

Fabrication of bulk Al-TiB₂ nanocomposite by spark plasma sintering of mechanically alloyed powder

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ABSTRACT

Production of Al-TiB₂ nanocomposite powder was studied by using mechanical alloying (MA). The target was to obtain Al-20 wt. % TiB₂ metal matrix nanocomposite by mechanical alloying of pure Ti, B and Al powder mixture. A double step process was used to prevent the formation of undesirable phases like Al₃Ti intermetallic compound, which has been described in our previous papers. The resultant powder was consolidated by spark plasma sintering (SPS). The structural characteristics of powder particles and sintered samples were studied by X-ray diffractometry (XRD), scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Hardness measurements were conducted on the cross section of powder particles and sintered sample.

Keywords: aluminum matrix nanocomposite, mechanical alloying, in situ TiB₂, spark plasma sintering

1 INTRODUCTION

TiB₂ is known as a suitable reinforcing phase for Al-base composites because of its thermodynamic stability. It also presents a high modulus, excellent refractory properties and a high resistance to plastic deformation even at high temperatures [1-3]. In the last decade in situ metal matrix composites (MMCs) have been developed, in which the reinforcements are synthesized in the metal matrix during fabrication, by chemically reacting the constituents. In comparison to traditionally fabricated materials, in situ MMCs undergo less degradation at high temperatures. This is because of their superior thermodynamic stability, stronger interface bonding resulting from the clean reinforcement-matrix interfaces and finer and more uniform distribution [4]. Several techniques including exothermic dispersion, reactive-gas injection, reactive sintering, reactive milling and mechanical alloying have been developed to fabricate in situ MMCs [4-5]. MA as a solid state powder processing method has been extensively used to fabricate in situ ceramic particle reinforced MMCs. MA has an advantage over other in situ fabrication routes as it is capable of producing nanostructured composite powder with high uniformity [5].

In previous investigations it has been reported that during the in situ synthesis of Al-TiB₂ composites form different starting powder mixtures, Al₃Ti intermetallic compound is also formed, which is brittle and has been reported to considerably reduce the fatigue life of composites [6-7]. Therefore it is of interest to eliminate the formation of Al₃Ti intermetallic compound.

A challenge in processing of nanostructured materials is that long time exposure at high temperature sintering, often results in severe grain growth. Several consolidation techniques such as hot-pressing, shock consolidation, sintering with the application of ac currents, pulsed electric current and spark plasma sintering (SPS) are introduced to overcome these difficulties [8]. SPS is a newly developed rapid sintering technique with a great potential for achieving fast densification results with minimal grain growth in a short sintering time. It is proven that enhanced sinterability of powders subjected to SPS mainly associated with particle surface activation and increased diffusion rates on the contact zones caused by applied pulse current [9].

In this study, Al-20 wt. %TiB₂ nanostructured composite powder was successfully synthesized by double step mechanical alloying without the formation of unwanted phases. Consolidation of the powder was conducted via SPS technique and microstructure the powder and sintered bulk nanocomposite was also investigated.

2 EXPERIMENTAL PROCEDURE

Elemental Al, Ti and B powders were used as starting materials. The aluminum powder with an average particle size of 63 µm was supplied from ECKA Granulate, Velden. The titanium powder with a particle size of 40-60 µm was obtained from GKN Sinter Metal Filters, Radevormwald, and the Merck boron powder had a particle size of about 2 µm. Powder mixtures were milled by a Fritsch planetary ball mill with a rotating speed of 360 rpm. The ball to powder weight ratio was chosen to be 10 and the diameter of the chromium steel balls was 15 mm. The hard chromium steel vial was evacuated and filled with argon to prevent oxidation during the mechanical alloying process. To avoid severe adhesion of aluminum powder to the balls and the vial, 1 wt. % zinc stearate was added to the mixture as the controlling agent.

A double step process was used to obtain an in situ Al-TiB₂ metal matrix powder. In the first step Ti and B powders were milled with the composition of Al- 62.01 wt. % Ti-27.99 wt. % B. This powder was then used in further milling with additional aluminum powder to achieve the Al- 20 wt. % TiB₂.

