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MICROSTRUCTURE FORMATION OF Al-5 MASS%Mg ALLOY MELTS BY INTERACTION WITH SILICA

Dissolution of Si in Al-5 mass%Mg alloy melt by the reduction of SiO_2 and its effect on microstructure formation of the alloy after solidification were investigated. Al-5 mass%Mg alloy without silica powder had approximately 0.05 mass%Si as an impurity. No significant difference in Si content was observed after the reaction with silica for 10 min, while the Si content increased up to about 0.12 mass% after 30 min. From the microstructure analysis and calculation of Scheil-Gulliver cooling, it was considered that as-cast microstructures of Al-5 mass%Mg-1 mass% SiO_2 alloys had the distribution of eutectic phase particles, which are comprised of β-Al_3Mg_2 and Mg_2Si phases. Based on the phase diagrams, only limited amount of Mg can be selectively removed by silica depending on the ratio of Si and Mg. Addition of silica of more than approximately 1.5 mass% in Al-5 mass%Mg alloy led to the formation of spinel and removal of both Mg and Al from the melt.

Keywords: Al-Mg system, Silica, Reduction, Mg_2Si, Phase diagram

1. Introduction

The secondary production of Al is more environmentally sound and energy efficient than primary production. In Al scrap, Mg is one of the main impurity elements since a great quantity of Mg is added to Al alloys to improve mechanical properties and corrosion resistance [1,2]. Moreover, Mg enrichment in Al alloys can increase the oxidation tendency during melting process for a long duration [3,4]. The use of silica-based powders is considered a viable option to remove Mg in Al scrap [2,5]. According to the previous reports on effect of SiO_2 on Mg removal in Al scrap [2,5], MgO and $MgAl_2O_4$-spinel are formed by reduction of SiO_2, leading to loss of Mg and dissolution of Si in Al melt occurring simultaneously. Si is also a main alloying element used in majority of commercial Al alloys and co-existence of Mg and Si in Al alloys possibly provides a strengthening effect by Mg_2Si precipitations [1]. In addition, Si can also increase the oxidation resistance of Al-Mg alloys containing high contents of Mg at high temperatures [6]. The aim of this study is to investigate dissolution of Si in Al-Mg alloy melt containing a high content of Mg by the reduction of SiO_2 and its effect on microstructure formation of the alloy after solidification.

2. Experimental

The composition of the base metal examined in this study is Al-5 mass%Mg, in which the Mg content is similar to its maximum level in Al 5xxx alloys [1]. Alloys were made in an induction furnace under ambient atmospheric conditions. Pure Al was melted in a graphite crucible at 750°C and then, 5 mass%Mg in the form of pure Mg ingots was added into the melt. At 700°C, 1 mass%SiO_2 in a powder form was added into the Al-5 mass%Mg alloy melts with no additional equipment and maintained for a while to examine the reduction of SiO_2 powder by naturally occurring reactions in conventional melting processes. After that, each alloy melt was also cast into a steel mold that was preheated to 200°C. In this study, the maximum duration for the reaction between the melt and silica powder was 30 min, which is generally considered as a holding time for melt stabilization at each alloying process step in Al foundry. The dissolution amount of Si in the base metal was examined by inductively coupled plasma optical emission spectroscopy (ICP-OES). As-cast microstructures were observed using optical microscope after etching the surface of polished samples with Keller's reagent. The presence of the phases containing Si in the as-cast samples was analyzed using field emission (FE) – scanning electron

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microscopy (SEM) equipped with an energy dispersive X-ray spectroscopy (EDS) detector. Mg oxidation and SiO_2 reduction in Al-5 mass%Mg alloy melt were predicted on the basis of calculations of phase diagrams by *FactSage 7.3* [7]. Inter-products between Al-5 mass%Mg alloy melt and silica were also examined based phase equilibria diagrams calculated by *FactSage 7.3*.

