

PRODUCTION OF TiAl/Al₂O₃ AND TiAl/Ti₂AlC/Al₂O₃ COMPOSITES BY EXPLOSION SYNTHESIS

A. Amirkaveei^{1,*} and A. Saidi²

* abolfazlamirkaveh@gmail.com

Received: May 2012

Accepted: November 2012

¹ Department of Research and Development, Hormozgan Steel Complex, HOSCO, Hormozgan, Iran.

² Department of Materials Engineering, Isfahan University of Technology, Isfahan, Iran.

Abstract: Thermal explosion mode of combustion synthesis was used to fabricate TiAl-Al₂O₃ and TiAl-Ti₂AlC-Al₂O₃ composites from elemental powder mixtures of TiO₂, Al and C and characterized by XRD and Scanning Electron Microscopy (SEM) coupled with Energy-Dispersive Spectroscopy (EDS). The experimental results showed that thermite reaction of Al with TiO₂ caused TiAl-Al₂O₃ composite formation. By adding carbon powder to the thermite mixture, ternary carbide Ti₂AlC and TiAl-Ti₂AlC-Al₂O₃ composite were formed. With low carbon in thermite mixture, direct formation of Ti₂AlC without intermediate TiC was observed. After TiAl formation, Ti₂AlC precipitated from molten TiAl in the vicinity of carbon particles. SEM micrographs show that addition of carbon to thermite mixture changes the microstructure to a laminated form with plate-like grains.

Keywords: TiAl/Al₂O₃ composite; TiAl/Ti₂AlC/Al₂O₃ composite; Combustion synthesis; Microstructure

1. INTRODUCTION

TiAl-based composites are materials for high temperature applications. Due to their relatively low densities, excellent oxidation and corrosion resistance, composites such as TiAl-Al₂O₃ have received considerable interest. TiAl has a low density (3.8 g/cm³) and a high melting point (about 1773 K). Ternary carbides such as Ti₃AlC₂ and Ti₂AlC are promising ceramics because of their properties combining the merits of both metals and ceramics. They are good electrical and thermal conductors, they resist thermal shock and they have high flexural strength, thermal stability and high temperature oxidation resistance. Also, Al₂O₃ has been chosen as a ceramic reinforcement because of its advantageous thermo-mechanical behavior.

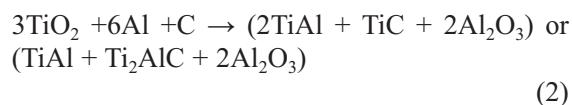
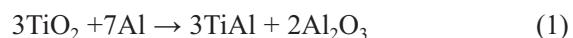
Recently, formation of different composites including TiAl-Ti₅Si₃, TiAl-Ti₂AlC and Ti₂AlC/Al₂O₃ by combustion synthesis was investigated by Yeh et al. [1-3]. Synthesis of Al₂O₃/TiAl/Nb₂O₅ using hot pressing reaction has also been reported [4]. Combustion synthesis was mostly used for the production of these composites [5-8]. For the production of bulk sample and for mechanical properties investigation, many researchers used hot press aided reaction synthesis [5, 9]. Mechanical

alloying was another method employed for the synthesis of TiAl/α-Al₂O₃ nanocomposite [10] and Ti₃AlC₂ powders [11].

In this work, TiAl/Al₂O₃ and TiAl/Ti₂AlC/Al₂O₃ composites were produced via thermal explosion synthesis. Formation of composites was investigated by XRD, SEM and EDX analysis.

