PRODUCTION OF $TiAl/Al_2O_3$ AND $TiAl/Ti_2AlC/Al_2O_3$ COMPOSITES BY EXPLOSION SYNTHESIS

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Abstract: *Thermal explosion mode of combustion synthesis was used to fabricate $TiAl-Al_2O_3$ and $TiAl-Ti_2AlC-Al_2O_3$ composites from elemental powder mixtures of TiO_2, 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 TiO_2 caused $TiAl-Al_2O_3$ composite formation. By adding carbon powder to the thermite mixture, ternary carbide Ti_2AlC and $TiAl-Ti_2AlC-Al_2O_3$ composite were formed. With low carbon in thermite mixture, direct formation of Ti_2AlC without intermediate TiC was observed. After TiAl formation, Ti_2AlC 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: *$TiAl/Al_2O_3$ composite; $TiAl/Ti_2AlC/Al_2O_3$ 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 $TiAl-Al_2O_3$ have received considerable interest. TiAl has a low density (3.8 g/cm^3) and a high melting point (about 1773 K). Ternary carbides such as Ti_3AlC_2 and Ti_2AlC 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, Al_2O_3 has been chosen as a ceramic reinforcement because of its advantageous thermo-mechanical behavior.

Recently, formation of different composites including $TiAl-Ti5Si_3$, $TiAl-Ti_2AlC$ and Ti_2AlC/Al_2O_3 by combustion synthesis was investigated by Yeh et al. [1-3]. Synthesis of $Al_2O_3/TiAl/Nb_2O_5$ 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 $TiAl/\alpha-Al_2O_3$ nanocomposite [10] and Ti_3AlC_2 powders [11].

In this work, $TiAl/Al_2O_3$ and $TiAl/Ti_2AlC/Al_2O_3$ composites were produced via thermal explosion synthesis. Formation of composites was investigated by XRD, SEM and EDX analysis.

2. EXPERIMENTAL PROCEDURES

Commercially available TiO_2 (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:

$$3TiO_2 + 7Al \rightarrow 3TiAl + 2Al_2O_3 \quad (1)$$

$$3TiO_2 + 6Al + C \rightarrow (2TiAl + TiC + 2Al_2O_3) \text{ or } (TiAl + Ti_2AlC + 2Al_2O_3) \quad (2)$$

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 cruse 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/Al_2O_3 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 Al_2O_3 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 Al_2O_3 layer formation. Also, furnace temperature was higher, which accelerated the reaction.

During the reaction, TiO_2 was reduced by molten Al (Al_2O_3 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 TiO_2 in the reacted sample. Main peak of TiAl was detected at 2Θ angle equal to 38.825°. The main peak of TiO_2 in 2Θ=25.275° vanished and a peak in 2Θ=25.575° corresponding to Al_2O_3 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. % Al_2O_3) is shown in Fig. 3. Lighter particles are Al_2O_3 and darker areas are TiAl. Most of the Al_2O_3 particles (corundum) have a grain size smaller than 1μm 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

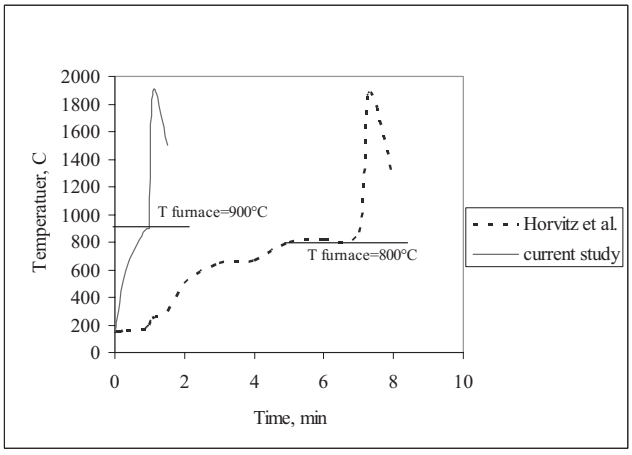


Fig.1. Comparison of temperature change curve of $3TiO_2$+7Al with reference [8]

