

## PREPARATION AND TESTING OF PROTOTYPE Mg<sub>2</sub>Si-Mg-TiC AND Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> COMPOSITES

### PRIPRAVA IN PREIZKUŠANJE PROTOTIPNIH KOMPOZITOV Mg<sub>2</sub>Si-Mg-TiC/TiB<sub>2</sub> IN Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub>

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Prejem rokopisa – received: 2009-05-25; sprejem za objavo – accepted for publication: 2009-07-07

In this work, the preparation of various light weight Mg-Mg<sub>2</sub>Si-TiC metal matrix composites and Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> ceramic composites has been described and the influence of their structure on mechanical response was discussed.

Mg-Mg<sub>2</sub>Si-TiC composites with continuous magnesium matrix densified to >95 % T.D. were fabricated by pressureless reactive infiltration of preforms made from Mg<sub>2</sub>Si and TiC powders. Infiltration was performed in an argon atmosphere at temperatures 700, 800 and 900 °C for 1 h. Trials made with Mg<sub>2</sub>Si preforms reinforced with TiB<sub>2</sub> were unsuccessful.

Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> ceramic composites densified to >97 % T.D. were prepared by pressureless reactive sintering of tablets made from Mg<sub>2</sub>Si and TiC or TiB<sub>2</sub> powders. The reactive sintering was performed at 1020 °C for 0.5–1 h under a protective argon atmosphere.

The phases present in the obtained composite samples have been identified by scanning electron microscopy/energy dispersive X-ray spectroscopy. In addition, room temperature tensile tests ( $R_m$ ,  $R_{p0.2}$ ,  $A$ ) and hardness measurements ( $HV$ ) were also undertaken.

The results have shown that Mg-Mg<sub>2</sub>Si-TiC composites are with tensile properties superior to that of conventional magnesium alloys while Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> samples combined high hardness (9–10 GPa) and low density (2.2–2.5 g/cm<sup>3</sup>).

Key words: Mg-Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> composites, reactive pressureless infiltration, reactive pressureless sintering, microstructural examination, tensile test, advanced, low-weight engineering materials

V delu opisujemo pripravo lahkih kompozitov Mg-Mg<sub>2</sub>Si-TiC s kovinsko matrico in keramične kompozite Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> ter preučevanje vpliva njihove strukture na mehanske lastnosti.

Kompozite Mg-Mg<sub>2</sub>Si-TiC s kontinuirno matrico iz magnezija, zgoščene do >95 % T.G., smo izdelali po postopku reakcijske infiltracije pri atmosferskem tlaku predoblik, stisnjenih iz prahov Mg<sub>2</sub>Si in TiC. Infiltracija je potekala v atmosferi argona, 1h pri temperaturah (700, 800 in 900) °C. Poskusi infiltracije predoblik Mg<sub>2</sub>Si, ojačenih s TiB<sub>2</sub> niso bili uspešni.

Keramične kompozite Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub>, zgoščene do >97 % T.G., smo izdelali z reakcijskim sintranjem pri atmosferskem tlaku tablet, stisnjenih iz prahov Mg<sub>2</sub>Si in TiC ali TiB<sub>2</sub>. Vzorce smo reakcijsko sintrali pri 1020 °C 1h v zaščitni atmosferi argona.

Mikrostrukturo in fazno sestavo pripravljenih vzorcev smo analizirali z vrstičnim elektronskim mikroskopom in XRD. Mehanske preiskave: natezni preizkus ( $R_m$ ,  $R_{p0.2}$ ,  $A$ ) in merjenje trdot ( $HV$ ) smo izvajali s standardnimi metodami pri sobni temperaturi.

Rezultati nateznega preizkusa so pokazali, da imajo kompoziti Mg-Mg<sub>2</sub>Si-TiC veliko boljše mehanske lastnosti kot navadne magnezijeve zlitine, medtem ko vzorci Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub> združujejo visoko trdoto (9–10 GPa) in nizko specifično maso (2.2–2.5 g/cm<sup>3</sup>).

