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Synthesis and consolidation via spark plasma sintering of nanostructured Al-5356/B_4C composite

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ABSTRACT

The combination of two versatile processing routes: cryomilling and spark plasma sintering (SPS) was employed in the present study for fabrication of bulk Al5356-B_4C nanocomposites. This approach resulted in large microhardness and flexural strengths values in the consolidated samples of 244 ± 2.06 HV and 707 MPa respectively. While cryomilling provided the combined advantage of homogenous dispersion of the reinforcement in the matrix as well as nanostructuring of the Al-5356 phase, the SPS facilitated the solid state consolidation of the metal matrix composite (MMC) in one single step at very high sintering rates and very short dwelling times. No additional processing steps such as forging or hot extrusion were necessary to achieve powder consolidation. The resulted samples exhibited two to three times higher microhardness values than the monolithic micron sized un-reinforced alloy.

1. Introduction

Metal matrix composites (MMCs) are thriving in popularity due to their superior mechanical performance over the conventional metal counterparts. In this group of materials, Al based MMCs engage a distinct position due to the advantage of lightweight. Introducing a hard phase in the particulate form in an Al-matrix triggers significant increase in wear and erosion resistance, as well as higher stiffness, hardness and strength [1]. The reinforcement phases are usually ceramic in nature; some in the oxide form: Al_2O_3 [2–4], and others in covalent bonded materials such as SiC [5,6], AlN [7], Si_3N_4 [7,8], and B_4C [9–14].

With an elevated hardness value, ranking third, after diamond and BN, while being lighter than the two, B_4C grants the advantage of great wear resistance, second only to diamond [15], as well as good chemical stability. All these characteristics render B_4C an excellent candidate as reinforcement phase for MMCs used in various structural and mechanical applications. There have been several studies in the literature which illustrate the influence of B_4C addition to aluminium matrix with respect to mechanical properties. Oñoro et al. [16], showed that an addition of only 5% B_4C to different aluminium alloys, triggered an increase in hardness of the resulted MMCs. Using a combination of processing techniques such as mixing in a planetary mill, followed by cold uniaxial pressing and hot extrusion they were able to obtain an increase in hardness values from 80 HV to 130 HV for Al-6061 MMC and from 138 HV to 154 HV for Al-7015 MMC, respectively. A similar trend was seen for the room temperature ultimate tensile strength (UTS) for both MMCs. Additionally, at temperatures as high as 500 °C the UTS values exhibited by the MMCs were still higher than the monolithic alloys counterparts at analogous temperature. As it was indicated in the study all the improvements in mechanical properties were attributed exclusively to B_4C addition, since insignificant grain refinement of the matrix was observed [16]. These results are in good agreement with the findings obtained by Cambronero et al. [17], which working with the same Al-7015 matrix and B_4C concentration, have obtained, by a combination of mechanical alloying, cold pressing and extrusion, a homothetic increase in microhardness value, as well as two to four times higher wear resistance when compared with the monolithic alloy. Further additions of B_4C in the matrix triggers consequent increase in hardness values [18,19]. As recorded by Topcu et al. [18], for pure Al matrix exhibiting a hardness value of 25.8 HV, higher B_4C contents, increasing from 5 wt% to 20 wt%, resulted in an enhancement of hardness from 45.6 HV to 82.1 HV for the MMC specimens obtained by a combination of attrition milling and cold isostatic pressing.

Reducing the grain size of the matrix to the nanometric regime provides the opportunity to additional increase in strength and hardness. The strengthening mechanism in nanostructured materials is related to the increased grain boundary density through the known Hall–Petch relationship. When compared to micron-

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sized counterparts, nanostructured materials exhibit two to nine times increased hardness. For Al samples with 30 nm grain size, hardness values of 357 HV were obtained, i.e. almost eight times higher than nominal 46 HV value, exhibited by 5 μm grain size counterparts [20,21]. Therefore ultra high strength composites can be obtained through the optimization of two concepts: reinforcement properties and simultaneous nanostructuring the metal matrix.

A limited number of publications have been reported in the literature regarding fabrication of aluminium MMCs with B_4C as reinforcing phase via cryogenic milling [10,11]. In all studies, an increase in strength was recorded. Li et al. [11] have reported an exceptional high value of 1065 MPa yield strength for a 10% B_4C Al based MMCs, as obtained by a combination of cryomilling, cold isostatic pressing (CIP) and hot extrusion techniques.

One of the challenges encountered in working with nanostructured powders is their consolidation into bulk part. The long time exposure to high temperature needed for sintering favours significant grain growth and loss of the nanostructure. Recently, a relatively new technique, spark plasma sintering (SPS), has been successfully used in nanostructured powder consolidation [22,23]. A unique characteristic of SPS technique is that a pulsed DC current is applied to the sample and the die, resulting in Joule heating, hence facilitating very high heating and cooling rates, and consequently sintering is achieved within a few minutes [24]. The short processing times allows grain growth to be controlled and makes SPS an attractive choice over the conventional powder sintering methods such as hot or isostatic pressing or extrusion.

