

Removal of primary iron rich phase from aluminum-silicon melt by centrifugal separation

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Abstract: Recycling is a major consideration in continued aluminum use due to the enormous demand for high quality products. Some impurity elements gradually accumulate through the repetitive reuse of aluminum alloy scrap. Of them, the iron content should be suppressed under the allowed limit. In the present research, a novel separation method was introduced to remove primary iron-rich intermetallic compounds by centrifugation during solidification of Al-Si-Fe alloys. This method does not use the density difference between two phases as in other centrifugal methods, but uses the order of solidification in Al-Si-Fe alloys, because iron promotes the formation of intermetallic compounds with other alloying elements as a primary phase. Two Al-Si-Fe alloys which have different iron contents were chosen as the starting materials. The iron-rich phase could be efficiently removed by centrifuging under a centrifugal force of 40 g. Coarse intermetallic compounds were found in the sample inside the crucible, while rather fine intermetallic compounds were found in the sample outside the crucible. Primary intermetallic compounds were linked to each other via aluminum-rich matrix, and formed like a network. The highest iron removal fraction is 67% and the lowest one is 7% for Al-12Si-1.7Fe alloy. And they are 82% and 18% for Al-12Si-3.4Fe alloy, respectively.

Key words: Al-Si alloys; iron removal; centrifugal separation

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Most of the aluminum components for cars are made by high pressure die casting. High pressure die casting is an efficient and economical process which is appropriate for high speed production and high dimensional accuracy of the components. Practically all die cast aluminum alloys are produced using recycled materials^[1]. However the casting yield in high pressure die casting is rather low because the portion of biscuit, runner and overflow compared to the casting itself is high due to its thin and complicated shape. Lots of scrap is generated, which is used for subsequent melting.

Therefore recycling is a major consideration in continued aluminum use due to the enormous demand for high quality products in connection with the rising amount of secondary raw materials. More than half of all the aluminum currently produced in the European Union (EU-25) originates from recycled raw materials and that trend is on the increase.

Growing environmental concerns and heightened social responsibility, especially during the last decade, have served to boost the recycling activity, since recycling requires as little as 5% of the energy needed for primary aluminum production^[2].

However, some impurity elements gradually accumulate through the repetitive reuse of aluminum alloy scrap. Of them, the iron content should be suppressed under the allowed limit. Iron comes from the bauxite ore and ferrous metals such as melting tools and dies, and is one of the most troublesome impurities in aluminum cast materials. Iron also gradually accumulates through the reuse of aluminum alloy scrap. During the solidification of aluminum-silicon alloys, iron promotes the formation of intermediate phases such as AlFeSi intermetallic compounds, which may damage the properties of the final cast products. In secondary aluminum industries, iron contamination caused by high amounts of iron scrap is always possible^[3].

Iron is also present in most traditional die casting alloys as an impurity, yet a very useful impurity. Iron in the alloy may lead to a decrease in die soldering [4] which occurs since molten aluminum is injected into the die by very high pressure of over 49 MPa. Iron in concentrations of 0.8wt.% or more greatly reduces the tendency of an alloy to solder in die casting tooling. Iron in a moderate range intended to reduce soldering

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is also credited with improving somewhat the resistance of die casting alloys to hot tearing during solidifications in rigid die cast tooling. That characteristic may, however, be overstated as some die casters say that iron at the upper end of the typical range actually increases hot-tearing issues [5]. The highest allowed content of iron in the specification of high pressure casting alloys is 1.3wt.% for A380 and A390 alloys [6]. However, repetitive use of aluminum alloy scrap is a cause of increasing the iron content in the castings. It can be increased 3wt.% to 4wt.% by repetitive use of scrap.

