in various industries (automotive, aerospace and electrical engineering). These are most often sub-eutectic (exceptionally eutectic) alloys with a content of 6 to 13 wt. % Si and 1 to 5 wt. % Cu. The aim of the paper is to point out the importance of the evaluation of input raw materials that determines the overall properties of the casting and the costs invested in its production. A negative impact on performance can be expected when using an alloy made up of a high proportion of recycled material, despite its economic benefits. Experimental alloys were evaluated based on the results of crystallization process and a combination of scanning electron microscopy (SEM), energy dispersive X-ray analysis (EDX), and deep etching. The effect of remelting and increasing the remelted returnable material in the batch was manifested especially in the crystallization of iron-rich phases. The negative effect of remelting on the structural components was manifested after the fourth remelting. Gradual increase of remelted returnable material in the batch causes harmful changes in the crystallization process.

**Keywords:** Al-Si-Cu, Remelting, Returnable material, Structural components, Scanning electron microscope

## 1. Introduction

The remelting process involves influencing the structure of the alloy, which can be caused by the remelting itself (without a significant change in chemical composition) or by a change in chemical composition (especially metal-loss/remelt-loss and diffusion of various elements). The most frequently observed change, which must be given special attention, is the increased iron content when comparing the remelted aluminum alloy with the primary alloy. [1-3].

When using recycled alloys it is therefore important to pay attention to the chemical purity of the alloy, which depends on the overall quality of the input materials, i.e. returnable materials. A

returnable or remelted (recycled) material may contain, in addition to the above-mentioned higher iron content, also a larger amount of oxide inclusions that are introduced into the melt with the batch and impair its purity. It is therefore important to determine in the manufacture of castings the appropriate ratio of the primary alloy and the returnable alloy in the batch, with respect to the required properties of the casting. It is therefore recommended to melt castings with the highest quality requirements with a maximum of 50 % returnable material. In addition to determining the proportion of batch material, it is also crucial to determine the shape or "piece number" of returnable material. Qualitatively more suitable are compact, larger pieces (such as risers and gating system) that have a relatively small surface area relative to their volume, because small proportions of returnable material or metal spray parts have

a larger surface area and a smaller volume, and thus introduce a large amount of inclusions into the melt. Recycled AlSi9Cu3 alloys are mainly used for the automotive and electrical industry for castings such as cylinder heads, shaft housings or various parts of electric motors. [3-6].

The solidification of Al-Si-Cu alloys begins with the primary  $\alpha$ -phase particles formation. As the temperature decreases, the volume of the liquid phase decreases and the solid phase increases, which begins the primary precipitation of the Al-Si eutectic. Exceeding the critical iron content of the alloy increases the precipitation of intermetallic phases that form during solidification of the eutectic. Lastly, the copper-rich eutectic phases precipitate in the final stage [7-9].

## 2. Materials and methodology

The evaluation of the change in crystallization and solidification of the structural components due to multiple remelting and the use of multiple-remelted returnable material in the batch in combination with a commercial grade (purity) alloy was performed using a sub-eutectic AlSi9Cu3 alloy.

Table. 1

Chemical composition of all alloys used in the first and second stages of the experiment (wt. %)

Elements	Si	Cu	Mg	Zn	Ni	Ti	Mn	Cr	Fe
<b>Z1</b>	9.441	2.174	0.429	1.158	0.090	0.035	0.174	0.024	1.414
<b>Z3</b>	9.316	2.114	0.423	1.157	0.097	0.037	0.186	0.043	1.475
<b>Z5</b>	9.313	2.104	0.407	1.144	0.115	0.033	0.181	0.061	1.51
<b>Z7</b>	9.286	2.097	0.394	1.173	0.133	0.031	0.187	0.103	1.612
<b>Commercial purity</b>	<b>9.563</b>	<b>2.206</b>	<b>0.426</b>	<b>1.160</b>	<b>0.092</b>	<b>0.038</b>	<b>0.184</b>	<b>0.027</b>	<b>1.081</b>
<b>Returnable material</b>	<b>9.294</b>	<b>2.074</b>	<b>0.348</b>	<b>1.016</b>	<b>0.129</b>	<b>0.034</b>	<b>0.184</b>	<b>0.113</b>	<b>1.674</b>
<b>20-80</b>	9.507	2.197	0.391	1.044	0.122	0.035	0.231	0.049	1.294
<b>50-50</b>	9.418	2.173	0.361	1.041	0.134	0.033	0.223	0.072	1.419
<b>70-30</b>	9.245	2.02	0.344	0.961	0.108	0.031	0.209	0.112	1.569
<b>80-20</b>	9.415	2.08	0.358	1.07	0.126	0.032	0.206	0.101	1.617
<b>90-10</b>	9.291	2.143	0.357	1.046	0.127	0.032	0.199	0.106	1.643

The melting of the experimental alloys was performed in both stages in a T15 electric resistance furnace. The batch weight at each melt was 12 kg. The casting temperature was in the temperature range  $750 \pm 10$  °C. The crystallization process of the experimental alloys was evaluated using a K-type thermocouple (NiCr-Ni). The scheme of metal mold and thermocouple location is shown in the Fig. 1. LabView 2 Hz software was used to record data.

