Influence of cooling rate on morphology evolution of primary Mg_2Si in Al–15Mg_2Si-1.0Gd in-situ composite

Hamidreza Ghandvar, Tuty Asma Abu Bakar*, Mohd Hasbullah Idris

School of Mechanical Engineering, Faculty of Engineering, Universiti Teknologi Malaysia (UTM),
81310 Johor Bahru, Johor, Malaysia

Abstract

The effects of cooling rate on morphology of primary Mg_2Si in Al–15Mg_2Si composite modified with 1.0 wt. % Gd during solidification process were studied. To obtain different cooling rates, the step casting with four different thicknesses was used. The results showed that with increasing the cooling rate in Al-15%Mg_2Si-1.0% Gd composite, the absorption of Gd atoms as external factor on {100} facets of primary Mg_2Si crystal increases, leads to retarding of the growth rates of primary Mg_2Si along the <100> directions; results in the formation of primary Mg_2Si with various morphologies. With decreasing the thickness of mold from 50 to 40, 30 and finally to 20mm, the morphology of primary Mg_2Si altered from a dendrite to an octahedral and then truncated octahedral and finally to a truncated octahedral with larger {100} faces. This study can be beneficial to offer a simple method to control the morphology of primary Mg_2Si crystals in Al-Mg_2Si composite to tailor new composites with high mechanical properties.

Keywords: Al-Mg_2Si composite, Gadolinium, Cooling rate, Modification

---

Article Info

Received 23th December 2019
Accepted 15th May 2020
Published 1st December 2020
*Corresponding author: Tuty Asma Abu Bakar; email: tuty@utm.my

ISSN: 1823-7010, eISSN: 2600-7444

Introduction

Aluminum metal matrix composites (AMCs) have achieved considerable attention due to their superior properties such as high wear resistance, high strength to weight ratio, low thermal expansion, and lower costs of production, which make them useful for high-performance applications especially in the manufacture of automotive parts [1]. Recently, Al-Mg_2Si composites have receive considerable attention for applications requiring significant lightweight structural components owning to the existence of the intermetallic compound Mg_2Si phase with excellent physical properties make these composites as a high potential candidate for replacement of Al-Si alloys [2]. However, the formation of brittle and coarse morphology of Mg_2Si in the in-situ composite during the solidification process leads to localization of stress concentration at sharp ends and angles of Mg_2Si particles which induce low mechanical properties and consequently, limitation in the application of the composites [2,3].Therefore, modification of the Mg_2Si particles is necessary for the enhancement of the mechanical properties. Several studies have been taken into account to modify the structure of Mg_2Si particles with a melt treatment approach using different types of additional elements such as Sr [4], Gd [5], and Sb [6]. Other than the effect of additional elements, changing the cooling rate is another efficient method to induce refinement/modification of Mg_2Si particles. Hadian et al. reported a considerable decrease in primary Mg_2Si particle size from 16μm to 10μm with increasing the cooling rates of the composite melt by decreasing the thickness of the test bar from 9mm to 3mm [7]. Nevertheless, there are limited studies to examine the effect of cooling rates on the morphology evolution of primary Mg_2Si in RE modified Al-Mg_2Si composites. Therefore, the aim of the present study is to explore the impact of various cooling rates by changing the mold thickness on the microstructure of 1.0 wt. % Gd modified Al-15%Mg_2Si composite.

Materials and Methods

Al–15%Mg_2Si composites were in-situ synthesized using pure Al (99.98 wt. % purity), pure Mg (99.85 wt. % purity), pure Si (98.85 wt. % purity) and pure Gd (99.86 wt. % purity). The chemical composition of the composite illustrated in Table 1. Pure Al and Si were melted at 850 °C in an induction furnace and when temperature was reduced to 750 °C, pure Mg and 1.0 wt. % Gd were added into the melt. After homogenization the melt was poured into a sand mold without preheating (25 °C). The procedure was repeated for 2.0 wt. % Gd. Particularly, to control the cooling rate of the Al-15%Mg_2Si alloy, 1.0 wt. % Gd-modified Al–Mg_2Si alloy was poured into the cast iron mold with different thickness bars of 50, 40, 30, and 20mm (Figure. 1). This configuration allowed us to acquire a range of solidification rates resulting in different microstructures in the casting. After solidification and removing each specimen was cut and a metallography procedure was carried out on the samples through grinding with SiC papers and polishing using colloidal silica suspension. To obtain the three dimensional morphology of primary Mg_2Si crystals, composite samples were deeply etched with a solution of 20% HCl and 80% ethanol. To observe the microstructure of the samples and 3D morphologies of primary Mg_2Si, scanning electron microscopy (SEM) equipped with an energy dispersive spectrometer (EDS) analyzer was used.

