Effect of intensive melt shearing on the formation of Fe-containing intermetallics in LM24 Al-alloy

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Effect of intensive melt shearing on the formation of Fe-containing intermetallics in LM24 Al-alloy

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Abstract. Fe is one of the inevitable and detrimental impurities in aluminium alloys that degrade the mechanical performance of castings. In the present work, intensive melt shearing has been demonstrated to modify the morphology of Fe-containing intermetallic compounds by promoting the formation of compact α-Al(Fe,Mn)Si at the expense of needle-shaped β-AlFeSi, leading to an improved mechanical properties of LM24 alloy processed by MC-HPDC process. The promotion of the formation of α-Al(Fe, Mn)Si phase is resulted from the enhanced nucleation on the well dispersed $MgAl_2O_4$ particles in the melt. The Fe tolerance of LM24 alloy can be effectively improved by combining Mn alloying and intensive melt shearing.

1. Introduction

In diecast aluminium alloys, although a small amount of Fe is usually beneficial for preventing the die sticking during casting, it is often recognized as an impurity because of the significantly detrimental to their mechanical properties [1,2]. In fact, Fe is easily picked up during the melting and casting process because of the inevitable use of steel tools and scrap materials. An excess of Fe is thus a major problem in industrial application, especially for the use of recycled alloys. Therefore, efforts to understand Fe in aluminium alloys and develop an economical approach to diminish its detrimental effects are very significant in industry.

The detrimental effects of Fe to aluminium alloys are mainly due to its low equilibrium solubility in aluminium and the formation of Fe-containing intermetallic compounds. Depending on the content of silicon, iron, and manganese in aluminium alloys, there are two types of Fe-containing intermetallics formed during solidification as a primary phase in aluminium alloys, one is α-Al(Fe, Mn)Si with a bcc structure, the other is β-AlFeSi with monoclinic structure of up to 0.5at.% Mn. α-Al(Fe, Mn)Si is less harmful for the mechanical properties of aluminium alloys, in comparison with β-AlFeSi, due to the different crystal structures [3]. Traditionally, an approach to reduce the harmful effect of Fe is to restrict its content in the alloy to avoid the formation of primary Fe-containing compounds and/or to modify the crystal structures of Fe-containing compounds by adding elements to the alloys [4].

It has been the subject of extensive studies to mitigate the negative effect of Fe impurity in aluminium alloys by addition of elements [5,6] that include manganese, chromium, cobalt, beryllium, and molybdenum [7,8]. Of these elements, manganese is the most effective one and extensively utilized. Barlock and Mondolfo [9] established a quaternary section running through Al_3Fe, Al_6Mn and Si and found that the α-Al(Fe, Mn)Si phase forms via continuous substitution of Mn with Fe in the α-AlMnSi lattice, and the Mn contents in α-Al(Fe, Mn)Si and β-AlFeSi are rather limited. Mondolfo [10] further confirmed that α-Al(Fe, Mn)Si exhibits the same crystal structure as $Al_{15}Mn_3Si_2$ and an equilibrium quaternary phase, $Al_{15}(Fe,Mn)_3Si_2$ could stabilize the ternary $Al_{15}Mn_3Si_2$ compound. It has been found that the two types of intermetallics are unlikely to form a continuous solid solution phase in between because of their different structures [11]. Recently, Shabestari [12] found the effect of Mn/Fe ratio on the primary intermetallic phases in an Al-12.7wt.%Si alloy with Fe and Mn levels up to 1.2wt.% and 0.5wt.%, respectively. Crepeau [13] and Couture [14] have found that Mn/Fe≤0.5 in weight percentage provides best results in casting. Therefore, it is generally believed that the cubic α-Al(Fe, Mn)Si solidifies into compact morphologies due to its high-symmetric crystal structure, which can significantly reduce the detrimental effect of Fe on the mechanical properties [11,12].

Although the works of Barlock and Mondolfo have been widely used to explain Mn effect, there are practically no commercial Al-alloys falling within the field of their Al_3Fe-Al_6Mn-Si pseudo-section. Meanwhile, the mixture of primary α-Al(Fe, Mn)Si usually presents in the form of agglomerates and β-AlFeSi exhibits large needle shapes. It is logically anticipated that the detrimental effect will be minimised if the needle morphology can be altered and the agglomerates can be dispersed uniformly into the melt, when Fe is not avoidable in the alloys. Furthermore, the addition of corrector elements such as Mn and/or Cr should be as low as practically possible for the sustainable management of materials recycling, the inevitable Fe during recycling will lead to a continual build-up of transition metals (Fe, Mn, Cr etc), hence increase the quantities of intermetallic phases. It is therefore desirable that the detrimental effects of Fe can be minimised or removed with minimal addition of corrector elements by combining alloy chemistry with an approach that can maximize the effect of the alloy chemistry.