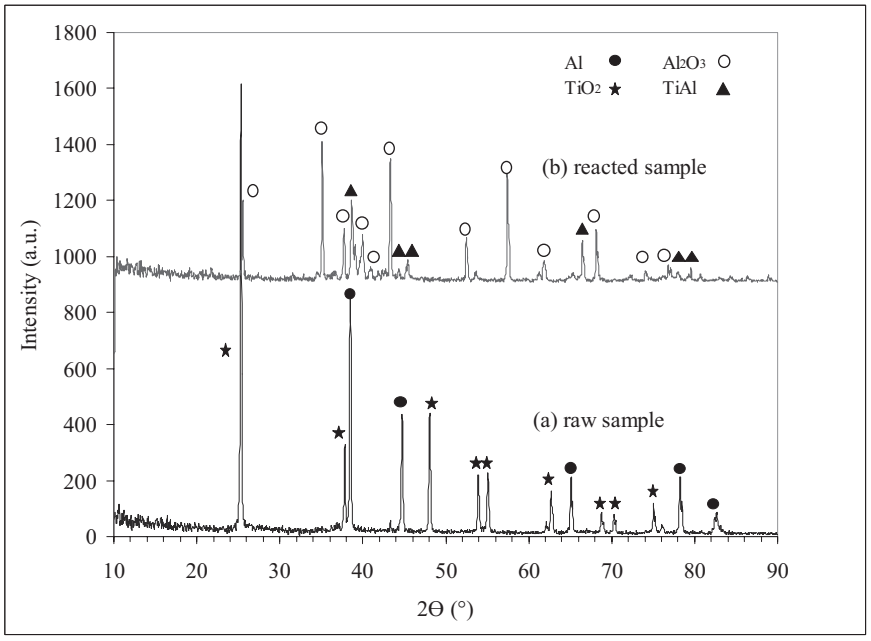


Fig. 2. Comparison of X-ray diffraction patterns of raw sample ($3TiO_2$+7Al) and reacted sample

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.

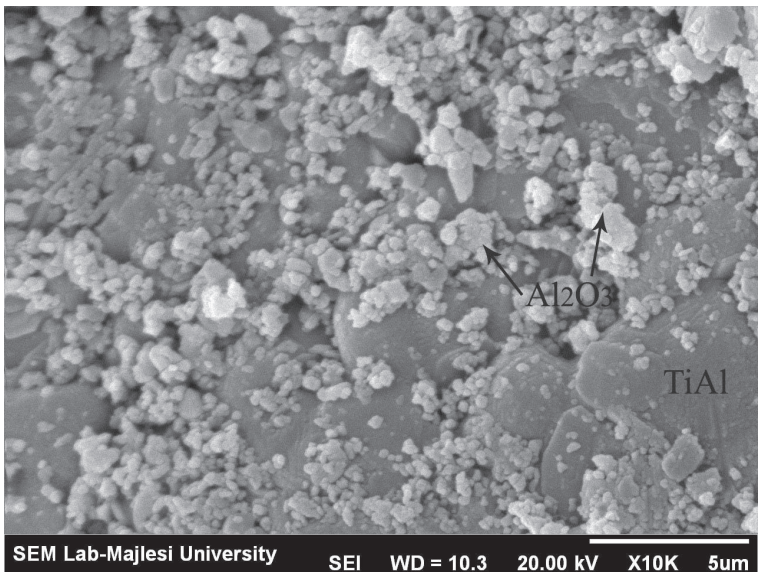
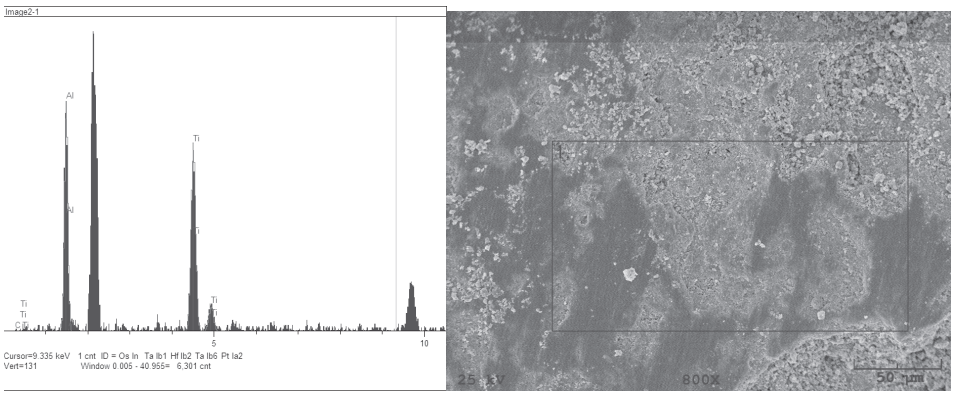


