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Application of Natural and Artificial Ageing on Multiply Remelted AlSi9Cu3 Alloy

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Abstract

This article focuses on the study of the influence of remelting and subsequent natural and artificial ageing on the structure of recycled AlSi9Cu3 alloy with increased iron content. The assessed changes in eutectic silicon and iron-based intermetallic phases were carried out using optical and scanning electron microscopy. The degradation of the eutectic silicon morphology due to remelting occurred only at the highest numbers of remelting. The effect of remelting the investigated alloy, which is accompanied by a gradual increase in wt. % Fe, began to manifest significantly through a change in the length of the ferric phases after the fourth remelting. As expected, the artificial ageing process has proven to be more effective than natural ageing. It has led to a change in the eutectic silicon morphology and has been beneficial in reducing the lengths of adverse ferric phases. The use of alloys with higher numbers of remelting, or with greater "contamination", for the manufacture of shape-challenging castings is possible when using a suitable method of eliminating the negative factors of the remelting process. The results of our investigation show a suitable method of the above elimination the application of heat treatment T5 – via artificial ageing.

Keywords: Remelting, Fe phases, Artificial ageing, Natural ageing, Eutectic silicon

1. Introduction

Al-Si-Cu alloys are the most widely used type of aluminium alloys. Because of their low weight and excellent casting and mechanical properties, such alloys are applied in various industries, with the largest part of their production being castings for the automotive industry. At present, more than half of the aluminium castings manufactured in the European Union come from recycled material. The growing interest in aluminium recycling and the manufacture of castings from secondary alloys is associated with benefits that are obtained by using recycled material. Recycled aluminium remelting can save up to 95% of the energy needed to produce primary aluminium, thus avoiding the processes associated with mining, ore refining and melting [1, 2]. For this reason, foundries in batches nowadays use increasing amounts of remelted material (inlet systems, chunks or non-complying castings), which can account for several dozen percent of the total batch. However, with the use of remelted material, the question arises as to whether it is possible to expect the same quality of the resulting castings as with the use of primary aluminium. The multiple remelting process also brings about certain changes in the structure of aluminium alloys, which can lead to a degradation of the casting performance properties, and this is the main reason for the importance of investigating the remelting process – how and to what extent it can affect the performance properties [3,4].

Due to increasing use of recycled aluminium alloys for difficult castings, especially for the automotive industry, castings quality is considered a key factor. Mechanical and casting properties of aluminium alloys depend to a large extent on the chemical composition. Actually, it is the chemical composition that is often affected by the remelting process and it changes [5, 6]. As a result of remelting, there can occur a reduction in wt. % of some elements due to their overburn, or an increase by diffusion from the surrounding environment. Iron is considered the most damaging element, whose content in the alloy can increase as a result of remelting. With increasing iron content, precipitation of intermetallic phases increases in the form of long needles, which results in the degradation of mechanical properties. For recycled alloys, the value of the so-called critical Fe_{crit} content in wt. % is important. This value is directly proportional to the silicon concentration in the alloy and determines the amount of iron permitted prior to the onset of an adverse β - Al_5FeSi phase. The Al_5FeSi phase is most often formed in Al-Al_5FeSi-Si eutectic in the form of thin needles and significantly reduces mechanical properties, strength, yield strength and fracture toughness. The critical Fe content in wt. % is calculated from:

$$Fe_{crit} \approx 0{,}075 \text{ x } (\text{wt. \% Si}) - 0{,}05 \quad (1)$$

Greater influence than chemical composition on mechanical and casting properties has the size and morphology of excluded intermetallic phases. The purpose of this article is to investigate the influence of remelting and also that of natural and artificial aging on excluded phases and their morphology in alloys with varying numbers of remelting [7-9].

Castings from AlSi9Cu3 alloys are used most frequently in the state without heat treatment. A certain degree of self-curing occurs due to the presence of copper. The ageing process is slow and spontaneous. Final properties are achieved after 100 – 150 hours. Artificial ageing takes place in foundry alloys at 150 to 200 °C for 1 to 10 hours [9,10].