3. Results and discussion

Fig. 1 demonstrates the relationship between analyzed silicon content in the as-cast ingot and reaction time. Al-5 mass%Mg alloy without silica powder has approximately 0.05 mass%Si as an impurity element, which is considered to be from pure Al raw material. After the reaction with silica for 10 min, no significant difference in Si content is observed. However, the Si content increases up to about 0.12 mass% after 30 min, indicating that the reduction of silica occurred in this condition. As a result of thermodynamic calculation by *FactSage 7.3*, 0.47 mass%Si is soluble in the liquid based on the phase equilibrium of Al-5 mass%Mg-1 mass%SiO_2 alloy at 700°C. This is assuming that the entire amount of added SiO_2 is reduced in Al-5 mass%Mg alloy. Thus, the maintenance for 30 min without any additional process such as agitation would be an insufficient condition to reach the phase equilibrium from a kinetic point of view. On the other hand, the trade-off of about 0.12 mass% between Mg and Si contents only for 30 min in conventional melting and alloying processes may be meaningful in case that the alloy melt has Mg contents slightly over the compositional range. Moreover, a small amount of Si in Al-Mg alloys can decrease oxidation rates according to a previous report [6].

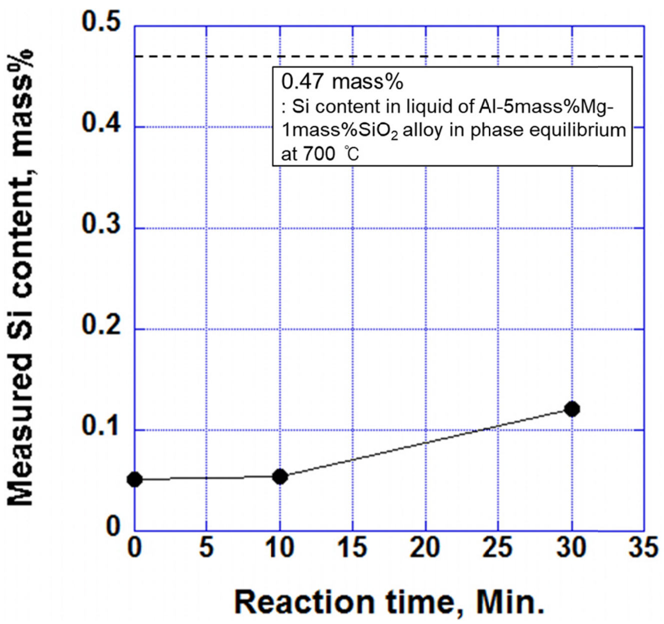

Fig. 1. Si content change in Al-5 mass%Mg-1 mass% SiO_2 alloys depending on reaction time between Al-5 mass%Mg alloy melt and SiO_2 powder at 700°C

As-cast microstructures of Al-5 mass%Mg-1 mass% SiO_2 alloys after different reaction times are shown in Fig. 2. The microstructure of silica-free alloy was not examined as it contained the Si content similar as that of the Al-5 mass%Mg-1 mass% SiO_2 alloy after the reaction for 10 min. The two as-cast microstructures show the distribution of small amounts of eutectic phase particles because of the high solubility of Mg in Al, but no significant difference due to their similar compositions. As a result of Scheil-Gulliver cooling calculation by *FactSage 7.3*, the solidification sequence is explained as Liquid → FCC_Al as the primary reaction and Liquid (remaining) → FCC_Al + β-Al_3Mg_2 two-phase eutectic reaction at approximately 451°C in Al-Mg binary system. With addition of a small amount of Si, FCC_Al + β-Al_3Mg_2 + Mg_2Si three-phase eutectic reaction occurs. The slight increase of Si in Al-Mg alloy leads to the increase of Mg_2Si fraction. However, no significant change between the two alloys in the fractions of eutectic phases is shown due to the small difference of Si content.

SEM-backscattered electron (BSE) image of as-cast microstructure after reaction for 30 min. with the numbered areas analyzed by EDS is given in Fig. 3. The eutectic phase areas