A lot of research on how to reduce the iron content in the aluminum alloy scrap has been reported so far, which is mainly applicable for the aluminum alloys which contain more iron impurities. These methods mainly include the dilution method, gravity sedimentation, filtration, centrifugal separation, electromagnetic purifying method, fractional solidification, flux method, electro-slag refining, etc. However the operation process and equipment for these methods are relatively complicated [7-16]. Of them, centrifugal separation process seems to be attractive, because it can be applied to directly remove iron-rich phases from the partially solidified aluminum alloy melts without the addition of any other elements. The principle of centrifugal separation is to use the density difference between the iron rich phase and the aluminum liquid. The iron-rich phases moved to the edge side of the melt and the central part was purified [7]. However, iron-rich phases coexist with aluminum rich phases in one solid, so it needs another step to separate them mechanically and chemically.

So we tried to develop a novel method to decrease the iron content effectively using a vertical centrifugal separator which needs no more effort for further separation since each part of the high and low iron content is divided automatically into two pieces after centrifugation. This method does not use the density difference between the two phases as in the other centrifugal methods, but uses the order of solidification in Al-Si-Fe alloys. Excessive iron

in aluminum melt produces coarse AlFeSi intermetallic compounds during solidification. This intermetallic compound is the primary phase during solidification of Al-Si-Fe alloy. If they form a network at the initial stage of solidification, the unfrozen aluminum-rich phases can be moved outside the crucible leaving the iron-rich phases, including AlFeSi intermetallic compounds, inside during rotating. We have already proven the possibility of realization of the above assumption with Al-Si alloys [17]. Two Al-Si-Fe alloys which have different contents of iron were chosen as the starting materials.

1 Experimental detail

Alloys in the Al-12Si-Fe system with iron contents of 1.7wt.% and 3.4wt.% were prepared in an electrical resistance furnace. Master alloys of Al-25wt.%Si and Al-75wt.%Fe were mixed with pure aluminum ingots to obtain the targeted alloys. A vertical type centrifugal separation apparatus was designed to push the unfrozen aluminum-rich melt outside the crucible leaving AlSiFe intermetallic compounds inside during rotation as shown schematically in Fig. 1. The pouring temperature was set at 100 °C above the liquidus temperature of each alloy. Before rotating the centrifugal apparatus, about 0.045 kg of alloy melt was left in the crucible to crystallize the primary intermetallic compound after being poured into the crucible. After the desired waiting time, the crucible was quickly rotated and cooled in ambient air. The waiting time after pouring the melt into the crucible was determined based on the measured