2. Experimental methods and materials

The AlSi9Cu3 alloy was used as the experimental material. The alloy was prepared in the form of ingots with the total batch weight of 100kg. To investigate the effect of a higher iron content at multiple remelting, we intentionally increased wt. % of iron in the experimental alloy. Intentional "contamination" was achieved by adding AlFe10 pre-alloy into the melt at 750 ± 5 °C, thereby increasing the iron content from the original value of 1.08 wt. % (primary alloy) to approximately 1.4 wt. %. For the further progress of the experiment, the newly formed alloy with a higher iron content was used as a reference alloy designated D1. Its chemical composition is shown in Tab. 1. In Tab. 1 can be seen that copper, which already has a minor wt. % in the basic alloy from the supplier, as specified by the standard. The remelting process was carried out in an electric resistance furnace in a steel crucible treated with a protective coating.

Remelting consisted of pouring ingots into the pre-prepared metal moulds. After solidification and cooling, the ingots were used again as a batch for the next melting without additional chemical treatment. The process was repeated 6 times, which resulted in six-fold remelting of the reference alloy. Samples for micro-structural analysis and mechanical tests were cast from every second melt (designated D3, D5, D7) into a metal mould with a minimum temperature of 100 ± 5 °C. The casting temperature ranged from 750 to 770 °C, and 12 samples were cast for each tested alloy. The chemical composition of the alloys, and the calculated critical iron content according to Eq. (1), are shown in Tab.1. We did not inoculate, modified or refined the alloy whatsoever. We only mechanically removed oxide coats before casting. The resulting alloys D1, D3, D5 and D7 were assessed in three different states. In the cast (mould) state, where we did not perform any additional heat treatment, after natural ageing (TS – approximately 160 hours at 20 ± 5 °C), and after heat treatment (TS – T5 artificial ageing at 200 ± 5 °C for 4 hours and cooling in water with a temperature of 60 ± 3 °C) [7].

3. Results of experimental work

In order to perform structural analysis, we used a Neo-foto 32 light microscope, and SEM observation with EDX analysis using a VEGA LMU II scanning electron microscope. The samples for experimentation were prepared for structural analysis using a standard metallographic procedure. To use the light microscope for observation, we etched the samples in a solution of 20 ml H_2SO_4 and 100 ml water. For SEM and EDX analysis, we used 0.5% HF solution. Deep etching samples were etched for 20 – 30 seconds in a solution of 36 ml HCl + 100 ml H20 and intensively rinsed in alcohol to remove residues of the dissolved aluminium matrix. After natural and artificial ageing, each sample was subjected to measuring the length of the Al5FeSi ferric phase at 500-fold magnification.

Effect of multiple remelting on the alloy in the cast state

The analysed microstructure of the D1 reference alloy is composed of primary α-phase dendrites, eutectic silicon in unmodified form, and iron-based platelet formations and intermetallic phases in needle morphology that are evenly distributed in inter-dendritic regions (Fig. 1a). Two-fold remelting allowed us to observe a change in the D3 alloy. There were: a refinement (spheroidization) of eutectic silicon and partial fragmentation of iron-based ferric phases excluded in the form of shorter needles (Fig. 1b). After the fifth remelting the eutectic silicon did not undergo a significant change, but the increased Fe content to 1.738 wt. % in D5 alloy resulted in the increased number and size of the Fe needle phases (Fig. 1c). Eutectic silicon is present in the form of thickened plate formations in the D7 microstructure alloy. At the Fe content of 1.889 wt. %, we can observe in the D7 alloy a large number of long and thin Fe-phase needles (Fig. 1d). Increasing the iron content in the alloy may also have been caused by the insufficient treatment of the steel crucible, using coating after each remelting, which could lead to the mould being "impurified" by iron, since molten aluminium cannot dissolve iron from unprotected steel tools.

