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EFFECT OF COMPACT DENSITY AND PREHEATING TEMPERATURE OF THE Al-Ti-C PREFORM ON THE FABRICATION OF IN-SITU Mg–TiC COMPOSITES

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Abstract

Magnesium reinforced *in-situ* TiC particulates was successfully synthesized utilizing the self-propagating high temperature synthesis (SHS) process .The result showed that preform temperature and compact density have a significant effect on the SHS reaction. When the compact density was 68 % of the theoretical density, no SHS reaction was observed. However, with an increase in density from 68 to 72 %, the successful thermal explosion reaction was observed in the Mg- melt. Besides, the effect of pre-heat temperature on the fabrication of Mg/TiC composite was extensively studied and it was found that the preheat temperature of 300 ^{0}C failed to give rise to SHS reaction. However, the increased pre-heat temperature of 450, 500 and 550 ^{0}C favors the reaction inside the liquid melt, but when the temperature is 600 ^{0}C, the ignition reaction occurred in the preheating furnace itself

Introduction

In recent years, magnesium alloys are becoming more important for the industrial applications due to their relatively low density, high-damping capacity, good castability and machinability. However, the low strength, elastic modulus and wear resistance at elevated temperature have restricted their applications as engineering materials [1]. Metal matrix composite (MMC) technology significantly improves the wear resistance, elastic modulus, and tensile strength of unreinforced metals and alloys. Among numerous MMC systems under development, composites with aluminum and magnesium matrices and ceramic particulate reinforcements are of commercial interests to the automotive and aerospace industries [2]. Because of the formation of clean, ultra fine and stable ceramic reinforcements the *in-situ* MMCs exhibit excellent mechanical properties [3]. In magnesium matrix, TiC is particularly useful in reinforcement because of its hardness and stiffness, wettability, high-temperature stability and lower weight. Magnesium-based MMCs are currently being explored for a number of automotive and aerospace applications, such as automotive pulley, cog-tooth sprockets, oil-pump cover, cylinder liner, and aircraft engine casting [4-6].

Self-propagating high temperature synthesis (SHS) is drawing an increasing attention as a technique for synthesis of refractory materials due to its attractive advantages such as high purity of products, low processing cost, energy and time efficiency [7,8]. A wide variety of materials, such as carbides, borides, nitrides, intermetallics and composites have been produced by this method. Recently, SHS reaction in the Al-Ti-C system has been widely investigated because of its lower ignition temperature, about 450-600 ^{0}C, which is approximately the same as the temperature of magnesium alloy [9,10]. Literature also reports that the particle size, density and percentage aluminium in the preform has great effect on ignition temperature of the preform [2,3]. Many researchers have worked on Al-Ti-C system and demonstrated greater effect of compact density and pre-heat temperature on ignition of SHS reaction [1,6]. Limited studies have been carried out on the reaction in molten magnesium.

In this study, an attempt is made to study the feasibility of the *in-situ* synthesis of Mg-TiC and impact of compact density and the pre-heat temperature on the thermal explosion synthesis reaction of the preform of Al, Ti and C powders in molten magnesium.
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Experimental

The preforms in this work were made from commercial powders as given in Table 1. The reactants (powders) were mechanically blended by a ball mill. Then the resulting mixtures were pressed into cylindrical compacts of 20mm in diameter and 15 mm in length by using a stainless-steel die with two plungers. The compacts were pressed at pressures of 110–140 MPa to obtain densities of 75±2% theoretical density. Commercial magnesium ingot (purity 99.6% approximately) was selected as the matrices for the composites.

