Effects of Mg content on primary Mg$_2$Si phase in hypereutectic Al–Si alloys

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Abstract: Hypereutectic Al–Si alloy with variant Mg contents were fabricated by casting, and the effects of Mg content on the microstructure of primary Mg$_2$Si particles in hypereutectic Al–Si alloys were investigated. The results show that the volume fraction of primary Mg$_2$Si particles increases linearly with raising the Mg content, but the average size of Mg$_2$Si particles does not exhibit a corresponding change. When the Mg content is 3%, $\langle 1 0 0 \rangle$ directions have the fastest growth velocity, so that Mg$_2$Si particles are likely to form octahedron shape. When gradually increasing the Mg content, the distributions of Mg and Si atoms on the solid–liquid interface become inhomogeneous, which results in the formation of irregular octahedron structures. Finally, when the Mg content is about 10%, the morphology of primary Mg$_2$Si particles changes from the octahedron shape into various complex structures with a large size.

Key words: hypereutectic Al–Si alloys; primary Mg$_2$Si particles; growth pattern; growth velocity

1 Introduction

Hypereutectic Al–Si alloys have widespread applications in automotive, aerospace and general engineering industries due to their excellent properties, such as low thermal expansion coefficient, satisfied castability, good corrosion resistance and wearing resistance [1–3]. However, the primary Si particles in hypereutectic Al–Si alloys are usually coarse, which leads to poor mechanical properties [4]. Therefore, many efforts have been made to control primary Si particles in an ideal shape and distribution in alloys, for instance, adding rare earth elements into Al–Si alloys [5–7] or increasing the solidification rate [8].

In recent years, researchers have found that Mg plays a vital role in improving properties of hypereutectic Al–Si alloys. When a certain amount of Mg is added to hypereutectic Al–Si alloys, primary Mg$_2$Si phase would form [9]. Mg$_2$Si is an intermetallic compound with a low density (1.90 g/cm$^3$), a high melting point (1087 °C), a high hardness (4.5×10$^9$ N/m$^2$), a high elastic modulus (120 GPa) and a low coefficient of thermal expansion(7.5×10$^{-6}$ K$^{-1}$) [10]. It is obvious that Mg$_2$Si has excellent properties for hypereutectic Al–Si alloys. FROMMEYER et al [11] found that Mg$_2$Si–Al composites have a higher yield strength and elastic modulus than Al–Si composites. THIRUGNANAM et al [12] found that the strength and hardness of Al–Si–Mg alloys became higher when increasing the Mg content. Another study indicated that the distribution of eutectic Si became homogeneous as well by adding Mg in alloys [13]. For expanding the application of Mg$_2$Si–Al composites, it is valuable to understand what the changes are in the microstructure of hypereutectic Al–Si alloys and how primary Mg$_2$Si particles grow as the Mg content increases. However, few efforts have been made to research the effects of Mg content on the primary Mg$_2$Si phase.

The present investigation was carried out to research the effects of Mg content on the microstructure of primary Mg$_2$Si phase in hypereutectic Al–Si alloys. Furthermore, the relationship between Mg content and growth mechanisms of primary Mg$_2$Si was discussed.

2 Experimental

Commercial pure Al (99.7%), Si (98%), Cu (99.7%) and Mg (99%) were used as raw materials to prepare Al–16Si–xMg (x=1, 3, 6 and 10) alloys. The chemical
compositions of alloys are presented in Table 1. The raw materials were thoroughly melted by a 30 kW electrical resistance furnace to prepare the Al–16Si–xMg alloys.

The total mass of raw materials was 13 kg. Four groups of alloys with different chemical compositions were melted and cast, respectively. The mass of Al, Si and Mg was accurately calculated according to Table 1. The crucible and the furnace were firstly preheated to 300 °C for 1 h. Secondly, Si and Al were put into the crucible subsequently after the furnace was heated to 500 °C. Thirdly, the temperature of furnace was increased to 800 °C. When Si was completely melted, Cu, Mg and other metals were added into the melted liquid. The whole smelting process was accompanied with slight stirring. At last, the Al–Si–Mg melted liquid was poured into a preheated mould and the ingots were obtained. M1, M3, M6 and M10 were used to present the alloys with the Mg contents of 1%, 3%, 6% and 10% (mass fraction), respectively.

