On grain refinement and titanium segregation in Al-Si alloy

*MI Guo-fa¹, WANG Kuang-fei¹, WEN Tao¹, ZHANG Ming², WANG Hong-wei², ZENG Song-yan²

(¹School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo 454000, China; ²School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China)

Al-Si alloys exhibit highly desirable combined properties, such as excellent castability, high specific strength and toughness, outstanding fatigue resistance and corrosion resistance. As a result, Al-Si alloys are widely used in automotive, aviation and military industries. The tensile strength of Al-Si alloys can reach 320 MPa and elongation 5%. But the tensile strength of Al-Si alloys produced by automotive industry or aviation industry in China can only achieve 300 MPa and elongation about 3%, which cannot meet the demand of the automotive and aviation industries. Many problems need to be solved to improve the properties of the widely-used A357 alloy, starting with an improved processing prior to casting of the melt of this alloy. The effect of melt processing is improved with an increase in holding time after adding the master alloy into Al-Si alloy. Initial studies show that the density of TiAl₃ is relatively high (about 3.37 g/cm³) when mass fraction of Ti is between 0.18%-0.23%. The strong trend of localized aggregation and deposition of Ti in the casting and melting process lead to a decrease in Ti concentration in castings. In addition, gravity segregation of Ti in ingot during the process of cooling has an adverse effect on grain refinement. Therefore, it is important to study the segregation of Ti and determine effective methods to control segregation. The effect of Ti on grain refinement of Al-Si alloy was studied by analyzing the segregation of Ti and some solutions were proposed.

1 Experiment

The alloy was prepared using high purity aluminum (> 99.99%), purity Mg, Al-12%Si master alloy and Al-3.8%Be master alloy, with A357 composition as shown in Table 1. The refinement of the alloys samples was conducted using a Φ 10 Al-5Ti-1B rod refiner produced by LSM Corporation (UK). The crucible was made from silica sand with 20 mass% of silica sol and formed in seamless thin-walled steel tube (Φ 90 mm × 250 mm). The effect of Al-5Ti-1B addition and holding time on Ti segregation was studied by using EDS at different positions of A357 samples at the same temperature. Four groups of samples were poured from the crucible and then put into an electrical resistance furnace for holding at 720°C. The samples were removed after 2 h, 4 h, 8 h and 16 h, respectively. Four samples were chosen in every group for measuring their compositions. Ti segregation was studied with different holding times and different positions of the sample, and with the addition of master alloy at constant melt temperature. Samples were etched by 20% NaOH after polishing and the microstructure was observed by using optical microscopy.

Table 1 Chemical composition of experimental alloy

<table>
<thead>
<tr>
<th>Element</th>
<th>Si</th>
<th>Mg</th>
<th>Be</th>
<th>Ti</th>
<th>Fe</th>
<th>Others</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>wt%</td>
<td>6.5</td>
<td>0.6</td>
<td>0.07</td>
<td>0.4</td>
<td>&lt;0.5</td>
<td>&lt;0.15</td>
<td>Balance</td>
</tr>
</tbody>
</table>

2 Results and discussion

2.1 Existent form of Ti in Al-5Ti-1B master alloy

Figure 1 shows the XRD pattern of Al-5Ti-1B master alloy. Al-5Ti-1B master alloy is constitutive of Al, TiB₂ (AlB₂), TiAl₃, according to Fig.1. The microstructure of Al-5Ti-1B master alloy is revealed by SEM morphology in Fig.2. Both coarse block phase and disperse granular phase exist in the master alloy. In addition, the disperse granular phase can be seen in a coarse block phase. The dimension of the block phase is 10 – 50 µm and 0.3 – 3 µm for disperse granular phase. Initial research indicates that the dispersed granular phase is TiB₂ with columnar morphology and
enriched Ti was found around TiB₂ by EDS analysis.

The process of refinement of Al-Si alloy is equivalent to the process in that phases dissolved in the melt. At common smelting temperature, TiB₂ exists in the melt mainly in the form of fine particles because of its high melting point. According to Fig.3, TiAl₃ can exist in stable form when the melt temperature is 723°C and mass fraction of Ti is 0.3%.

A graphite-like plane structure was formed because of the heavy interaction between B atom and B atom in TiB₂ crystal. This plane structure supports a proper plane substrate for nucleation of TiAl₃. In addition, the wetting angle between TiB₂ and TiAl₃ is relatively small; hence, low nucleating energy of TiAl₃ leads to heterogeneous nucleating between TiB₂ and TiAl₃. Many heterogeneous nuclei appeared because of the excellent lattice matching between α-Al and TiAl₃. All of the factors mentioned above lead to the refinement of aluminum alloy [3]. Therefore, the existent form of Ti is mainly TiB₂, TiAl₃ and their complexes after the master alloy dissolves into the melt.

2.2 Analysis of grain size and composition of Ti after melt processing

The relationship between grain size and position (the top marked “1” and the bottom marked “4”, four positions were chosen and the distance interval is 20 mm) of sample refined by Al-5Ti-1B is shown in Fig.4. Samples were prepared after holding for 2 h, 4 h, 8 h and 16 h, respectively. According to Fig.4, the grain is very fine at the bottom of sample. With increasing distance from the bottom, grain grows obviously for most samples. However, in the sample after 2 h heat preservation, the grain refining trend is not the same and the grain is finer at position 2 than position 3. It is not clear how this anomaly occurs. The refining trend is most obvious after 4 h holding and grain size is 1.0 mm at position 1, 0.25 mm at position 4. The refining ratio is 75%. Although the refining effect of others is not as high as those after 4 h heat preservation, their refining ratios also reach 65%.