Table 1.
Relationship between wt. % of selected elements and cast number (CN)

CN	Si [wt. %]	Fe [wt. %]	Cu [wt. %]	Mn [wt. %]	Mg [wt. %]	Cr [wt. %]	Fe_{crit} [wt. %]
D1	9.347	1.416	1.741	0.178	0.427	0.025	0.651
D3	9.306	1.51	1.741	0.186	0.417	0.04	0.648
D5	9.302	1.738	1.667	0.181	0.402	0.065	0.647
D7	9,179	1.889	1.633	0.187	0.339	0.106	0.638

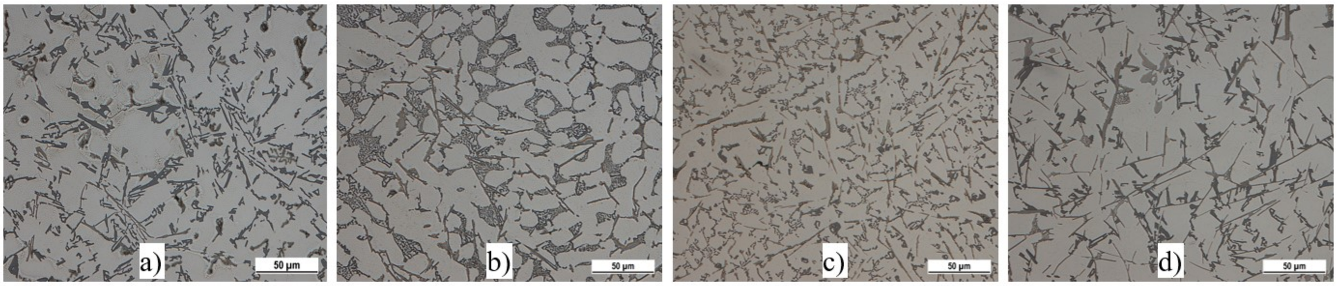
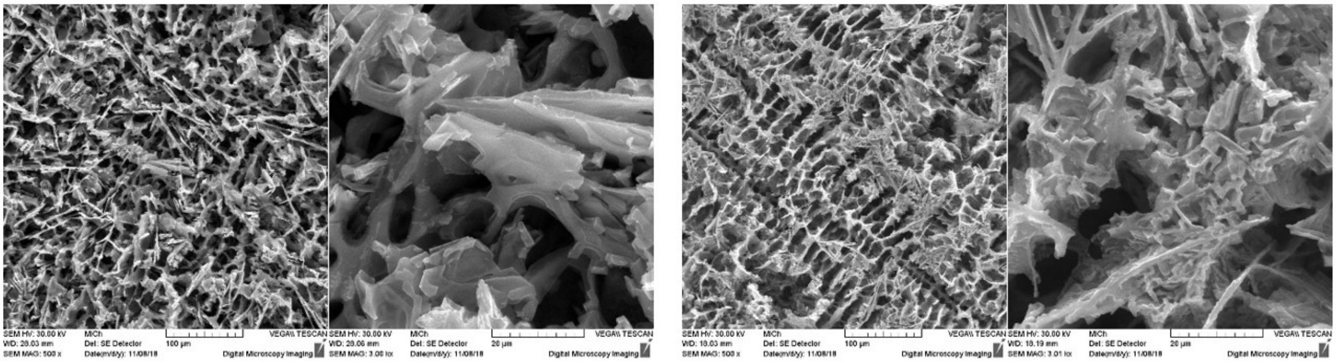


Fig. 1 Microstructure of AlSi9Cu3 alloy in dependence on number of remelting in cast state:
a) D1 alloy b) D3 alloy c) D5 alloy d) D7 alloy

