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# Mechanically activated combustion synthesis of $Fe_3Al$ composite powders reinforced with sub-micrometer TiC particles

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ABSTRACT

The aim of this work was to develop a process to synthesize $Fe_3Al$ matrix composite reinforced with nano scale TiC particles from pure elemental powders as reactants, employing combustion synthesis. The effects of particle size, pre-milling time of precursors on ignition temperature and final powder microstructure were studied. The synthesized composite powders were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM) equipped with energy dispersive X-ray spectrometry (EDX). The targeted powder chemistry, microstructure and optimum spatial distribution of TiC particles were obtained using a pre-milling time of 30 min and a furnace temperature of 1100 °C. Powders synthesized in this condition contained only $Fe_3Al$ and TiC phases. Because of its high hardness and the presence of nanosized reinforcements, this composite powder can be considered a good candidate for thermal spray applications in manufacturing wear resistant parts.



## 1. Introduction

Iron aluminides ($Fe_3Al$) have recently received a great deal of attention due to their low cost, low density, ease of fabrication and resistance to oxidation and corrosion [1]. This combination of properties makes them suitable for utilization in various applications such as heating elements, furnace fixtures, heat exchanger piping, sintered metal gas filters, etc. [2]. Nonetheless, adoption of this type of alloys is limited due to their poor ductility at ambient temperature, mediocre high-temperature strength and relatively low wear resistance [3–5]. Chromium (2 at.%) is usually added to improve ductility of $Fe_3Al$ at room temperature [6]. Nevertheless, if its wear resistance could be increased, $Fe_3Al$ would constitute an attractive coating material for protecting steel substrates.

Creating a two-phase metal matrix composite (MMC), by adding hard ceramic particles to a metallic matrix, is an efficient way to improve hardness and high-temperature strength. Among all the potential reinforcement materials for improving mechanical properties of iron aluminides, TiC has been found to be a suitable one, based on its high hardness, low density, low chemical reactivity and good compatibility with the iron aluminide matrix [7,8]. The reinforcement particles could be added to the matrix either in-situ or ex-situ. The former consists of the formation or precipitation of the particles directly from the matrix. The latter consists of embedding pre-formed solid particles into the matrix [9,10].

## 2. Theory

MMC produced by in-situ precipitation of TiC particles take advantage from clean and strong matrix-reinforcement interfacial bonding. Moreover, in-situ synthesized particles, compared to ex-situ ones, are usually finer and more homogenously distributed [11]. Recently, several studies were devoted to the in-situ synthesis of iron aluminide intermetallic composites by mechanical alloying [12,13]. However, the synthesis of iron aluminides by mechanical alloying from elemental powders requires long high-energy milling times (15–25 h) [14]. In one of the most recent studies, mechanical alloying in a high-energy ball mill was performed for 6 h following a heat treatment of 2 h at 1000 °C to precipitate TiC particles within the $Fe_3Al$ matrix [15]. Such a process is relatively expansive when significant quantities of powders are required for coating large industrial components.

A simple alternative production method for synthesizing ceramics and intermetallics is combustion synthesis [16,17]. Combustion synthesis has recently received particular attention due to

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its high efficiency, short process-time, and low production cost. Moreover, Deevi and Sikka [18] have shown that it is possible to benefit from the highly exothermic reaction between molten aluminum and iron to favour the formation of $Fe_3Al$. Similarly, Yeh et al. [19], have shown that although the adiabatic temperature of synthesis of $(3Ni+3Al+3Ti+B_4C) = 3NiAl+2TiB_2+TiC$ is 2627 °C (the ignition temperature of reactants was considered to be the melting point of aluminum 660 °C) and the combustion temperature measured in their study was 1200 °C. Although encouraging results have been recently achieved on combustion synthesis of $Fe_3Al$-TiC composites, incomplete conversion of reactants and the presence of the unreacted raw materials in the final products were reported [20]. Similarly, no detailed information regarding process control and optimum process conditions was provided. Hence, more detailed analysis of the microstructural evolution is necessary to determine the mechanism of formation of $Fe_3Al$-TiC by combustion synthesis. The present study investigates the synthesis of $Fe_3Al$-TiC powders in Fe-Al-Ti-C system through combustion synthesis in thermal explosion mode and discusses the formation mechanism of TiC.

## 3. Material and methods

### 3.1. Raw materials and preparation of pellets

The starting materials consisted of iron, aluminum, titanium, graphite and chromium powders. The suppliers, purity and particle size are listed in Table 1.