2. EXPERIMENTAL PROCEDURES

Commercially available TiO₂ (99.99%, 5μm), Al (99.9%, 45μm) and C (99.0%, 10μm) powders were utilized for the production of composites. The powder, were blended according to the following reactions:



With regard to the fact that a part of carbon reacts with the oxygen trapped in pellet porosity, the amount of carbon is in a respective excess of 10 wt. % compared to the stoichiometric composition. The powders were mixed in a planetary mill under argon atmosphere for 5 min. For better compactibility, the

powder was mixed with ethanol and then cold-pressed under 200 kg/cm² into cylindrical samples with a diameter of 15 mm and a height of 30 mm. After drying, the pellets were placed in a crucible and were covered by alumina powder so that they were protected from atmosphere. For temperature measurements and visual observation, some of the samples were placed in a furnace without protection. A muffle furnace was used for explosion synthesis. Measurement of temperature during the process was made by high temperature sensors (SensyTemp TSH200) connected to a PC for data acquisition. After processing, the microstructure and phase composition of the synthesized composites were characterized by employing X-ray diffraction (XRD) and scanning electron microscopy (SEM) with energy dispersive analysis (EDS).

3. RESULTS AND DISCUSSION

3. 1. Production of TiAl/Al₂O₃ Composite

Reaction (1) is exothermic with an adiabatic temperature of about 1500°C. But the recorded combustion temperature, as shown in Fig. 1, was 1890°C which is because of the preheating of the sample to about 900°C. Fig. 1 also shows a comparison between results of this work and that obtained by other researchers [8]. As can be seen from the heating curve in Fig. 1, the chosen furnace temperature is 900°C which was 100°C higher than that in the previous work [8]. The trend of curves is

the same but at 900°C the reaction was complete in less than 2 min. Horvitz et al. reported that delayed thermal explosion was because of Al₂O₃ layer formation during milling. This layer acts as a diffusion barrier. In comparison to this research, blending of the mixture of powders in a ball mill under argon atmosphere for 5 min can reduce Al₂O₃ layer formation. Also, furnace temperature was higher, which accelerated the reaction.

During the reaction, TiO₂ was reduced by molten Al (Al₂O₃ formation) and residual Al reacted with Ti (TiAl formation).

Fig. 2 shows the X-ray diffraction patterns of the raw sample and the reacted sample. As can be seen, titanium aluminide TiAl was formed completely and there was no residual TiO₂ in the reacted sample. Main peak of TiAl was detected at 2θ angle equal to 38.825°. The main peak of TiO₂ in 2θ=25.275° vanished and a peak in 2θ=25.575° corresponding to Al₂O₃ was detected in the reacted sample. On the basis of Fig. 2, there is no other titanium aluminide in the product.

Microstructure of the obtained composite (TiAl/47.5 wt. % Al₂O₃) is shown in Fig. 3. Lighter particles are Al₂O₃ and darker areas are TiAl. Most of the Al₂O₃ particles (corundum) have a grain size smaller than 1 μm and are distributed uniformly. EDX analysis of a chosen area of the product is shown in Fig. 4. This analysis confirms the formation of TiAl in composite.

In thermal explosion mode of synthesis, in a moment, the whole sample ignited and the

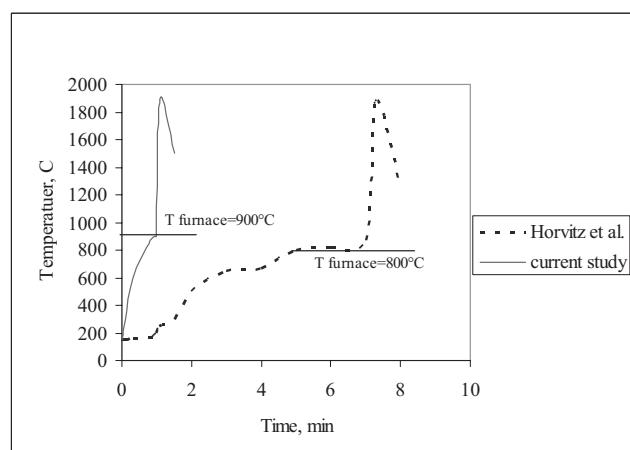


Fig.1. Comparison of temperature change curve of 3TiO₂+7Al with reference [8]

