

# Synthesis of SiC/SiO<sub>2</sub> core–shell powder by rotary chemical vapor deposition and its consolidation by spark plasma sintering

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## Abstract

SiC (core) and SiO<sub>2</sub> (shell) powders were synthesized via rotary chemical vapor deposition (RCVD). The SiC particles (3C, < 1 μm in diameter) were coated with a layer of SiO<sub>2</sub> (10–15 nm in thickness). Using spark plasma sintering, the SiC/SiO<sub>2</sub> nanopowders were then synthesized into SiC/SiO<sub>2</sub> composite bodies. Although a phase transformation from 3C to 6H was observed at above 2123 K in the sintered monolithic SiC bodies, sintered SiC/SiO<sub>2</sub> bodies did not display such phase transformation. In addition, SiC/SiO<sub>2</sub> bodies did not exhibit grain growth until the sintering temperature reached 2223 K. The density and Vickers hardness of the sintered SiC/SiO<sub>2</sub> bodies increased with increasing sintering temperature. The highest density and hardness of SiC/SiO<sub>2</sub> composite bodies were 98.1% and 24.4 GPa at 2223 K, respectively, which were higher than the corresponding values of 90% and 14 GPa for monolithic SiC bodies. © 2012 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

**Keywords:** Rotary chemical vapor deposition (RCVD); SiO<sub>2</sub> nano-layer; Silicon carbide (SiC); Spark plasma sintering (SPS)

## 1. Introduction

Silicon carbide (SiC) has been widely used to manufacture high-temperature materials because of its high oxidation resistance, high hardness and high thermal-shock resistance [1]. Monolithic SiC is hard to densify by conventional sintering techniques because of its highly covalent nature and the low self-diffusion of Si and C [2]. Therefore, high sintering temperature and the addition of sintering aids are normally required to enhance the densification process for SiC. During this densification process, the sintering additives form a second phase at grain boundaries. This second phase usually causes the mechanical and physical properties of the material to deteriorate, particularly at higher temperatures [3]. It has been shown that a SiC body can be fabricated by pressureless sintering with the aid of boron and carbon, and densified to more than 95% of the relative

density at temperatures higher than 2323 K [4]. However, the low fracture toughness of such bodies limits their use in many potential structural applications [5].

Spark plasma sintering (SPS) is capable of fabricating dense bodies at relatively low temperatures in a short amount of time [6]. Zhou et al. densified SiC powder (with 30 nm diameter particles) doped with an Al<sub>4</sub>C<sub>3</sub>–B<sub>4</sub>C additive to 99% of the relative density at 1873 K [7]. Ohyanagi et al. demonstrated the densification of a SiC powder (with 5–10 nm diameter particles) by high-energy mechanical milling to 98% of the relative density at 1973 K without using any additives [8].

Ye et al. densified a SiC powder with a 20 mass% SiO<sub>2</sub> additive to 94.2% of the relative density at 2133 K in a high-pressure CO gas atmosphere by liquid-phase sintering [9]. It is known that SiO<sub>2</sub> can be easily sintered [10], and can thus be used as a possible sintering additive to promote the densification of SiC. However, SiO<sub>2</sub> might degrade the mechanical properties of the SiC because of its low hardness and brittleness [11]. To use SiO<sub>2</sub> as a sintering aid for SiC such that its mechanical properties at least do not degrade, SiO<sub>2</sub> should be uniformly dispersed on the nanometer scale within the body. However, to the best of our knowledge, no such study has ever been performed. By

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sintering bodies from a powder consisting of particles having SiC core and SiO<sub>2</sub> shells (hereafter referred to as SiC/SiO<sub>2</sub>), this study demonstrates that SiO<sub>2</sub> can be a suitable additive for SiC and can enhance the mechanical properties of the sintered SiC body. We developed a rotary chemical vapor deposition (RCVD) technique to prepare the powder with the core–shell particle powder by depositing a thin layer of SiO<sub>2</sub> on the SiC cores. Previous studies reported that Ni nanoparticles could be prepared [11] and a layer of SiO<sub>2</sub> [12] could be deposited on cubic BN (cBN) powder by RCVD. The resulting composite powder layer could then be consolidated by SPS. The cBN (core)/SiO<sub>2</sub> (shell) powder could be effectively densified as the SiO<sub>2</sub> additive retarded the phase transformation of cBN into hexagonal BN (hBN). In this study, the SiC/SiO<sub>2</sub> powder particles were synthesized by RCVD, and the SiC/SiO<sub>2</sub> composites were produced by SPS.

## 2. Experimental

β-SiC powder (purity: 99%; average particle size=0.28 μm; Ibiden, Japan) particles were coated with a layer of SiO<sub>2</sub> by RCVD using tetraethyl orthosilicate ((C<sub>2</sub>H<sub>5</sub>O)<sub>4</sub>Si) as a precursor. Fig. 1 shows a schematic of the RCVD process. To suspend the SiC powder properly in the reactant gas, four blades were attached to the inner wall of the reactor as depicted in the inset of Fig. 1.

