PLASMA SINTERING OF CERAMIC MATERIALS

Kazunori KIJIMA
National Institute for Research in Inorganic Materials
Sakura-mura, Niihari-gun, Ibaraki 300, JAPAN

ABSTRACT

Silicon carbide, which is one of the most unsinterable ceramics, was successfully sintered in a thermal plasma of argon. This report will show how to obtain silicon carbide ceramics sintered to nearly theoretical density. Some merits of plasma sintering for ceramics will be demonstrated and in compared with normal sintering.

1. INTRODUCTION

The major merits in application of thermal plasmas to chemical processing and materials preparation are in its use of active species, optical emissions and thermal energy. There are, however, few reports about applications to ceramic sintering, which should become a very important field, as well as powder syntheses and film depositions. Alumina is one typical oxide ceramics which has been sintered in plasma\(^1\)\(^-\)\(^2\) and few reports about plasma sintering of non-oxide ceramics such as silicon carbide have been published.\(^3\)

Much of the interest in sintering silicon carbide has been created from the increasing demand for engineering materials in high temperature applications such as gas turbines and heat exchangers. It is difficult to obtain a highly densified product by sintering SiC.\(^4\)

2. EXPERIMENTAL

The starting powder was commercially available beta-silicon carbide whose major impurities are free carbon, free silicon\(_4\), aluminum and iron. The mean particle size is \(0.3 \times 10^{-6}\) m. The powder was mixed with amorphous boron and phenol resin in ethanol using a plastic ball mill for 24 hours. The amount of sintering additives were 0.5 wt % of boron and 1.0 wt % of residual carbon from phenol resin. The mixture was dried thoroughly in vacuum for several hours. A bar
(5x5x40 mm) was shaped in a die at a pressure of 300 kg/cm² and then isostatically pressed at 2000 kg/cm². The resultant green densities of the bar were between 1.90 and 2.10 g/cm³.

Fig. 1 shows a block diagram of the apparatus used for the present study. The plasma furnace is made of a silica tube with water cooling. The gaseous pressure and flow rate were controlled to keep constant while sintering. Argon gas of high purity grade was used without special purification. A high frequency generator can be generated 4 MHz of frequency and 15 kW of maximum output power, which was inductively coupled to argon gas by a work coil without an electrode. The argon plasma was ignited by evacuating the furnace to 7 Pa (0.05 torr) and a stable egg-shaped thermal plasma was obtained after pressurizing argon. The bar of silicon carbide to be sintered was introduced gradually into the plasma, since the plasma moved away from the cold bar. The bar was supported by a graphite rod and located concentrically within the furnace. The gaseous pressure was measured using a diaphragm type gauge and gas flow rates were measured by a rotameter without correction for pressure.

![Block Diagram](image)

**FIG. 1** A block diagram for plasma sintering

3. RESULTS

Fig. 2 shows a picture of plasma sintering occurring. The shape of the thermal plasma changes from whole egg, as shown in Fig. 2, to half egg by increasing the argon gas flow rate. The length of the egg decreased by pressurizing the argon gas. A sample to be sintered can be seen below the plasma. The argon gas flowed down stream and a sample was inserted into the plasma by moving it up slowly. A rapid insertion of a stream into the plasma cause plasma instability and thermal crack in the sample. Residual stress in a sample produced during the forming process also leads to cracks in the sample.

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during sintering. The insertion speed was controlled by observing the interaction between the plasma and a sample at red heat. Longer duration of plasma sintering caused evaporation of the sample lowering its density. A duration of 120 sec was sufficient to reach theoretical density in the present work.

Typical conditions required to obtain nearly theoretical density were: Ar gas flow rate = 600 ml/min (without calibration), gaseous pressure = 6.7x10^4 Pa (50 torr), anode voltage = 7 kV, anode current = 1.5 A, grid current = 70 mA, and sintering duration = 60 sec.

Silicon carbide compact powders were densified to 96-99% of theoretical density using these conditions. The surface of the sintered body became mirror-like after polishing. Fig. 3 and Fig. 4 are SEM microphotographs of fractured surface before and after plasma sintering, respectively. The green density of the sample was about 64% of theoretical. The grain sizes of a green body and a sintered body were measured to be 0.2-1 micron from Fig. 3 and 0.5-2 micron from Fig. 4, respectively. The grain sizes of traditionally sintered bodies are more than 5 micron SiC. The SEM observation indicated that plasma sintering could control grain growth during densification. Abnormal grain growth could not be observed from SEM observation. Plasma sintering may be suitable to obtain a fine microstructure for SiC ceramics. Fine grain and pore sizes are effective on mechanical strength as shown in Fig. 5.

The observation of fractography in Fig. 4 showed that fracture propagated along grain boundaries in the sintered body. Reasons for breaking down between particles are not clear at present. It could be due to fracture speed or relatively weak bonding between particles. It is necessary to study the fractography and mechanical properties in more detail.

There have been many discussions about the unsinterability of SiC. It is highly covalent bonded crystal and has low diffusivity. In the earliest stages, the major sintering mechanisms at normal sintering temperature (2300-2600 K) are considered to be surface diffusion and grain boundary diffusion.6) This well explains the sintering phenomena that silicon carbide can not be densified by normal sintering without additives. Plasma sintering has clearly promoted
FIG. 3 SEM microphotograph of SiC powder before sintering.
(2x10^-6 m: ___)

FIG. 4 SEM microphotograph of SiC ceramics after sintering.
(2x10^-6: ___)
densification and restrained grain growth in the present experiments, implying that the mechanism of plasma sintering differs from that of normal sintering.

Plasma sintering showed the following advantages compared with normal sintering: 1) sintering duration was very short (1/5-1/50 of normal sintering time); 2) densification was enhanced without grain growth; 3) long samples could be sintered by passing continuously and through the plasma; 4) energy efficiency was high. These advantages might be brought about by changing from diffusion rate and sintering mechanism from surface diffusion to volume diffusion.

REFERENCES