The objective of the present study was twofold: first to generate nanostructured Al-5356-20%B_4C powder via cryomilling and second to explore the SPS approach to consolidate bulk form while preserving the nanostructured features. To our best knowledge, to date there has been no investigation for these particular MMCs that combines the two processes, i.e. synthesis of the powder via cryomilling followed by SPS consolidation, although independently, both procedures have been explored to a certain extent for Al-based MMCs – B_4C reinforced. More in depth analyses was carried out for synthesis of Al-5083-B_4C. In some studies cryomilling was employed for MMCs powder generation [11,12,25,26] and the consolidation was carried out via cold isostatic pressure followed by hot extrusion [11,12,25] whereas in others, sintering was achieved via 1–3 steps of forging followed by cold rolling [26]. Regarding SPS consolidation of this category of material only a very recent study [27] explores briefly this sintering technique for pure Al matrix with B_4C reinforcement. However, different reinforcement concentration than the present study were used; the MMC powder was synthesized at room temperature in argon atmosphere using a Spex mill, and much attention was given to the solid state reactions between the reinforcement phase and the matrix in the sintering process.

In the current study we focus on the investigation of the microstructure (grain size and morphology) of the cryomilled powder as well as the influence of the hard phase on structure refinement of the matrix at both macroscopic (individual powders) as well as microscopic level (individual grains). A comparison with the mechanism that governs the structure refinement of ductile materials is performed. Consolidation profiles and optimum conditions are explored for the SPS compacts. The influence of various sintering parameters on the mechanical properties of the MMCs is being investigated. Microstructures of the bulk SPS samples, microhardness indentation as well as examination of the fracture surface are used for sintering evaluation.

2. Experimental procedure

2.1. Synthesis of the ns MMC powders

Aluminium 5356 alloy with Mg (5%) as major alloying element and boron carbide (B_4C) powders were used as starting material. The aluminium 5356 powder with an average particle size of 22 μm was supplied by Valimet Inc., Stockton, CA, USA. The B_4C powder ranging between 3 and 13 μm was provided by Atlantic Equipment Engineers, Bergenfield, NJ, USA. The powders were mixed together at a ratio of 20:80 B_4C: Al-5356 on a weight percent basis. The cryomilling experiments were conducted using a Union Process 1-S attritor, equipped with an air insulated stainless steel tank. Stainless steel balls, with a diameter of 4.85 mm, were used as grinding media at a ball to powder mass ratio of 32:1. The impeller rotation was set at 180 RPM. To prevent adhesion of the powder to the tank, balls, and attritor, and to control the fracturing events, 0.25 wt% of stearic acid was added as a process control agent. A total milling time of 8 h was employed. Samples were collected every hour for in-depth characterization. The changes in powder morphology throughout the process were examined with a Field Emission Gun Scanning Electron Microscope (FEG-SEM) S-4700 Hitachi. Particle size distributions were obtained using a Horiba Particle Size Analyzer LA-920. The X-ray diffraction pattern were acquired with a Phillips PW1710 diffractometer, using Cu Kα radiation ($\lambda = 0.1542$ nm) between 15° and 90° at 0.005° step size and a time of 0.5 s per step. Annealed Al-5356 powder was used for instrumental broadening correction. The grain size and lattice strain were determined from the XRD peak broadening using Williamson–Hall method [28]. The grain size evaluation was also performed on bright and dark field micrographs conducted on a JOEL JEM-2011 Transmission Electron Microscope (TEM) operating under 200 keV beam energy.

2.2. Spark plasma sintering of the ns MMC powders

The cryomilled powder was consolidated under vacuum in 20 mm dies using a Spark Plasma Sintering (SPS) apparatus Model SPS 10-3 from Thermal Technologies Inc., Santa Rosa, CA, USA. The sintering profiles involved 100 °C/min heating rate to a peak temperature that varied from 450 °C to 525 °C, according to the sample and dwell times between 15 s to 5 min, followed by furnace cool down. Concurrently with heating, a pressure of 50 MPa was applied and released at the end of the sintering cycle.

2.3. Post sintering characterization

The resulting samples were measured and weighted in order to calculate the relative density. A micro hardness tester (Clark CM-100AT) used in conjunction with an optical microscope equipped with a Clemex software was used to assess the hardness of the obtained bulk SPS compounds. A 500 g-force load was applied to perform indentation. Flexural tests were performed using a Tinius Olsen H25K-S universal testing machine via 3-point bending test on bars cut from the SPS samples. Fracture surfaces resulted from flexural test were examined with FEG-SEM. Grain size measurements were calculated from the XRD peak broadening and verified via TEM direct measurements.

3. Results and discussion

3.1. Synthesis of the MMC powders

3.1.1. Starting powders morphology and size

The morphology of the starting powders is presented in Fig. 1a for B_4C and Fig. 1b for Al-5356, respectively. The as received B_4C