Table 1. Particle size and purity Al, C, and Ti powders

Sl. No.	Reagent	Particle size (μm)	Purity (%)
1	Aluminum	<40	98.00
2	Graphite	<20	99.5
3	Titanium	<50	98.7
4	Magnesium	Ingot	Commercial

About 1kg of magnesium was melted in the electric resistance bottom pouring furnace at 800 ^{0}C under argon atmosphere. Sixty gram of the pellet of different density and pre-heat temperature were added to the melt. After about 20 minutes, the melt was stirred at 600 rpm for 15 min using stainless steel impeller to facilitate incorporation and uniform distribution of *in-situ* from TiC in the metallic matrix. Experiments were conducted under a protective atmosphere of argon. Subsequently, SHS reaction happened and TiC particles were formed in the liquid of magnesium alloy. Processing of the magnesium matrix

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composites also consisted of melt stirring and composite casting (metal die casting). The as-cast ingots were sectioned, polished, and examined under a scanning electron microscopy (SEM). Phase identification of the resultant products was confirmed by X-ray diffraction and Micro-Raman Spectroscopy (Renishaw invia U.K.).

Results and Discussion

Effect of Compact Density

In this part of study, the effect of compact density on ignition temperature of preform and fabrication on Mg-TiC composites was carried out. Preform compact of densities 68, 70, 75, 80 and 85 % of theoretical density was prepared by compacting the powder in a steel die. In order to study the effect of compact density on ignition temperature of the compact, the compact was kept in tube furnace having argon atmosphere and heating was started at a very slow rate (3-4 ^{0}C/ min). The ignition temperature was measured by a thermocouple and the results were plotted. Figure 1 shows that with increase in compact density, ignition temperature of the SHS reaction decreases and becomes optimum when the compact density is about 80% of the theoretical density.

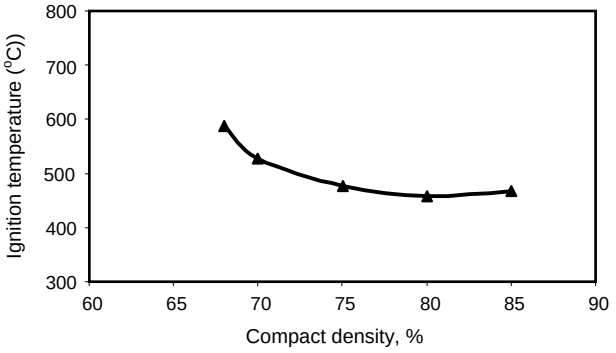

Figure 1. Effect of compact density on the ignition temperature of SHS reaction

In liquid-phase sintering**,** [9].the predominant mechanism leading to densification involves fluid flow and particle rearrangement. During this stage, the liquid wets the particles and flows among them. Thus, a more efficient packing occurs by the capillary forces acting on the particles. In the present study, the aluminum added to the titanium and carbon mixture is believed to exert a similar effect (i.e. particle rearrangement) on the reactant compact. However, the extent of the effect of capillary forces and subsequent particle rearrangement can be influenced by the density of the compact. If the density of the compact is low, capillary spreading and particle rearrangement is localized to where the liquid phase exists [9]**.** In contrast, if the density of the compact is high, capillary spreading and particle rearrangement is confined in the close among the particles. Therefore, an optimum compact density exists that allows the maximum extent of the capillary spreading and particle rearrangement to be achieved. Under such a circumstance, the maximum interfacial area between the aluminum melt and the graphite particles is obtained, and, consequently, the ignition temperature is at its minimum.

Figure 2 shows the XRD pattern of the Mg-TiC composites fabricated by using different compact density of preform. Form the XRD pattern it can be seen that when the compact density was 68 % or below, no TiC peaks were observed. It means that the SHS reaction could not start in the magnesium melt. The reason may be the effect of magnesium as diluent leading to the compact with 68 % density dispersed into the molten magnesium before the initiation of the SHS reaction. As a result, the compact density with 68 % failed to lead SHS reaction in the liquid magnesium. With the compact density of 72±1 %, the formation of TiC in the melt was observed.