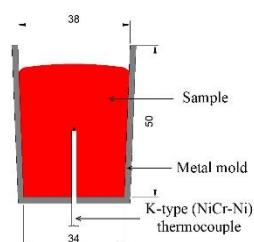


Fig.1. Scheme of metal mold and thermocouple location

The experiment was divided into two stages. In the first stage of the experiment, reference AlSi9Cu3 alloy with approx. 1.4 wt.% iron content was created (by adding AlFe10 master alloy) and was marked Z1. Subsequently, the newly formed reference alloy Z1 was subjected to 6-fold remelting (casting of ingots into metal molds and subsequent use of ingots as a batch for further melting). Specimens for evaluation of microstructures were cast from every second (other) melt (marked Z3, Z5 and Z7) into a metal mold ( $T_{\text{mold}} = 100 \pm 5$  °C).

In the second stage, which was dedicated to the evaluation of the returnable material amount increase impact in the batch, was used a commercial purity AlSi9Cu3 alloy and remelted returnable material from AlSi9Cu3 alloy. The returnable material consisted of risers and gating systems and remnants of ingots used in the first stage of the experiment. In the next part of the experiment, five experimental alloys with varying ratios and designations were successively cast, namely 20-80; 50-50; 70-30; 80-20; 90-10, where the first two digits indicate the percentage of remelted returnable material and the other two digits indicate the proportion of commercial purity alloy AlSi9Cu3 in the batch. Chemical composition of all alloys used in the first and second stages of the experiment is given in Tab. 1.

## 3. Results and Discussion

From the graphs at Fig. 2. it can be seen that due to the remelting process and the increase of the returnable material in the batch, a significant change occurred only in the nucleation of the iron phases. The nucleation temperatures of iron-based phases in the Z1 reference alloy were at the level of  $T_{\text{AlSiFe}} = 578$  °C, respectively  $T_{\text{AlSiFe}} = 574$  °C for the alloy with the lowest returnable material content 20-80. After applying 6-fold remelting, the Z7 alloy showed the  $T_{\text{AlSiFe}}$  value of 585 °C, and  $T_{\text{AlSiFe}} = 584$  °C for the maximum of returnable material content of 90-10.

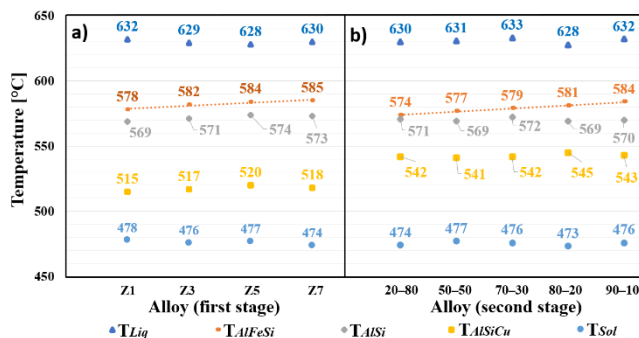


Fig. 2. Characteristic transformation temperatures of structural components of AlSi9Cu3 alloy a) with different degree of remelting b) with various amount of returnable materials in batch

### Precipitation of eutectic silicon

The eutectic Si particles are present in the Z1 alloy and the Z3 alloy after the second remelting in unmodified form. In the plane of the cut, it was possible to observe them as dark gray variously oriented sticks (Fig. 3a) and in 3D morphology as large hexagonal plates with visible twinning (detail: red ellipse). The the fourth and sixth remelting led to, the eutectic Si crystallized in the morphology of coarse hexagonal plates with an undirected distribution or like polyhedral grains (Fig. 3b). Irregular multiple doubling can be observed on the hexagonal plates (detail: red ellipse).