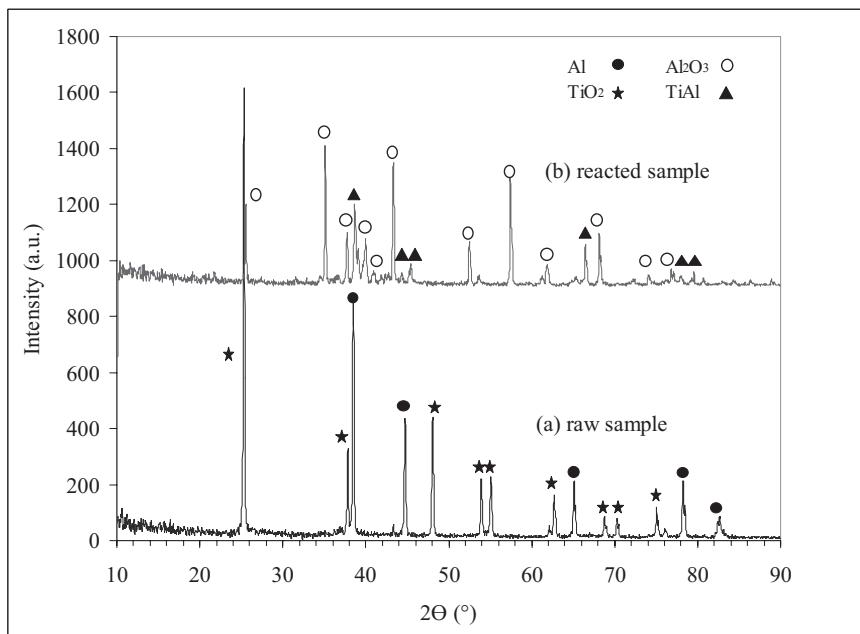


Fig. 2. Comparison of X-ray diffraction patterns of raw sample ($3\text{TiO}_2+7\text{Al}$) and reacted sample

reaction proceeded. After the formation of TiAl molten islands, Al_2O_3 was ejected forming round particles (Fig. 3). Because of fine raw materials (milled), Al_2O_3 forms around TiAl molten islands with a uniform size and distribution. It seems if an SHS (self propagating high temperature)

mode of synthesis were used, the movement of the molten front would cause to elongate Al_2O_3 form along to reaction front. It is clear that the round and uniform Al_2O_3 in the composite has an increased potential of mechanical properties.