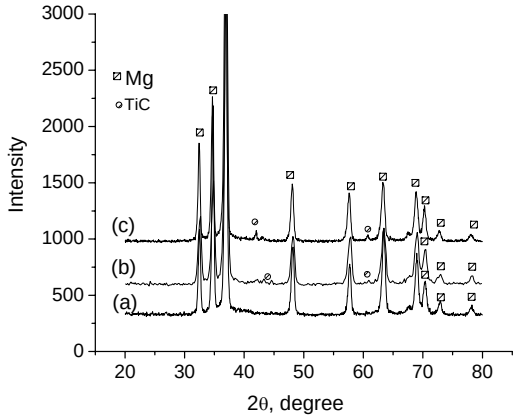

Figure 2. XRD pattern of the Mg-TiC composite fabricated using compact of density of preform (a) 68% (b) 70% (c) 75% of theoretical density

Effect of Preheat Temperature

When the preform preheat temperature was 450^{0}C, an exothermic reaction in the melts was immediately observed after the addition of the preforms to molten magnesium. However at temperature of 300 ^{0}C, the reaction in the melts did not occur, and there was a substantial amount of untreated carbon that floated on the surface of the melts during stirring. Furthermore, a large amount of residue was found in the bottom of the melts after stirring. XRD analysis (Cu Kα) showed the residue consisted of Ti and Mg. when the preheat temperature was increased to 450, 500 and 550 ^{0}C, an exothermic reaction in the melt occurred immediately after the addition of the preform to the molten magnesium. In case of preheat temperature of 600 ^{0}C, the SHS reaction started in the pre-heating furnace and TiC was formed in the preform.

The SHS reaction of the Al–Ti–C system can be ignited by heating the top surface of the preform by passing an electric current through a resistance coil [12,13]; however, the SHS reaction can also be ignited throughout the preform when it is heated to the ignition temperature by heating the entire preform at a constant rate [14]. In the present study, the exothermic reaction of the preform with a preheat temperature of 450 ^{0}C was initiated throughout the entire preform by the heat of molten magnesium, and the heat liberated. Consequently it can further raise the temperature, facilitating a faster reaction. Thus, it can be considered as SHS reaction with a simultaneous combustion mode.

Raman spectra analysis

Raman spectra were recorded by using the 514-nm radiation of an Ar laser excitation. The characteristic Raman spectra of the Mg-TiC composites fabricated with using pre-heated preform at 300, 450 and 600 ^{0}C are shown in Fig. 3. From the Fig. 3(a) it can be seen that when the pre-heat temperature was 300 ^{0}C only two peaks of carbon at 1320 and 1590 cm^{-1} were observed in the spectra. Since titanium, aluminium and magnesium do not have Raman active vibrational modes, they do not produce a Raman spectrum. So it is believed that at 300 ^{0}C, the preform gets dissolved before the ignition of the SHS reaction in the Mg- melt and as a result no TiC formation takes place. Fig. 3(b) and 3(c) show sharp TiC peaks with broad peaks of carbon. The carbon peaks are strong in Fig. 3(b) as compare to Fig. 3(c) and it shows that some unreacted carbon is present with TiC. Unreacted carbon is more when the pre-heat temperature is low and it decreases at higher pre-heat temperature; this may be due to the incompletion of TiC formation reaction.

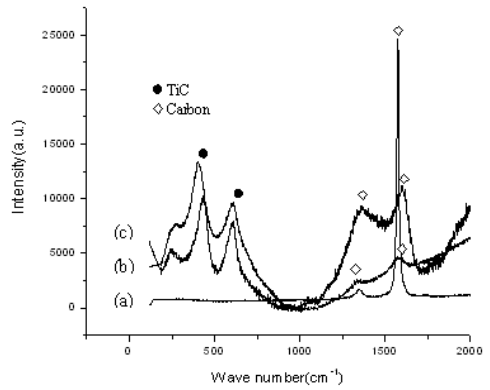

Figure 3. Micro-Raman pattern of Mg-TiC composite by using preform pre-heated at (a) 300 ^{0}C, (b) 450 ^{0}C, (c) 600 ^{0}C

X-ray diffraction analysis

Like Raman spectra, similar trend is observed in XRD pattern of compact perform quenched from 300, 450 and 600 ^{0}C, as shown in Fig. 4.