Table 1. Chemical compositions of Al–15Mg_2Si composites (wt. %)

Composite	Mg	Si	Gd	Fe	Cr	V	Mn	Ti	Al
Gd-00	9.71	5.09	0.00	0.18	0.02	0.01	0.01	0.01	Bal.
Gd-10	10.12	5.03	0.98	0.16	0.01	0.03	0.01	0.01	Bal.
Gd-20	9.85	5.06	1.97	0.13	0.02	0.01	0.01	0.01	Bal.

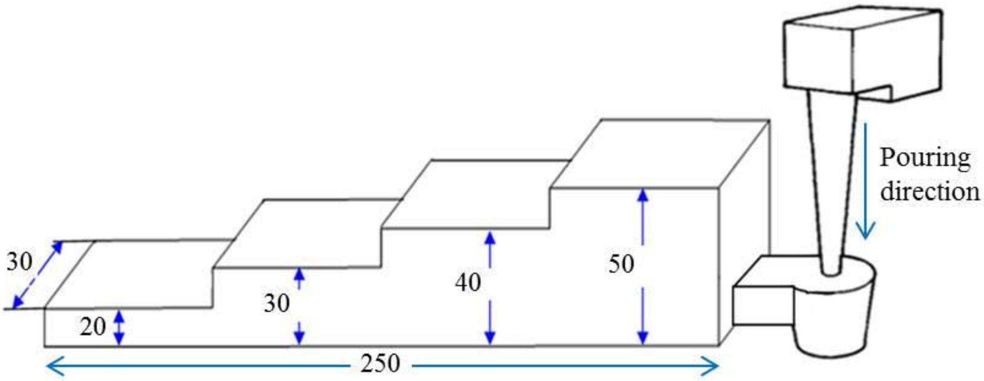


Figure 1. Schematic diagram of step casting (unit: mm)

Results and Discussion

Effect of Gd content on the composite microstructure

Figure.2 (a-f) illustrates the microstructure of the Al-15%Mg_2Si composites treated with 0, 1.0 and 2.0 wt. % Gd additions with corresponding three dimensional structures of primary Mg_2Si particles at mold thickness of 30mm. As observed, in the untreated condition, the primary Mg_2Si are dendritic with an average size of 65µm (Figure.2 (a, b)). When composite is treated with 1.0 wt. % Gd, the morphology of primary Mg_2Si changed to polyhedral and its average size decreased to 35µm (Figure. 2(c, d)). Refinement in the microstructure of the composite could be associated with the change in the phase diagram as a result of constitutional undercooling [8]. Moreover, the responsible factor in alteration in the growth morphology of Mg_2Si is the entering of Gd atoms in the lattice of the Mg_2Si crystal or changing its surface energy [3]. Nevertheless, with increasing the content of Gd to 2.0 wt. %, the morphology of primary Mg_2Si alters to bulges structure (Figure. 2 (e) and (f)), in which their sizes increase to 55 µm. It demonstrates that with increasing the Gd addition more than 1.0 wt. %, the modification effect of Gd addition is reduced, in which its mechanism will be discussed later. The BSE micrograph and corresponding line scanning spectra of the primary Mg_2Si in the composite treated with 1.0 wt. % Gd is depicted in Figure 3 (a, b). As seen the Mg and Si elements are spread across the crystal consistently; the Al element is barely detected in the Mg_2Si crystal; while, the Gd element is mostly concentrated in the interface of matrix and primary Mg_2Si (Figure 3 (b)). In the untreated composite, <100> is the preferential growth direction; therefore, the Mg_2Si morphology is dendritic (Figure.2 (b)). Nonetheless, the growth of Mg_2Si along the <100> direction is restricted seriously with the addition of 1.0wt. % Gd, leads to formation of truncated octahedral Mg_2Si crystal with six {100} faces and eight {111} faces (Figure 2(d)). Kubota et al [9] reported that the movement of the advancing surfaces could be restricted by absorbing the impurity atoms on the crystal surface, leads to the transition of crystal morphology. Hence, it is proposed that with preferentially absorption of the Gd atoms on {100} of Mg_2Si crystals, the growth along the <100> directions is restricted and primary Mg_2Si with truncated octahedral morphology is formed (Figure 2 (d)).