Powders were sintered by a FCT HP D 250 spark plasma sintering (SPS) equipment, at Fraunhofer Institute for Manufacturing and Advanced Materials, Powder Metallurgy and Composite Materials, Franhofer, Germany. Four sets of variables were examined from which one was chosen for consolidation of the powder. Powder was placed into a graphite die with a diameter of 40 mm and compacted under argon atmosphere with an applied pressure of 35 MPa. The whole process lasted 600 sec and the maximum temperature was 550 C with the dwell time of 0 sec. The changes of the SPS parameters during the consolidation process are shown in Fig. 1. Compacted disks of 40 mm diameter and 5 mm thickness were obtained from the SPS. The density of the SPS materials was determined using Archimedes' principle.

Investigation of the structural changes during mechanical alloying and after sintering was conducted by a SEIFERT 30033 PTS diffractometer employing monochromatic Cu K_α radiation ($\lambda = 0.15406$ nm). XRD scans were performed with a step size of 0.05° in 2 θ and a dwell time per step of 20 s. The cross-sectional microstructures of the powder particles and the sintered samples were studied by a LEO scanning electron microscope (SEM). Microhardness examination of Al-TiB₂ powders was conducted by a Leco testing machine while the hardness of compacted samples was measured by a Zwick microhardness machine.

3 RESULTS AND DISSCUSION

Fig. 2 shows the X-ray diffraction patterns of the Al- 62 wt.% Ti- 28 wt.% B and Al- 20 wt.% TiB₂ powders after different milling times. After 10 hours of milling the peaks related to TiB₂ ceramic phase are obvious, as well as Al and Ti, showing the gradual formation of TiB₂ (Fig. 2-b). After 20 hours of milling (Fig. 2-c) no evidence of remaining titanium or formation of undesired phases could be detected on the XRD pattern. The lack of Al peaks on XRD pattern can be due to the several effects including high strain level induced in the Al lattice, nanosized Al grains and lower x-ray scattering intensity of Al compared to the other constituents. Milling of the powder, resulted from first step, together with aluminum to achieve Al- 20 wt.% TiB₂ composition resulted in no clear structural change in the powder (Fig. 2-d). Detailed explanations of the milling process and results have been presented elsewhere [10-11]. The grain size of the aluminum matrix and TiB₂ was obtained from XRD analysis using Williamson-Hall (WH) equation [12]. In the resulting powder from the double step mechanical alloying, the grain

size of both TiB₂ particles and aluminum matrix was measured to be about 15 nm.

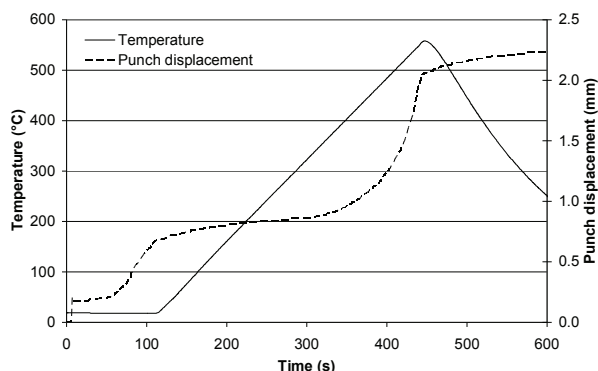


Figure 1: An example of the evolution of SPS conditions during the consolidation process.

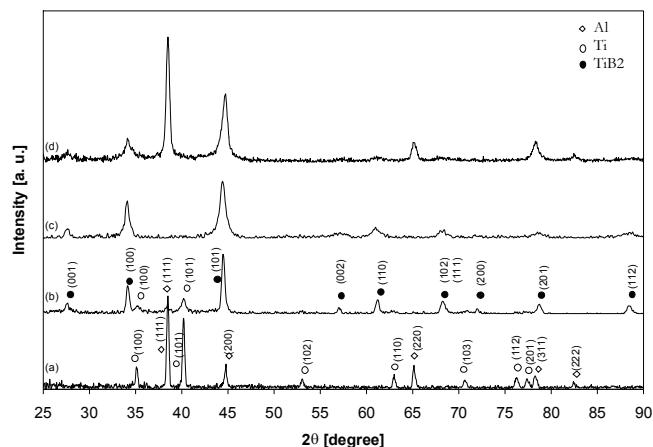


Figure 2: XRD patterns of Al- 62 wt.% Ti- 28 wt.% B powder mixture: (a) as-received, (b) as-milled for 10 h, (c) as-milled for 20 h and (d) final Al- 20 wt.% TiB₂ powder obtained from double step mechanical alloying.