Fig. 3. SEM microphotograph of TiAl/Al_2O_3 composite



	Units	Conc	Error sig-2	Intensity (c/s)	Line	.Elt
	%.wt	0.000	0.000	0.000	Ka	C
	%.wt	51.260	6.191	95.86	Ka	Al
	%.wt	48.740	6.429	103.38	Ka	Ti
Total	%.wt	100.000				

Fig. 4. EDX spectra of $TiAl/Al_2O_3$ composite from indicated area in SEM image

3. 2. Production of $TiAl/Ti_2AlC/Al_2O_3$ Composite

Based on reaction (2), samples of Al, TiO_2 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 Ti_2AlC was detected in 2Θ=39.475°. Other peaks corresponding to Ti_2AlC were found in 2Θ=13.00°, 34.12°, 53.22°, 60.82°, 70.60° and 71.92°. Also, TiAl and Al_2O_3 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:

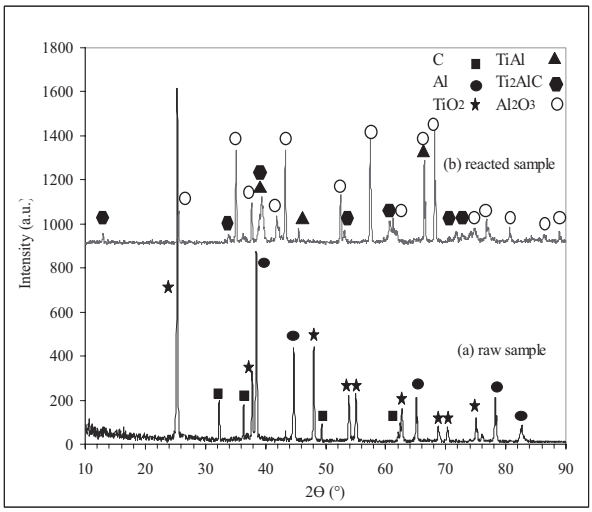
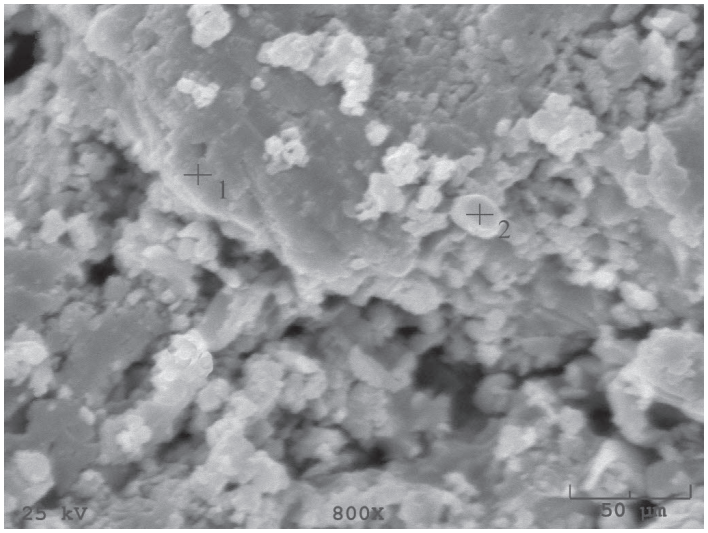


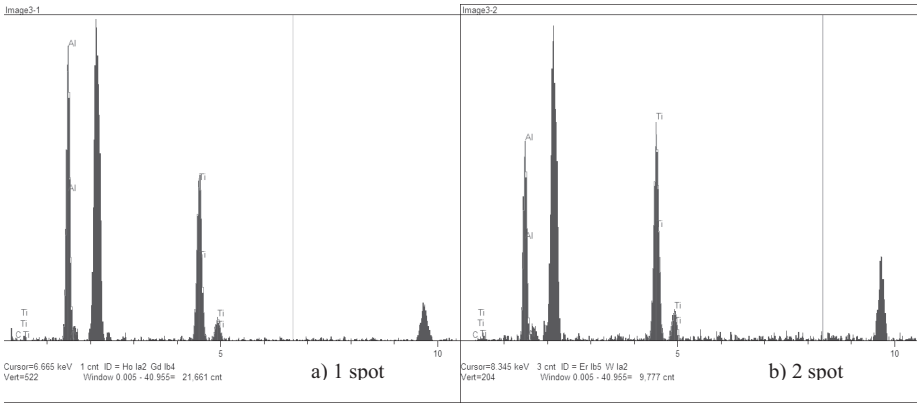
Fig. 5. Comparison of X-ray diffraction patterns of raw sample ($3TiO_2$+6Al+C) and reacted sample

$$TiAl + TiC \rightarrow Ti_2AlC \quad (3)$$

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





	Units	Conc	Error sig-2	Intensity (c/s)	Line	.Elt
	%.wt	6.262	0.839	1.76	Ka	C
	%.wt	54.335	13.706	469.74	Ka	Al
	%.wt	39.403	11.954	357.34	Ka	Ti
Total	**%.**wt	100.00				