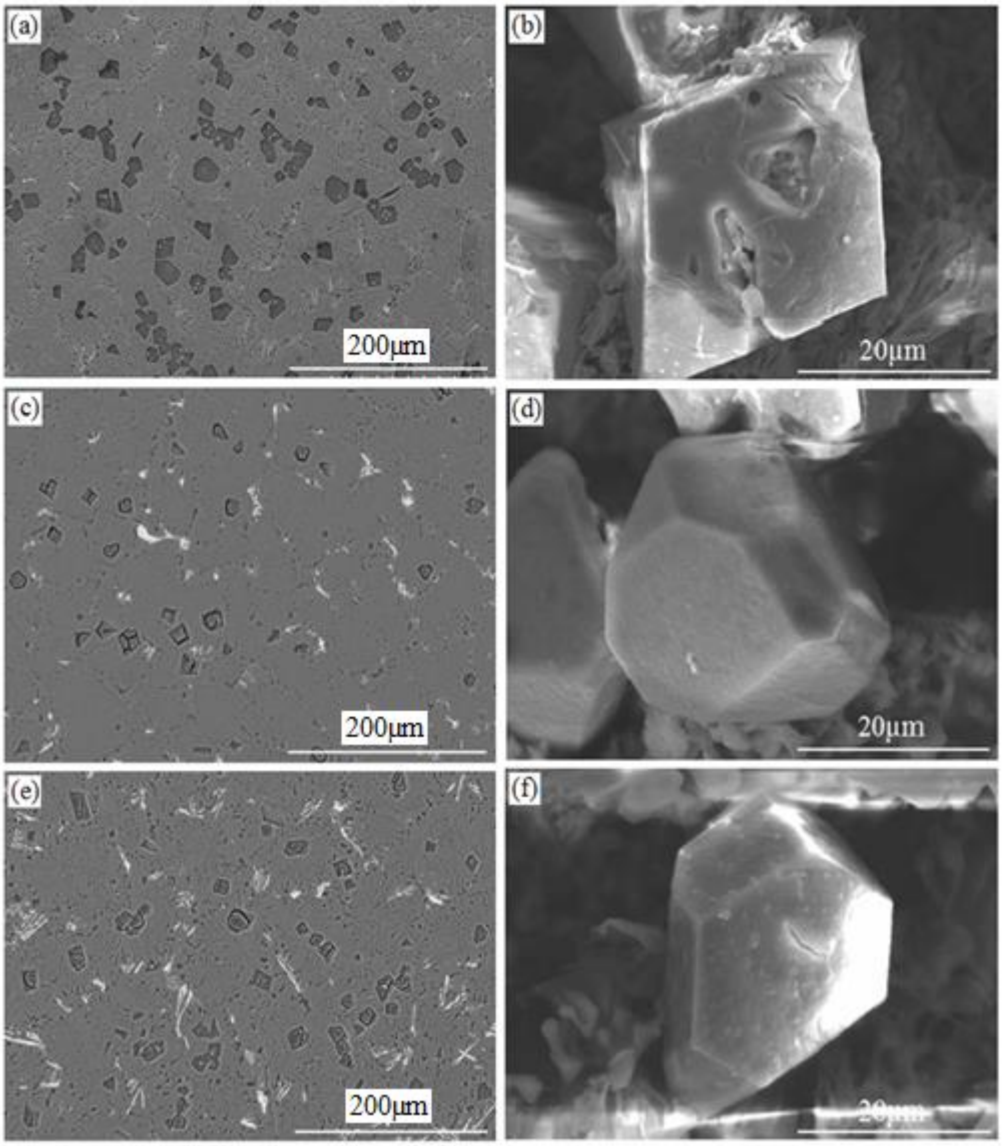


Figure 2. SEM micrographs and corresponding three dimensional morphologies of the primary Mg_2Si in (a, b) Al-15%Mg_2Si-0 wt. % Gd, (c, d) Al–15%Mg_2Si–1.0 wt. % Gd and (e, f) Al–15%Mg_2Si–2.0 wt. % Gd composites with a mold thickness of 30mm

