



## NUCLEATION PHENOMENON IN SiC PARTICULATE REINFORCED MAGNESIUM COMPOSITE

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### Introduction

Metal matrix composite (MMC) technology can improve the service performance of matrix metals and alloys. Most magnesium alloy based MMCs are produced via a casting process, therefore the casting microstructures are of vital importance to their performance in service. The formation of microstructure during solidification of MMCs is mainly influenced by four factors: nucleation (or its absence) on particles; particle pushing by the solidification front; particle settling in the melt; and chemical reaction between particles and matrix. So far many studies on the microstructures of MMCs have focused on identification of the interfacial chemical reaction between reinforcement and matrix [1–3], and there is only a limited and controversial understanding regarding the nucleation phenomena in MMCs.

It has been reported [4–6] that primary silicon can nucleate preferentially on the reinforcement particles in hypereutectic Al-Si alloy. However, it is also reported [5] that in hypoeutectic Al-Si alloy, primary  $\alpha$ -aluminum phase cannot nucleate on the reinforcement particles and that these particles tend to be pushed into the last freezing interdendritic regions. For magnesium based composites, both particle pushing (or capture) and grain refinement were observed in sand cast SiCp/Mg(AZ91) [7] and SiCp/Mg(ZCM630) composites [8] and permanent mold cast SiCp/Mg(AZ91) composites [9]. However, since it was not clear whether the primary  $\alpha$ -Mg phase could nucleate on SiC particles, the grain refinement effect was attributed to a possible growth retardation mechanism. Recently, Luo [10] reported that in a magnesium matrix composite with 10 vol.% SiC, the significant grain refinement (67%) was the combined result of heterogeneous nucleation and restricted grain growth. In the composite with 0.5 vol.% SiC, the moderate grain refinement (37%) was mainly attributed to a heterogeneous nucleation mechanism. The evidence of heterogeneous nucleation of primary magnesium on SiC particles was attributed to the small lattice disregistry (4%) between SiC and magnesium with an orientation relationship of  $(111)_{\text{SiC}}// (0001)_{\text{Mg}}$ . In a word, it can be noted that nucleation phenomenon plays a key role in determining solidification microstructures of MMCs.

The primary objective of the present research was to study the interfacial microstructure of SiC particulate reinforced magnesium matrix composite. Great effort was made to identify the orientation relationship between SiC particles and the matrix phase, because this relationship is very important in explaining the process of nucleation on the particles.

### Experimental

A Mg-9Al-Zn alloy was selected as the matrix for the composite. The chemical composition of the alloy is similar to that of AZ91. The reinforcement particles were high purity silicon carbides with an average diameter of 10  $\mu\text{m}$ . Composites with 15 vol.% SiC particles were prepared using compocasting. In the compocasting process, the preheated SiC particles were entrapped into the magnesium alloy heated to a highly viscous, semi-solid state; and then the slurry was remelted and mixed above the liquidus. About 1 kg of composite was fabricated in an electric resistance furnace using a steel crucible under argon protective atmosphere.

The as-cast composite ingots were sectioned into thin slices, mechanically ground on 1000 grit silicon carbide paper, polished to approximately 60  $\mu\text{m}$  in thickness, and subsequently thinned using argon-ion beam at 5 kV, 4 mA at angles 30° and 10°. The foils prepared were carefully examined using JEM-2000EX transmission electron microscope (TEM) equipped with a double-tilt holder and operated at 160 kV.

### Results and Discussion

A previous study [11] shows that the interfacial structure of SiC particulate reinforced magnesium composite is essentially featureless and there is no evidence of extensive chemical reaction at the interface, but further crystallographic orientation of the magnesium matrix/SiC particle interface has not been identified. It is generally believed that in metals and alloys, solid particles in a solidifying melt which have low energy solid-liquid interfaces provide favorable sites for heterogeneous nucleation. However, so far the nucleation phenomenon of a liquid phase in the presence of solid particle during solidification has not been fully established because of its complex nature, though heterogeneous nucleation is anticipated on the basis of the published work. One criterion of heterogeneous nucleation is that the disregistry of nucleant planes is at a minimum (less than 5%) [12]. According to the work by Bramfitt [12], the modified equation about disregistry ( $\delta$ ) is as follows:

$$\delta_{(hkl)_n}^{(hkl)_s} = \frac{1}{3} \sum_{i=1}^3 \frac{|d[uvw]_s^i \cos \theta - d[uvw]_n^i|}{d[uvw]_n^i} \cdot 100\%$$

where:

- (hkl)<sub>s</sub> = a low index plane of the substrate;
- [uvw]<sub>s</sub> = a low index direction in (hkl)<sub>s</sub>;
- (hkl)<sub>n</sub> = a low index plane in the nucleated solid;
- [uvw]<sub>n</sub> = a low index direction in (hkl)<sub>n</sub>;
- d[uvw]<sub>n</sub> = the interatomic spacing along [uvw]<sub>n</sub>;
- d[uvw]<sub>s</sub> = the interatomic spacing along [uvw]<sub>s</sub>;
- $\theta$  = the angle between the [uvw]<sub>s</sub> and [uvw]<sub>n</sub>

Table 1 shows the results of calculation for some possible crystallographic orientations for primary  $\alpha$ -Mg nucleation on the SiC particles. It can be noted that when the orientation relationship between Mg phase and SiC phase is  $(10\bar{1}0)_{\text{Mg}} // (0001)_{\text{SiC}}$ , the disregistry is the lowest (2.3%). Therefore, it is possible for primary Mg phase to nucleate on SiC particles by this orientation relationship.

Fig. 1 shows the most common type of interfacial structure in the magnesium composite. No reaction layer was observed at the particle/matrix interfaces. It can be also noted that the SiC particles are entrapped in a magnesium grain. This observation suggests that the SiC particle might act as the

TABLE 1  
Results of Calculation for Some Possible Orientation Relationships between Mg and SiC

	$(0001)_{\text{SiC}} \parallel (0001)_{\text{Mg}}$			$(0001)_{\text{SiC}} \parallel (10\bar{1}0)_{\text{Mg}}$			$(0001)_{\text{SiC}} \parallel (\bar{1}101)_{\text{Mg}}$		
$[uvw]_{\text{SiC}}$	[100]	[110]	[010]	[210]	[220]	[010]	$\bar{1}10$	[020]	[110]
$[uvw]_{\text{Mg}}$	[100]	[110]	[010]	[001]	[011]	[010]	$[01\bar{1}]$	$[12\bar{1}]$	[110]
$d[uvw]_{\text{SiC}}$	5.335	5.335	5.335	5.335	6.160	3.080	5.335	6.160	3.082
$d[uvw]_{\text{Mg}}$	5.210	5.210	5.210	5.210	6.111	3.210	6.119	7.619	3.210
$\theta$	0	0	0	10.89	10.89	10.89	10.89	4.973	4.337
$\delta$		4.0%			2.3%			12.69%	

substrate for heterogeneous nucleation of primary magnesium. Further selected area electron diffraction analysis at the interface between the magnesium matrix and SiC particle is shown in Fig. 2. After indexing, the orientation relationship between them is:

$$[0001]_{\text{Mg}} // [\bar{1}5\bar{4}0]_{\text{SiC}}$$

$$(10\bar{1}0)_{\text{Mg}} // (0001)_{\text{SiC}}$$

The crystallographic orientation relationship identified is consistent with the anticipation based on the theoretical calculation (Table 1) and confirms that magnesium can indeed nucleate on SiC particles with a small lattice disregistry (2.3%). However, the selected area diffraction pattern technique did not help to identify whether the magnesium phase is primary magnesium phase or eutectic magnesium phase. If the magnesium phase were formed by eutectic reaction,  $[L \rightarrow \text{Mg}(\alpha) + \text{Mg}_{17}\text{Al}_{12}(\beta)]$ , the eutectic magnesium phase would be expected to nucleate on the primary magnesium phase (rather than on a SiC particle) to decrease the surface energy. Therefore, it may be deduced that the magnesium phase nucleated on the SiC particles must be primary magnesium phase.

Another phase nucleated on SiC can also be seen from Fig. 1 (just below the SiC particle). Electron diffraction patterns for the phase are shown in Fig. 3. After index, this phase is identified as  $\text{Mg}_{17}\text{Al}_{12}$  eutectic phase, which has body center cubic structure and lattice parameter  $a = 10.56 \text{ \AA}$ .

In terms of eutectic phase features, as shown in Fig. 1, there is clear evidence that the wetting angle ( $\theta$ ) between eutectic and SiC is less than  $90^\circ$ . This clearly indicates that the  $\text{Mg}_{17}\text{Al}_{12}$  eutectic wets the

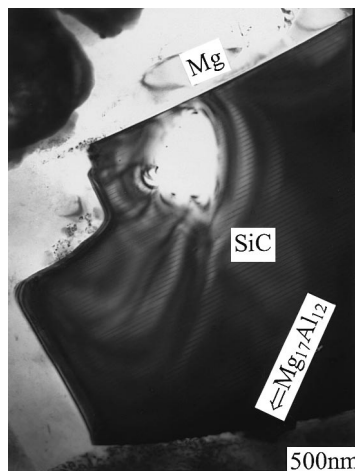


Figure 1. TEM micrograph showing a typical interface between SiC particle and Mg matrix.