The SiC powder was placed in the reactor chamber and heated to 898 K. The reactor chamber was rotated at 45 rpm. The liquid precursor (C<sub>2</sub>H<sub>5</sub>O)<sub>4</sub>Si was heated to 363 K and its vapors were introduced into the reactor chamber at a supply rate  $R_s$  of  $1.25 \times 10^{-6} \text{ kg s}^{-1}$  using Ar as a carrier gas. The flow rate of Ar was  $8.33 \times 10^{-7} \text{ m}^3 \text{ s}^{-1}$ . O<sub>2</sub> was separately introduced into the reactor chamber at a flow rate of  $3.33 \times 10^{-7} \text{ m}^3 \text{ s}^{-1}$ . The total pressure inside the chamber  $P_{\text{tot}}$  was maintained at 400 Pa, and the total deposition time was 7.2 ks.

The SiC/SiO<sub>2</sub> composite powder thus formed was consolidated by SPS (SPS-210LX, SPS Syntex, Japan).

A sample of uncoated monolithic SiC powder was also sintered for comparison purposes. The powder samples to be sintered were poured into a 10 mm inner diameter graphite die. This graphite die was covered with carbon wool that acted as a thermal insulator. The heating rate was  $1.7 \text{ K s}^{-1}$ , and the soaking time was 0.6 ks. The sintering temperatures ranged from 1723 to 2223 K. The temperature was measured by an optical pyrometer focused on a hole (diameter=2 mm and depth=5 mm) in the graphite die. The loading pressure was 100 MPa.

The densities of the sintered bodies formed from the SiC/SiO<sub>2</sub> composite as well as those from the monolithic SiC were measured using Archimedes' method and distilled water. Their values were converted to relative densities using the theoretical densities of β-SiC ( $3.21 \text{ Mg m}^{-3}$ ) [2] and SiO<sub>2</sub> ( $2.20 \text{ Mg m}^{-3}$ ) [11]. The crystal structures and phases of the sintered SiC/SiO<sub>2</sub> and SiC bodies were identified by X-ray diffraction (XRD) analyses (GRAD-C, Rigaku) using Cu Kα radiation. Their microstructures were observed by transmission electron microscopy (TEM) (EM-002B, TOPCON) and scanning electron microscopy (SEM) (S-3100H, HITACH). The Vickers hardness  $H_v$  and fracture toughness  $K_{\text{IC}}$  of the sintered SiC/SiO<sub>2</sub> and SiC bodies were determined at room temperature using a Vickers microhardness tester (HM-221, Mitutoyo) using loads  $P$  of 1.96 and 9.8 N. The hardness and toughness values were determined at 10 different points. The value of  $H_v$  (GPa) was calculated using Eq. (1):

$$H_v = 1.854 \times 10^{-9} \times \frac{P}{d^2} \quad (1)$$

where  $P$  (N) is the applied load and  $d$  (m) is the average value of the two diagonal lengths of the Vickers indentation. The value of  $K_{\text{IC}}$  (MPa m<sup>1/2</sup>) was found using Eq. (2) [13,14]:

$$K_{\text{IC}} = 0.073 \times 10^{-6} \times \frac{P}{c^{3/2}} \quad (2)$$

where  $c$  (m) is the average half length of the cracks formed around the corners of the indentations.

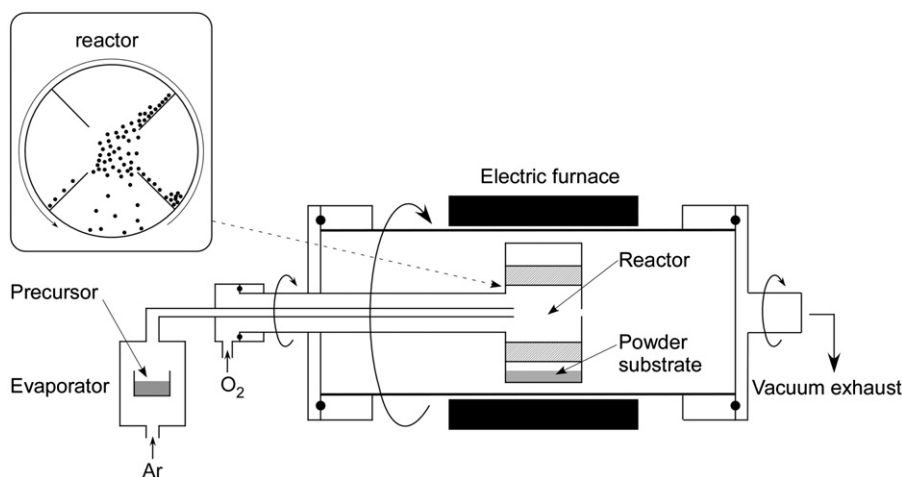


Fig. 1. Schematic of rotary chemical vapor deposition (RCVD) process.

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