Ključne besede: kompoziti Mg-Mg<sub>2</sub>Si-TiC in Mg<sub>2</sub>Si-TiC/TiB<sub>2</sub>, reakcijska infiltracija pri atmosferskem tlaku, reakcijsko sintranje pri atmosferskem tlaku, preiskave mikrostrukture, mehanske preiskave, lahki inženirski materiali prihodnosti

## 1 INTRODUCTION

Magnesium alloys and Mg-based composites are prospective candidates for light-weight structural materials<sup>1-2</sup>. However, most Mg alloys are of limited use in high performance applications due to their low mechanical properties<sup>3</sup>. Improvement of their mechanical properties could be achieved by reinforcement with different ceramic particulates, which has already been well demonstrated<sup>4,5</sup>, or by applying new magnesium-based compounds (such as Mg<sub>2</sub>Si) as the matrix constituent<sup>6</sup>. Among these, magnesium silicide (Mg<sub>2</sub>Si) is particularly attractive because of its superior characteristics such as high melting point (1085 °C), low density (1.99 g/cm<sup>3</sup>), high hardness (350–700 HV) and elastic modulus (120 GPa)<sup>7</sup>.

On the other hand, the major disadvantage of Mg<sub>2</sub>Si is its brittleness<sup>8-9</sup>, limiting the usage of bulk (sintered or hot pressed) Mg<sub>2</sub>Si as a structural material in engineering applications. A possible solution considered in this work is the formulation of ultra-light composite materials with a Mg<sub>2</sub>Si-Mg matrix reinforced with ceramic particulates (TiC, TiB<sub>2</sub>, B<sub>4</sub>C) in order to achieve an improvement in mechanical properties and brittleness.

## 2 EXPERIMENTAL

In the first set of experiments, Mg<sub>2</sub>Si-Mg-TiB<sub>2</sub> and Mg<sub>2</sub>Si-Mg-TiC composite samples were fabricated by pressureless infiltration of porous preforms with molten magnesium. Preforms were isostatically pressed from the various mixtures of commercial Mg<sub>2</sub>Si (99.5 %, 30 μm)

and TiC (99.5 %, 30 µm) or TiB<sub>2</sub> (99.5 %, 30 µm) powders, as listed in **Table 1**. Samples were cylinders 30 mm high and 20 mm in diameter. Infiltration was performed in a vacuum furnace in an argon atmosphere at temperatures of (700, 800 and 900) °C for 1 h.

**Table 1:** The volume fractions of various Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiB<sub>2</sub> mixtures used for preforms in infiltration, and tablets in sintering experiments

**Tabela 1:** Sestava različnih zmesi Mg<sub>2</sub>Si-TiC in Mg<sub>2</sub>Si-TiB<sub>2</sub> v volumenskih deležih (f%), uporabljenih za izdelavo predoblik za infiltracijo in tablet za sintranje

Mixture	Composition, f%		
	Mg <sub>2</sub> Si	TiC	TiB <sub>2</sub>
A	90	10	–
B	80	20	–
C	90	–	10
D	80	–	20

In the second set of experiments, composite samples were prepared by pressureless sintering of isostatically pressed tablets made from the same Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiB<sub>2</sub> mixtures listed in **Table 1**. Sintering was performed at 1020 °C, for 0.5–1 h in a protective argon atmosphere.

The as-synthesized composite samples were cut, machined and polished in accordance with standard procedures.

Microstructural characterization of fabricated composites was performed by optical and scanning electron microscopy (OM and SEM), whereas X-ray diffraction (XRD) measurements were applied to the samples to identify the phases and their crystal structure.

Quantitative determination of the volume percentage of Mg<sub>2</sub>Si, secondary phases and ceramic particles in the matrix, as well as the retained porosity, was performed by analysing the optical and scanning electron micrographs of as polished composite bars using the point counting method and image analysis and processing software.