Recently, a melt conditioning technique using integrated high pressure die casting process (MC-HPDC) has been developed at Brunel University and has been used for the production of high quality cast components at laboratory scale [6]. In the MC-HPDC process, intensive shearing provided by a specially designed twin screw mechanism is directly imposed on the melt prior to die-filling. The sheared melt is in such a state that its uniformity in chemistry and temperature is significantly improved before casting by the conventional HPDC process. The experimental results carried out by the authors have demonstrated that MC-HPDC process can offer unique solidification behaviour and form uniform and fine microstructure [15,16]. It would be desirable to explore the effect of melt shearing on the morphologies of Fe-containing intermetallic compounds. Therefore, the current work attempts to study the effect of melt shearing on the morphology and distribution of the Fe-containing compounds in the castings made by the widely used commercial LM24 (Al-Si-Cu based) cast alloy.

2. Experimental

The composition of the LM24 alloy used in this investigation is shown in Table 1, which was analysed using optical mass spectroscopy.

Table 1. Alloy composition used in the experiments

Element	Si	Fe	Mn	Cu	Mg	Zn	Ni	Al
wt. %	7.76	1.08	0.27	3.21	0.11	2.28	0.03	balance

A standard TP-1 test was used to evaluate the formation of Fe-containing intermetallics in the LM24 alloy under a well controlled cooling rate. The details of the TP-1 test can be found in ref. [17]. During the experiments, the ingot was melted within a graphite crucible up to 750°C for about 30 minutes. The melt was sheared at 645°C for 60s before being discharged into the TP-1 mould that was immediately cooled down with the controlled cooling rate. The casting was cut at a section of 38mm from the bottom for microstructural examination. In comparison, the melt without shearing was naturally cooled to 645°C and then immediately cast into the TP-1 mould under the same condition.

In order to investigate the intermetallics more effectively and efficiently in the alloy, a pressure filtration technique was used to make a casting with concentrated Fe-containing intermetallic phase formed during solidification. The pressure filtration technique can also offer a relatively low cooling rate due to the specially made crucible by high insulating fibrous material [18]. In the experiments for the melt without shearing, 1500g melt was introduced into the filtration crucible at 750°C and allowed its temperature to lower to 645°C before starting the filtration process at a pressure of 16psi on the top of the crucible. The melt was weighted during filtration and the process was terminated when the remaining weight was at a level of 400g, implying that the melt was concentrated by 3.75times. In the experiments for the melt shearing, the same amount of melt was intensively sheared at 645ºC for 60s and immediately discharged into the crucible for filtration under the same conditions.

To investigate the mechanical properties of the alloy processed by MC-HPDC, the melt was sheared at 645°C for 60s and then discharged into the shot sleeve of a 280-tonne cold chamber HPDC machine. 4pcs of standard tensile test specimens with ϕ=6.4mm in gauge diameter were made by one shot during the processing. In comparison with the result without shearing, the melt was naturally cooled to 645°C and then cast with the HPDC process with the die temperature and casting parameters the same as that for the MC-HPDC process.

Microstructures of the samples were observed at the middle of the tensile test bar. Scanning electron microscopy (SEM) was carried out with a field emission gun Zeiss Supera 35 instrument, equipped with energy dispersive spectroscopy (EDS) facilities and operated at an accelerating voltage of 15-20 kV. The mechanical properties were measured at room temperature by an Instron 5569 at a crosshead speed of 1mm/min (strain rate: 0.66×10^{-3}/s). At least 8 samples were tested each time and the average was taken as the properties.