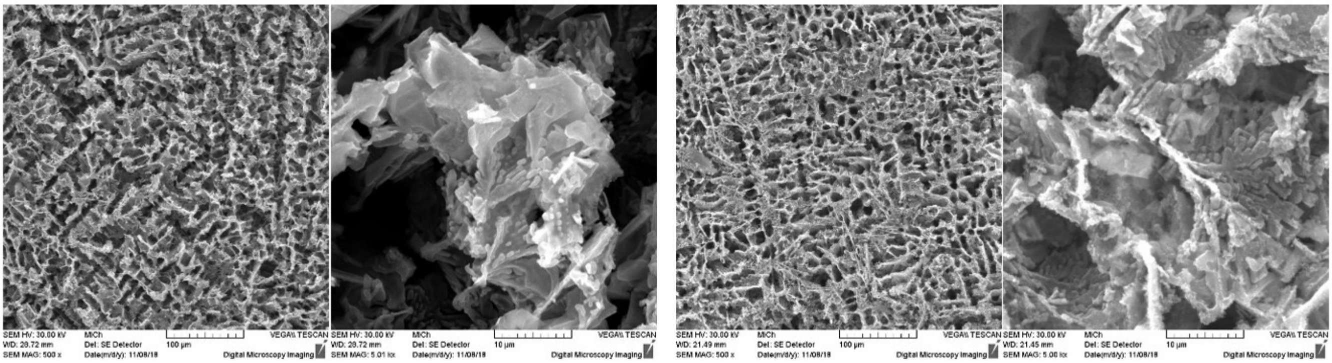
Influence of natural and artificial ageing

Eutectic silicone in the reference D1 alloy is, after natural ageing, excluded in the form of continuous pallet formations (Fig. 2a). However, artificial ageing has led to a partial fragmentation of the eutectic (Fig. 2b). Natural ageing, combined with two-fold remelting, has resulted in the 3D alloy in refining some, originally thick eutectic pallets, and their thickening and local fragmentation (Fig. 3a). After applying artificial ageing on the D3 alloy, the process of thickening and fragmentation ran to a larger extent, and we can observe even spheroidised separated (cleaved) fragments of eutectic silicon (Fig. 3b).



a) b)

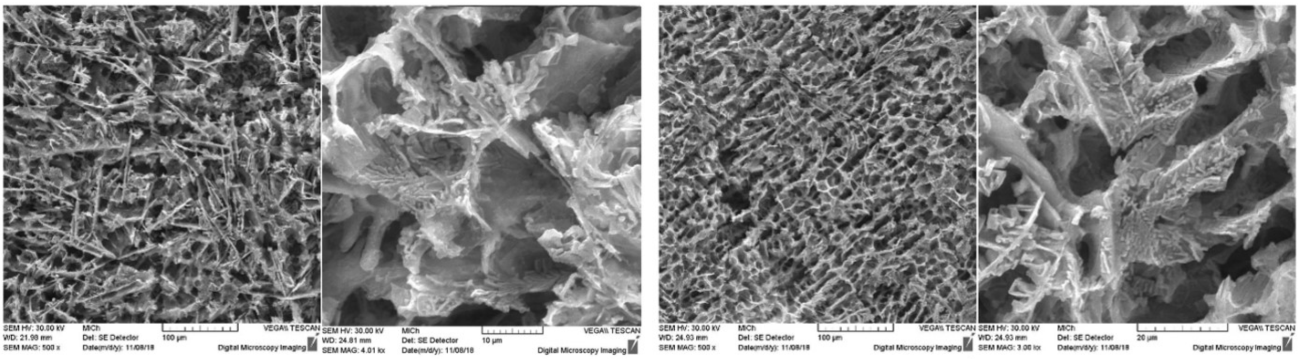
Fig. 2 Eutectic silicon after deep etching of reference alloy D1, SEM a) after natural aging b) after artificial aging



a) b)