Composite density measurements were carried out in accordance with Archimedes' principle, applying distilled water as the immersion fluid.

The initial density of the green compacts (preforms and tablets) was calculated from the mass and geometry of the samples.

Tensile tests were conducted on cylindrical tension-test specimens 5 mm in diameter and 25 mm gauge length using an automated servo-hydraulic tensile testing machine with a crosshead speed of 0.254 mm/60 s.

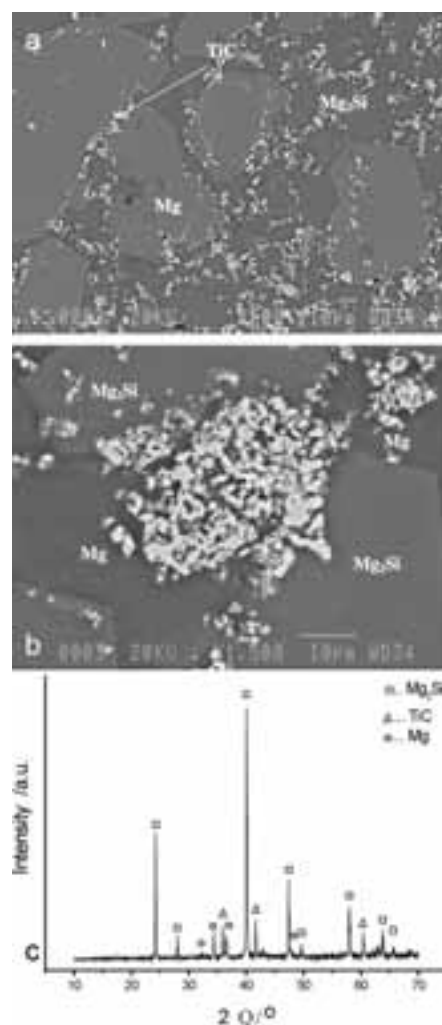
Vickers hardness (*HV*) measurements were performed at room temperature on polished composite samples as an average of 15 indentations. These measurements were made on an automatic digital tester using a pyramidal diamond indenter with a facing angle of 136° a 0.025 kg indenting load, 50 µm/s load applying speed, and a 15 s load holding time.

### 3 RESULTS AND DISCUSSION

#### Composites made by pressureless infiltration

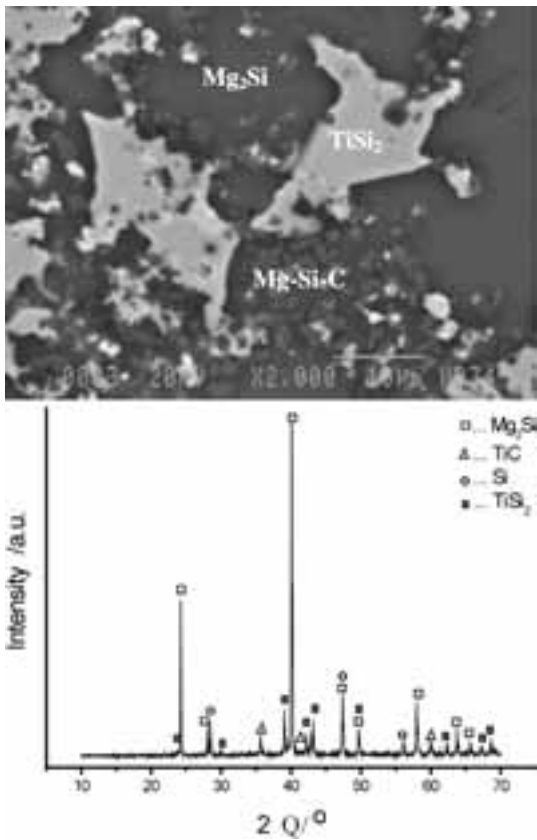
The calculated porosity of the preforms used was within the range of (30–35 ± 5) %. Based on the experimental findings, the pressureless infiltration of Mg<sub>2</sub>Si-TiC preforms with molten magnesium did not occur below 900 °C. At 900 °C, the infiltration was complete within 1h, resulting in composite samples with less than 5 % of retained porosity. At the same time, under the applied experimental conditions, the pressureless infiltration of Mg<sub>2</sub>Si-TiB<sub>2</sub> preforms was unsuccessful.