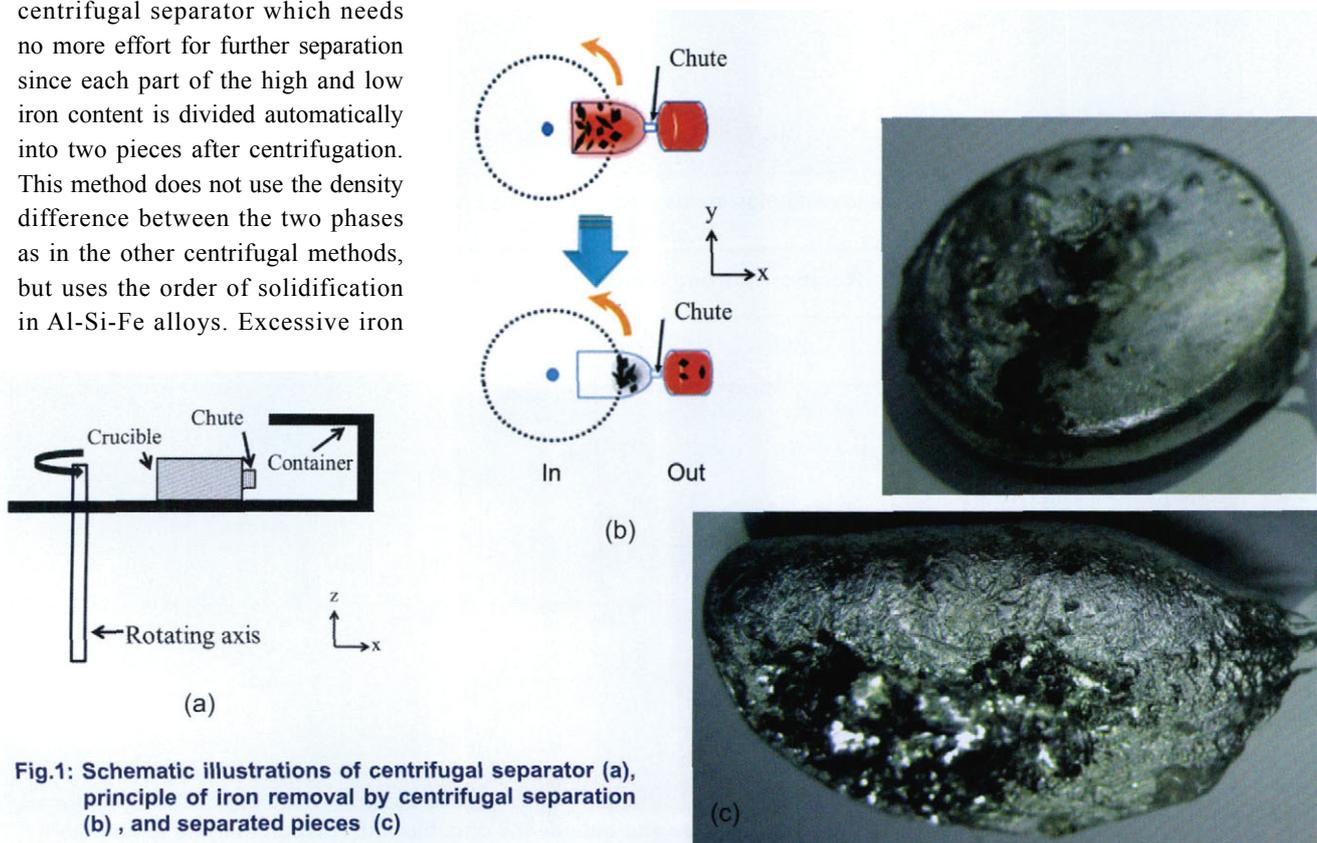


Fig.1: Schematic illustrations of centrifugal separator (a), principle of iron removal by centrifugal separation (b), and separated pieces (c)

cooling curves. The chute diameter of 6.5–8.5 mm and rotation starting temperature of 580–630 were chosen as experimental variables. The centrifugal acceleration was fixed at 40 g.

Microstructures were observed using an optical microscope. Quantification of the intermetallic compounds was made by analyzing optical micrographs of the alloys. Some of the specimens were also studied using SEM-EDS to obtain more information on their phases. The iron content was analyzed using an XRF spectrometer.

2 Results and discussion

2.1 Microstructure

The equilibrium fractions of phases of the Al-12Si-1.7Fe alloy (1.7 Fe alloy) and Al-12Si-3.4Fe alloy (3.4 Fe alloy) with temperature were calculated using the software JMatPro. AlSiFe compounds are the primary phase during solidification of Al-Si-Fe alloys [18].

The Al-Fe-Si ternary eutectic composition occurs at about 0.8wt.% Fe [5]. The solidification ranges are at 699 – 577 for 1.7 Fe alloy and 651 – 577 for 3.4 Fe alloy, respectively. The calculated fractions of the primary phase at the rotation starting temperatures of 623, 614, 606 and 597 are 5.8, 6.9, 7.5, and 8.3%, respectively, for the 3.4 Fe alloy. Those at the rotation starting temperatures of 587, 585, 583 and 580 are 2.5, 2.7, 2.8 and 3%, respectively, for the 1.7Fe alloy.

