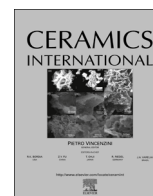




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# Silicon carbide ceramics: Mechanical activation, combustion and spark plasma sintering



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## ABSTRACT

Single-stage fabrication of SiC ceramics by a combination of self-propagating high temperature synthesis (SHS) and spark plasma sintering (SPS) is reported. SHS+SPS is demonstrated to be an efficient method for production of SiC ceramics with density 3.1 g/cm<sup>3</sup>, hardness of 24 GPa and toughness of 5 MPa m<sup>1/2</sup>. The starting material for the process is fine (50–300 nm in size) highly reactive powder, which involves composite particles of elemental carbon and silicon. This powder was prepared using a high-energy ball milling (HEBM). To optimize precursor preparation conditions, the structure transformation in nano-composite Si/C particles at different HEBM stages is also investigated.

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## 1. Introduction

Silicon carbide (SiC) ceramics have a set of unique physical-chemical properties, such as high hardness and mechanical stability at high temperatures, excellent thermal conductivity and low coefficient of thermal expansion, high resistance to corrosion and oxidation, wide bandgap and others [1]. This low density ceramic has been widely used for a variety of application in the automotive and aerospace industries, in high power electronics devices and bulletproof vests. Pressure-less sintering with additives, reaction-sintering and hot pressing are the main technologies for mass-production of SiC ceramics. However, the density of the ceramics produced by reaction sintering is relatively low (typically less than 85–90%). Hot pressing requires very high temperatures (> 2000 °C) and consequently, special and expensive equipment, furthermore, production of ceramics with density above 2.9 g/cm<sup>3</sup> without additives is still a challenge [2,3].

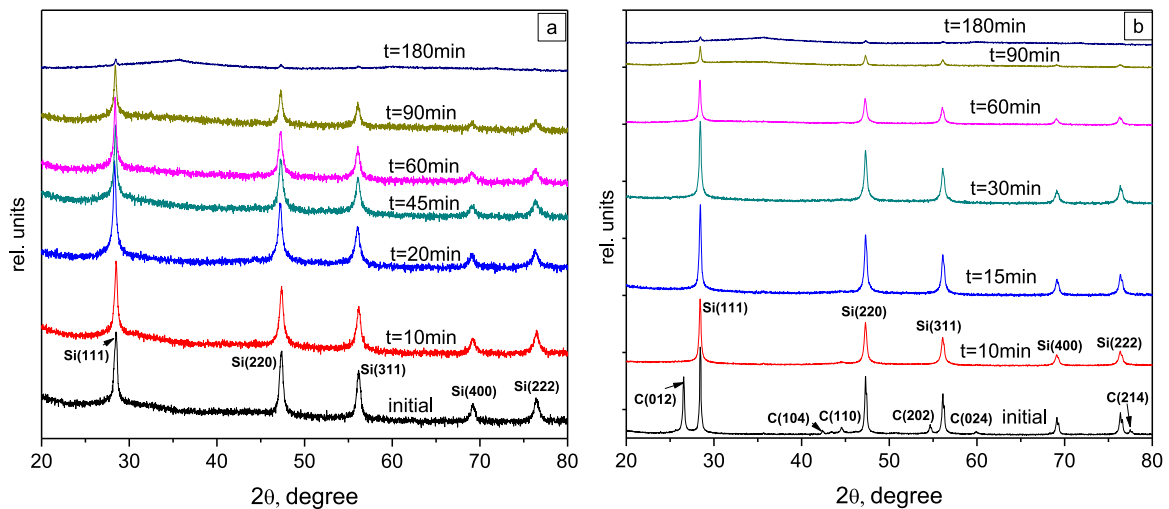
Recently it was demonstrated that fabrication of high quality (pore-free with nm-size grains) SiC ceramics requires use of sub-micron or nano SiC powders [4], as well as employment of advanced densification technologies, such as spark plasma sintering

(SPS) [5]. During SPS, the pulsating DC current passes through the graphite die, as well as the powder compact, when using conductive samples. This Joule heating and applied uniaxial load plays a critical role in the densification of powder compacts, which results in near theoretical density at lower temperatures and reduced processing times compared to conventional sintering techniques [6]. These conditions lead to a lower rate of grain growth, allowing one to preserve the sub-micron scale of SiC grains.

In our previous work, we have investigated SPS of different sub-micron SiC powders produced by combustion synthesis (CS) [7]. Here we report the fabrication of SiC ceramics by a one-step method that combines SPS and CS. Production of high-density ceramics by the SPS+CS approach requires the use of composite reactive initial powder. This composite reactive powder is obtained by use of High Energy Ball Milling (HEBM). HEBM is an intensive mechanical treatment of a powder mixture in an energetic planetary ball mill, which results in milling of the brittle components and plastic deformation of the ductile particles [8]. In addition, under optimum HEBM conditions, the composite particles are formed by cold welding of the initial precursors [9]. It was also found that HEBM of reactive systems (i.e. mixtures of two or more powders that can react in a self-sustained mode) results in a significant increase of their reaction rate [10]. Thus, it is concluded that this mechanical treatment “activates” the reactive systems, which enables the use of the mixtures for the one step process,

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**Fig. 1.** XRD spectra of Si-C composite particles prepared by HEBM with different milling time,  $t$ : (a) Si + amorphous carbon lampblack mixture; (b) Si + crystalline graphite mixture.

involving reactive sintering under SPS conditions [11–13]. Thus, we also report the mechanism of composite Si/C particles formation during HEBM process.