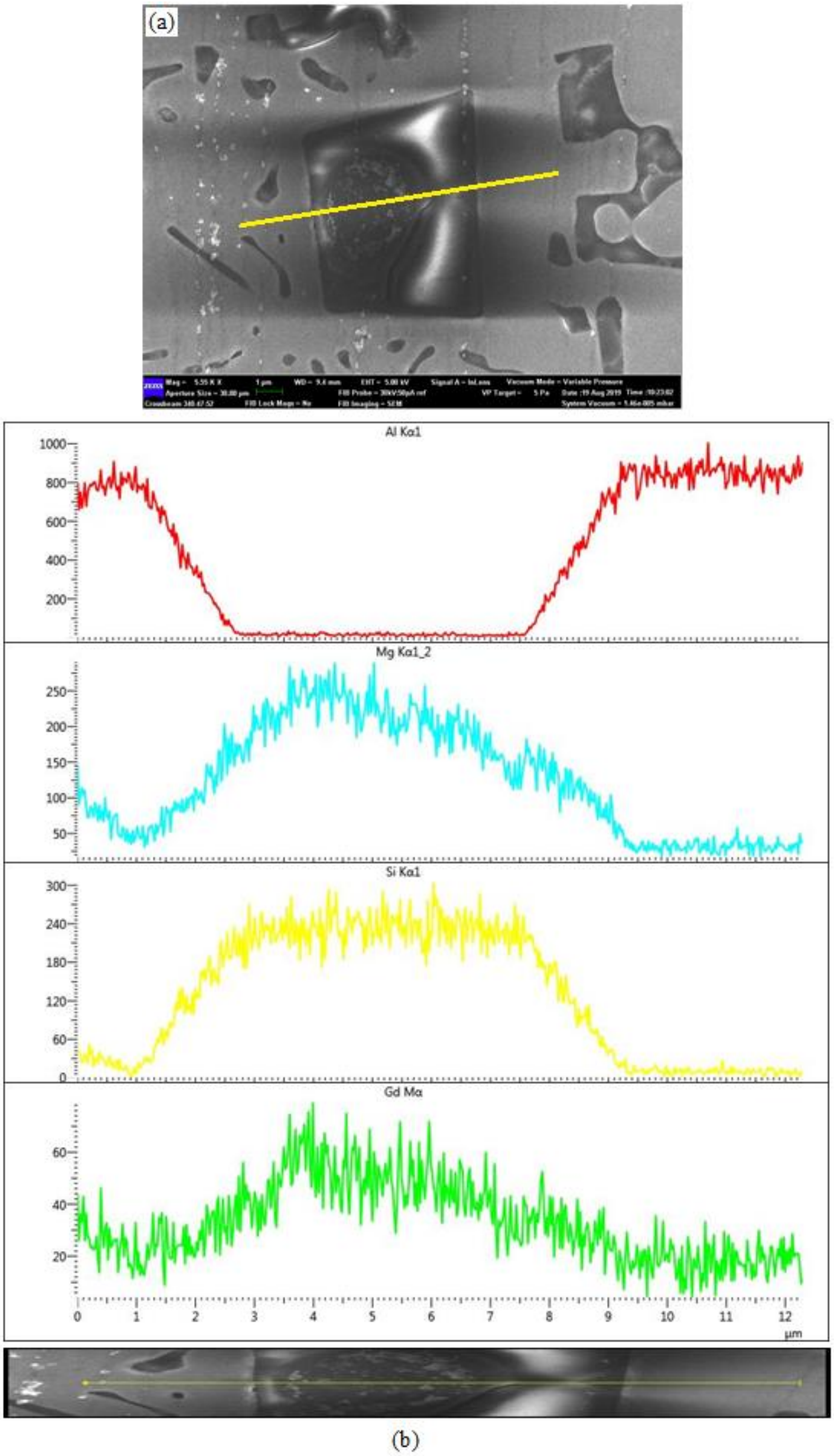