3. Results

The typical microstructures of LM24 alloy in TP-1 samples are shown in figure 1 for the solidification of the melt without shearing and for the solidification of the melt with the intensive shearing at 645°C for 60s. The α-Al solid solution phase exhibited a typical dendritic morphology. There was no significant change in the morphologies of α-Al and eutectic structures for alloys in the two processes. The interdendritic regions are characterised with a eutectic-like microstructure. The eutectic structures are made of typical Al/Si eutectic phases and transition-metal-bearing intermetallic compounds. Intermetallic phases could be easily identified by EDX quantification, due to their significantly different compositions. Obviously, there was little intermetallic compound present in the LM24 alloy that contained 1.08wt.%Fe. In the casting made by the melt without shearing, a small amount of intermetallic compounds were in needle shape, as shown in figure 1a. There were very few intermetallics in the shape of tiny particles. However, when the melt was sheared intensively before casting, the intermetallic compounds exhibited equiaxed polyhedral particles and very rare needle-shaped intermetallics were found in the sample, as shown in figure 1b. EDX quantification has identified the needle-shaped intermetallic compounds as β-AlFeSi phase (18.7wt.%Si, 13.9wt.%Fe, 0.6wt.%Mn) and the compact and equiaxed intermetallic compounds as α-Al(Fe, Mn)Si phase (9.7wt.%Si, 12.1wt.%Fe, 3.2wt.%Mn). The primary α-Al(Fe, Mn)Si phase existed as particles when they were small and could develop into more complex morphologies with increasing sizes. This observation is consistent with the α-Al(Fe, Mn)Si morphologies reported in previous work [9, 11]. By analysing the morphological difference, it seems that the intensive shearing promotes the formation of α-Al(Fe, Mn)Si and suppresses the formation of β-AlFeSi in LM24 alloy. However, it needs more evidence to draw the conclusion, as too few intermetallics were observed in the LM24 alloy. In order to consolidate the observation, we conducted a concentrated sample using a pressure filtration technique.

In the pressure filtration technique, the melt is forced to flow through a ceramic filter at a given temperature, which was originally used to characterise the cleanness of melt. However, this technique has also been sucessfully used to concentrate intermetallics in magnesium alloys [19]. The results of the filtrated LM24 samples are shown in figure 2 for both the intensively sheared melt and the melt without shearing. For the microstructure of LM24 alloy made by the melt without shearing in figure 2a, both large dendritic α-Al(Fe, Mn)Si and needle-shaped β-AlFeSi were observed. However, in the microstructure of LM24 alloy made by the intensivly sheared melt in figure 2b, many polyhedral α-Al(Fe, Mn)Si particles and a little fine β-AlFeSi were found. One important observation is that more α-Al(Fe, Mn)Si were found in figure 2b than in figure 2a. This implies that the intensive melt shearing cannot only alter the intermetallics morphology and supress the formation of β-AlFeSi, but also increase the number of intermetallics in the microstrucutre. Therefore, the variation in microstructure will potentially lead to the improvement of mechanical properties of LM24 die casting.

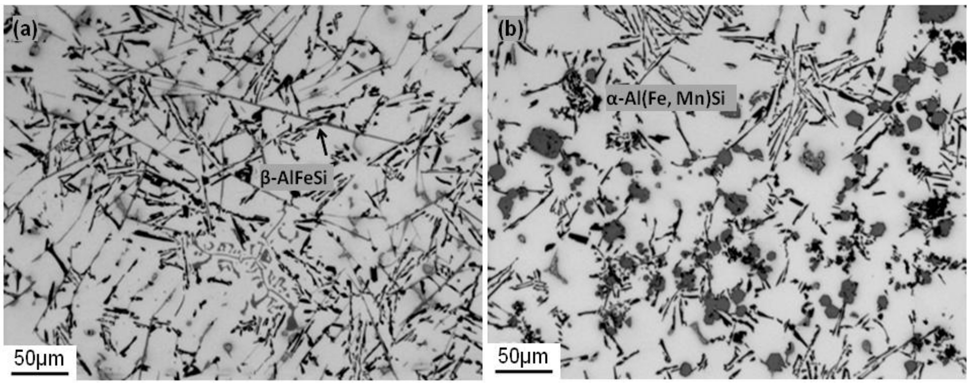

Figure 1. Optical micrographs showing the microstructure at the centre of the cross section of LM24 alloy cast at 645ºC in the TP-1 sample, (a) β-AlFeSi in conventional casting, and (b) compact α-Al(Fe, Mn)Si in the casting made by the intensively sheared melt.