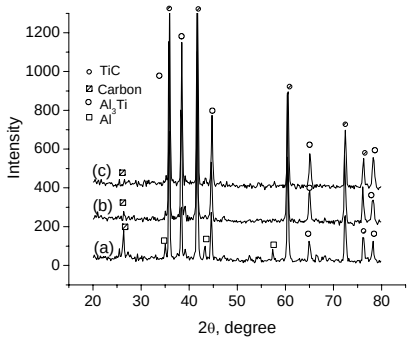

Figure 4. XRD pattern of compact perform quenched from (a) 300 ^{0}C, (b) 450 ^{0}C, (c) 600 ^{0}C

From the XRD pattern it can be seen that in the preform quenched at 300 ^{0}C, Al_3Ti, Al and C peaks are present but when quenched at 400 and 600 ^{0}C no Al peaks have been observed. This shows that at 300 ^{0}C reaction is not completed and support to the mechanism of thermal explosion synthesis in Al-Ti-C system reported by Guan *et al* [2] as given below:

$$3Al + Ti \rightarrow TiAl_3 \quad (1)$$

$$TiAl_3 + C \rightarrow TiC + 3Al \quad (2)$$

$$Ti + C \rightarrow TiC \quad (3)$$

Figure 4(b) and 4(c) show strong TiC peaks along with some weak Al_3Ti peaks, which indicates that as the preform temperature increases titanium carbide formation reaction is completed.

Optical and scanning electron microscopy

Optical micrographs of the *in-situ* processed as cast composites fabricated by using preform of different pre-heat temperature are shown in Figure 5(a)-(d).

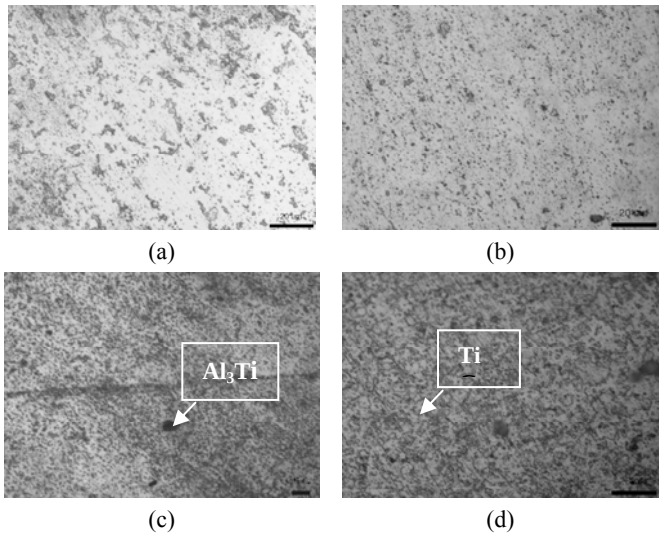

Figure 5. Optical micrograph of Mg-TiC composite prepared by heating preform at (a) 300 ^{0}C, (b) 450 ^{0}C, (c) 550 ^{0}C and (d) 600 ^{0}C

Fig. 5(a) shows only α-Mg and carbon which indicates that preform does not take part in the reaction. Fig. 5(c) and (d) reveal a relatively uniform distribution of TiC particulates of the size 2-3 μm in the as cast composite samples. Because the reinforcement phases were *in-situ* formed in the molten magnesium, the interface between TiC particulates was free from oxides.

Fig. 6(a) shows the SEM micrograph of the fabricated Mg-TiC composite in which small size, nearly 1-2 μm, TiC particulates are distributed in the Magnesium matrix. When the preform is heated above 600^{0}C temperature in Ar atmosphere, the SHS reaction starts in the preform itself and it can be seen in Fig. 6(b).

(a) (b)

Figure 6. SEM micrograph showing (a) Mg-TiC composite (b) Al-Ti-C preform quenched at 700 ^{0}C

Conclusions

The following conclusions can be drawn from the present study.

1. An *in-situ* TiC particulate reinforced magnesium matrix composite is successfully synthesized utilizing the exothermic reaction of the preform consisting of Al, Ti and C powders.
2. SHS reaction does not take place in the magnesium melt when the preform temperature below 450^{0}C.
3. Compact density below 68% of theoretical density fails to give rise to SHS reaction. Nearly 75% compact density is found to be optimum for SHS reaction and TiC particle distribution.

Acknowledgements

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