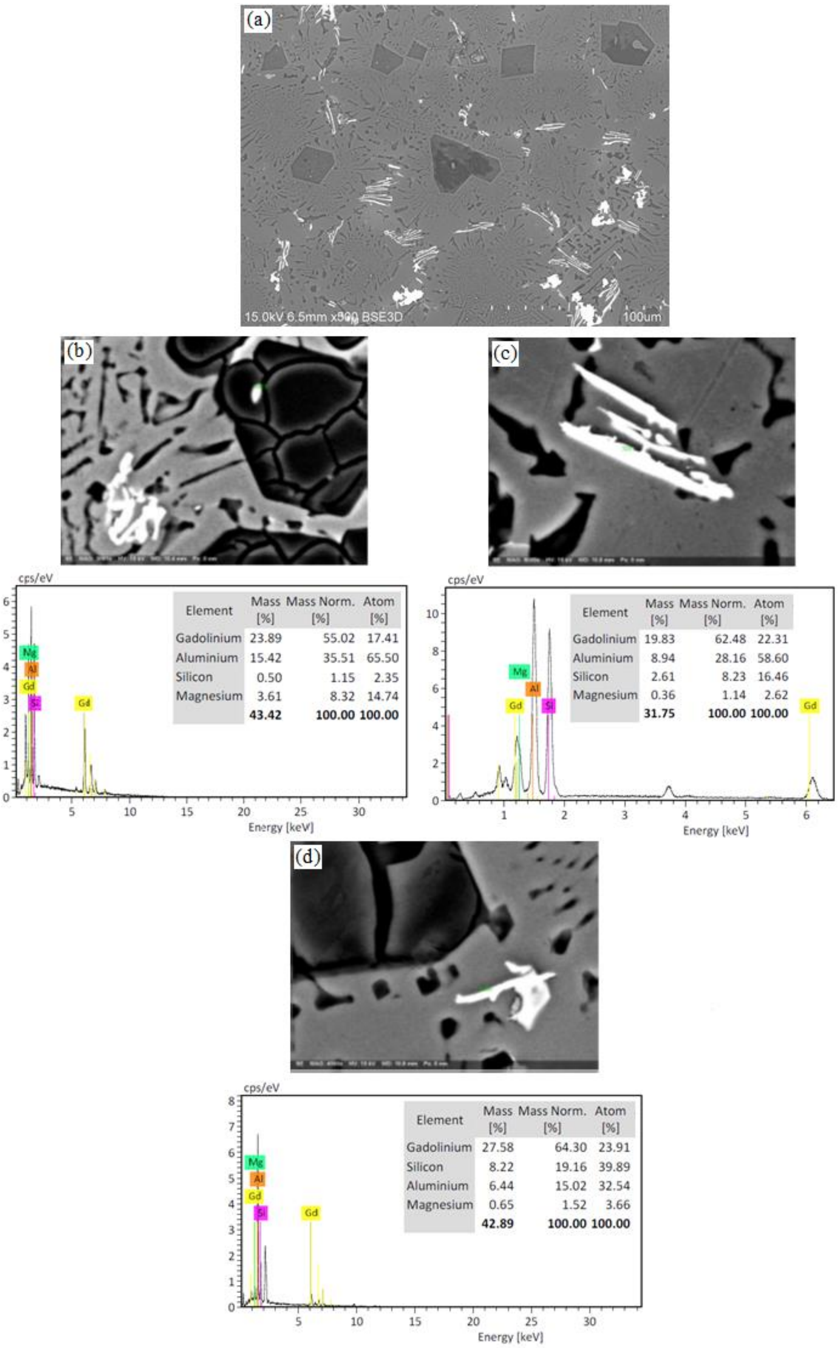