Table 1 Chemical compositions of alloys

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Mass fraction/%</th>
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<tr>
<td></td>
<td>Si</td>
</tr>
<tr>
<td>M1</td>
<td>16</td>
</tr>
<tr>
<td>M3</td>
<td>16</td>
</tr>
<tr>
<td>M6</td>
<td>16</td>
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<tr>
<td>M10</td>
<td>16</td>
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Four groups of metallographic specimens were taken out at the same position of ingots in the middle of ingots. The sodium hydroxide solution (15%, mass fraction) and the hydrofluoric acid aqueous solution (0.5%, mass fraction) were used as two kinds of etchant. The microstructures of the specimens were analyzed using a Nikon–L150 optical microscope and Tescan scanning electron microscope.

3 Results

3.1 Microstructures of Al–Si alloys with different Mg contents

Figure 1 shows the microstructures of M1, M3, M6 and M10. Only coarse primary Si particles and eutectic phase can be observed when the Mg content is 1%. As the Mg content rises to 3%, a small number of primary Mg2Si particles can be found. With the increase of Mg content, as shown in Figs. 1(c) and (d), there are an increasing number of Mg2Si particles and the average size of Mg2Si rises as well. Generally, primary Mg2Si can be found in the alloys after the reaction of Mg (the Mg content is at least 3%) with Si. When the Mg content is up to about 6%, coarse Si particles can hardly be found in the alloys.

The volume fraction of primary Mg2Si particles in the hypereutectic Al–Si alloys was measured by image processing software, and the detail is shown in Fig. 2. When the Mg contents are 1%, 3%, 6% and 10%, the volume fractions of primary Mg2Si particles are 0, 3.88%, 10.66% and 22%, respectively. It is obvious that the volume fraction of primary Mg2Si particles rises with raising the Mg content.

The average size of primary Mg2Si particles was measured by quantitative metallography method and is shown in Fig. 3. The average size of Mg2Si particles rises with increasing the Mg content. It changes from 30 to
Fig. 2 Volume fraction of primary Mg$_2$Si particles with different Mg contents

Fig. 3 Average size of primary Mg$_2$Si particles with different Mg contents

Fig. 4 Morphologies of primary Mg$_2$Si particles in alloys with different Mg contents: (a) 3%; (b) 6%; (c) 10%

32 μm as the Mg content increases from 3% to 6%. Once the Mg content rises to 10%, the average size reaches 95 μm.

In Figs. 2 and 3, it is worth noting that the volume fraction of primary Mg$_2$Si particles almost exhibits a linear increase with increasing the Mg content in alloys, but the size of Mg$_2$Si particles does not show the same growth tendency with the variation of Mg content. So, further study was carried out to explain this phenomenon.

3.2 Morphology of primary Mg$_2$Si particles with different Mg contents

Figures 4(a), (b) and (c) show the morphologies of primary Mg$_2$Si particles when the Mg content is 3%, 6% and 10%, respectively. It is clear from Figs. 4 (a) and (b) that most primary Mg$_2$Si particles exhibit polygon shape when the Mg contents are 3% and 6%, such as triangle, rectangle and trapezoid. However, with raising the Mg content to 10%, as shown in Fig. 4(c), the shape of primary Mg$_2$Si particles changes to Chinese character, polygon shapes with hole and so on.

Then, the three-dimensional examination was used to observe the morphology evolution of primary Mg$_2$Si particles, as shown in Fig. 5. When the Mg content is 3%, the primary Mg$_2$Si particles exhibit complete octahedron, as shown in Fig. 5(a). With increasing the Mg content to 6%, Mg$_2$Si particles show incomplete octahedron or octahedron structure with hoppers, as shown in Figs. 5(b) and (c). Once the Mg content reaches 10%, Mg$_2$Si particles tend to be various complex structures, as shown in Fig. 5(d). The evolution of the morphology of Mg$_2$Si particles indicates that the growth behavior of Mg$_2$Si particles may change when the Mg content increases from 3% to 10%.

