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Letter

An exploratory investigation on the in-situ synthesis of SiC/AlN/Al composites by spark plasma sintering

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ABSTRACT

The powder mixtures of aluminum and silicon carbonitride (Al–SiCN) have been consolidated into bulk materials by spark plasma sintering. XRD patterns indicated the in-situ formation of SiC and AlN. The density of the SiCN–Al composite sintered by SPS at 500°C is 2.65 g/cm^3 or higher in a weight range of SiCN powder particles up to 2.5%. The formation of new phases and the relatively absence of porosities in the matrix composites are attributed to the co-existence of solid–liquid state that leads to the effective densification of the mixed powders. The breakdown of SiCN network structure was considered to result from a reaction between partially aluminum melt and SiCN particles. An improvement in density and hardness is observed.

1. Introduction

Non-oxide ceramic reinforced aluminum matrix composites offer significant performance advantages over pure metals and metallic alloys for advanced structural and technological applications. Silicon carbonitride (SiCN) and composites are remarkable materials for their combination of mechanical and thermal properties at high temperatures [1]. Considered as an emerging sintering technique, spark plasma sintering (SPS) has the potential for rapid and efficient consolidation of a broad spectrum of powder materials at relatively low temperatures and within short processing times. This technology combines simultaneously the application of pressure and electric current directly on the sample. SPS has been utilized for the preparation of dense compacts of Si_3N_4-based ceramics, β–sialon, and $\alpha \pm \beta$–sialon composite material, as demonstrated by the work of Shen and Nygren [2]. They also expanded their research to investigate consolidation mechanisms and the phase transformation sequences during the sintering process. The application of the SPS technique is not only to produce a dense bulk material within short processing periods, but also to fabricate nanomaterials and to obtain special microstructures [3–15].

A considerable investigation has been conducted by Mizuuchi et al. [16] on the fabrication of Al–matrix composites containing dispersed diamond particles in pure Al and Al–Si alloy by spark plasma sintering (SPS) process. The relative packing density of the diamond-Al composite was $\geqslant$99% in a volume fraction range of diamond between 35% and 50%. A preliminary work was carried out by Zhang and his co-workers [17] to study the microstructures and mechanical properties of spark plasma sintered Al–SiC composites containing 80 wt% SiC. The X-ray diffraction results showed the presence of Al, SiC, Si, and Al_4C_3 phases. Other authors [18–20] demonstrated the feasibility of making aluminum–carbon nanotube (Al–CNT) composite materials by a combination of spark plasma sintering (SPS) and hot extrusion processes. While much research has been reported on characterizing metal–matrix composites produced from Al alloys combined with SiC, Si_3N_4, sialon or non-oxide ceramic particulates, no reports has been published regarding the processing of Al–silicon carbonitride (SiCN) composites via SPS technique, despite the obvious attractions of this approach. Therefore, in the present preliminary work, we report for the first time the formation of in-situ SiC/AlN/Al ceramic composite by SPS from pyrolysed SiCN powder particles and Al metal. It is believed that the presence of crystalline phases (SiC and AlN) in the synthesized composites would constitute a good choice to compete with current non-oxide ceramic materials, for example for armored vehicles or other specialized applications.

2. Experimental procedure

The dispersed phase silicon carbonitride (SiCN) was prepared from a commercially available liquid polymer precursor with the trade-name PURS (poly-ureasilazane). The liquid polymer was cross-linked at 300 °C, then crushed

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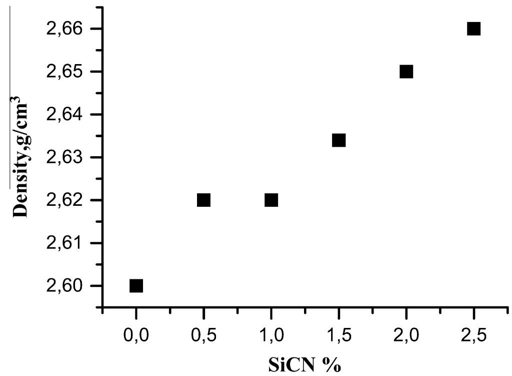

Fig. 1. Variation of density with SiCN percentages.

and ball milled into powder followed by pyrolysis at 1000 °C for 1 h in nitrogen atmosphere. The amorphous SiCN ceramic phase will not contribute any peaks to the X-ray diffraction results.