Figure 3. (a) SEM micrograph of primary Mg_2Si and (b) corresponding line scanning spectra of Al, Si, Mg and Gd elements in the Al–15%Mg_2Si–1.0%Gd composite

It has been reported that for some extent modifiers elements such as Sr could absorb onto the {110} and {111} faces of the Mg_2Si crystal, but by their absorption preferentially onto the {100} facets of Mg_2Si, considerable morphology evolution occurs which leads to the final formation of cubic Mg_2Si [4]. Likewise, Gd atoms can absorb preferentially on the {100} faces; nevertheless, the responsible mechanism for the preferential absorption of the modifier elements onto the {100} faces of primary Mg_2Si crystals is still unclear and requires further investigation.

Another feature of the composite micrographs in Fig. 1 is the existence of white intermetallic compounds in the matrix of the composites, particularly when 2.0 wt. % Gd is added. The EDS results in Figure.4 demonstrate the presence of Gd (IMCs) with different atomic% correspond to MgGd, Al_2Si_2Gd and AlSiGd after the treating of the composite with 2.0 wt. % Gd. This result is consistent with our findings in the previous study regarding the formation of the aforementioned Gd (IMC) in Al-15%Mg_2Si-2.0%Gd composite [4].



Element	Mass [%]	Mass Norm. [%]	Atom [%]
Gadolinium	23.89	55.02	17.41
Aluminium	15.42	35.51	65.50
Silicon	0.50	1.15	2.35
Magnesium	3.61	8.32	14.74
	43.42	100.00	100.00

Element	Mass [%]	Mass Norm. [%]	Atom [%]
Gadolinium	19.83	62.48	22.31
Aluminium	8.94	28.16	58.60
Silicon	2.61	8.23	16.46
Magnesium	0.36	1.14	2.62
	31.75	100.00	100.00

Element	Mass [%]	Mass Norm. [%]	Atom [%]
Gadolinium	27.58	64.30	23.91
Silicon	8.22	19.16	39.89
Aluminium	6.44	15.02	32.54
Magnesium	0.65	1.52	3.66
	42.89	100.00	100.00

Figure 4. (a) SEM micrograph of Al-15%Mg_2Si-2.0% Gd composite with (b-d) corresponding EDS point analysis of white intermetallic compounds

As mentioned previously, increasing the Gd content to 2.0 wt. %, over-modification happens which leads to decreasing the refinement of primary Mg_2Si particles. This phenomenon is that, Gd intermetallic compounds are formed prior to precipitation of primary Mg_2Si which leads to a reduction in the amount of Gd atoms to be absorbed on the growth ahead of primary Mg_2Si [3]. As a result, with increasing the Gd concentration more than 1.0 wt., the effect of poisoning on the primary Mg_2Si is considerably weakened, leading to increase in particle size.

Effect of different cooling rates on morphology evolution of primary Mg_2Si

In order to investigate the morphology evolution of primary Mg_2Si crystals as a function of various cooling rates, Al-15%Mg_2Si-1.0wt. % Gd was chosen because of observed pronounce modification effect of 1.0wt. % Gd compared to 2.0 wt. % (Figure.2). In order to control the cooling rate of the Al-15Mg_2Si-1.0 Gd composite, the thickness of the mold was increased from 20mm to 30mm and then to 40mm and finally to 50mm. Figure.5 illustrates the deep-etch micrographs of primary Mg_2Si crystals in Al-15Mg_2Si-1.0Gd prepared with various mold thickness. As seen, when the thickness of the mold is 20mm the structure of primary Mg_2Si is a perfect truncated octahedral (Figure 5(a)). With increase of the mold thickness to 30mm, morphology of primary Mg_2Si changed to truncated octahedral (Figure. 5(b)). With further increasing of the mold thickness to 40 and 50mm, octahedral and dendritic morphologies of primary Mg_2Si crystals are formed respectively as observed in Figure. 5(c, d). Hence, it can be seen that the variation of the cooling rate affects the morphology of primary Mg_2Si, in which with increasing the cooling rate the influence of adsorption of Gd atoms (as external factor) on {100} faces of Mg_2Si is strengthened resulted in morphology alteration of Mg_2Si crystals in Al-15%Mg_2Si composite. In fact, external factors force the crystal to develop different morphologies from equilibrium condition. However, the intrinsic crystal structure (as internal factors) resulted in equilibrium crystal morphology with minimum total surface free energy [10,11]. In the Mg_2Si crystal, the {111} owns the slowest growth rate and will be retrained as the crystal surfaces; while, under equilibrium condition, the {100} facets with the fastest growth rate will disappear. It has been reported that for Mg_2Si crystal, the ratio of growth rates along the <100> and <111> decides the crystal morphology, in which R= V<100>/V<111>, if, $R \geq \sqrt{3}$, a dendrite and an octahedron can be achieved; whereas; if, $1<R\leq\sqrt{3}$, a truncated octahedral is obtained; if $\sqrt{3.3} <R\leq 1$, a truncated cube can be formed and if $R=\sqrt{3.3}$, a cube can be prepared [12].

