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Hot-spots generation, exaggerated grain growth and mechanical performance of silicon carbide bulks consolidated by flash spark plasma sintering

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ABSTRACT

Flash Spark Plasma Sintering (FSPS) that combines flash sintering and electric field assisted sintering was used to densify SiC ceramics. FSPS was compared with 'conventional' SPS which used a moderate 100 °C·min^{-1} heating rates. Dense SiC specimens were obtained despite being at temperature of 1850–2050 °C for few seconds. FSPS lead to generation of hot-spots and thus caused localized exaggerated grain-growth. This allows producing silicon carbide ceramics with bimodal grain size distribution. Analysis of microstructure and strength of silicon carbide ceramics consolidated using flash heating allowed proposing several optimization routes of the FSPS process.

1. Introduction

There is an unstoppable trend for quicker fabrication of ceramic materials. Within last few decades methods like microwave sintering or spark plasma sintering (SPS) showed that conventionally hard to sinter materials can be heat-up to sintering temperature using heating rates exceeding 100 °C·min^{-1} and consolidated within minutes [1–6]. Flash sintering (FS) approach squeezes processing time to seconds at relatively low-temperatures [7]. Originally FS experiments were focused mainly on consolidation of various oxide ceramics and had a limitation of size and shape [7]. Recent studies show that by using modification to original FS techniques non-oxide compounds such as SiC, ZrB_2 or B_4C ceramics may be consolidated using 'flash' regime [8–12]. Grasso et al. [11], in particular, showed that consolidation of large size specimens of SiC (60 mm in diameter) can be completed within minutes using the flash SPS (FSPS) method. Due to abandoning of graphite mold-punches set-up, heating during FSPS is mainly controlled by ceramic sample properties. Furthermore, FSPS approach showed that temperature distribution inside flashed specimens can be lead to creation of hot-spots. A temperature gradient between center and surface of the SiC specimen during flash SPS was estimated as 200–300 °C using finite-elements simulation [11]. Using FSPS approach Vasylkiv at al. successfully reported consolidation of the 3YSZ ceramics with size of 20 mm in diameter [13]. Taking into account a possibility of producing relatively large size specimens by both works [11,13], it is natural to seek whether the consolidation by FSPS of such advanced ceramics as SiC [14–17] may lead to improvement of mechanical properties.

Hence, the aim of this study was to consolidate commercially available SiC powders by the FSPS method targeting large size specimens. Secondly, to explore the difference between mold-free 'flash' SPS and regular SPS set-up that uses graphite die-punches set [13]; performing both procedures using identical hardware [13]. Finally, to verify whether FSPS can be utilized to consolidate bulk SiC specimens and lead to increase in mechanical properties such as flexural strength at room temperature and at 1600 °C.

2. Materials and methods

Commercially available beta silicon carbide (β-SiC UF, Ibiden Co., Gifu, Japan) and amorphous boron (aB, 97%, Wako Pure Chemical

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Industries, Ltd., Osaka, Japan) [18] powders were used as the starting materials (Fig. 1). Silicon carbide with 1 wt% aB mixture was homogenized using wet mixing in alcohol, followed by drying at about 100 °C. The resultant powders were screened through a 60, 400 and 1250 mesh screens.

The homogenized powder mixture was loaded into a graphite die with an inner diameter of 30 and 50 mm and subjected to SPS. The outer surface of the die was wrapped in 5-mm-thick graphite felt to homogenize the temperature distribution and reduce heat loss by radiation. The mold system containing the powder mixture was placed in an SPS furnace ('Dr. Sinter', SPS 1050, Sumitomo, Japan) [11].

A two-step consolidation process was used to produce FSPS and SPS specimens. First, specimens with diameter of 30 or 50 mm and high of 5–7 mm were prepared by the preliminary SPS consolidation at 1200 °C. The samples were heated under vacuum to 1200 °C at 100 °C·min^{-1} under axial pressure of 60 MPa. After 20 min of dwell at this intermediate step, samples for the FSPS were cooled down to room temperature at a rate of 100 °C min^{-1} (Fig. 2(a)). The specimens were evacuated from the graphite die and were subjected to the mold-free [11–13] FSPS consolidation. This step consisted of wrapping the pre-consolidated SiC specimens with additional graphite foils and putting them into set-up in depth described in Ref. [13].

The temperature during FSPS experiments was probed by the side pyrometer focused on the side of the graphite felt using an emissivity of 0.90. In FSPS experiments, a constant uniaxial pressure of 20 MPa was applied. The samples were discharged under a peak power of about for about 5–20 s (Fig. 3). The power was switched off after selected discharge time, and specimens were allowed to cool to room temperature under unchanged pressure conditions. FSPS experiments were performed in argon gas with a flow rate of 2 L min^{-1}.