Fig. 3-a shows the cross sectional SEM micrograph of an Al-20wt.%TiB₂ particle obtained from the third series. A fine and uniform distribution of TiB₂ particles within the Al matrix is obtained with many particles being too small to be measured on SEM. A higher magnification of the final Al-20 wt.% TiB₂ composite powder obtained from STEM is presented in Fig. 3-b. The approximate average TiB₂ particle size measured using image analysis software was about 90 nm, ranging from 10 nm to maximum 1 μ m. The mean particle size seems to be smaller than that of previous studies on in situ Al-TiB₂ composite produced by MA [6]. After 40 hours of MA the TiB₂ particles and aluminum matrix achieved a mean grain size of about 20 nm, according to According to the dark-field image (Fig. 4).

This is in good agreement with the values obtained from the WH equation.

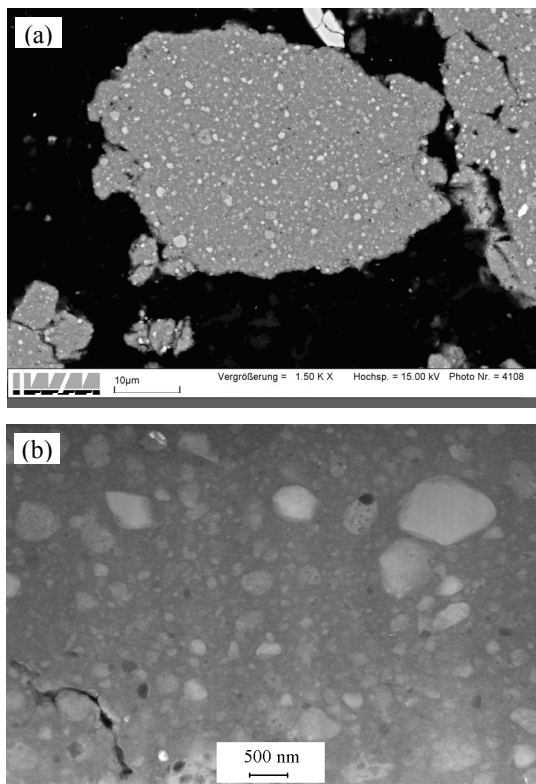


Figure 3: a) Typical cross sectional SEM micrograph of Al-20 wt.% TiB₂ powder particles, b) STEM image of an Al-20 wt.% TiB₂ powder particle.

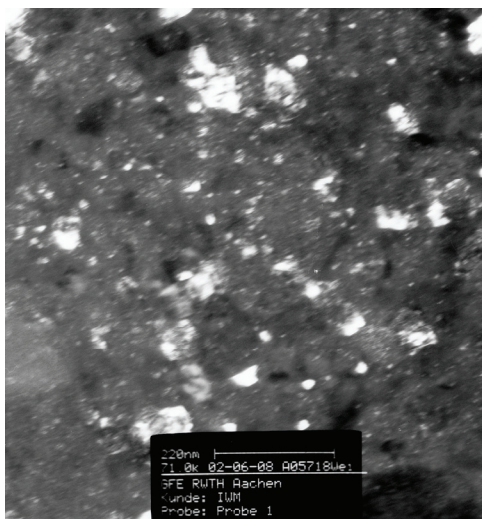


Figure 4: Dark field image of Al-20 wt.% TiB₂ powder particles obtained from double step MA.

Final powder from the double step mechanical alloying consisting 20 wt.% TiB₂ was sintered using SPS technique.

According to the XRD results, after SPS at 550 °C no obvious structural change occurred in the consolidated Al-TiB₂ nanocomposite (Fig. 5). It can be concluded that by the double stage synthesis process, the formation of undesirable compounds even after exposure to sintering temperature, is prevented. Lu et al. reported that during the high temperature exposure of as milled Al-Ti-B powder mixture, Al₃Ti phase is formed in the aluminum matrix along with TiB₂ [7].