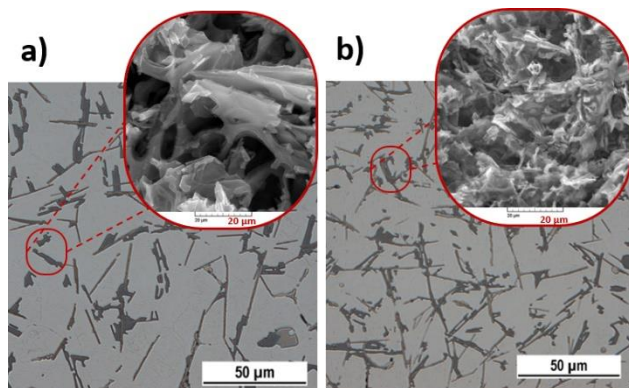


Fig. 3. Morphology of eutectic silicon, optical microscope and deep etching SEM a) Z1 reference alloy b) Z7 alloy

The morphology of the eutectic silicon grains in the alloy with a 20 % of returnable remelted material (20–80) was preserved and it was the same as in the Z1 alloy. Eutectic Si is precipitated in an unmodified form as non-oriented distribution hexagonal plate crystals with a TPPE (twine plane re-entrant edge) growth mechanism [10]. The degraded morphology of eutectic Si, which is characteristic of alloys with a higher number of remelts (after the fourth and sixth remelts), began to manifest itself as early as in the alloy with a balanced, 50–50 ratio (Fig. 4). A 50 % share of the returnable remelted material in the batch caused that non-oriented thick hexagonal plates of eutectic Si predominated in the microstructure. In alloys containing 70% and more of returnable material, the eutectic Si crystallized exclusively in the morphology of coarse hexagonal plates and polyhedral grains, exactly as in the Z7 alloy after the sixth remelting.

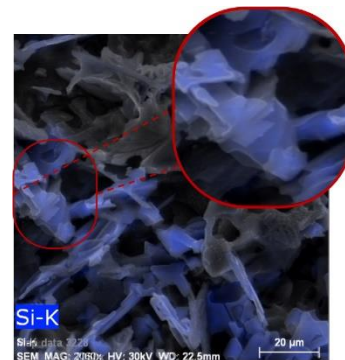


Fig. 4. Morphology of the 50–50 eutectic Si alloy with mapping focused on Si, deep etching SEM

### Precipitation of iron and copper-rich intermetallic phases

Iron-rich phases are present in acicular morphology in all alloys subjected to remelting as  $\beta$ -Al<sub>7</sub>FeSi phases (Fig. 5a). The application of multiple remelting did not significantly change the morphology, but there were visible increases in the lengths of the needles (acicular formations) of the iron-based phases (Fig. 5b) after the fourth remelting. The preferential nucleation of the iron phases over the eutectic silicon phases allowed for the growth of acicular formations up to lengths more than 70  $\mu$ m (for Z5 and Z7 alloys). Changes in the average lengths of the iron phases are given in Tab. 2.

The copper-rich Al<sub>2</sub>Cu phases were observed in alloys subjected to remelting and Al<sub>7</sub>FeCu phases in alloys after the fourth (Z5) and sixth (Z7) remelts (Fig. 5b). The Al<sub>2</sub>Cu phases crystallized in the form of the ternary eutectic Al–Al<sub>2</sub>Cu–Si and as continuous "blocks". The intermetallic Al<sub>2</sub>Cu phase in the form of a continuous block is shown in Fig. 5a (red ellipse).

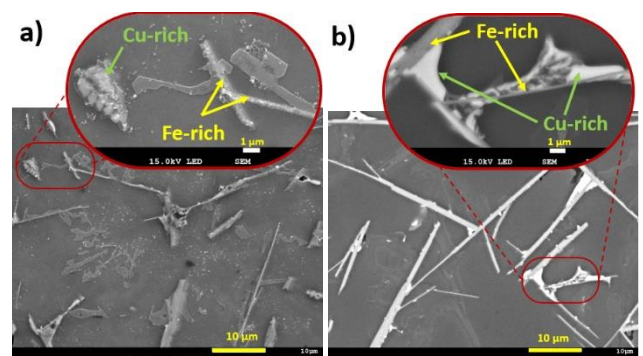


Fig. 5. Distribution of iron and copper-rich phases, SEM a) the Z3 alloy b) the Z5 alloy

Iron-based intermetallic phases in alloys with different ratios of commercial purity alloy and returnable remelted material crystallized in interdendritic sites as acicular formations in metallographic cutting (Fig. 6). The measurement of the average lengths of the acicular formations confirmed their expected growth due to the increase in the ratio of remelted returnable material. The measured values of acicular formation lengths are given in Tab. 2.