Figures 2 and 3 show the microstructures of samples inside and outside the crucible with rotation starting temperature for 3.4 Fe and 1.7 Fe alloy samples. Coarse intermetallic compounds are found in the sample inside the crucible, while rather fine intermetallic compounds are found in the sample outside the crucible. Although the density of AlSiFe compound is higher by 1.5 times than that of the aluminum alloy melt, these compounds were not moved outside but remained inside. This is different from the other centrifugal separation methods [7]. Primary intermetallic compounds are linked to each other

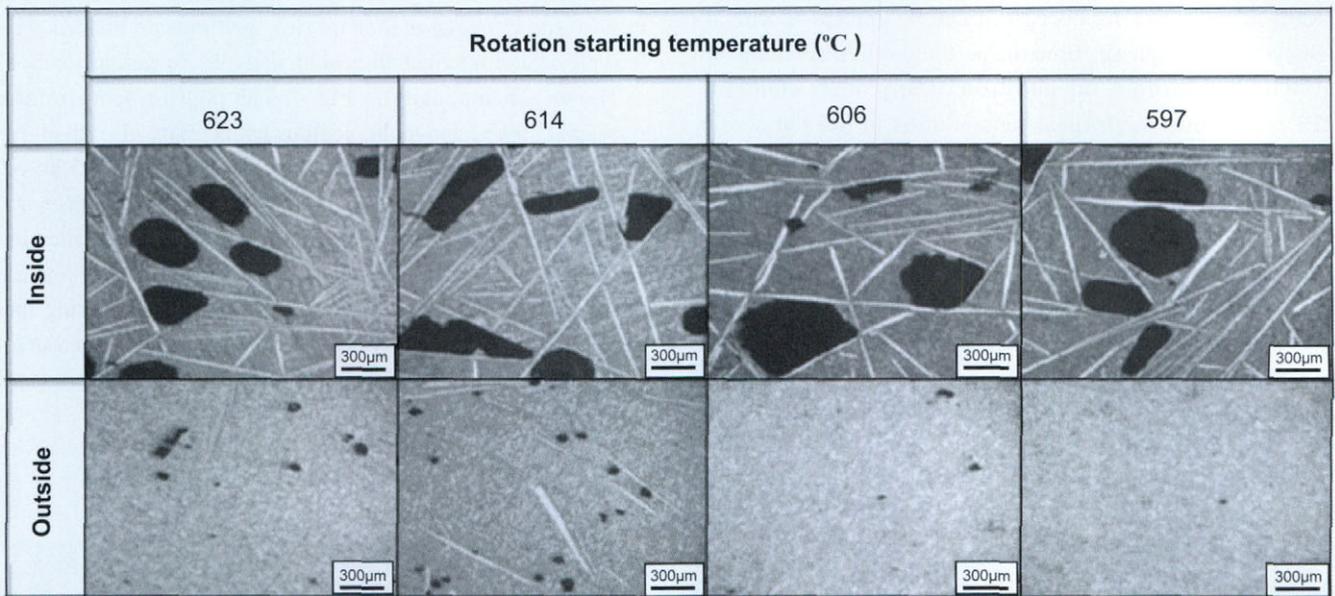


Fig. 2: Microstructures of Al-12Si-3.4Fe alloy samples inside and outside the crucible with rotation starting temperature