The microstructure and phase composition of the composite samples obtained is presented in **Figure 1 a, b, c**.



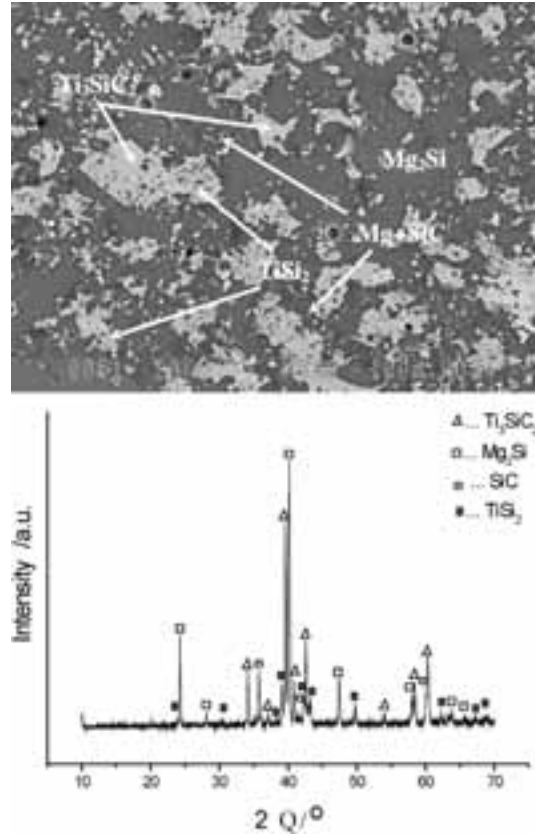
**Figure 1:** (a, b) SEM micrograph of a pressurelessly infiltrated preform with the initial composition of the preform skeleton of the volume fraction of 70 % Mg<sub>2</sub>Si-20 % TiC and an initial porosity of (30 ± 5) %. The phases detected are Mg, Mg<sub>2</sub>Si and TiC; (c) XRD of the sample shown in the **Figure 1a-c**.

**Slika 1:** (a, b) SEM posnetek mikrostrukture pri atmosferskem tlaku infiltrirane predoblike začetne sestave v volumenskih deležih 70 % Mg<sub>2</sub>Si-20 % TiC ter začetne poroznosti (30 ± 5) %. Ugotovljene faze so: Mg, Mg<sub>2</sub>Si in TiC; (c) XRD vzorcev, prikazanih na sliki 1 a-c.



**Figure 2:** (a) SEM micrograph of pressurelessly sintered composite sample with the initial volume composition 90 % Mg<sub>2</sub>Si and 10 % TiC, (b) XRD of the sample

**Slika 2:** (a) SEM-posnetek mikrostrukture vzorcev kompozitov začetne volumenske sestave 90 % Mg<sub>2</sub>Si in 10 % TiC, sintranih pri atmosferskem tlaku, (b) XRD vzorca



**Figure 3:** (a) SEM micrograph of pressureless sintered composite sample with the initial volume composition 80 % Mg<sub>2</sub>Si and 20 % TiC, (b) XRD of the sample

**Slika 3:** (a) SEM-posnetek mikrostrukture vzorca kompozita začetne volumenske sestave 80 % Mg<sub>2</sub>Si in 20 % TiC, (b) XRD vzorca

**Table 2:** Average room temperature tensile properties and Vickers hardness of pressurelessly infiltrated composite samples