The trend of nucleation of copper-rich phases in close proximity to iron phases has also been confirmed for alloys with different ratios of multiple remelted returnable material. The mapping shown in Fig. 6 (in the upper part) shows the iron-phase acicular formations as nucleation sites for copper-rich phases.  $Al_2Cu$  phases were observed in both forms in experimental  $AlSi9Cu3$  alloys with an increasing returnable material ratio. Equally,  $Al_7FeCu_2$  phases with increased Fe content were observed (mainly in alloys 70–30, 80–20 and 90–10), being most frequently precipitated in small formations directly on the plate (acicular formation) of iron-based phases.

As can be seen in Tab. 2, the first application of two-fold remelting resulted in a decrease of the average lengths of acicular formations of iron-based phases from 56  $\mu m$  to 27  $\mu m$ . The shortening of the average lengths can be explained by the fact that the long acicular formations were "broken" into phases with smaller dimensions due to remelting. Subsequent application of the fourth and sixth remelting resulted in a rapid increase in acicular lengths to 71  $\mu m$  (Z5) and 89  $\mu m$  (Z7). The average acicular formation lengths in alloys with varying ratios of secondary alloy and returnable material in the batch were in a narrower range (from 35  $\mu m$  to 51  $\mu m$ ) than those in remelted alloys. It follows from the above that the very long dimensions of the iron phase acicular formations of the in the Z5 and Z7 alloys were not preserved in the alloys where their batches also contained 90 % of the remelted returnable material.

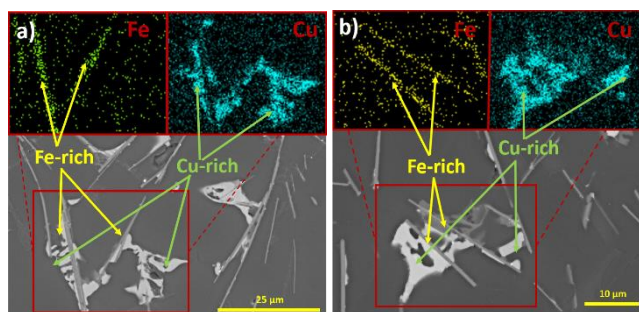


Fig. 6. Mapping of the site rich in the presence of copper and iron-based phases, SEM a) the 50 – 50 alloy b) the 80 – 20 alloy

Table 2.

Average acicular Fe phases lengths of experimental alloys ( $\mu m$ )

Alloy	Z1	Z3	Z5	Z7	
Acicular	56	28	71	89	
Alloy	20-80	50-50	70-30	80-20	90-10
Acicular	34	39	45	44	51

## 4. Conclusions

Multiple remelting as a form of aluminum alloys recycling is a very complex and complicated issue despite the fact that it is a common and integral part of the cycle of aluminum alloy castings.

The presented paper aims to provide an insight into the issue of multiple remelting of alloys and their subsequent use in a batch with an alloy of commercial purity from the point of view of crystallization of individual structural components.

By evaluating the cooling curves for all experimental alloys and their first derivatives were observed changes in both stages of experiment occurred during the nucleation of iron phases as an increase in their nucleation temperatures. The nucleation of iron phases in all alloys was preferred to the nucleation of eutectic Si. The preferential crystallization created ideal conditions for the growth of harmful  $\beta-Al_5FeSi$  acicular formations, which was confirmed especially in the alloys Z5 and Z7. Subsequent use of remelted returnable material, even in the amount of 90 %, did not prove the "inheritance" of large lengths of iron phases. For the 90 – 10 alloy, the average acicular formation lengths of 51  $\mu m$  were measured, which is 20  $\mu m$  shorter than those in the Z5 alloy, or 40  $\mu m$  shorter than those in the Z7 alloy. On the contrary, negative "inheritance" was observed in the morphology of eutectic Si. Coarse hexagonal plates with undirected distribution or in the form of polyhedral grains (characteristic for Si in the Z5 and Z7 alloys) were already present in the structure in the alloy with a balanced ratio (50–50 alloy). Exclusively coarse morphology and polyhedral grains of eutectic Si were observed in alloys with 70%, 80% and 90% of returnable material. The  $Al_7FeCu_2$  phase was present in the alloys with increasing wt. % of iron.

The presented paper was created as part of extensive experimental work focused on the recycling of aluminum alloys with increased iron content. Experiments show that the change in microstructure due to remelting and increasing of the return material in the batch leads to a decrease in mechanical properties (especially ductility) as well as selected foundry properties (porosity and increased susceptibility to hot tears).

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