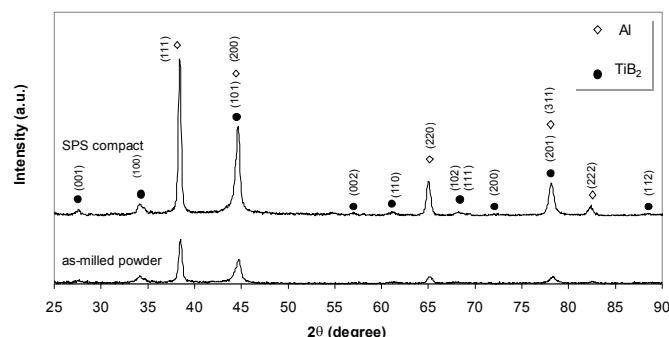


Figure 5: XRD pattern of the SPSed 20 wt.% TiB₂ nanocomposite compared to the as MAed powder.

Fig. 6 shows the SEM micrograph of SPSed Al-20wt.%TiB₂ nanocomposite. The size of TiB₂ particles in the sintered sample was measured from about 50 nm to 1 μm, which has not increased obviously in comparison with the as MAed powder. From TEM examination of the sintered samples (Fig. 7) the mean grain size of phases in the composite was measured about 25 nm. It can be concluded that the in situ synthesized Al-TiB₂ nanocomposite powder has a desirable stability to grain growth at high temperatures.

The density of the sintered sample was determined 2.8 g. cm⁻³ by using Archimedes' principle, which is 95.2 % of theoretic density and porosity was measured 3.8 %. Microhardness measurements showed an almost large decrease in the hardness from powder to sintered state. Hardness measurement of the samples showed a large decrease from powder to sintered sample. Microhardness of the powder was measured about 480 VHN, while the hardness of sintered sample was about 206 VHN. Reduction in hardness of the SPSed sample in comparison with the as-MAed powder can be related to the recovery, and relief of work hardening effects during sintering process. However hardness of the compacted sample in this study is about two times higher than that reported in previous findings [6]. This is proposed to be the consequence of nanostructure composite obtained in the present study.

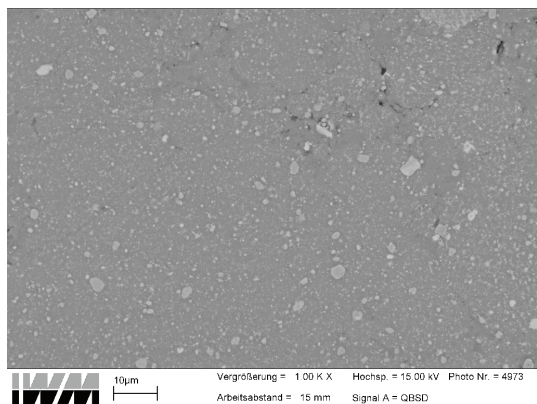


Figure 6: Cross sectional SEM micrograph of the SPSed Al- 20 wt.% TiB₂ nanocomposite

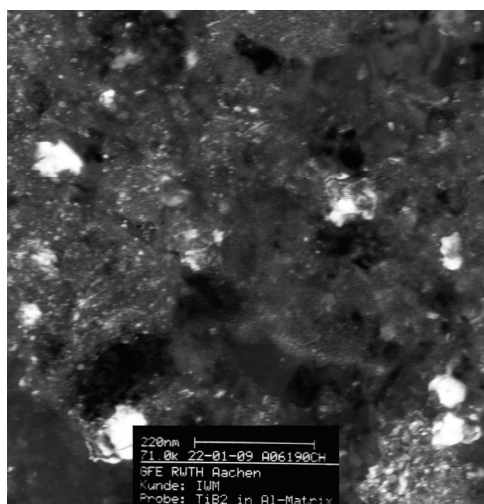


Figure 7: Dark field images of (a) Al- 20 wt.% TiB₂ powder particles and (b) sintered sample obtained from TEM experiments

CONCLUSIONS

Al-TiB₂ nanocomposite powder was synthesized by a double-step MA process of elemental powders. TEM observations showed formation of TiB₂ particles with a mean size of about 90 nm and uniform distribution in the Al matrix after a total milling time of 40 h. No traces of undesirable phases such as titanium aluminides were observed even after consolidation of the powder by SPS. The fabricated powder showed a suitable stability against grain growth during the sintering process.

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