



Contents lists available at ScienceDirect

Ceramics International

journal homepage: [www.elsevier.com/locate/ceramint](http://www.elsevier.com/locate/ceramint)

## Sintering behaviour of silicon carbide matrix composites reinforced with multilayer graphene

M. Petrus\*, J. Wozniak, T. Cygan, B. Adamczyk-Cieslak, M. Kostecki, A. Olszyna

Faculty of Materials Science and Technology, Warsaw University of Technology, Woloska 141, Warsaw, Poland

### ARTICLE INFO

#### Keywords:

Sintering  
Composites  
SiC  
Carbon

### ABSTRACT

The scope of this paper includes preparation and characterisation of dense silicon carbide matrix composites reinforced with multilayer graphene (MLG). Application of graphene as a reinforcement phase should simultaneously improve mechanical properties of SiC matrix composites and act as one of the sintering activators. In the present work the mechanical properties and the microstructure changes of samples sintered with different additions of graphene (0.5, 1, 2, 3, 4 wt%) and boron (0.3, 1 and 2 wt%) were examined. The composites were consolidated at two different temperatures (1800 °C and 1900 °C) using the Spark Plasma Sintering method (SPS). Reference samples with the addition of graphite as a source of carbon (1 and 3 wt%) were also sintered in the same conditions. The abovementioned amounts of graphite are an optimal content which is essential to obtain high density of samples [1–9]. The influence of MLG on density, mechanical properties and phase structure of the sintered samples were investigated. A high rate of densification for the composites with 0.3 wt% of B and 1 wt% of MLG sintered at 1900 °C was observed. Moreover, these composites showed the highest average of microhardness (2663 HV0.5) and single-phase structure.

### 1. Introduction

Silicon carbide offers favourable properties owing to its covalent bond, such as high hardness and strength at high temperatures. However, such a bond causes retarding of solid-phase sintering due to slow bulk diffusion [1,2]. Prochazka et al. [3] showed that dense SiC can be sintered with the use of boron and carbon densification aids (solid state diffusion process). The first hypotheses explaining the influence of carbon on the improvement of sinterability were based on the assumption that carbon reduces the SiO<sub>2</sub> present on the grain surface of silicon carbide. The subsequent research did not confirm the mentioned hypotheses due to the fact that this oxide is being eliminated at temperatures above 1400 °C [4–6]. Other explanations stated that an excessive amount of carbon may diffuse inside the SiC grains and cause the silicon deficiency, which would contribute to an increase of the diffusion level in these places [7]. On the other hand, the present research confirms the fact that carbon limits these mass transports, i.e. diffusion in the gaseous phase and the surface diffusion, which are ineffective in eliminating pores. The carbon, present in the process of sintering, bonds the volatile silicon compounds, which are the products of the thermal decomposition of SiC. The presence of the abovementioned silicon compounds has a negative influence on the process of densification [8,9]. Furthermore, a small amount of boron

activates the sintering process [10–12]. The SiC may be densified also to high relative density by liquid phase sintering process where oxides like alumina, silica and rare earth oxides are used as sintering aids. These materials are characterized by a higher value of fracture toughness [13].

Graphene shows unique combinations of mechanical, electrical and thermal properties [14,15]. Due to this fact, it is widely used in many branches of technology [16,17]. Because of its two-dimensional geometry and large specific surface area graphene seems to be a perfect reinforcement for metal, ceramic and polymer matrix composites. According to the results reported by Liu et al. [18] and Porwal et al. [19], the addition of graphene to an alumina matrix can improve the mechanical properties e.g. fracture toughness and hardness. Graphene was also used as a reinforcement in a non-oxide ceramic. Kvetkova et al. announced a significantly higher fracture toughness for silicon nitride composites containing 1 wt% of multilayer graphene [20]. Kun et al. reports an increased value of bending strength and elastic modulus for composites Si<sub>3</sub>N<sub>4</sub>+1 wt% MLG in comparison to non-reinforcement monolithic silicon nitride [21]. However, the experiments carried out by Rutkowski et al., did not show any increase of the fracture toughness for silicon nitride composites up to 4 wt% of graphene [22]. The main problem in producing composites reinforced with graphene is the creation of agglomerates, with increasing the

\* Corresponding author.

E-mail address: [matpetrus92@gmail.com](mailto:matpetrus92@gmail.com) (M. Petrus).

<http://dx.doi.org/10.1016/j.ceramint.2017.01.010>

Received 8 November 2016; Received in revised form 21 December 2016; Accepted 4 January 2017  
0272-8842/ © 2017 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

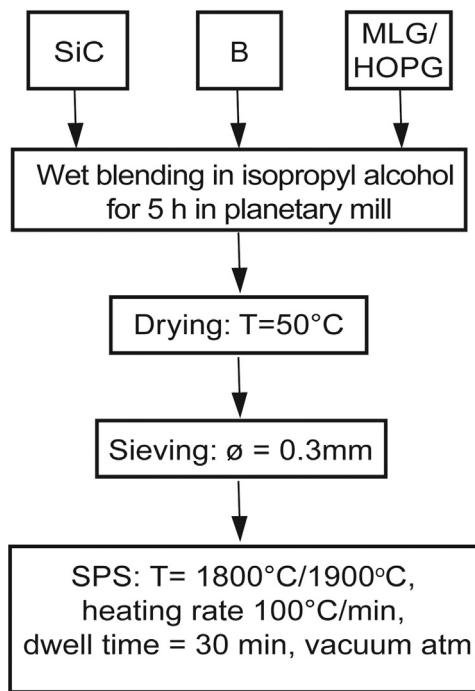


Fig. 1. Production scheme of composites.

amount of reinforcement [23]. As suggested above, the use of graphene as a reinforcement phase in ceramic matrix composites can bring numerous advantages. Moreover, carbon is a sintering activator which is essential to obtain a dense SiC composite. Taking both of these facts into consideration it may be concluded that the use of graphene in the SiC matrix enables to fabricate a dense composite characterized with unique mechanic properties.