Fe and Al were blended using a molar ratio of 3:1 and 2 at.% of Cr was added to the powder mixture. Similarly, Ti and graphite were mixed while respecting a molar ratio of 1:1. All the blends were prepared in a 2L V-blender. Some of the premixes were milled during 30 min using a high-energy ball mill (Zoz GmbH, Wenden, Germany) to evaluate the effect of mechanical activation on the synthesis process. Hardened steel balls of 10 mm in diameter were used as the milling media. Each premix was prepared to obtain composite powders of $Fe_3Al$ containing 30-50-70 mol% of TiC. The TiC concentrations were selected based on the recent investigation on the wear behavior of $Fe_3Al$-TiC coatings prepared by High-Velocity Oxygen Fuel (HVOF) [15]. The resulting premixes were compacted at room temperature using a uniaxial pressure of 200 MPa. As such, green cylindrical pellets, having an outside diameter of 35 mm and a height of 4 mm, were pressed to a green density corresponding to 75% of the theoretical density [21].

### 3.2. Combustion synthesis process

The combustion reaction was carried out inside a preheated horizontal tube furnace. Combustion synthesis was performed by swiftly placing the pellets in the hot section of the preheated furnace to achieve the highest possible heating rate since low heating rates lead to pre-combustion diffusion reactions, forming intermediate and undesirable phases [22]. The samples were placed in the high temperature section of the furnace for 5 min. This approach provided a heating rate of approximately 500 °C/min, which was sufficiently high to trigger the thermal explosion mode of combustion synthesis. A reducing atmosphere, made of 95 vol% of argon and 5 vol% of hydrogen, was used for all the tests. Upon completion of the synthesis reaction, the samples were cooled in air. Contrary to what was put forward by Merzhanov et al. [23], annealing of the final powder was not carried out in this study. The rational behind this decision was to make sure that the synthesized powder could be easily ground back to a powder form for HVOF coatings. Moreover, as it will be seen below, no sign of supersaturated solid solution could be identified in the final product. Since the synthesis products had a fragile and sponge-like structure, they could be easily ground to powder form, using a mortar and pestle.

**Table 1**
Raw materials used in this study.

| Materials | Supplier | Particle size (μm) | Purity % |
|---|---|---|---|
| Fine iron powder | Micronmetals | <44 | 99.9 |
| Coarse iron powder | | <80 | |
| Fine titanium powder | Micronmetals | <20 | 99.7 |
| Coarse titanium powder | | <80 | |
| Aluminum powder | Ampal | <20 | 99.0 |
| Graphite powder | Fisher Scientific | <10 | 99.0 |
| Chromium powder | ACuPowder Int. | <20 | 99.0 |

### 3.3. Material analysis

Particle size analysis was carried out using a particle size analyzer (MALVERN; Mastersizer 2000). Characterization of the phases present in the synthesized powders was performed by X-ray diffraction (SIEMENS, D5000, Karlsruhe, Germany) using Cu K-α radiation ($\lambda = 1.5406$ Å). Microhardness of the synthesized powders was measured with a Vickers microhardness indenter (MMT-X7A, MATSUZAMA CO., Japan) utilizing a load of 50 gf for 13s. To this end, the powders were mounted in epoxy resin, ground using 180–600 grit emery paper and finally polished with 6 μm diamond paste and alumina suspension to obtain a mirror-like finish. The microstructure and elemental distribution of the synthesized powders were characterized by scanning electron microscopy (JEOL, 840, Tokyo, Japan) equipped with an energy-dispersive X-ray spectrometer (EDS) (PGT Avalon). A microprobe (Cameca SX-100)) was also utilized to perform elemental mapping in wavelength dispersive X-ray spectrometry (WDS). Differential scanning calorimetry (DSC) (NETZSH STA 449 F3) was employed to follow phase transformations during synthesis using a maximum temperature of 1200 °C and a heating rate of 20 °C/min. These tests were carried out in a flowing atmosphere of purified argon at a flow rate of 50 mL/min. Samples were weighed before and after synthesis and no noticeable differences were observed, indicating no mass loss or gain during synthesis.

## 4. Results and discussion

### 4.1. Powder production

Fig. 1 presents the typical XRD pattern of the synthesized products through the combustion synthesis of the un-milled materials at 1200 °C. Relatively coarse particles of both titanium and iron ($D_{50} \approx 80$ μm) were used for this synthesis. As seen, along with FeAl, the final products also contain un-reacted Fe. The presence of sharp Ti and Fe peaks as well as titanium aluminides, such as $Ti_3Al$ and $Ti_2Al$, indicates that the reaction was incomplete. No TiC was detected in the products.

To investigate the effect of pre-milling on the formation of $Fe_3Al$-TiC, a mixture of relatively coarse powders ($D_{50} \approx 80$ μm) was pre-milled for 30 min prior to combustion synthesis. Fig. 2 shows the XRD pattern of the products, pre-milled for 30 min and synthesized at 1200 °C. It can be seen that $Fe_3Al$ and TiC were formed, no remaining Ti and Fe are present but the final product still contained undesirable $Ti_3AlC$ and $Ti_3Al$ phases. Therefore, 30 min pre-milling of coarse particles is not an appropriate process to produce the desired composite. This is in accordance with data reported in literature. For instance, it has been found that large Fe particle size leads to the formation of non-stoichiometric