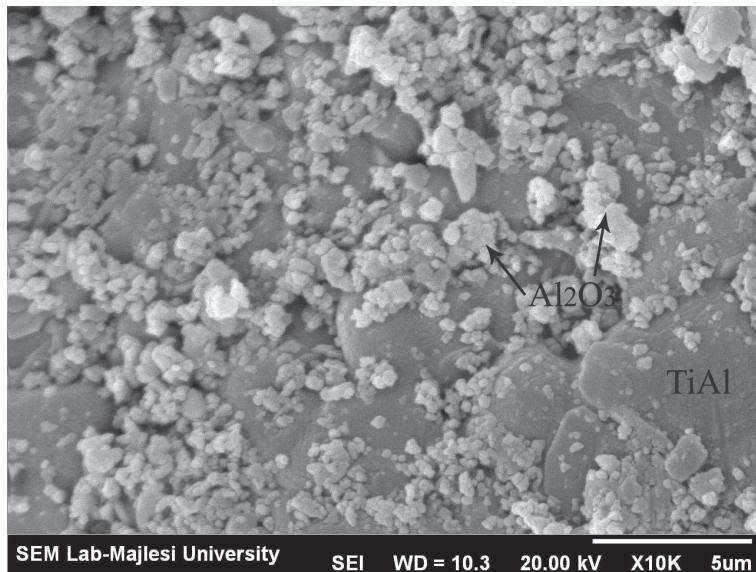


Fig. 3. SEM microphotograph of $\text{TiAl}/\text{Al}_2\text{O}_3$ composite

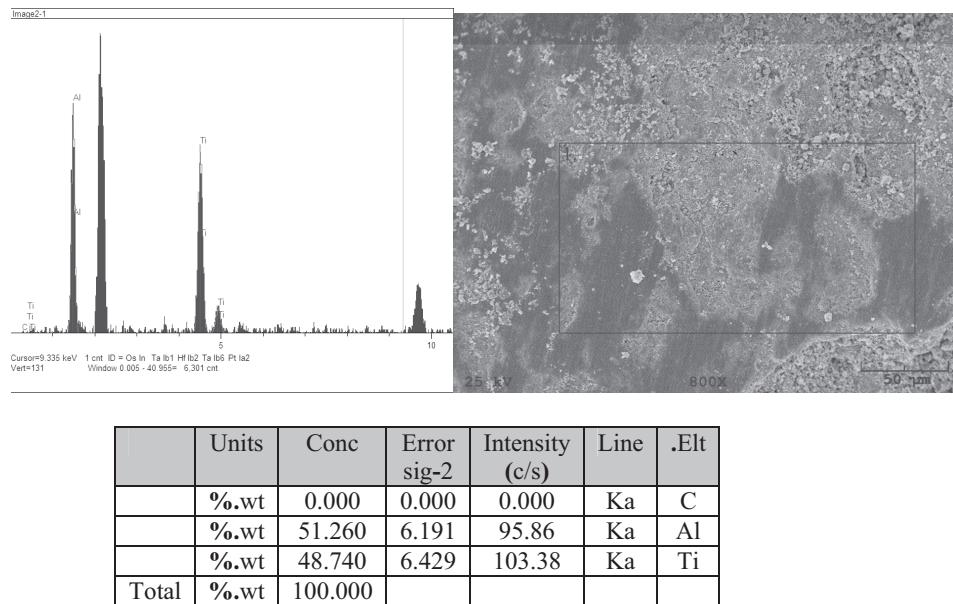


Fig. 4. EDX spectra of TiAl/Al₂O₃ composite from indicated area in SEM image

3. 2. Production of TiAl/Ti₂AlC/Al₂O₃ Composite

Based on reaction (2), samples of Al, TiO₂ and C were also prepared and ignited under argon atmosphere (The same procedure as reaction 1). After combustion, a porous and spongy pellet was obtained which is the general appearance of SHS product. X-ray patterns of raw and reacted samples are shown in Fig. 5. Main peak of Ti₂AlC was

detected in $2\Theta=39.475^\circ$. Other peaks corresponding to Ti₂AlC were found in $2\Theta=13.00^\circ$, 34.12° , 53.22° , 60.82° , 70.60° and 71.92° . Also, TiAl and Al₂O₃ were formed as can be seen in Fig. 5. In Fig. 5, there is no indication of the TiC phase. Insufficient carbon in reagent sample and conversion of some carbon to gas phase can be a reason for this situation. Also, TiC might be produced and then react with TiAl as follows:

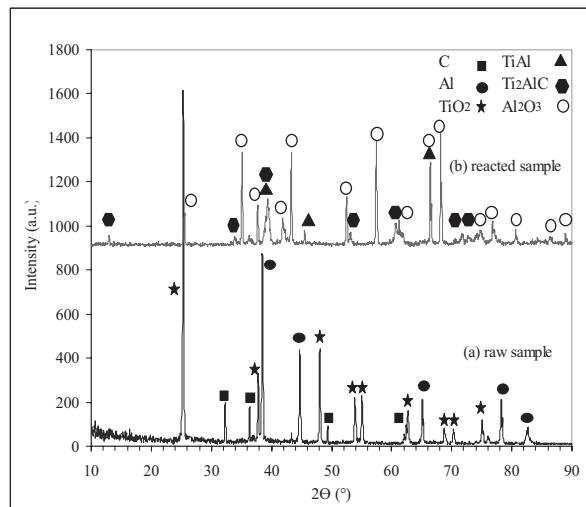


Fig. 5. Comparison of X-ray diffraction patterns of raw sample (3TiO₂+6Al+C) and reacted sample