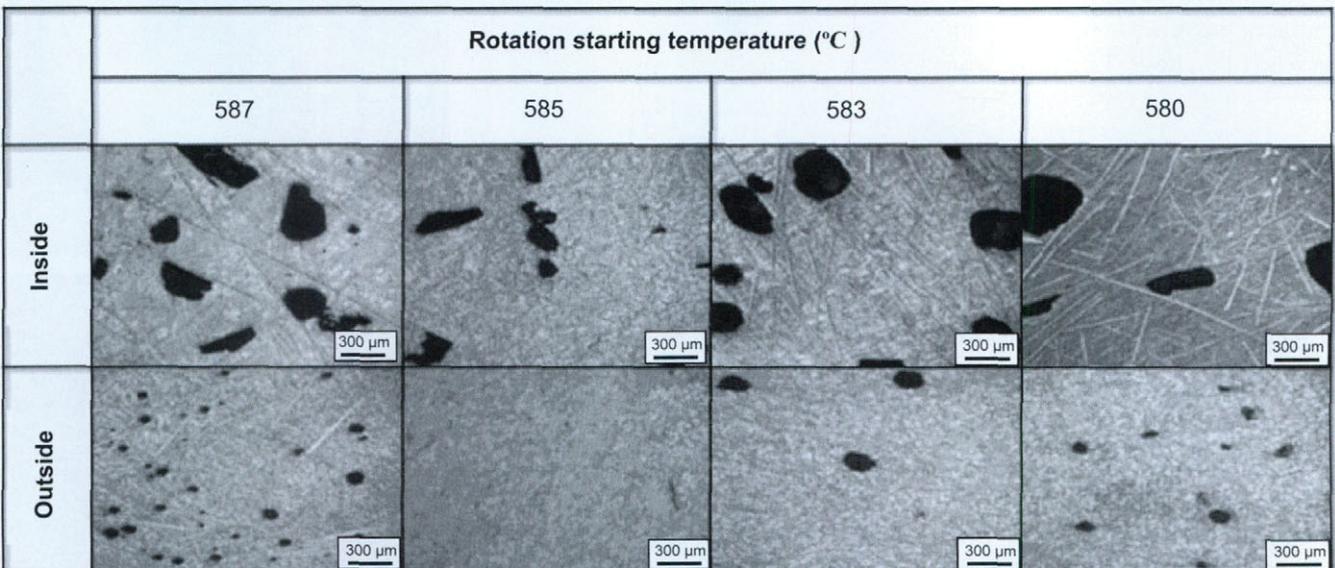


Fig. 3: Microstructures of Al-12Si-1.7Fe alloy samples inside and outside the crucible with rotation starting temperature

via aluminum-rich matrix and formed like a network. So it seems that they cannot escape the chute hole which connects the crucible with the outer container. The black parts shown in Figs. 2 and 3 are large pores, in which the aluminum rich phase existed at the initial stage of solidification. And this aluminum rich phase was pushed outside by centrifugal force.

Figure 4 shows the mapping images of the intermetallic compound and matrix. The intermetallic compounds in the 3.4 Fe sample are much larger than those in the 1.7 Fe sample. The average length and spacing are 580 and 280 μm , respectively, for the 3.4 Fe sample. The formation of $\beta\text{-Al}_5\text{FeSi}$ phase can be suppressed in two ways: by addition of sufficient manganese and, in alloys without manganese, by high cooling rates [19]. However the $\beta\text{-Al}_5\text{FeSi}$ phase is easily formed, since the manganese content is not high and the cooling rate is slow in this study. Dinnis et al [20] reported that high iron levels result in the formation of an Al-Si- Al_5FeSi ternary eutectic, instead of the Al-Si binary eutectic; and the large platelets form prior to the ternary eutectic, while the smaller platelets probably form as part of the ternary eutectic. The platelets form a complex and interconnected network in an Al-9%Si-1%Fe alloy.

Much iron remains in the type of AlSiFe compound inside the crucible and it shows that the iron content can be lowered by using the order of solidification, which is expected. This was also proven by the iron content analysis of the samples by the XRF spectrometer. The hardness of the AlFeSi compound was 525 HV, ten times greater than that of the matrix.