In this work silicon carbide – multilayer graphene composites were fabricated. MLG should play the role of a sintering aid and reinforcement. The influence of MLG on density, microstructure, mechanical properties (microhardness, fracture toughness) and phase structure of the sintered samples was investigated. The results were compared with the samples, where the source of carbon was graphite.

## 2. Experiment

SiC-B-Gn composites were produced using the powder metallurgy technique (Fig. 1). The following substrates were used: commercial  $\beta$ -SiC powder (99.8% chemical purity), commercial MLG powder (Graphene Supermarket, 99.2% chemical purity, 4.5  $\mu\text{m}$  average flakes size, 12 nm average flakes thickness), commercial amorphous boron powder (International Enzymes Limited, 96% chemical purity) and highly oriented pyrolytic graphite (HOPG). Powders were wet-blended

at planetary type mill for 5 h in isopropyl alcohol. Composites SiC-xB-yGn (where  $x=0.3, 1, 2$  wt% and  $y=0.5, 1, 2, 3, 4$  wt%) were sintered using the SPS method, in the following parameters:  $T=1800/1900$  °C,  $t=30$  min,  $V_t=100$  °C/min,  $p=50$  MPa. In the same conditions samples with graphite (1 and 3 wt%) and 0.3 wt% of boron were also sintered.

The density of the specimens was examined using the Archimedes method, microhardness and fracture toughness were measured with a Vickers Hardness Tester (FV-700e) using the indentation method under the load of 49.05 N. The Young's Modulus of the composites was tested with the use of the ultrasonic method (Optel Refractometer). The microstructure observations were performed on a scanning electron microscope (SEM Hitachi 5500). The Raman spectra was measured with a Nicolet Almega XR-Dispersive Raman spectrometer (laser with an excitation wavelength of 532 nm) and for each sample at least 3 scans were performed. The qualitative phase composition was analysed using XRD diffraction (Bruker D8Advance, Cu  $K\alpha$   $\lambda=0.154056$  nm, voltage – 40 kV, current – 40 mA, angular range 0–85°, step  $\Delta 2\theta$  – 0.05°, counting time – 3 s).

## 3. Results and discussion

The morphology and the Raman spectra of multilayer graphene are presented in Fig. 2a and b respectively. Fig. 2a exhibits regular flake-like shape. In addition, the Raman spectrum is typical for MLG, which suggested 2D-band shape and position of D-band and G-band [24]. Fig. 3 shows the morphology of highly oriented pyrolytic graphite. A few  $\mu\text{m}$  thick graphite flakes can be observed.

Measurements of the relative density show better results for the samples sintered at 1900°C than for those sintered at 1800°C, irrespective of graphene and boron content (Fig. 4a, b and c). The highest value of relative density (98.5%) is obtained for the samples with 1 wt% of graphene and 0.3 wt% of boron. The same phenomenon may be observed in the case of all samples with 1 wt% of graphene and different amounts of boron. However, the composites with the content of boron over 0.3 wt% were characterized by lower values of density. Higher density was also observed in the case of the samples sintered at 1900 °C, where the source of carbon was graphite. However, in order to obtain a comparable level of density, the addition of graphite should be significantly higher than the one of graphene (3% and 1% accordingly). Therefore it may be inferred that even a low addition of graphene provides a higher densification level of the composites than the conventional carbon sources.

The Raman spectra of the composites (Fig. 5) shows carbon bands, which can be identified as originating from graphene (Fig. 2b). This observation proves that during the sintering process the structure of graphene was preserved. Taking into consideration the high density of the obtained composites it can be concluded, that graphene may act as the source of carbon, which is essential to achieve a dense composite [3–9]. Furthermore, owing to the fact that it preserved its structure, graphene may also function as reinforcement. Band TO and LO were

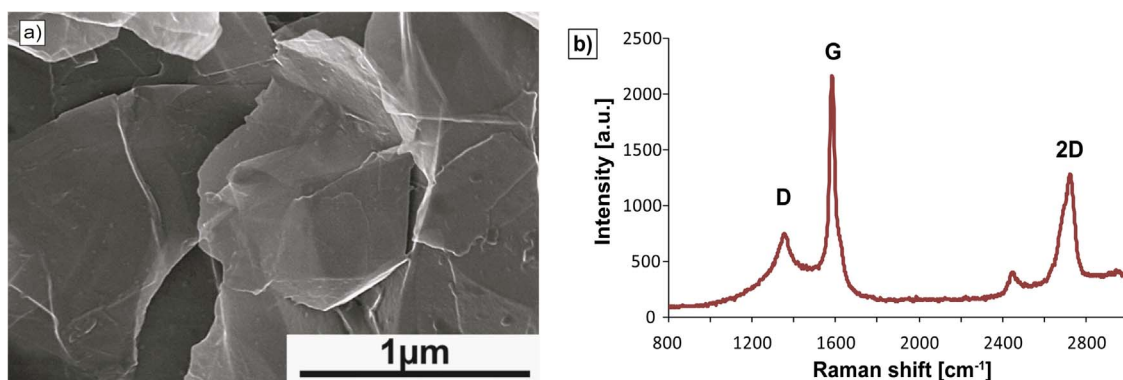


Fig. 2. Morphology (a) and the Raman spectra (b) of multilayer graphene.

Download English Version:

<https://daneshyari.com/en/article/5438633>

Download Persian Version:

<https://daneshyari.com/article/5438633>

[Daneshyari.com](https://daneshyari.com)