The process of refinement of Al-Si alloy is equivalent to the process in that phases dissolved in the melt. At common smelting temperature, TiB₂ exists in the melt mainly in the form of fine particles because of its high melting point. According to Fig.3, TiAl₃ can exist in stable form when the melt temperature is 723°C and mass fraction of Ti is 0.3%.

A graphite-like plane structure was formed because of the heavy interaction between B atom and B atom in TiB₂ crystal. This plane structure supports a proper plane substrate for nucleation of TiAl₃. In addition, the wetting angle between TiB₂ and TiAl₃ is relatively small; hence, low nucleating energy of TiAl₃ leads to heterogeneous nucleating between TiB₂ and TiAl₃. Many heterogeneous nuclei appeared because of the excellent lattice matching between α-Al and TiAl₃. All of the factors mentioned above lead to the refinement of aluminum alloy [3]. Therefore, the existent form of Ti is mainly TiB₂, TiAl₃ and their complexes after the master alloy dissolves into the melt.

2.2 Analysis of grain size and composition of Ti after melt processing

The relationship between grain size and position (the top marked “1” and the bottom marked “4”, four positions were chosen and the distance interval is 20 mm) of sample refined by Al-5Ti-1B is shown in Fig.4. Samples were prepared after holding for 2 h, 4 h, 8 h and 16 h, respectively. According to Fig.4, the grain is very fine at the bottom of sample. With increasing distance from the bottom, grain grows obviously for most samples. However, in the sample after 2 h heat preservation, the grain refining trend is not the same and the grain is finer at position 2 than position 3. It is not clear how this anomaly occurs. The refining trend is most obvious after 4 h holding and grain size is 1.0 mm at position 1, 0.25 mm at position 4. The refining ratio is 75%. Although the refining effect of others is not as high as those after 4 h heat preservation, their refining ratios also reach 65%.

Macrostructures of samples are shown in Fig.5. There is significant refining effect at the bottom of the sample, but this effect was seldom observed at the top. The refining of grain progresses from top to bottom. According to Fig. 5a, grain at the top is quite coarse and its size is about 2 mm, but at the bottom, grain is so fine that its size decreases to about 0.2 mm (Fig. 5d).
2.3 Analysis of Ti segregation

With the increasing distance from the top, the content of Ti increased markedly according to Fig.6. Some complexes are going to deposit after a long holding time. Because of the high density of TiAl3 formed on TiB2, a large number of heterogeneous nuclei formed at the bottom and seldom at the top and this phenomenon causes a greater refining effect at the bottom than the top of the sample. Generally speaking, the grain refining effect is not effective because of the nonuniform Ti concentration and this phenomenon should be avoided in practical production. Ti segregation always appeared in the process of cooling or holding after handling by Al-5Ti-1B master alloy in Al-Si alloys. Therefore, the higher the amount of Ti (0.3% Ti is added in this experiment) in Al-Si alloys, the easier Ti segregation occurred at the bottom. The overall refining effect is strongly influenced by the deposit of complexes in the process of holding.

In addition, TiAl3 is not a stable phase if the content of Ti is less than 0.1% in the melt, thus it is very important to add proper quantities of Ti in the melt for the refinement of aluminum [4]. Many Ti compounds were deposited at the bottom due to gravity segregation in long holding times. If the holding time is too long, with the increase of Ti in bottom, nucleation begins to decline when Ti content is larger than the minimum Ti content in the forming of the TiAl3 layer. The reason for the decline is probably that with the thickening of the TiAl3 layer, the poor lattice matching between TiAl3 crystal plane and α-Al makes it more difficult for nucleating [5]. Therefore, melt processing time is also an important factor. This may be the reason that the refining effect after 4 h holding is better than others.

2.4 Methods of preventing Ti segregation

The grain refining effect is not favorable because of the nonuniform Ti concentration. This phenomenon should be avoided in practical production. If there is a large density variation between initial eutectic and melt in hypoeutectic and hypereutectic aluminum alloys, the initial eutectic will rise or fall at a low cooling rate. This may lead to composition variation in castings, namely, gravity segregation. Gravity segregation is a localized phenomenon caused by the variation of density in the melt. Gravity segregation is closely related to density variation, crystallizing component interval of phase diagram and temperature interval. With an increase in density variation, the crystallizing component interval is lengthened and density variation between primary crystal and residual liquid phase is also increased. Large crystallizing temperature interval or small cooling rate may lead to serious Ti segregation, because in this condition there is enough time for a rising or falling of primary crystals. Generally speaking, Ti segregation is mainly caused by gravity; thus, three ways to prevent Ti segregation are put forward. First, improving the cooling rate, so as not to allow sufficient time for initial precipitated phase’s rise or fall. Second, new elements can be added in the melt. These elements must have similar density with the melt and must be separated at the primary stage of solidification to prevent the rise or fall of initial phase. Third, convection or stirring are effective methods to prevent Ti segregation.

3 Conclusions

(1) The best refining effect of Al-5Ti-1B was achieved in A357 alloy after 4 h holding and the refining ratio reaches 75%.
(2) The distribution of Ti in castings is not uniform and Ti content in the bottom is higher than in the top. With increasing distance from the top, Ti segregation increases obviously and the content variation reaches 25.6% between top and bottom.
(3) The phenomenon of Ti segregation (gravity segregation) appeared in the process of holding and cooling of the melt after treating with Al-5Ti-1B master alloy.

References


The project is supported by the Innovation Fund for Outstanding Scholar of Henan province (No.0621000700).