In the present study, when the mold thickness is 20mm, the cooling rate is fast and the solidification process is non-equilibrium. In this condition, the {100} faces of primary Mg_2Si appear and extend. Furthermore, absorption of the Gd atoms on {100} facets of primary Mg_2Si is the highest which leads to restriction of growth rate along the <100> seriously and the ratio of growth rate (R) reduces. As a result, the growth rate of {100} faces of Mg_2Si is moderately retained and truncated octahedral Mg_2Si with large {100} faces is formed (Figure.5 (a)). With increasing the mould thickness to 30mm, cooling rate decreases and the solidification process moves into the equilibrium state, meanwhile, Gd atoms influences the morphology of Mg_2Si crystal, which leads to restriction of the growth along the <100> directions. Hence, truncated octahedral morphology of primary Mg_2Si consists of both {100} and {111} faces are achieved (Figure.5 (b)). With further increase of the mold thickness to 40mm, the solidification process moves more to the equilibrium state and the growth pattern of primary Mg_2Si is faceted. In this condition the influence of Gd atoms as the external factor decreases and the {100} facets of the Mg_2Si crystal with fastest growth rate disappear; while, {111} facets of Mg_2Si with lowest growth rate are remained which results in formation of octahedral primary Mg_2Si As a result, octahedral morphologies of primary Mg_2Si formed by retaining the {111} faces (Figure. 5 (c)). Finally with increasing the thickness of the mold to 50mm, the solidification of the Al-Mg_2Si composite is nearly equilibrium, in which the facet planes would be trunk in {110} and dendritic tip in {111} along <100> direction. The growth facet in other directions, <110> and <111> are slow and this phenomenon leads to anisotropic growth of dendritic Mg_2Si structure as shown in (Figure 5 (d)).

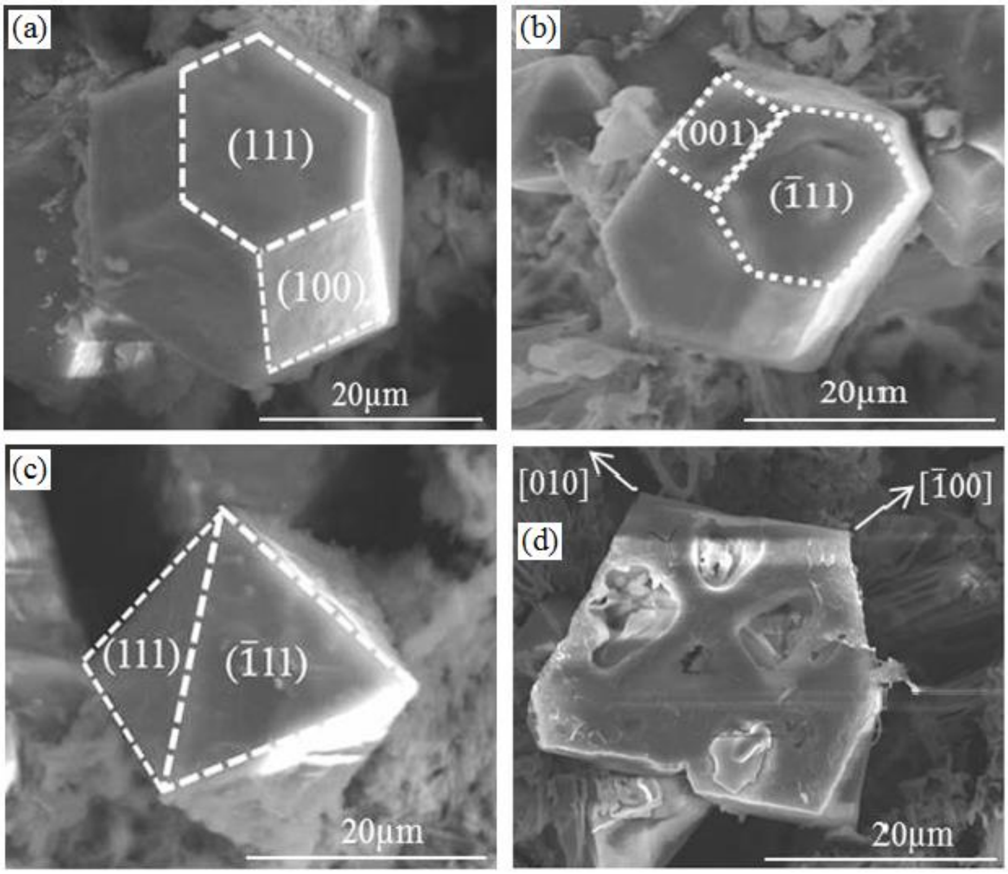


Figure 5. Three dimensional morphologies of primary Mg_2Si crystal in Al–15Mg_2Si–1.0Gd composite with various mold thickness: (a) 20, (b) 30, (c) 40 and (d) 50mm.