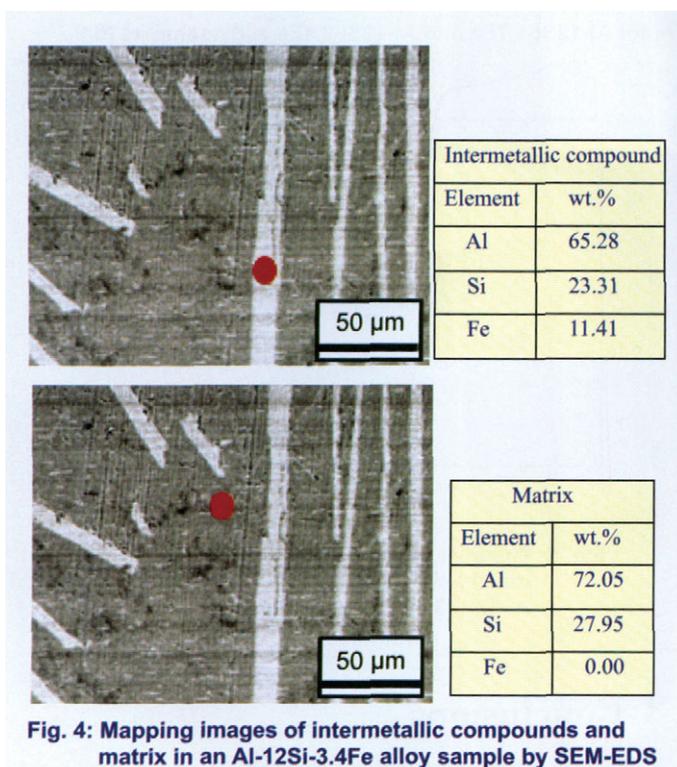


Fig. 4: Mapping images of intermetallic compounds and matrix in an Al-12Si-3.4Fe alloy sample by SEM-EDS

2.2 Yield and iron removal fraction

Figure 5 shows the yield plotted against the rotation starting temperature with iron content. Yield is the ratio of outside

weight to total poured weight expressed as a percentage. Since most of the alloy melt went outside the crucible at high rotation starting temperature (over 585 $^{\circ}\text{C}$) for the 1.7 Fe alloy, separation was not successful. Also most of the alloy melt froze inside the crucible at lower rotation starting temperature, since the solidification proceeded too fast. There was no clear relationship between the yield and the rotation starting temperature for the 1.7 Fe alloy. Since the freezing range was not so wide, precise control of the solid fraction with temperature was difficult. Therefore at high rotation starting temperature (over 585 $^{\circ}\text{C}$), the yield was high, while it decreased rapidly at low rotation starting temperature irrespective of the chute diameter. For the 1.7 Fe sample, the amount of intermetallic compounds was not enough to make an intermetallic compound network. In some cases, needle-like compounds went out, together with the aluminum-rich melt, through the chute during the initial stage of centrifugation. For the 3.4 Fe sample, the yield decreased with a decrease in rotation starting temperature. It is thought that the high solid fraction at low starting temperature prevents the melt from going outside. As solidification proceeded, the network of compounds became stronger and the viscosity increased very rapidly closing to its solidification end. On the other hand, yields were much higher than the solid fractions of primary phase at rotation starting temperatures for all samples, because the samples that remained inside the crucible consisted of primary intermetallic compound and aluminum-rich phase.

Figure 6 shows the removal fraction of iron plotted against the rotation starting temperature. Removal fraction is the ratio of initial content of iron to its final content after centrifugation expressed as a percentage. For the 1.7 Fe sample, the removal fraction is low. It seems that much needle-like intermetallic compound was pushed out due to their weak connection. However it was increased at slightly lower rotation starting temperature. For the 3.4 Fe sample, the removal fraction increases with a decrease in rotation starting temperature. It means that more iron was remaining inside the crucible, since the network of intermetallic compound was strong. The average removal fraction of iron is 47.1% and 54.1% for the 1.7 Fe and 3.4 Fe samples, respectively. The highest iron removal fraction is 65%, and the lowest one is 7% for Al-12Si-1.7Fe alloy. And they are 82% and 18% respectively for Al-12Si-3.4 Fe alloy. Previously reported iron removal fractions for various aluminum alloys by various methods were in the range of 35% to 93% [15].