	Units	Conc	Error sig-2	Intensity (c/s)	Line	.Elt
	%.wt	0.000	0.000	0.000	Ka	C
	%.wt	45.994	7.080	125.34	Ka	Al
	%.wt	54.006	8.406	176.65	Ka	Ti
Total	**%.**wt	100.00				

Fig. 6. EDX spectra of $TiAl/Ti_2AlC/Al_2O_3$ composite, a) spot 1 corresponding to Ti_2AlC and b) spot 2 correspond to TiAl

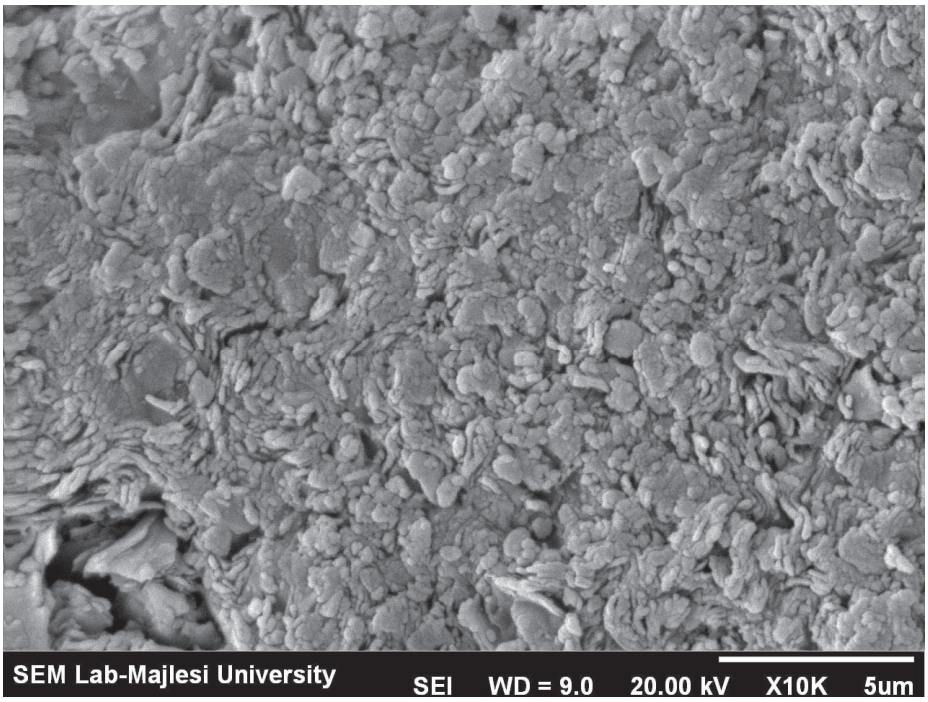


Fig. 7. SEM microphotograph of $TiAl/Ti_2AlC/Al_2O_3$ composite

in the vicinity of carbon particles.

In accordance with X-ray results, EDX analysis confirms the formation of TiAl and Ti_2AlC 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 $TiAl/Ti_2AlC/Al_2O_3$ is formed by explosion synthesis at 900 °C. Fig. 7 shows the microstructure of $TiAl/Ti_2AlC/Al_2O_3$ composite. As can be seen, microstructure has a laminated form with plate-like grains. This structure is like Ti_3AlC_2/Al_2O_3 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 $TiAl/Al_2O_3$ and $TiAl/Ti_2AlC/Al_2O_3$ from TiO_2, Al, C powders and without metallic titanium is feasible by explosion synthesis at 900°C.
2. During reaction, TiO_2 is reduced by molten Al and TiAl is formed by reaction of metallic Ti and Al. Al_2O_3 was produced as a by-product.
3. With low carbon in thermite mixture, direct formation of Ti_2AlC without intermediate TiC was observed. After TiAl formation, Ti_2AlC precipitated from molten TiAl in the vicinity of carbon particles.
4. SEM micrographs show that most of Al_2O_3 particles have a grain size range smaller than 1 μm and are distributed uniformly in $TiAl/Al_2O_3$ composite. It is observed that $TiAl/Ti_2AlC/Al_2O_3$ composite has a laminated form with plate-like grains.

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