Conclusion

Addition of 1.0 wt. % Gd to Al-15%Mg_2Si composite resulted in refinement/modification of primary Mg_2Si particles, in which the morphology of primary Mg_2Si altered from dendritic to truncated octahedral shape with decreasing in its size from 65μm to 35μm. Furthermore, the morphology of primary Mg_2Si crystals in Al–15Mg_2Si–1.0Gd composite varies with changing the cooling rate. With increasing the cooling rate, the effect of an external factor (adsorption of Gd atoms on the {100} facets of Mg_2Si) is strengthened in which the perfect truncated octahedral morphology of Mg_2Si is formed in the highest cooling rate when the mold thickness is 20mm. The present study offers a cheap method to control the morphologies of primary Mg_2Si, which is critical to achieving industrial production of morphology-controllable Al-high Mg_2Si alloys with high strength and toughness.

Acknowledgments

The authors gratefully acknowledge the research funding under the Research University Grant using vote number 4F945 and 06G22.

Author contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of conflict of interest

The authors have no disclosures to declare.

Compliance with ethical standards

The work is compliant with ethical standards

References

[1] Fallah,V., Langelier, B., Opoku, N. O., Raeisinia, B., Provatas, N., & Esmaeili, S. (2016). Cluster evolution mechanisms during aging in Al–Mg–Si alloys. *Acta. Mater*.103 290-300.
[2] Srinivas, V. & Singh, V. (2012) Development of in situ as cast Al-Mg_2Si particulate composite: Microstructure refinement and modification studies. *T Indian I Metals*. 65(6) 759–764
[3] Ghandvar, H., Idris, M.H., Ahmad, N., & Emamy, M. (2018) Effect of gadolinium addition on microstructural evolution and solidification characteristics of Al-15%Mg_2Si in-situ composite. *Mater. Charact* 135 57-70
[4] Chen, L., Wang, H. Y. Li, Y. J., Zha, M. &Jiang, Q. C. (2014) Morphology and size control of octahedral and cubic primary Mg_2Si in an Mg–Si system by regulating Sr contents. *CrystEngComm*, 16, 448-454
[5] Ghandvar, H., Idris, M.H., & Ahmad, N. (2018) Effect of hot extrusion on microstructural evolution and tensile properties of Al-15% Mg_2Si-xGd in-situ composites. *J. Alloys Compd*. 751 370-390
[6] Farahany, S. Ghandvar, H., Nordin, N.A. Ourdjini, A. (2017) Microstructure characterization, mechanical, and tribological properties of slow-cooled Sb-treated Al-20Mg_2Si-Cu in situ composites. *J. Mater. Eng. Perform*. 26 1685-1700.

[7] Hadian, R., Emamy, M., Varahram, N. & Nemati, N. (2008) The effect of Li on the tensile properties of cast Al–Mg_2Si metal matrix composite. *Mater. Sci. Eng., A*. 490 250-257
[8] Kim, J. J., Kim, D. H., Shin, K. S. & Kim, N. J. (1999) Modification of Mg_2Si morphology in squeeze cast Mg-Al-Zn-Si alloys by Ca or P addition. *Scr. Mater*. 41 333-340.

[9] Kubota, N. & Mullin, J. W. (1995) A kinetic model for crystal growth from aqueous solution in the presence of impurity. *J. Cryst. Growth*. 152 203-208.

[10] Xu, C. L. & Jiang, Q. C. (2006) Morphologies of primary silicon in hypereutectic Al–Si alloys with melt overheating temperature and cooling rate. *Mater. Sci. Eng., A*. 437 451-455.
[11] Qin, Q. & Zhao Y.G. (2008) Nonfaceted growth of intermetallic Mg_2Si in Al melt during rapid solidification. *J. Alloy. Compd*, 462, L28-L31.
[12] Wang, R., Lu, W. & Hogan, L.M. (1997) Faceted growth of silicon crystals in Al-Si alloys, Metall. Mater. Trans: A. 28 1233-1243.