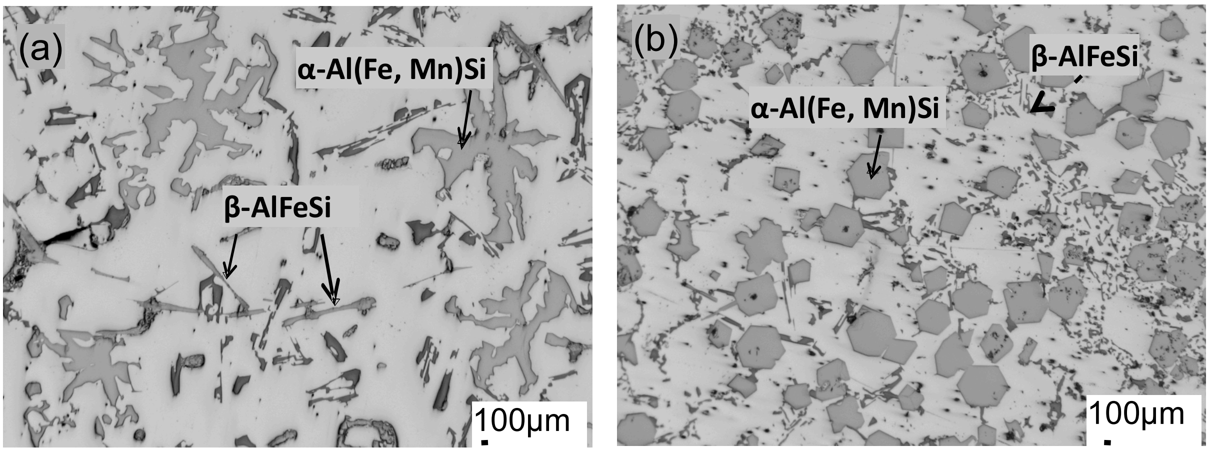

Figure 2. Optical micrographs showing the microstructure of a concentrated LM24 casting at a position close to the filter, (a) star-like α-Al(Fe, Mn)Si and needle-shaped β-AlFeSi in the sample cast at 645ºC with the melt without shearing, and (b) equiaxed polyhedral α-Al(Fe, Mn)Si and tiny β-AlFeSi in the sample cast with the melt intensively sheared at 645ºC for 60s.

The die casting was made under the same condition for both intensively sheared melt and the melt without shearing. Figures 3 and 4 show the resultant microstructure and mechanical properties of LM24 alloy, respectively. In the HPDC samples (figure 3a), both agglomeration of polyhedral α-Al(Fe, Mn)Si and needle-shaped β-AlFeSi are clearly shown in the microstructure. However, only individual dispersed polyhedral α-Al(Fe, Mn)Si particles are observed in the MC-HPDC sample (figure 3b). EDX quantification has identified the phase with the same composition as that in the TP-1 sample. The particle density of Fe intermetallics was at a level of 3pcs/mm^2 for HPDC sample, but it increased to 10pcs/mm^2 for the MC-HPDC sample, showing an apparent increase of particle density after intensive melt shearing. Figure 4 indicates that an improvement of the yield strength, the tensile strength and the elongation of LM 24 alloy can be achieved in the MC-HPDC sample.

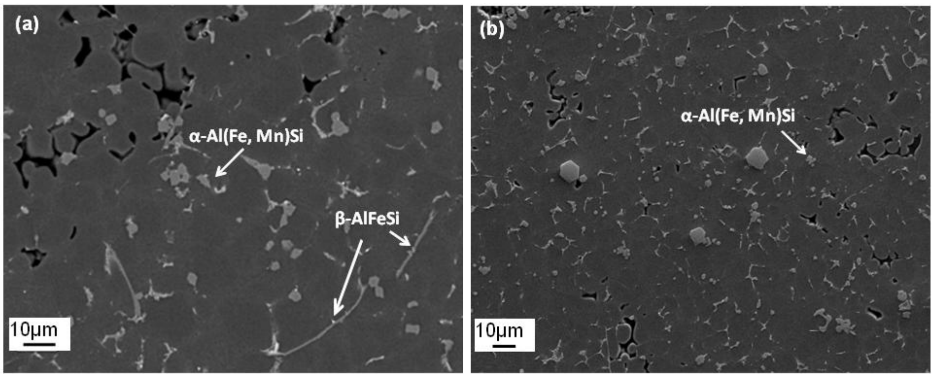

Figure 3. Backscattered SEM micrographs showing (a) the clusters of α-Al(Fe, Mn)Si and the needle-shaped β-AlFeSi in HPDC sample cast with the melt without shearing, and (b) the α-Al(Fe, Mn)Si in MC-HPDC sample cast with the melt sheared at 645ºC for 60s.

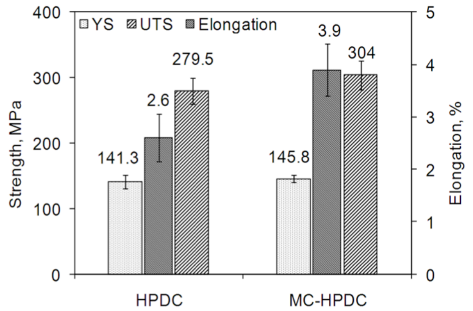

Figure 4. The comparison of the tensile strength and the elongation of LM24 casting made by conventional HPDC process and MC-HPDC process with a melt shearing at 645ºC for 60s.