Yan-Lin et al. [9] verified, via experiment, that when temperature reached 1100°C, carbon began to participate in TiC formation. At the same time, reaction (3) occurred. It is commonly believed that the reaction mechanism of this process is solution-precipitation [9]. Ti reacted with Al and released abundant heat which caused the systematic temperature to rise and C to react with unreacted Ti to form TiC. As the systematic temperature was raised higher, TiC was dissolved

into TiAl matrix and as the exothermic reaction finished, the systematic temperature dropped down and ternary phase Ti_2AlC precipitated [9].

This mechanism might be correct in the case of Ti, Al and C elemental powders. Nevertheless, in this research, the presence of TiO_2 and the formation of Al_2O_3 as a by-product was a barrier for carbon access to Ti and TiC formation. Also, as noted earlier, with low carbon in thermite mixture, direct formation of Ti_2AlC without intermediate TiC is reasonable. After TiAl formation, Ti_2AlC precipitated from molten TiAl

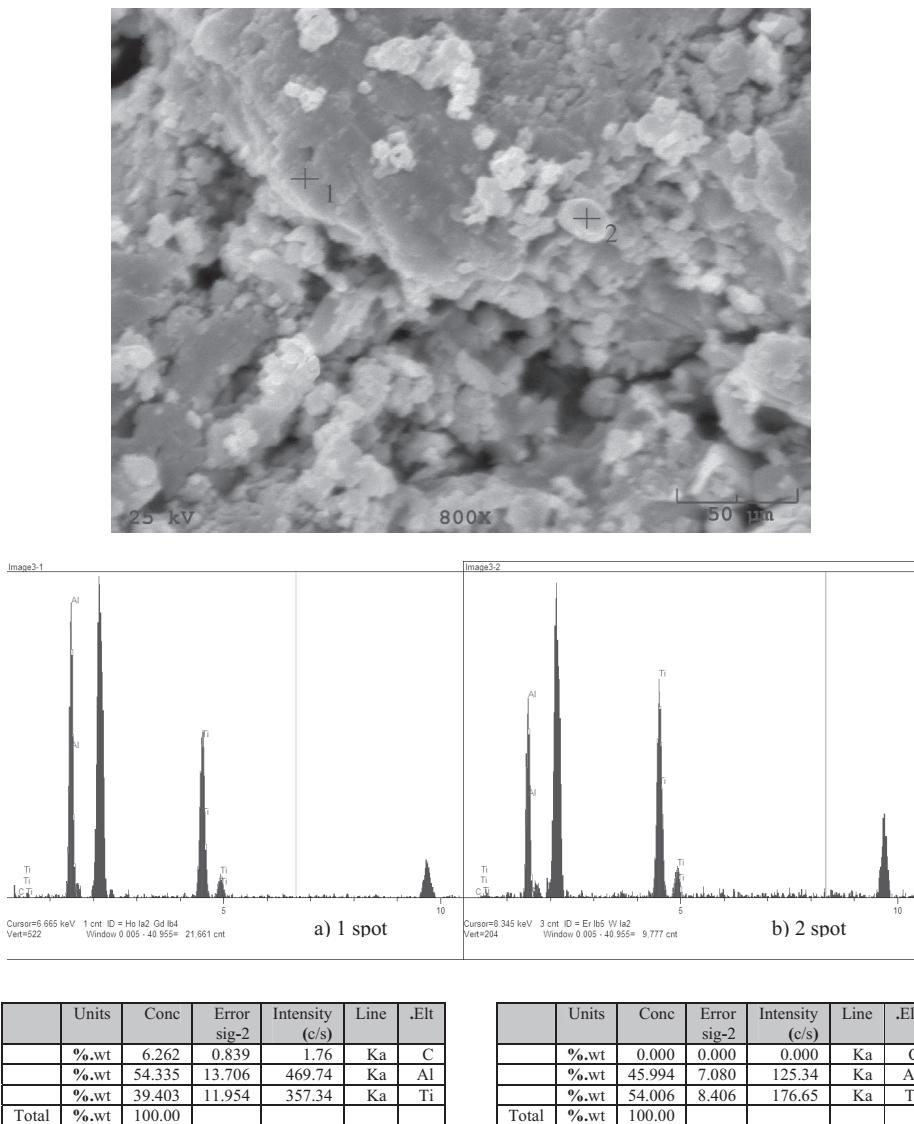


Fig. 6. EDX spectra of $\text{TiAl}/\text{Ti}_2\text{AlC}/\text{Al}_2\text{O}_3$ composite, a) spot 1 corresponding to Ti_2AlC and b) spot 2 correspond to TiAl

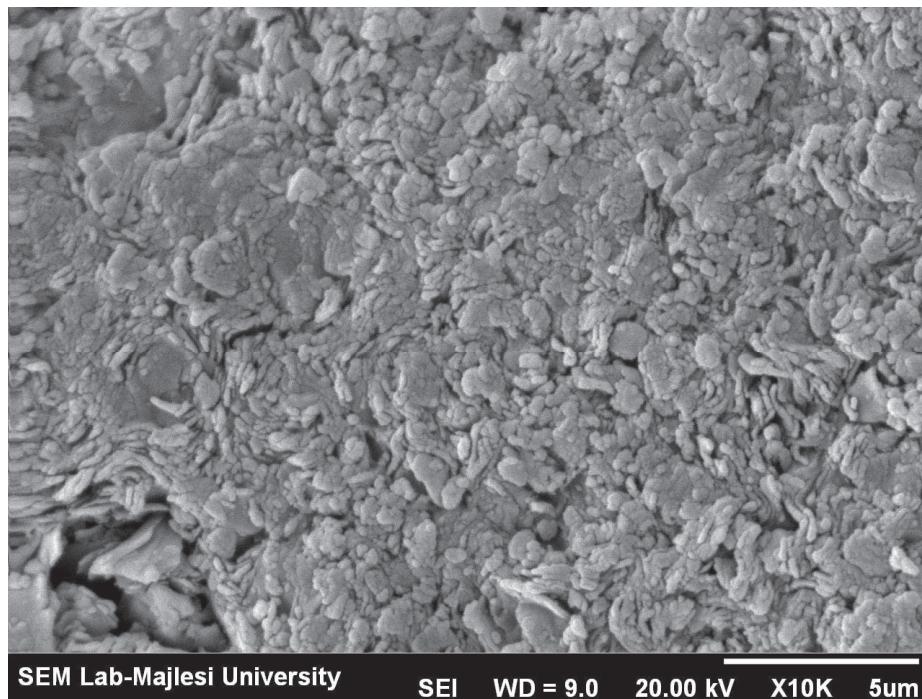


Fig. 7. SEM microphotograph of TiAl/Ti₂AlC/Al₂O₃ composite

in the vicinity of carbon particles.

In accordance with X-ray results, EDX analysis confirms the formation of TiAl and Ti₂AlC phases (Fig. 6). Chemical analysis of two spots indicates the formation of composite.

On the basis of this observation and XRD results, it can be concluded that TiAl/Ti₂AlC/Al₂O₃ is formed by explosion synthesis at 900 °C. Fig. 7 shows the microstructure of TiAl/ Ti₂AlC/Al₂O₃ composite. As can be seen, microstructure has a laminated form with plate-like grains. This structure is like Ti₃AlC₂/Al₂O₃ that was reported earlier [3].

Formation of carbon base gases in micro-scale can push different in situ molten islands outward and form a semi laminated structure with plate-like grains.