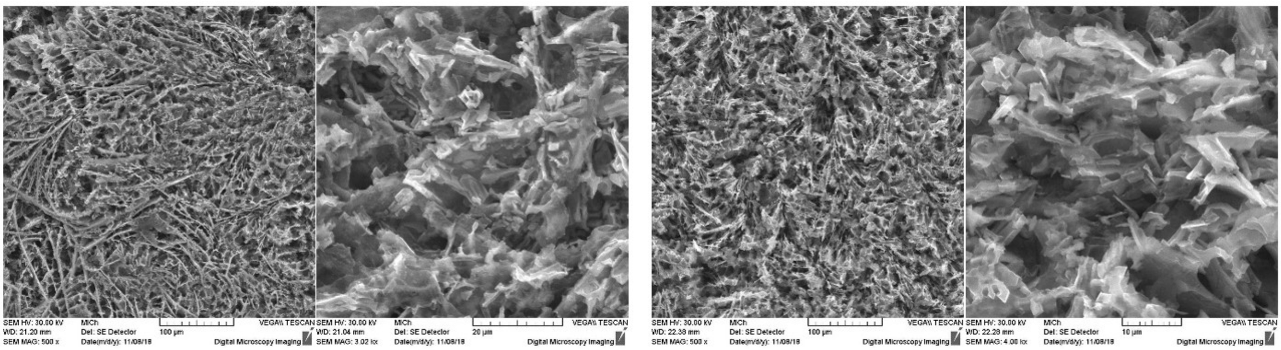
Fig. 3 Eutectic silicon after deep etching of alloy after 3rd remelting D3, SEM a) after natural aging b) after artificial aging

Fig. 4a shows eutectic silicon in the D5 alloy after the fifth remelting and natural ageing. Similarly to the D3 alloy, it crystallized in finer and, here and there, fragmented forms. By introducing artificial ageing, a larger amount of fragmented particles with subsequent spheroidization (Fig. 4b) can be observed. The seven-fold remelting of the alloy resulted in exclusion of eutectic silicon in non-oriented and irregular plate-like structures. Irregular morphology of eutectic silicon is observed in the D7 alloy after natural as well as artificial ageing (Fig. 5).



a) b)

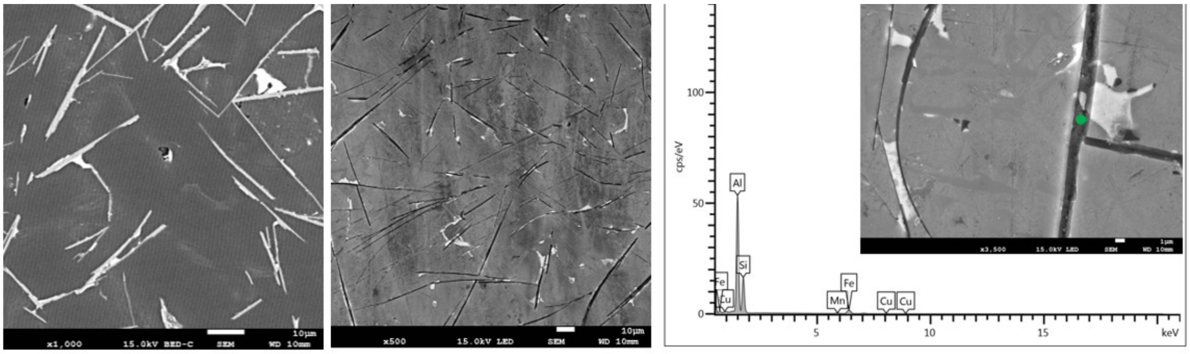
Fig. 4 Eutectic silicon after deep etching of alloy after 5th remelting D5, SEM a) after natural aging b) after artificial aging



a) b)

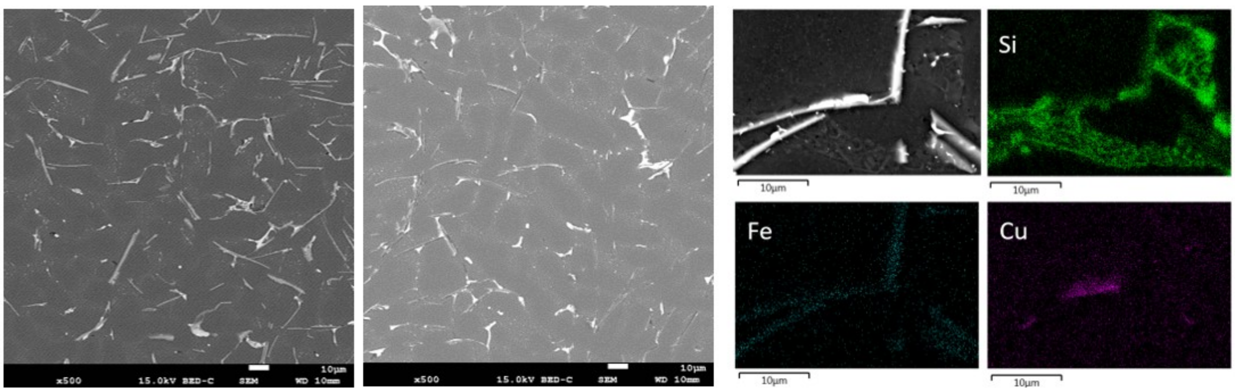
Fig. 5 Eutectic silicon after deep etching of alloy after 7th remelting D7, SEM a) after natural aging b) after artificial aging

