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Influence of spark plasma sintering parameters on densification and mechanical properties of boron carbide

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ABSTRACT

The densification behavior of boron carbide without sintering additives is reported for temperatures in the range of 1100 °C to 1800 °C by spark plasma sintering (SPS) technique together with the sintering parameters (Holding Time and Pulsed DC). The influence of porosity on mechanical properties (hardness, fracture toughness and elastic modulus) of boron carbide prepared by SPS is measured. Pulsed DC current is found to play a dominant role in the densification of boron carbide and in achieving near theoretical density at lower sintering temperature compared to conventional sintering techniques. Hardness, fracture toughness and elastic modulus of fully dense B_4C are measured as 37.2 GPa, 2.8 MPa.m$^{1/2}$ and 570 GPa respectively. Microstructural analysis indicates the presence of deformation twins in boron carbide grains.

1. Introduction

Boron carbide (B_4C) is one of the candidate materials for many high performance applications due to its combination of attractive properties like low density, high hardness, high elastic modulus, high melting point, high neutron absorption cross-section, good thermo-electric properties and chemical inertness. However, the application of this material is limited by its poor sinterability due to the existence of high degree of covalence of bonds between B and C atoms in B_4C. High sintering temperatures close to the melting point of boron carbide is required to achieve a complete densification of B_4C. This high sintering temperature in turn results in grain coarsening and residual porosities that deteriorate the material's performance [1–6]. Under these circumstances, several attempts have been made to develop high dense compacts by pressure-assisted compaction techniques like hot-pressing or hot-isostatic pressing with and without addition of sintering aids [1–3,7–16]. Sintering aid additions are reported to lower the consolidation temperatures of B_4C ceramics, but at the expense of purity of final product. Purity is of prime concern for applications in nuclear industries [1,2]. Thus, there is a need of new sintering techniques which would alleviate the sintering conditions for attaining dense B_4C ceramics without sinter additions. Spark plasma sintering (SPS) or Plasma Pressure Compaction is one such technique for densifying any class of materials, especially for materials that are difficult to sinter with conventional techniques. The presence of plasma in spark plasma sintering system is still unproven. Specific advantages of SPS over conventional sintering techniques are (1) faster heating rate, which avoids those mass transport mechanisms that do not contribute to densification (2) shorter dwell time, that retains finer microstructures and (3) DC pulse voltage that contributes for enhanced mass transport through electro-migration [17–22]. The present work was aimed at better understanding of processing conditions (time, temperature and

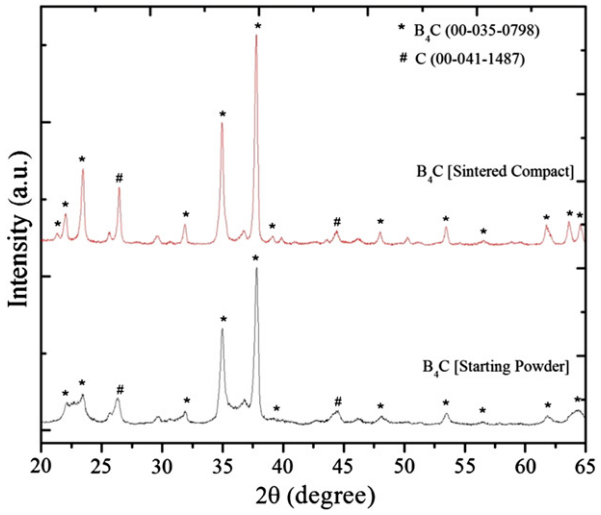

Fig. 1. XRD of starting B_4C powder and SPS processed full dense B_4C pellet.

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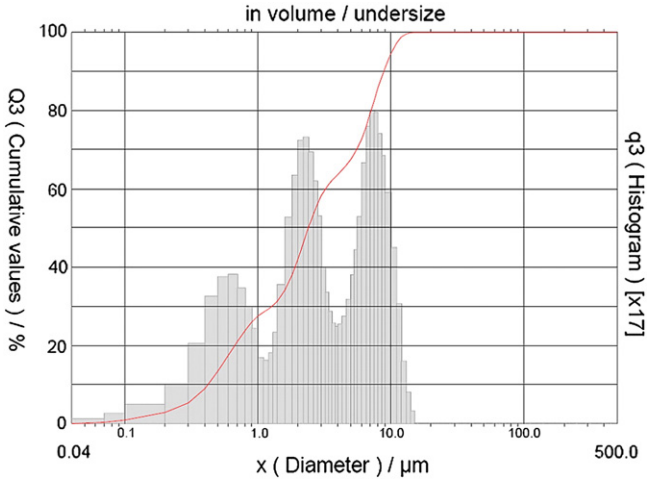

Fig. 2. Particle size distribution of starting B_4C powder (tri-modal).