4. CONCLUSIONS

On the basis of experimental works in this investigation, the most important results are:

1. Production of TiAl/Al₂O₃ and TiAl/Ti₂AlC/Al₂O₃ from TiO₂, Al, C powders and without metallic titanium is feasible by

explosion synthesis at 900°C.

2. During reaction, TiO₂ is reduced by molten Al and TiAl is formed by reaction of metallic Ti and Al. Al₂O₃ was produced as a by-product.
3. With low carbon in thermite mixture, direct formation of Ti₂AlC without intermediate TiC was observed. After TiAl formation, Ti₂AlC precipitated from molten TiAl in the vicinity of carbon particles.
4. SEM micrographs show that most of Al₂O₃ particles have a grain size range smaller than 1 μm and are distributed uniformly in TiAl/Al₂O₃ composite. It is observed that TiAl/Ti₂AlC/Al₂O₃ composite has a laminated form with plate-like grains.

REFERENCES

1. Yeh, C. L., Li, R. F., "Formation of TiAl-Ti₅Si₃ and TiAl-Al₂O₃ in situ composites by combustion synthesis", *Intermetallics* 16 (2008) 64-70.
2. Yeh, C. L., Shen, Y. G., "Formation of TiAl

– Ti_2AlC in situ composites by combustion synthesis”, *Intermetallics* 17 (2009) 169–173.

- 3. Yeh, C. L., Kuo, C. W., Chu, Y. C., “Formation of Ti_3AlC_2/Al_2O_3 and Ti_2AlC/Al_2O_3 composites by combustion synthesis in $Ti-Al-C-TiO_2$ systems”, *J. Alloys Compd* 494 (2010) 132–136.
- 4. Taotao, A., “Microstructure and Mechanical Properties of In-situ Synthesized $Al_2O_3/TiAl$ Composites”, *Chin. J. Aeronautics* 21(2008) 559–564.
- 5. Peng, L. M., Li, Z., Li, H., Wang, J. H., Gong, M., “Microstructural characterization and mechanical properties of $TiAl-Al_2Ti_4C_2$ – Al_2O_3 – TiC in situ composites by hot-press-aided reaction synthesis”, *J. Alloys Compd* 414 (2006) 100–106.
- 6. Mas-Guindal, M. J., Turrillas, X., Hansen, T., Rodríguez, M. A., “Time-resolved neutron diffraction study of $Ti-TiC-Al_2O_3$ composites obtained by SHS”, *J. Eur. Ceram. Soc.* 28 (2008) 2975–2982.
- 7. Ramaseshan, R., Kakitsuji, A., Seshadri, S. K., Nair, N. G., Mabuchi, H., Tsuda, H., Matsui, T., Morii, K., “Microstructure and some properties of $TiAl-Ti_2AlC$ composites produced by reactive processing”, *Intermetallics* 7 (1999) 571–577.
- 8. Horvitz, D., Gotman, I., Gutmanas, E. Y., Claussen, N., “In situ processing of dense Al_2O_3 – Ti aluminide interpenetrating phase composites”, *J. Eur. Ceram. Soc.* 22 (2002) 947–954.
- 9. Yan-Lin, Ch., Ming, Y., Yi-Ming, S., Bing-Chu, M., Jiao-Qun, Z., “The phase transformation and microstructure of $TiAl/Ti_2AlC$ composites caused by hot pressing”, *Ceram. Inter.* 35 (2009) 1807–1812.
- 10. Forouzanmehr, N., Karimzadeh, F., Enayati, M. H., “Synthesis and characterization of $TiAl/\alpha$ - Al_2O_3 nanocomposite by mechanical alloying”, *J. Alloys Compd* 478 (2009) 257–259.
- 11. Yang, C., Jin, S., Xu, C., Jia, S., “Fabrication of high-content Ti_3AlC_2 powders by mechanical alloying”, *Key Engineering Materials, Key Eng. Mater.* 368-372 (2008) 983-985.