Figure 7 shows the removal fraction of iron plotted against the yield. The removal fraction of iron is inversely proportional to the yield. Yields of the 1.7 Fe samples are higher than those of the 3.4 Fe samples due to the weak connection of the intermetallic compounds. However the removal fraction of the 1.7 Fe sample is lower than that of the 3.4 Fe sample.

The rotation starting temperature must be the most important process parameter; however it may depend strongly on the size of the crucible. So the rotation starting temperature was converted into the initial solid fraction in order to generalize it. The best efficiency in iron removal as a function of the initial

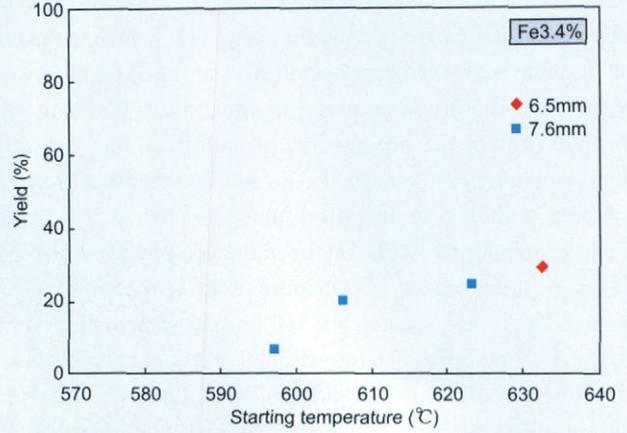
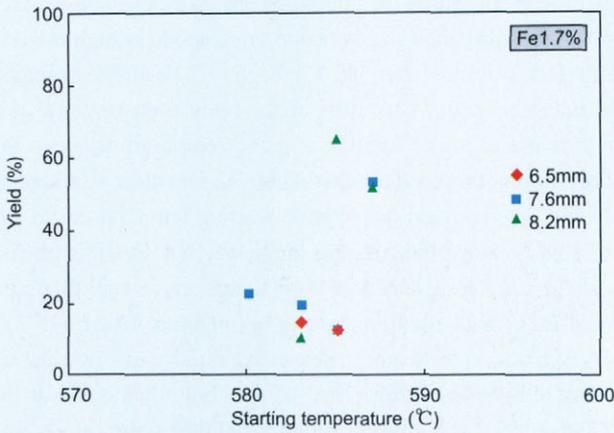


Fig. 5: Yield vs. rotation starting temperature for Al-12Si-1.7Fe and Al-12Si-3.4Fe alloy samples (6.5, 7.6 and 8.2 mm denote the chute diameter)

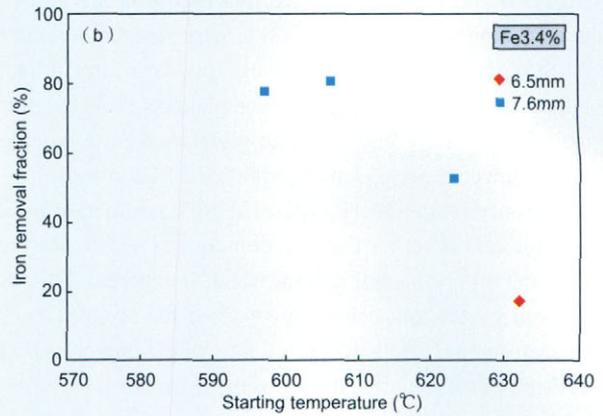
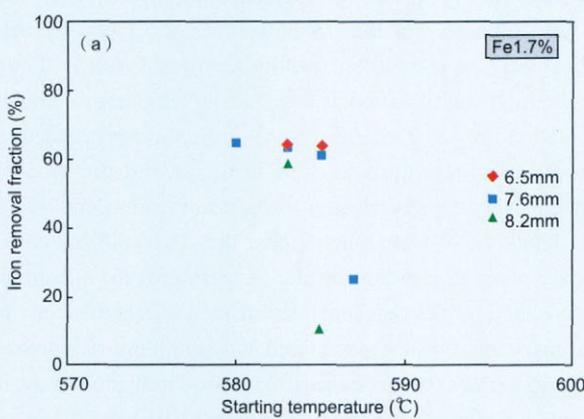