Six batch mixtures containing different dosage of SiCN powder particles (up to 2.5 wt%) in the aluminum matrix were prepared. These mixtures were mixed and milled to distribute the SiCN particles in the corresponding amount of aluminum powder. The powder precursors were compacted in a cylindrical graphite die with an inner diameter of 15 mm. The SPS lets a high pulsed electrical current pass through a graphite die containing the powders' mixture. Heat is thus internally generated and the reaction took place in few minutes. The powder mixtures (Al–SiCN)was sintered at 500 °C, with a pulse duration of 2.8 ms, a holding time of 2 min, a heating rate of 200 °C/min, and a pressure of 50 MPa in a 15 mm carbon mold, using a spark plasma sintering apparatus (Dr. Sinter 515S SPS) manufactured by SPS Syntex Inc., Kanagawa, Japan. The diameter and height of the resulting specimens were about 15 mm and 3 mm, respectively. The indentation parameters for hardness measurements were a 5 kg load with a dwell time of 15 s. The density of compacted specimens was determined by Archimedes principle by taking the Al matrix and dispersed SiCN densities as 2.699 and 2.42 g/cm^3 respectively. The macro and micro-structures of as-sintered composites containing various weight percentages of SiCN were examined by means of optical microscopy and scanning electron microscopy on polished surfaces using a JEOL equipped with an energy-dispersive X-ray spectroscopy (EDS). Crystalline phases were determined from X-ray powder diffraction patterns performed on a diffractometer (D8 Brucker, Germany) using Cu Kα radiation (λ = 1.54060 Å).

3. Results and discussion

The variation of the density with weight percentages of SiCN for the Al–SiCN composites was illustrated in Fig. 1. The density increases slightly with increasing weight percentages till achieving 2.66 g/cm^3 at 2.5 wt% SiCN. In the case of pure Al, the sintered body reached ~2.60 g/cm^3 at 500°C. The slight increase in density is due to the more formation of SiC (3.2 g/cm^3) and AlN (3.26 g/cm^3) in the matrix. The average density value, obtained by measuring the actual composite density by Archimede's method and comparing the result with its theoretical value for the given component content ratio, is lower than 2% experimental error. The achievement of relatively good densification at 500°C should be due to the combination of rapid rates of Joule heating and intrinsic field effects on mass transport [21]. It should be noted that previous works have been performed on synthesizing and controlling functionalities in the polymer derived SiCN ceramic–matrix composites for potential applications [22–25]. The spark plasma reaction sintering between Al and SiCN is actually a liquid–solid reaction where the Al partially melts, and then reacts with the nearby SiCN particles. It is worth mentioning that the formation of silicon carbide out of a SiCN matrix at this low temperature is quite unusual, as the crystallization of SiC from polymer-derived SiCN materials was reported to occur at temperatures above 1500–1600 °C [26,27]. We can conclude that the reaction of the amorphous SiCN phase with aluminum metal to form SiC and AlN at 500 °C, should be mainly attributed to the co-existence of solid–liquid state. It was reported by German [28] that the presence of a liquid phase improves mass transport rates during the sintering process. Others [29,30] emphasized the impact of heating rates on the densification kinetics during sintering. Furthermore, Roura et al. [30] discussed the validity of the concept of shrinkage rate for any transport mechanism.

In order to verify the phase composition of the SPS composites, X-ray diffraction patterns revealed clearly the presence of two new phases besides the starting Al powder, Fig. 2. However, the crystalline fraction of SiC and AlN phases is rather small. Their weak intensities are associated with the low addition of SiCN particles. This result indicates that although material transfer may be promoted by the SPS process, the existence of a liquid–solid state is a must to breakdown the SiCN network structure and produce SiC/AlN/Al composites. It is clear from the inset in Fig. 2, the existence of both SiC and AlN phases. It can be concluded that the presence of more liquid Al surrounding SiCN particles under the effect of electric current gives rise to the increase of the diffusion path, and thus promotes the formation of AlN during the sintering process. The whole SPS process took 270 s and the

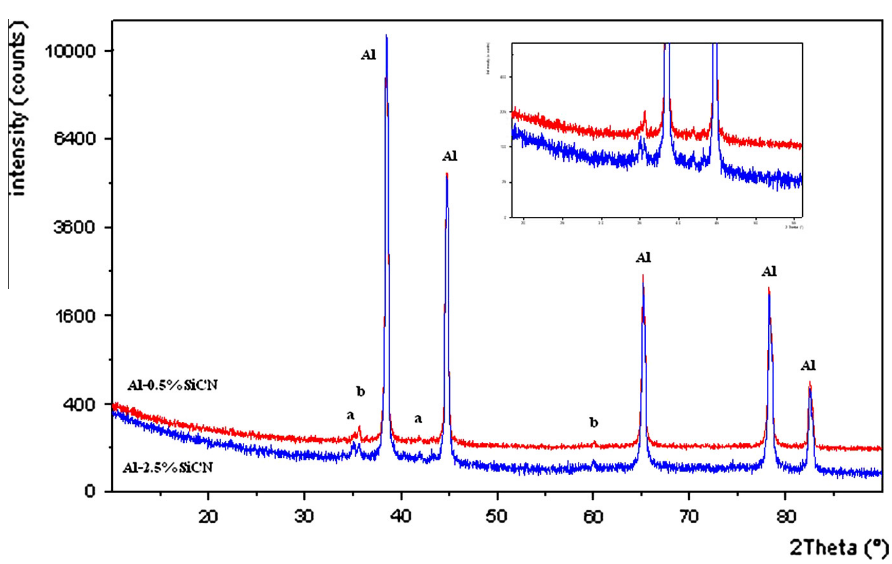

Fig. 2. XRD patterns of samples obtained by spark plasma sintering Al/SiCN mixtures containing 0.5 and 2.5 wt% of SiCN. (a) SiC, and (b) AlN.