Ferrous β-Al_5FeSi phases, just like in cast-state alloy, are present in the reference D1 alloy after natural aging in needle morphology with the average needle length of 55.7µm (Fig. 6a). Artificial ageing resulted in a significant shortening of iron phase needles, the average length of which ranged around 37.8 µm (Fig. 6b). Fig. 6c shows a point EDX analysis of the β-Al_5FeSi phase of the reference D1 alloy after artificial ageing, which confirmed the presence of Fe and Si elements. The effects of both natural and artificial ageing of the alloy after the third remelting of D3 led to the local refinement and shortening of the Fe phase needles (Fig. 7a, b) as compared to the casting state. The average length of needles in both cases reached approximately 27 µm. In Fig. 7c, the mapping shows the re-composition of elements in the intermetallic β - Al_5FeSi phase that are present in the alloy after the third 3D remelting and natural ageing with the content of 1.51 wt. % Fe. As can be seen, near these phases there is a certain amount of Cu excluded (probably it is one of the curing phases of Al_2Cu). Tab. 2 shows the average measured length of the β-Al_5FeSi phase needles for each alloy.



a) b) c)

Fig. 6. Distribution of intermetallic iron-based phases in reference alloy D1; SEM a) natural aging b) artificial aging c) EDX analysis of needle particles β - Al_5FeSi phase in D1 alloy after artificial aging



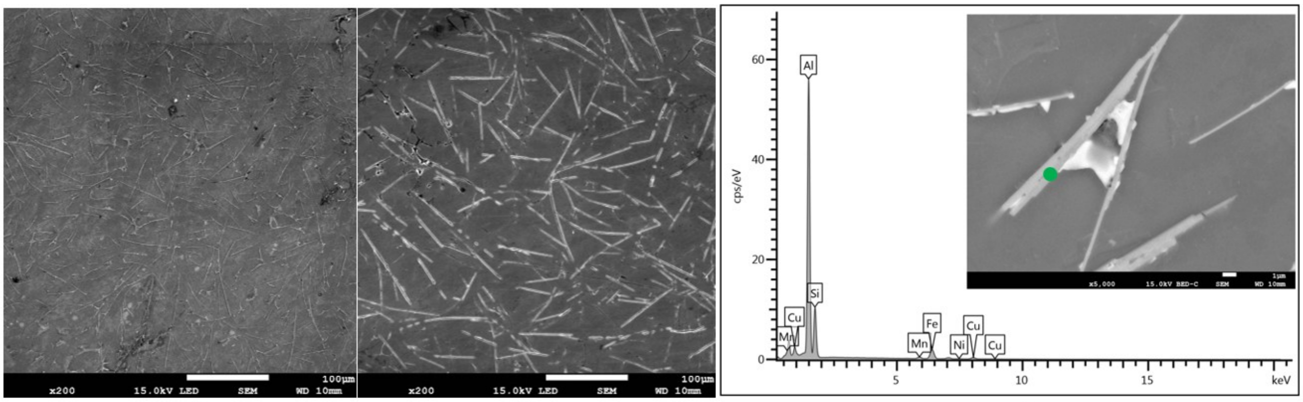
a) b) c)

Fig. 7. Distribution of intermetallic iron-based phases in alloy after 3rd remelting D3; SEM a) natural aging b) artificial aging c) mapping elements of needle particles β - Al_5FeSi phase in alloy D3 after natural aging