4 Discussion

In this section, the effects of Mg content on the growth mechanism of primary Mg$_2$Si particles will be explained. The primary Mg$_2$Si particles change from the octahedron structure to various complex shapes with increasing the Mg content in Al–Si alloys. The growth
pattern of primary Mg$_2$Si may change as the Mg content increases.

4.1 Growth mechanism of octahedron Mg$_2$Si

Octahedron Mg$_2$Si is mainly observed in the Al–Si alloys with approximately 3% Mg content. It is well known that Mg$_2$Si crystal displays a face-centered cubic crystal structure, {1 0 0}, {1 1 0} and {1 1 1} are three vital planes. Compared with {1 1 0} and {1 1 1} planes, {1 0 0} planes have the lowest lattice density, which are easier for atoms to adhere to. So, it clearly suggests that {1 0 0} planes have the fastest growth velocity.

When some Mg is added into hypereutectic Al–Si alloys, Mg–Si atomic clusters can easily extend to the crystal nucleus for the structure fluctuation and the energy fluctuation in the melt. While the primary Mg$_2$Si particles grow to a certain size, they will lose stability. The primary arms emerge and rapidly grow along six $\langle1 0 0\rangle$ directions, corresponding to {1 0 0} planes mentioned above. Subsequently, the secondary arms grow up along some directions such as $\langle1 1 0\rangle$ and they will expand, linking with each other and finally become the edges of octahedron [14]. As the growth proceeds, there are many chances for atoms to pile up on {1 1 1} planes [15]. Finally, {1 0 0} planes disappear, the Mg$_2$Si crystal is covered by {1 1 1} planes and complete octahedron Mg$_2$Si forms. For a complete octahedron Mg$_2$Si particle, as shown in Fig. 6, its corners and edges grow up along $\langle1 0 0\rangle$ and $\langle1 1 0\rangle$ directions, respectively, while its planes are parallel to {1 1 1} planes.

4.2 Growth mechanism of incomplete-octahedron Mg$_2$Si and complex-structure Mg$_2$Si

According to the microstructure of M3 shown in Fig. 5(a), the morphology of Mg$_2$Si particle exhibits complete octahedron. This suggests that the growth velocity of {1 1 1} planes is properly matched with that of {1 0 0} planes. However, the microstructure of M6 reveals that Mg$_2$Si particles show incomplete octahedron and octahedron with hopper (Figs. 5(b) and (c)), which means that {1 1 0} edges and {1 1 1} planes may not grow completely due to the high growth velocity of {1 0 0} planes. When the Mg content increases, the growth pattern may change.
On the basis of the crystal growth theory [16], it is known that when the polyhedral crystal grows in facet-growth form, the solute concentration will not be uniform on the solid–liquid interface. And according to Ref. [17], as Mg$_2$Si particles grow, the distributions of Mg and Si atoms obviously become uneven on the solid–liquid interface. So, it is well founded that as the Mg content increases, the concentrations of Mg and Si atoms along some \langle 1\,0\,0 \rangle directions of Mg$_2$Si particles are high. The directions with high solute concentrations grow faster than other \langle 1\,0\,0 \rangle directions, which leads to the formation of incomplete octahedron. It is found that the growth velocity of \langle 0\,0\,0 \rangle directions is lower than that of other \{1\,1\,1\} planes. Thus, \{0\,0\,1\} and \{0\,1\,0\} planes are exposed on the final morphology. However, other \langle 1\,0\,0 \rangle directions exhibit complete corners, as shown in Fig. 7.

Based on the discussion above, when the Mg content reaches about 6%, some \langle 1\,0\,0 \rangle directions grow relatively fast, which is difficult for Mg and Si atoms to fill the full \{1\,1\,1\} planes. As a result, \{1\,1\,1\} planes cannot grow completely, thus, a hopper-structure is left.