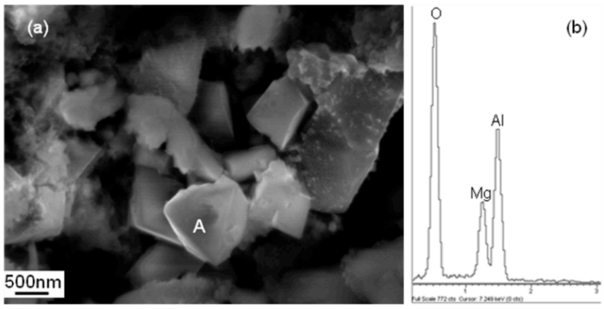

Figure 5. SEM micrograph showing (a) the clusters of $MgAl_2O_4$ spinel particles in the deep etched LM24 alloy sample solidified above the filter, (b) SEM-EDX spectrum of the particle A in (a).

4. Discussion

The formation of the α-Al(Fe, Mn)Si promoted by intensive melt shearing of the LM24 alloy can be explained by the enhanced heterogeneous nucleation of the α -Al(Fe, Mn)Si phase on the oxide particles dispersed by intensive shearing. Extensive observations have suggested the existence of an association between oxide particles and the Fe-containing intermetallic phase in Al-Si based alloys [20,21]. The authors have also observed the similar association and have confirmed that oxide agglomerates can be dispersed into fine individual particles and subsequently distributed uniformly into the melt under intensive shearing for both Mg- and Al-based alloys [15, 16]. In the present study, the oxides in the liquid LM24 have been identified to be $MgAl_2O_4$ spinel (figure 5). The lattice misfit between the $MgAl_2O_4$ and α-Al(Fe, Mn)Si phases is 4.85% at room temperature, calculated by matching the closest crystal planes and directions. The good lattice match indicates that $MgAl_2O_4$ spinel can be potential nucleation sites for the α-Al(Fe,Mn)Si phase. Since sufficient oxide particles are available in the melt after intensive shearing, the nucleation of the α-Al(Fe, Mn)Si phase can be enhanced, resulting in a fine microstructure with a better distribution of the intermetallic phase. This argument is also supported by the observation of significantly increased particle density of α-Al(Fe,Mn)Si phase in the casting made by the intensively sheared melt in both the pressure filtered TP-1 and MC-HPDC samples (figures 2 and 3).

The existence of agglomerations of α-Al(Fe, Mn)Si and β-AlFeSi are harmful to the mechanical properties of LM24 alloy due to the stress concentration and the resultant cracks for premature failure. However, with the casting processed by the intensive melt shearing in MC-HPDC process, the agglomeration of the primary Fe-containing intermetallic phases is minimised and they exist mainly as individual particles of the α-Al(Fe, Mn)Si phase that uniformly distribute throughout the entire cross section of the sample. Therefore, the stress concentration will be reduced and the resultant cracks for premature failure will be retarded, leading to an improvement in ductility and the strength of MC-HPDC sample (figure 4).

It is worth to emphasis that the microstructural examination has confirmed that the oxides usually exist in the form of films in aluminium melt and are detrimental on the mechanical properties of alloys because the oxide films initial cracks of fracture. However, the oxide films are broken up and separated into tiny agglomerates or individual particle and dispersed into the melt under intensive shearing. Therefore, the detrimental effect of oxides on the mechanical properties is significantly reduced. More recently, the experimental evidence has shown that the typical oxides may exist as heterogeneous nuclei for solidification and result in the formation of the refined microstructure in casting [15,19]. Consequently, on top of the modification of intermetallics, the shearing can improve the mechanical properties in several aspects. Although the mechanical properties of castings are affected by a number of factors in HPDC process, once other factors are fixed, the variations of the amount and the morphology of intermetallics become critical for the mechanical properties.

Conclusions

(1) The intensive melt shearing is effective in modifying morphologies of Fe-containing intermetallic compounds by promoting the formation of compact α-Al(Fe, Mn)Si compounds at the expense of monoclinic β-AlFeSi phase.
(2) The intensive melt shearing can improve the mechanical properties of LM24 alloy cast with MC-HPDC process.
(3) The promotion of the formation of α-Al(Fe, Mn)Si phase is probably resulted from the enhanced nucleation on the well dispersed $MgAl_2O_4$ particles in the melt of LM24 alloy.
(4) The Fe tolerance of LM24 alloy can be effectively improved by combining Mn alloying and melt shearing. The application of MC-HPDC process is expected to be an effective way in combating the detrimental effect of Fe impurity and significantly beneficial in recycling of aluminium alloys.

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