Table 2
Results of measurements of length of iron-based particles in dependence on number of remelting

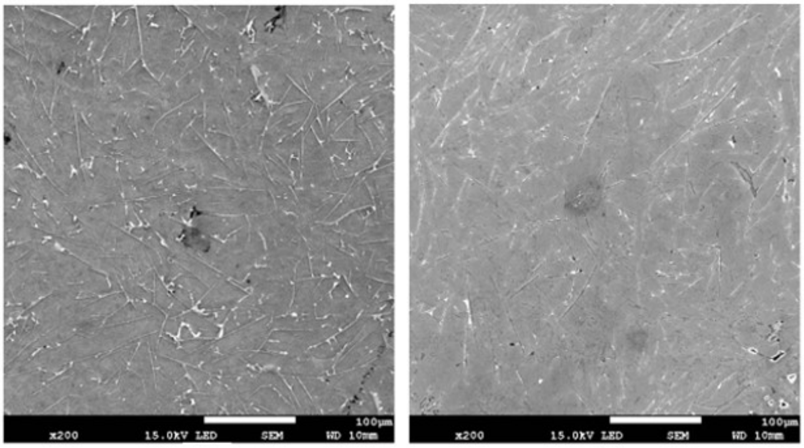
Alloy		D1	D3	D5	D7
Average length of iron phases [µm]	Natural aging	55.7	27.7	71.4	89.4
	Artificial aging	37.8	26.3	55.1	72.6

The alloy structure after the fifth remelting of D5 and after natural ageing is characterized by the presence of β – Al_5FeSi phases in the needle-like morphology, with the average length of the needles being 71.4 µm (Fig. 8a). Artificial ageing resulted in crystallization of iron phases with significantly shorter lengths (55.1 µm) (Fig. 8b), which represents a reduction by 23 % compared to natural ageing. A more beneficial effect of artificial ageing on the length of the iron phase needles manifested also in the alloy at the seventh remelting of D7. The average measured length of the β – Al_5FeSi phase needles after natural ageing is 89.4 µm (Fig. 9a), and after artificial ageing we measured the value of 72.1 µm (Fig. 9b). In this case, the length reduction is by 19 %. Point EDX analysis of the needle phase in alloy D5 after natural ageing shows the presence of Fe and Si elements, confirming that it is the β – Al_5FeSi phase (Fig. 8c). EDX point analysis confirmed that the iron phase needles in the monitored cases serve as nucleation sites for crystallization of the Al_2Cu curing phases.



a) b) c)

Fig. 8. Distribution of intermetallic iron-based phases in alloy after 5th remelting D5; SEM a) natural aging b) artificial aging c) EDX analysis of needle particles β - Al_5FeSi phase in D5 alloy after natural aging



a) b)

Fig. 9. Distribution of intermetallic iron-based phases in alloy after 7th remelting D7; SEM a) natural aging b) artificial aging

5. Conclusions

The fourth remelting can be described as critical based on observing the remelting affecting the iron-based intermetallic phases. Paradoxically, eutectic silicon even after applying the fifth remelting was present in a more favourable form than in the reference alloy. However, the degradation expected occurred in the repeated increase in the number of remelting.

The eutectic silicon morphology was not significantly influenced by natural aging. The introduction of artificial ageing has led, excluding the D7 alloy with the highest number of remelting, to a positive change in the eutectic silicon morphology in the investigated alloys. An equally positive influence on Fe phases was recorded due to heat treatment by artificial ageing. A significant shortening of needles was observed in alloys D1, D5 and D7.

The use of alloys with higher numbers of remelting, or with greater "contamination", for the manufacture of shape-challenging castings is possible when using a suitable method of eliminating the negative factors of the remelting process. The results of our investigation show a suitable method of the above elimination the application of heat treatment T5 – via artificial ageing.

At present, the aim of using remelted material is to achieve a substitution for primary alloys, without reducing the end-product final properties. Although various scientific studies have been carried out that had dealt with the above-discussed issue, yet despite the knowledge available now no precisely and well-defined way of influencing the remelting effect on the performance properties has been defined.

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