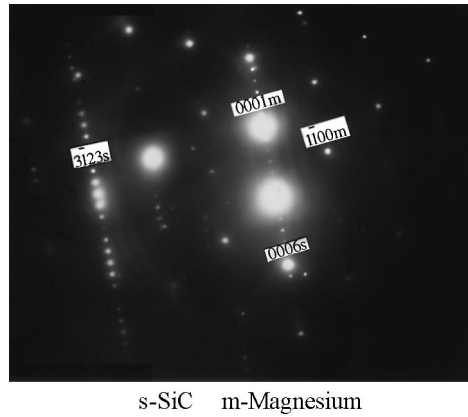


Figure 2. Composite electron diffraction pattern at the SiC/Mg interface.

SiC particle. In this regard the  $Mg_{17}Al_{12}$  eutectic can nucleate and spread on the SiC particulate and then grow into the melt. Fig. 4 is the composite electron diffraction pattern at the SiC/ $Mg_{17}Al_{12}$  interface. After indexing, the orientation relationship between SiC and  $Mg_{17}Al_{12}$  phase is as follows:

$$[1\bar{1}01]_{SiC} // [1\bar{1}1]_{Mg_{17}Al_{12}}$$

$$(01\bar{1}1)_{SiC} // (110)_{Mg_{17}Al_{12}}$$

This crystallographic orientation relationship has further strong evidence to prove the nucleation of eutectic phase on the SiC particulate. During the solidification process, the solute elements (such as Al) are rejected into the liquid at the solid-liquid interface because of their low solubility in magnesium. When the Al solute content reaches the eutectic composition, the eutectic reaction  $[L \rightarrow Mg(\alpha) + Mg_{17}Al_{12}(\beta)]$  will occur in the magnesium melt. On the other hand, when the SiC particle surface satisfies the substrate condition for  $Mg_{17}Al_{12}$  heterogeneous nucleation, then  $Mg_{17}Al_{12}$  phase will nucleate on the SiC particulate and grow into the melt.

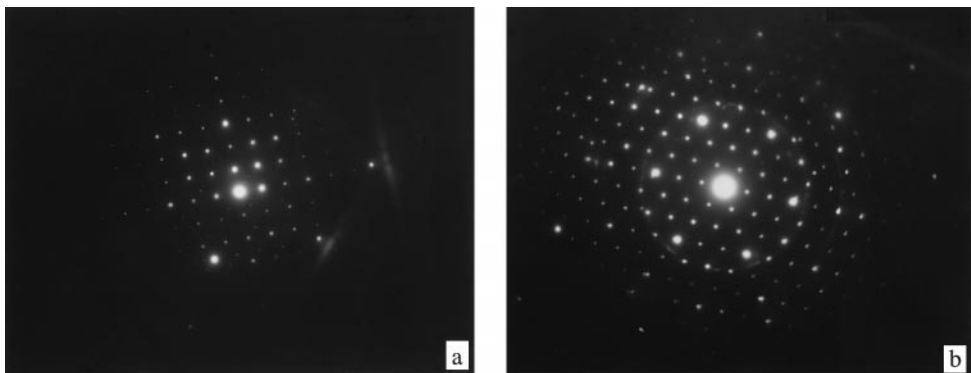


Figure 3. Electron diffraction patterns of  $Mg_{17}Al_{12}$  eutectic phase (a) [001]; (b) [111].

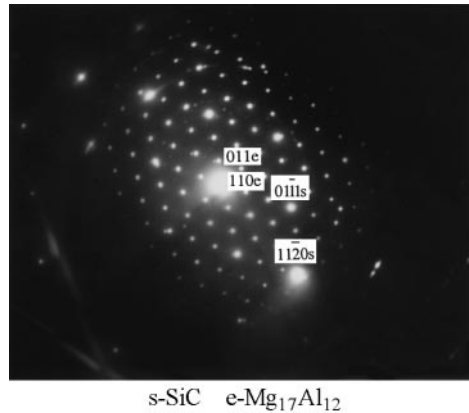


Figure 4. Composite electron diffraction pattern between SiC and  $Mg_{17}Al_{12}$ .

### Conclusions

During solidification process of SiC particulate reinforced magnesium composite, SiC particles can act as heterogeneous nucleation substrates for primary magnesium and eutectic phases with crystallographic orientation relationships of  $(10\bar{1}0)_{Mg} // (0001)_{SiC}$  and  $(01\bar{1}1)_{SiC} / (110)_{Mg_{17}Al_{12}}$  respectively.

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