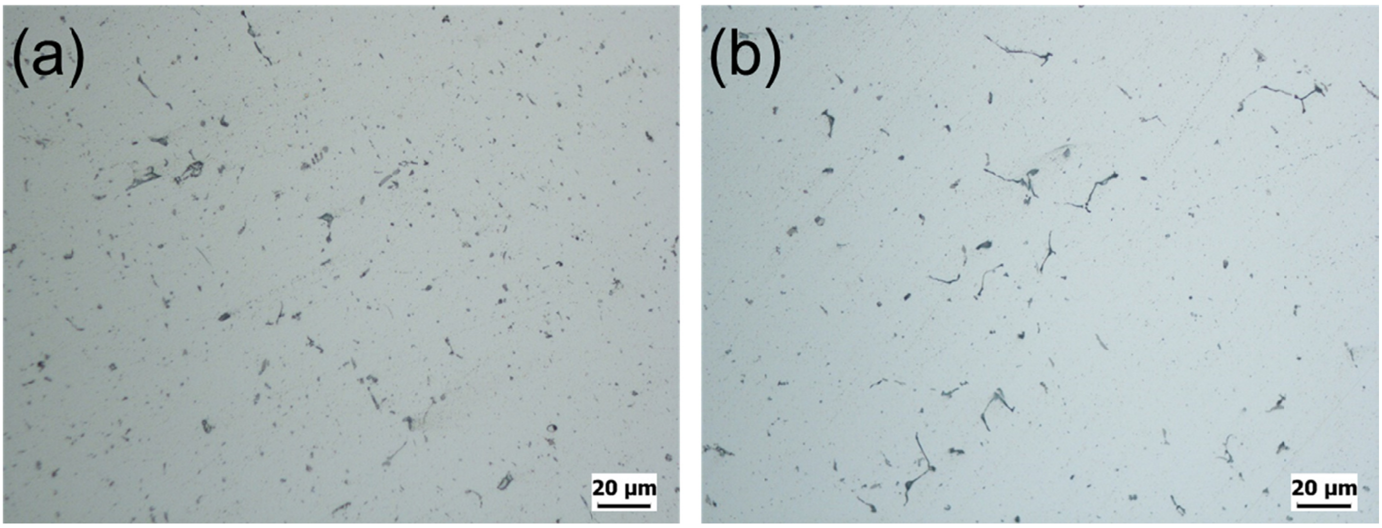

Fig. 2. As-cast microstructures of Al-5 mass%Mg-1 mass% SiO_2 alloys after different reaction times (a) 10 min and (b) 30 min

numbered 1 and 2 contain certain amounts of Si as shown in Table 1. From the SEM-EDS results, it is considered that the Mg_2Si phase is formed by Al + β-Al_3Mg_2 + Mg_2Si three-phase eutectic reaction, following Si dissolution from the reduction of SiO_2.

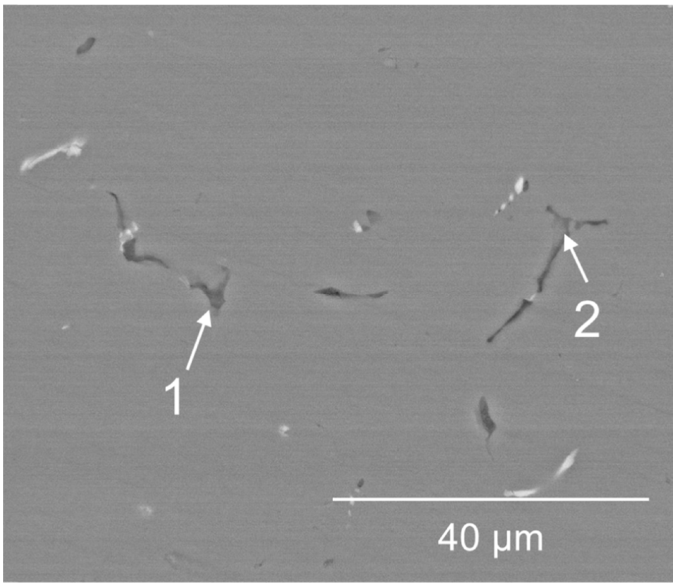

Fig. 3. SEM-backscattered electron (BSE) image of as-cast microstructure after reaction for 30 min

TABLE 1

Compositional analysis of areas shown in Fig. 3 by SEM-EDS

Area	Analyzed composition (mass%)		
	Al	Mg	Si
1	80.84	9.99	9.18
2	82.29	10.78	6.93