For the reference, the samples were also sintered using a conventional SPS configuration. For these studies, SPS was continued after a preliminary consolidation step at 1200 °C. Thus, after the 20 min dwell at 1200 °C, the SPS chamber was backfilled with argon, and pre-consolidated SiC specimens were heated up to 1900 °C at a rate of 100 °C min^{-1} and were held for 15 min (Fig. 2(b)). The pressure of 60 MPa was maintained during consolidation and cooling stages. Each specimen was gradually cooled to 600 °C at a rate of 100 °C min^{-1} and then naturally to room temperature in the furnace. Argon gas with a flow rate of 2 L min^{-1} was used.

The sintered specimens were ground with diamond disks with a particle size of up to 0.5 μm. Then, the density of the samples was measured by the Archimedes method using ethanol as a medium in accordance with ASTM B 963–08.

The three-point flexural strength was determined using rectangular bars (2 × 2.5 × 20 mm) cut from specimens with a diameter of 30 and 50 mm using electric discharge machining. Their lateral surfaces were ground and polished using diamond pastes.

The flexural strength tests were conducted at room temperature and at 1600 °C in argon using a Shimadzu AG-X plus system (Shimadzu, Japan). The loading speed was 0.5 mm min^{-1}. Twelve bars were tested at room temperature, and five specimens at 1600 °C. For the flexural strength tests at 1600 °C, the following heating schedule was used: from room temperature to 200 °C in 10 min and from 200 °C to 1600 °C at a rate of 18 °C min^{-1} [19]. A dwell time of 5 min was employed before the flexural test at 1600 °C. After testing, cooling from 1600 °C to room temperature was performed at a rate of 20 °C min^{-1}.

Microstructural observations and analyses were carried out on the fracture surfaces using scanning electron microscopes (SEM) SU 8000 cold-emission FE-SEM Hitachi and JEOL 6500F equipped with energy dispersive spectroscopy (EDS) detectors. Observations were made on fractured surfaces after flexural tests.

3. Results and discussion

Fig. 3 shows analysis of raw data obtained for SiC tile #1f consolidated using the flash spark plasma sintering method. Fig. 3(a) shows the output of the Sumitomo SPS machine recorded during FSPS. Flash consolidation does not start from the moment

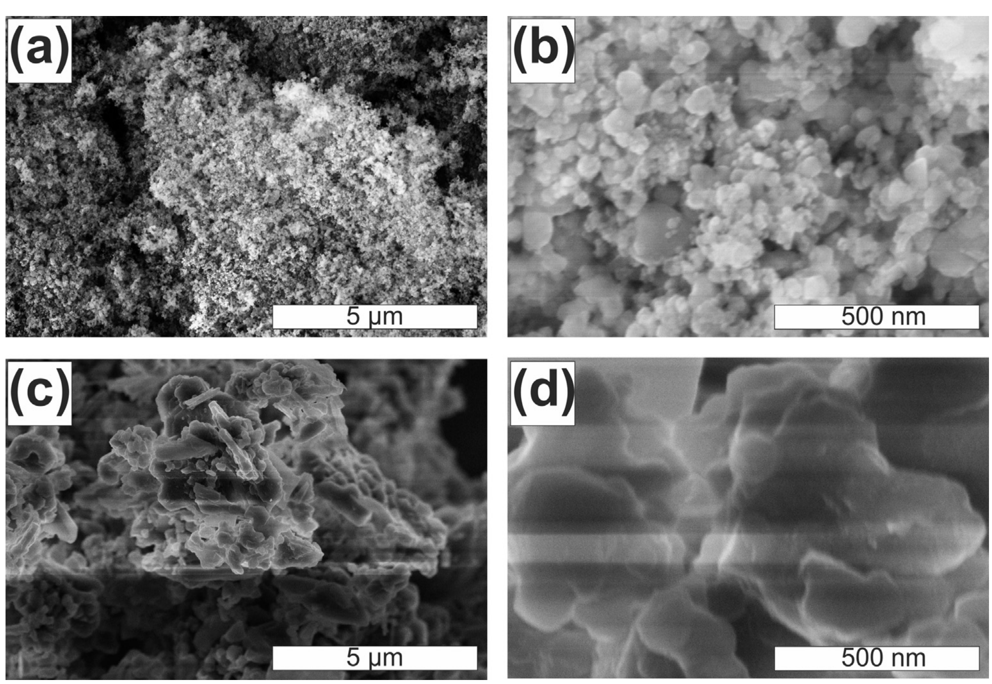

Fig. 1. SEM images of initial β-SiC (a,b) and amorphous boron (c,d) powders.