Fig. 6: Iron removal fraction vs. rotation starting temperature for Al-12Si-1.7Fe and Al-12Si-3.4Fe alloy samples (6.5, 7.6 and 8.2 mm denote the chute diameter)

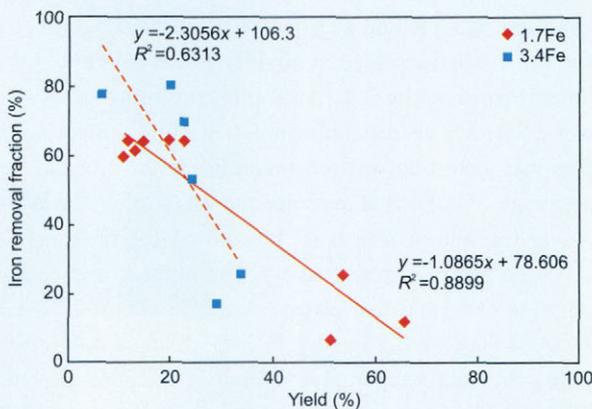


Fig. 7: Iron removal fraction vs. yield for Al-12Si-1.7Fe and Al-12Si-3.4Fe alloy samples

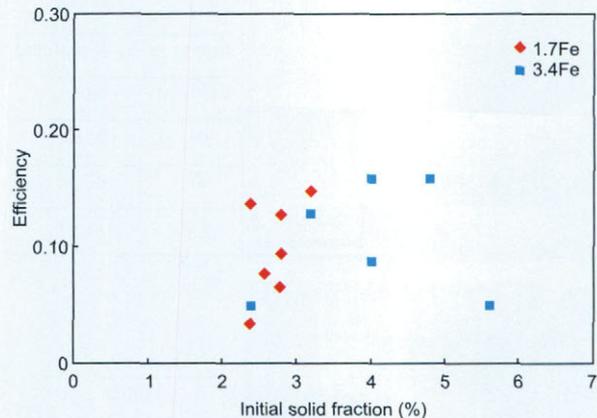


Fig. 8: The best efficiency of iron removal vs. initial solid fraction for Al-12Si-1.7Fe and Al-12Si-3.4Fe alloy samples

solid fraction is plotted in Fig. 8. The efficiency is evaluated by the product of removal fraction and yield. Higher removal fraction and higher yield are preferred in this process. Efficiency in removal of iron is increased with an increase in the initial solid fraction, but has a peak value. It shows the highest value of 0.16 when the initial solid fraction is 5% for the 3.4 Fe sample. The efficiency drops again at higher initial solid fraction because the removal fraction is high but the yield is too low.

3 Conclusions

A novel centrifuging method is introduced to separate iron-rich intermetallic compounds during solidification. This method does not use only the density difference between two phases as in the other centrifugal methods, but also uses the order of solidification in Al-Si-Fe alloys. An attractive feature of this method is that it needs no more effort to separate further since

each part of high and low iron content is divided automatically into two places after centrifugation.

Coarse intermetallic compounds were found in the sample inside the crucible, while rather fine intermetallic compounds were found in the sample outside the crucible. Primary intermetallic compounds were linked to each other via aluminum-rich matrix and formed like a network. Much iron remained in the type of β -Al₃FeSi compound inside the crucible and it showed that the iron content could be lowered using the order of solidification.

Iron-rich phases could be efficiently removed by centrifuging under the centrifugal acceleration of 40 g. The highest iron removal fraction is 65%, and the lowest one is 7% for the Al-12Si-1.7Fe alloy. And they are 82% and 18% respectively for the Al-12Si-3.4Fe alloy. The highest efficiency in the iron removal is 0.16 for the Al-12Si-3.4Fe sample when the initial solid fraction is 5%.

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