## 2. Experimental

Silicon (99.8%,  $d < 45 \mu\text{m}$ , Alfa Aesar, MA) and carbon lampblack (99.9%,  $d < 0.2 \mu\text{m}$ , Alfa Aesar, NJ) or graphite (99.5%,  $d < 45 \mu\text{m}$ , Alfa Aesar, NJ) powders were used as precursors.

The 5-g batches of initial stoichiometric Si:C (1:1 mol) mixtures were used for HEBM treatment in a PM100 single-station planetary ball mill (Retsch GmbH, Germany). The milling ball (stainless steel, 2 mm in diameter) to powder mixture weight ratio was 40:1. After the milling jar (stainless steel, 250 mL in capacity) was sealed, it was vacuumed and pressurized to  $\sim 2$  atm of 99.999% pure argon. A rotational speed of 650 rpm, was used, with the internal sun wheel rotation speed being 1300 rpm. HEBM time was varied from 10 to 90 min.

The prepared powders were placed into a cylindrical graphite die with 15.4 mm inside diameter, 30 mm outside diameter, and 30 mm height. The sample was also wrapped in graphite foil, 0.2 mm thick. The powder was sintered using a spark plasma sintering system (Labox 650, Sinter Land, Japan) under the following conditions: heating rate 200 °C/min, maximum temperature 2000 °C, pressure up to 50 MPa, dwell time 10 min. The parameters of the SPS set-up and schematic of the device are shown in Table 1S and Fig. 1S, correspondingly. The experimental parameters such as sample temperature, applied pressure, current, voltage, and sample displacement (shrinkage) were measured continuously during the consolidation cycle. Specifically, temperatures were measured by optical pyrometer focused on a hole drilled into the die surface to a depth of 4.5 mm. An emissivity value of 0.9 was used, which was obtained in calibration experiments with a two-color pyrometer.

The initial as-milled powders and as-produced materials were analyzed by a variety of characterization methods. Brunauer-Emmett-Teller (BET) specific surface area (SSA) measurements were carried out on a Coulter SA3100 analyzer at 77 K, with nitrogen as the adsorbent gas. All powders were vacuum outgassed at 298 K for 6 h before the SSA measurement. The microstructure of the powders was analyzed by field-emission scanning electron microscopy (FESEM; Magellan 400, FEI, Hillsboro, OR). Their phase compositions were determined by an X-ray diffraction (XRD)

analyzer (Scintag, X1 Advanced Diffraction System, Scintag Inc., Cupertino, CA) operated at 40 kV and 30 mA. TEM samples were prepared using a Ni grid. First, 0.05 g of the powder was put into a vial with ethanol and sonicated for 2 min. Next, two drops of the sonicated solution was put onto a Ni grid and dried for 2 min. Atomic level structure analysis was performed with a FEI-Titan 80–300 Transmission Electron Microscope (TEM) at 300 kV using both High Resolution Transmission Electron Microscopy (HRTEM) and High Angle Annual Dark Field (HAADF) Scanning Transmission Electron Microscopy (STEM) modes. The chemical composition of the samples was characterized by Energy Dispersive X-ray Spectroscopy (EDS) with spectral resolution of 130 eV (INCA, Oxford Instruments).

## 3. Results and discussion

### 3.1. Fabrication of composite reactive particles by high energy ball milling

A stoichiometric (1:1 mol) mixture of Si and amorphous carbon lampblack powders was subjected to HEBM under the aforementioned conditions. Fig. 1(a) shows XRD patterns for a set of Si/C composite materials obtained after different duration of the milling process. It can be seen that observed peaks belong to silicon phase. Even after 90 min of milling, no traces of the SiC phase can be detected. One can also see that the silicon x-ray peaks broaden with increased milling time, while their intensities simultaneously decrease. After 180 min of HEBM one can reach essentially complete *amorphization* of the mixture (Fig. 1(a)).

Fig. 1(b) shows the corresponding XRD spectra for the Si-graphite mixture. While the XRD spectrum of the initial silicon and graphite mixture contains peaks from both species, the graphite peaks completely disappeared after 10 min HEBM. It means that the milling conditions lead to extremely rapid *amorphization* of the graphite. Quantitative comparison of Fig. 1 a and b reveals that the XRD spectra of Si-C composites materials obtained with amorphous carbon and crystalline graphite after 60 min of high energy ball milling become almost identical.

Data on the specific surface area (SSA) of the powder is summarized in Fig. 2. The SSA values of the initial mixtures were found to be quite comparable, at 7 m<sup>2</sup>/g and 10 m<sup>2</sup>/g, for Si + crystalline graphite, C(c), and Si + amorphous carbon lampblack, C(a), respectively. This is due to the comparable SSAs of the crystalline

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