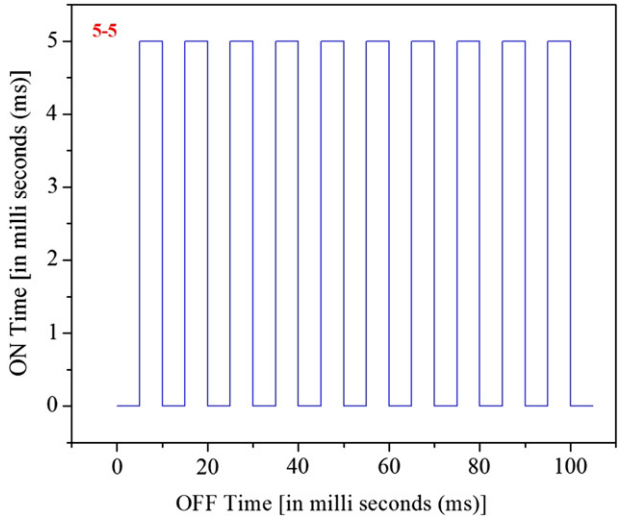

Fig. 3. Schematic of Pulsed DC waveform showing 5–5 (ms–ms) pulse sequence during the holding period.

Pulsed DC) in the densification and on mechanical properties of monolithic B_4C ceramic by spark plasma sintering technique.

2. Experimental procedure

2.1. Characterization of starting material

Boron carbide synthesized in-house, (B_4C, Particle size $D_{50} = 2.4$ µm) was used as a starting powder in the present study. The method adopted to produce powders was direct elemental reaction of B and C at 1800 °C under high vacuum condition, its detailed synthesis procedure is reported elsewhere [23]. The B_4C powders were characterized for phase identification by XRD (Cu–Kα, XRG3000, Inel, France) (Fig. 1) and particle size analysis by Laser Scattering Particle Size Analyzer (1064 liquid, Cilas, France) (Fig. 2). The chemical composition of the starting material was analyzed as 77.7 wt.% B, 22 wt.% C and 0.2 wt.% O.

2.2. SPS operation

Boron carbide powder was placed in a high density graphite die of internal diameter 17 mm. A piece of grafoil sheet (0.45 mm thick) was lined internally in the graphite die and also placed in between powder and plunger in order to avoid the interaction with the graphite material. The die set filled with the B_4C powder was then placed inside the SPS processing chamber. A minimum load of 2 kN was applied at the start, in order to ensure the firm contact of electrodes with the plunger/sample/die set up. Standard DC pulse ON–OFF sequence (5 ms-5 ms) was chosen for all the experiments and the pulse was considered as square waveform type (Fig. 3). The densification was performed in vacuum chamber between 1100 °C and 1800 °C with an interval of 100 °C and a soaking time of 15 min at the set temperature. The heating rate above 1000 °C was maintained as 100 °C/min. Sintering temperature was measured by focusing the optical pyrometer at the graphite plunger which is in direct contact with the powder compact as shown in Fig. 4. A maximum load of 12.5 kN (50 MPa) was applied at a rate of 0.25 kN/s when the temperature reached the set point. Holding time was maintained for 5, 10 and 15 min at that temperature. After completion of experiment, furnace was allowed to cool down to room temperature and the pellets were ejected from the die. The ejected pellets were cleaned on both the edges and circumference by removing the grafoil sheets.

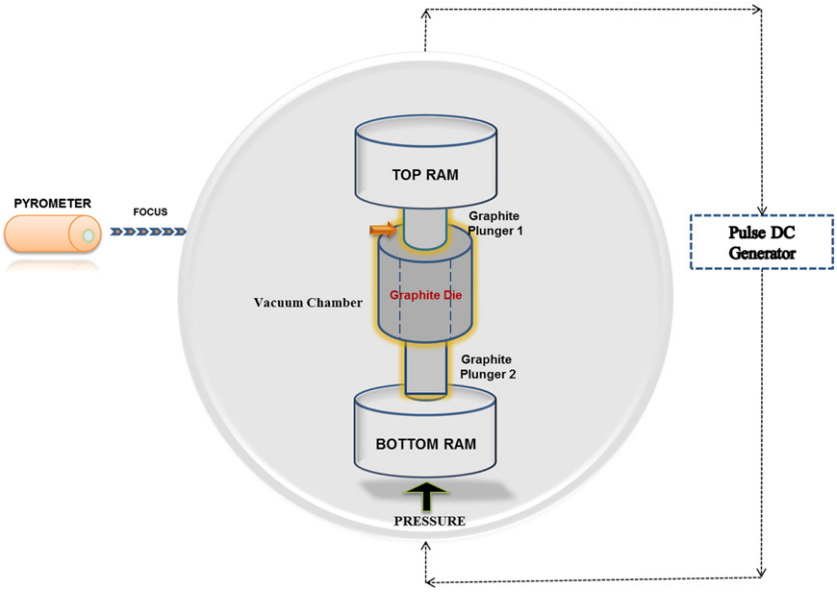

Fig. 4. Schematic of Spark Plasma Sintering (SPS) Facility showing the location of temperature measurement by optical pyrometer.