When the Mg content reaches 10%, various complex Mg$_2$Si structures can be observed in the alloys. Figure 8 shows kinds of complex primary Mg$_2$Si particles. As the Mg content is approximately 10%, the distributions of Mg and Si atoms on the solid–liquid interface become more inhomogeneous [16]. Therefore, a
few preferential growth directions may grow so fast that even the \( \{110\} \) directions cannot link with each other to form octahedron skeleton, let alone Mg and Si atoms to fill the full \{1 1 1\} planes. In this case, the primary Mg\(_2\)Si particles tend to grow to enormous octahedron-skeleton structure, as shown in Fig. 8(a). Other Mg\(_2\)Si particles develop one main branch only along the preferential growth direction (mark I in Fig. 8(b)) and perpendicular to the stem, and several secondary arms grow up (mark II in Fig. 8(b)). From Fig. 8(c), it can be seen that a few Mg\(_2\)Si particles still develop in the octahedron-growth form. No \( \{100\} \) direction has a very high growth velocity. Nevertheless, some edges are not able to grow completely and some planes are not full. At last, Mg\(_2\)Si grows to irregular structure. In general, when the Mg content reaches 10\%, most Mg\(_2\)Si particles exhibit enormous and complex structures due to seriously inhomogeneous distribution of solutes.

It can be deduced from the above discussion that the morphology change of primary Mg\(_2\)Si particle can be attributed to the Mg content. The growth of primary Mg\(_2\)Si goes through several stages. Firstly, when the Mg content is approximately 3\%, \{1 0 0\} planes grow preferentially, meanwhile, the edges grow along \{1 1 0\} directions and the atoms pile up on the \{1 1 1\} planes, which results in the formation of octahedron structure. Then, as the Mg content reaches 6\%, the growth velocity of \{1 0 0\} planes is too fast for atoms to fill the full \{1 1 1\} planes, so some octahedron structures exhibit hopper morphology. As the Mg content continues to increase, the differences of growth velocity among three vital planes become greater, so the primary Mg\(_2\)Si particles tend to develop to various complex shapes.

In terms of improving the properties of hypereutectic Al–Si alloys, adding 6\% Mg may be an ideal choice. Because the primary Mg\(_2\)Si has a moderate size and amount at this Mg content. As the Mg content reaches 10\%, most primary Mg\(_2\)Si particles exhibit complex shape and large size, which needs to be avoided.

5 Conclusions

1) The addition of Mg leads to an increase in both size and amount of primary Mg\(_2\)Si particles. When the Mg contents are 1\%, 3\%, 6\% and 10\%, the volume fractions of primary Mg\(_2\)Si particles are 0, 3.88\%, 10.66\% and 22\%, respectively. The average size of Mg\(_2\)Si particles rises from 30 to 95 \(\mu\)m as the Mg content increases from 3\% to 10\%.

2) Most primary Mg\(_2\)Si particles exhibit octahedron shape when the Mg content is low. \{1 0 0\} directions usually have the highest growth velocity as \{1 0 0\} planes have the lowest lattice density. Finally, the octahedron’s corners and edges grow up along \{1 0 0\} and \{1 1 0\}, respectively, and the facets are parallel to \{1 1 1\}.

3) Some \{1 0 0\} directions become the preferential growth ones as the Mg content gradually increases. The primary Mg\(_2\)Si particles change to incomplete octahedron, hopper-like structure and kinds of complex shapes. The content of solute is a main factor to influence the morphology of Mg\(_2\)Si particles.

References

Mg 含量对过共晶 Al–Si 合金中初生 Mg2Si 相的影响

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摘 要: 采用铸造方法制备不同 Mg 含量的过共晶 Al–Si 合金，并研究 Mg 含量对合金中初生 Mg2Si 显微组织的影响。结果表明: 随着 Mg 含量的增大, Mg2Si 的体积分数呈线性增加, 但是平均晶粒尺寸没有呈线性增加。当 Mg 含量为 3% 时, Mg2Si 晶粒的(1 0 0)晶向生长速度最快, Mg2Si 易生长成八面体状。随着 Mg 含量的升高, 晶粒固液界面溶质分布不均匀, 合金形成不规则八面体结构。当 Mg 含量约为 10% 时, Mg2Si 生长成尺寸较大的复杂结构。

关键词: 过共晶 Al–Si 合金; 初生 Mg2Si 相; 生长方式; 生长速度

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