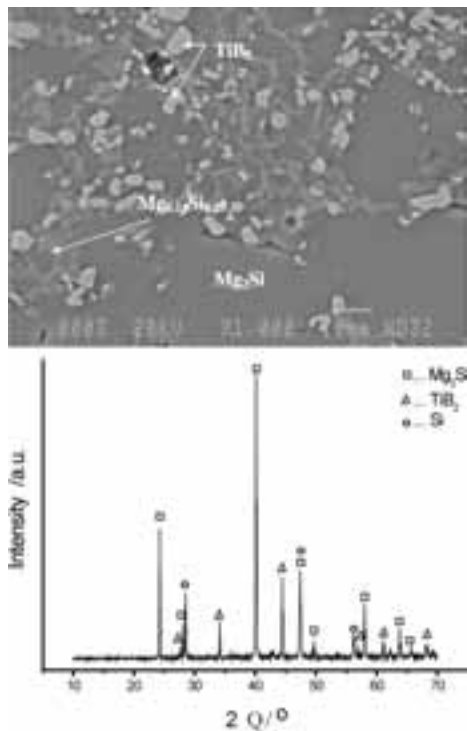
**Tabela 2:** Povprečne vrednosti nateznih lastnosti in Vickersove trdote, izmerjenih pri sobni temperaturi, pri vzorcih kompozita, infiltriranih pri atmosferskem tlaku

Composite initial composition fl%	Retained porosity (%)	Density $\rho/(g/cm^3)$	$E/$ (GPa)	Tensile strength $\sigma/MPa$	Vickers Hardness HV/GPa
63%Mg <sub>2</sub> Si+30%Mg+7%TiC	3.6 ± 0.4	2.03 ± 0.1	88 ± 9	186 ± 19	4.9 ± 0.5
56%Mg <sub>2</sub> Si+30%Mg+14%TiC	4.7 ± 0.5	2.32 ± 0.1	97 ± 10	197 ± 20	5.1 ± 0.5

**Table 3:** Average room temperature tensile properties and Vickers hardness of pressureless sintered composite samples

**Tabela 3:** Povprečne vrednosti nateznih lastnosti in Vickersove trdote, izmerjenih pri sobni temperaturi, pri vzorcih kompozita, sintranih pri atmosferskem tlaku

Initial composition fl%	Retained porosity (%)	Density $\rho/(g/cm^3)$	$E/$ (GPa)	Tensile strength $\sigma/MPa$	Vickers Hardness HV/GPa
Mg <sub>2</sub> Si+10%TiC	1.8 ± 0.2	2.25 ± 0.1	132 ± 13	487 ± 49	9.1 ± 1
Mg <sub>2</sub> Si+20%TiC	2.2 ± 0.2	2.53 ± 0.1	141 ± 14	532 ± 53	9.9 ± 1
Mg <sub>2</sub> Si+10%TiB <sub>2</sub>	2.3 ± 0.2	2.22 ± 0.1	134 ± 13	477 ± 48	9.6 ± 1
Mg <sub>2</sub> Si+20%TiB <sub>2</sub>	2.9 ± 0.3	2.43 ± 0.2	146 ± 15	528 ± 53	10.3 ± 1



**Figure 4:** (a) Microstructure of pressurelessly sintered composite sample with the initial volume composition 90 % Mg<sub>2</sub>Si and 10 % TiB<sub>2</sub>, (b) XRD of the sample

**Slika 4:** (a) Mikrostruktura vzorca kompozitov začetne volumenske sestave 90 % Mg<sub>2</sub>Si in 10 % TiB<sub>2</sub>, sintranih pri atmosferskem tlaku, (b) XRD vzorca

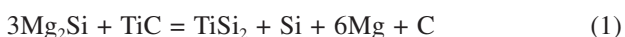
Large block-shaped Mg<sub>2</sub>Si particles of about 50 μm in size can be observed. The distribution of Mg<sub>2</sub>Si particles is in principle homogeneous with no agglomeration. The Mg matrix is continuous with dispersed fine TiC particles.