Phase diagram plotted for Mg versus SiO_2 mass fractions at 700°C is shown in Fig. 4(a). In the examined compositional ranges, MgO, spinel, and Al_2O_3 are present without any Si-based products. This result implies that a large amount of Si is soluble in Al-Mg alloy liquid and consequently, SiO_2 can be reduced by Mg and Al from the thermodynamical point of view. However, addition of silica of more than approximately 1.5 mass% in Al-5 mass%Mg alloy leads to the formation of spinel. Moreover, the spinel exists in the most regions in the examined ranges. Therefore, silica addition over a certain amount can remove not only Mg, but also Al by its reduction to form oxides. It also can be said that only limited amount of Mg is selectively removed by silica depending on the ratio of Si and Mg. Fig. 4(b) shows phase equilibria depending on fraction ratio between Al-5 mass%Mg and SiO_2 at 700°C calculated by *FactSage 7.3*. Silica can be reduced throughout a wide range of its fraction in Al-5 mass%Mg alloy melt. As aforementioned in Fig. 4(a), the formation of MgO to remove Mg, selectively, can occur only by a small fraction of silica. Therefore, it is considered that addition of silica is desirable only in case the Mg content in Al alloy slightly exceeds compositional specification to slightly strengthen by Mg_2Si formation improve oxidation resistance [6].

4. Conclusions

Al-5 mass%Mg alloy with no silica powder contained approximately 0.05 mass%Si as an impurity. A significant difference in Si content was not observed even after the reaction with silica for 10 min., while the Si content increased up to about 0.12 mass% after 30 min. From the microstructure analysis and calculation of Scheil-Gulliver cooling, it was confirmed that the as-cast microstructures of Al-5 mass%Mg-1 mass% SiO_2 alloys had the distribution of eutectic phase particles, which are comprised of β-Al_3Mg_2 and Mg_2Si phases. According to the phase diagrams of the examined alloy system, only limited amount of Mg can be selectively removed by silica depending on the ratio of Si and Mg. Addition of silica of more than approximately 1.5 mass% in Al-5 mass%Mg alloy led to the formation of spinel and removal of both Mg and Al from the melt. From the results in this study, it was considered that addition of silica is desirable only when the Mg content in Al alloy slightly exceeds composi-

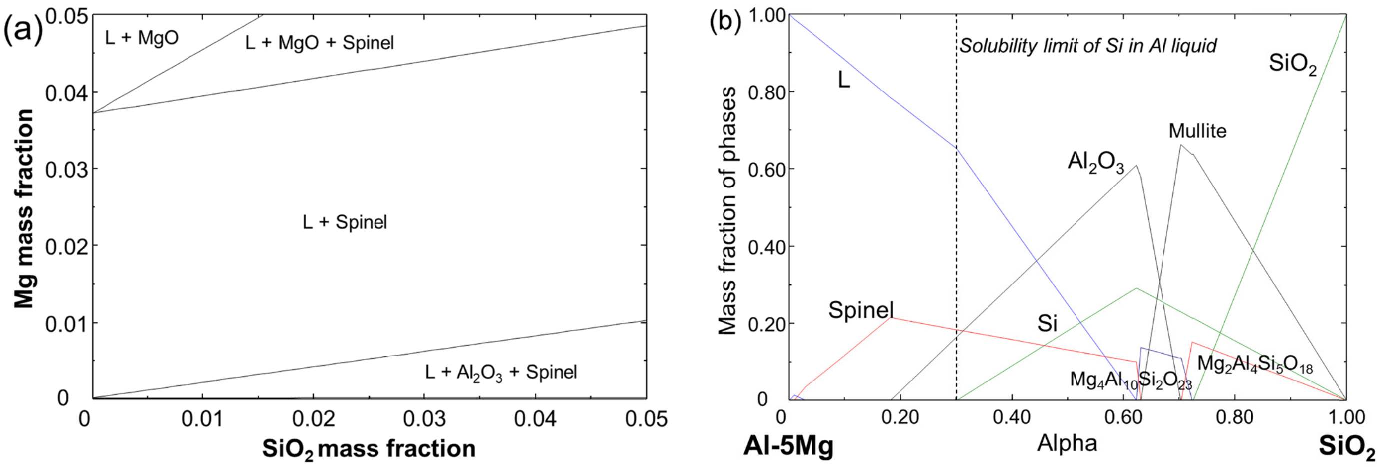

Fig. 4. (a) Phase diagram plotted for Mg versus SiO_2 mass fractions and (b) Phase equilibria between Al-5 mass%Mg and SiO_2 at 700°C calculated by *FactSage 7.3*.

tional specification to control the Mg content and provide slight strengthening effect by Mg_2Si formation and improvement of oxidation resistance, simultaneously.

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