#### Composites made by pressureless sintering

Pressureless sintering at 1020 °C for 1 h of Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiB<sub>2</sub> samples made from mixtures A, B, C and D (**Table 1**) resulted in almost fully dense composite species with a retained porosity of less than 3 %.

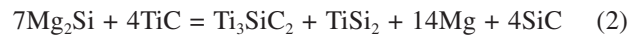
The microstructure of the composite samples obtained is presented in **Figures 2, 3 and 4**.

As illustrated in **Figure 2**, in pressureless sintered samples with the initial volume composition 90 % Mg<sub>2</sub>Si and 10 % TiC, the ceramic reinforcement reacted with Mg<sub>2</sub>Si transforming completely the initial TiC aggregates to dense TiSi<sub>2</sub> secondary grains. According to the X-ray diffraction patterns (**Figure 2 b**) of pressureless sintered Mg<sub>2</sub>Si samples with 10 % of TiC reinforcement, the main product of chemical reaction between Mg<sub>2</sub>Si and TiC is TiSi<sub>2</sub>. The presence of elemental silicon was also confirmed, while magnesium and carbon did not detected by XRD, Reaction 1:



The additional SEM investigation confirmed the presence of Mg-Si-C precipitates, most probably formed by further chemical reactions between elemental silicon, magnesium and carbon.

However, by increasing the amount of TiC reinforcement to 20 %, the reaction path was changed resulting in the formation of Ti<sub>3</sub>SiC<sub>2</sub>, TiSi<sub>2</sub> and SiC phases, **Figure 3a, b**, as well as the elemental magnesium, Reaction 2:



In pressurelessly sintered composite samples reinforced with TiB<sub>2</sub> particles large chunky Mg<sub>2</sub>Si particles were detected, **Figure 4**. During reactive pressureless sintering, these Mg<sub>2</sub>Si particles melt incongruently, forming a peritectic. Further densification of the samples proceeds via pressureless reactive liquid sintering. On cooling the samples, the molten phase crystallizes in the form of a continuous lace network with an average composition of Mg<sub>0.15</sub>Si<sub>0.85</sub>, with dispersed, fine TiB<sub>2</sub>, **Figure 4 a**.

## 4 CONCLUSION

The effect of fabrication techniques (reactive infiltration or pressureless reactive sintering) and processing conditions on the phase formation, microstructures and mechanical properties of Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiB<sub>2</sub> composites was examined.

It was found that pressureless reactive infiltration is effective in production of Mg<sub>2</sub>Si-Mg-TiC composites, whereas in the case of Mg<sub>2</sub>Si-Mg-TiB<sub>2</sub> samples, it was unsuccessful. The composite samples obtained by pressureless reactive infiltration of molten magnesium into a porous preform of Mg<sub>2</sub>Si with TiC ceramic reinforcement were designed to consist of a continuous magnesium matrix discontinuously reinforced with Mg<sub>2</sub>Si and TiC. Such a design was selected in order to reduce the well known brittleness of the Mg<sub>2</sub>Si phase, thereby creating an ultra-light structural material with excellent tensile properties.

On the other hand, pressurelessly reactive sintered Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiB<sub>2</sub> composites were designed to provide the improved hardness (9–10 GPa).

Although further experimental work is necessary to identify the real mechanism of pressureless densification of Mg<sub>2</sub>Si-TiC and Mg<sub>2</sub>Si-TiB<sub>2</sub> samples as well as the mechanical response of reinforced samples, the results obtained clearly demonstrate that routinely performed pressureless densification resulted in samples with almost theoretical density, proving at the same time the great industrial potential of this low-cost and highly productive fabrication method.

## Acknowledgement

This work was supported by funding from the Public Agency for Research and Development of the Republic

of Slovenia, as well as the Impol Aluminium Company and Bistral, d. o. o., from Slovenska Bistrica, Slovenia, under contract No. 1000-07-219308.

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