



## Original Research Paper

## Spark plasma sintering of silicon nitride using nanocomposite particles

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## ABSTRACT

The spark plasma sintering (SPS) of silicon nitride ( $\text{Si}_3\text{N}_4$ ) was investigated using nanocomposite particles composed of submicron-size  $\alpha$ - $\text{Si}_3\text{N}_4$  and nano-size sintering aids of 5 wt%  $\text{Y}_2\text{O}_3$  and 2 wt%  $\text{MgO}$  prepared through a mechanical treatment. As a result of the SPS,  $\text{Si}_3\text{N}_4$  ceramics with a higher density were obtained using the nanocomposite particles compared with a powder mixture prepared using conventional wet ball-milling. The shrinkage curve of the powder compact prepared using the mechanical treatment was also different from that prepared using the ball-milling, because the formation of the secondary phase identified by the X-ray diffraction (XRD) method and liquid phase was influenced by the presence of the sintering aids in the powder compact. Scanning electron microscopy (SEM) observations showed that elongated grain structure in the  $\text{Si}_3\text{N}_4$  ceramics with the nanocomposite particles was more developed than that using the powder mixture and ball-milling because of the enhancement of the densification and  $\alpha$ - $\beta$  phase transformation. The fracture toughness was improved by the development of the microstructure using the nanocomposite particles as the raw material. Consequently, it was shown that the powder design of the  $\text{Si}_3\text{N}_4$  and sintering aids is important to fabricate denser  $\text{Si}_3\text{N}_4$  ceramics with better mechanical properties using SPS.

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## 1. Introduction

Silicon nitride ( $\text{Si}_3\text{N}_4$ ) ceramics have been widely used in the industrial field because of their excellent mechanical, thermal, and chemical properties. However,  $\text{Si}_3\text{N}_4$  ceramics are very difficult to densify using solid state sintering because of their strong covalent bond and low self-diffusion constant. Liquid phase sintering by adding sintering additives is considerably more effective at fabricating fully dense  $\text{Si}_3\text{N}_4$  ceramics [1–6]. In liquid phase sintering, densification occurs as a result of particle rearrangement due to the formation of the liquid phase, followed by solution precipitation due to  $\alpha$ - $\beta$  phase transformation and Ostwald ripening to develop an elongated  $\beta$ - $\text{Si}_3\text{N}_4$  grain structure. It is well-known that the composition and characteristics of the liquid phase have a significant effect on the densification and microstructure development [7–9].

Spark plasma sintering (SPS) is a recently developed field-assisted sintering technique. It can produce rapid heating and complete densification in a short period of time at low temperatures in comparison with conventional sintering methods. Using the SPS

technique, hard-to-sinter oxides, nitrides, and carbides have been densified by applying a pulsed DC current passing through a graphite mold, leading to significant consolidation [10–14]. Many researchers have already reported the results of analyses of the sintering mechanism and microstructure evolution in  $\text{Si}_3\text{N}_4$  during SPS [15–17]. For example, Shen et al. suggested the formation of tough interlocking microstructures in  $\text{Si}_3\text{N}_4$  ceramics through a dynamic ripening mechanism during SPS [18]. Suganuma et al. reported the sintering behavior of submicron  $\alpha$ - $\text{Si}_3\text{N}_4$  powder by comparing the microstructures after SPS and HP [19]. Belmonte et al. reported the results of a reliability evaluation of  $\text{Si}_3\text{N}_4$  ceramics subjected to SPS. Their report showed the microstructures and  $\alpha$ - to  $\beta$ - $\text{Si}_3\text{N}_4$  phase transformation in relation to the depth from the surface of the sintered  $\text{Si}_3\text{N}_4$  bodies [20]. Hotta et al. and Lee et al. studied ways to control the microstructure using different heating rates and a nano-size amorphous  $\beta$ - $\text{Si}_3\text{N}_4$  powder [21,22]. Nishimura et al. reported that dense  $\text{Si}_3\text{N}_4$  ceramics were obtained via a high-energy mechanical milling process and subsequent SPS [16,23]. As a result, milled powders could be sintered using SPS at a relatively low temperature, and homogeneous dense  $\text{Si}_3\text{N}_4$  ceramics with a grain diameter of approximately 70 nm could be fabricated. Hu et al. recently investigated the rapid anisotropic grain growth of  $\alpha$ - and  $\beta$ - $\text{Si}_3\text{N}_4$  grains in a  $\text{Si}_3\text{N}_4$  microstructure using SPS. This report revealed that  $\alpha$ - to  $\beta$ - $\text{Si}_3\text{N}_4$  phase

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transformation at a high temperature favors the high supersaturation of dissolved  $\text{Si}_3\text{N}_4$  in the formed oxynitride melt, which promotes the ordered coalescence of precipitated  $\beta\text{-Si}_3\text{N}_4$  nanocrystallites. As a result, elongated  $\beta\text{-Si}_3\text{N}_4$  was formed by the ordered coalescence of nanocrystallites [24]. In addition, we have been reported the fabrication and evaluation of  $\beta\text{-SiAlON}$  using the SPS technique. Its superior properties were due to the much higher dispersion of raw materials and the absence of a sintering aid [25].

As previously stated, SPS is characterized by very rapid heating. Therefore, the location of the sintering aids in the green body should have a significant influence on the sintering behavior of  $\text{Si}_3\text{N}_4$  because the time required for the diffusion and reaction of the sintering aids should be relatively shorter than the conventional sintering process. In our previous studies, nanocomposite particles were prepared to obtain a uniform dispersion of nanoparticles in ceramics using a mechanical treatment as a dry process, in which high shear and compressive stresses were applied to the powders [26–32]. Although the sintering behavior of  $\text{Si}_3\text{N}_4$  using  $\alpha\text{-Si}_3\text{N}_4$ /sintering additive nanocomposite particles has been reported for gas pressure sintering [32], the behavior when using the SPS technique has not yet been investigated. In particular, we anticipated that the presence of the sintering aids in the starting powders would affect the densification, especially when the heating rate was high. The purpose of this study was to determine the sintering behavior of  $\text{Si}_3\text{N}_4$  through the SPS technique using nanocomposite particles prepared by a mechanical treatment.

## 2. Material and methods

A commercial  $\alpha\text{-Si}_3\text{N}_4$  powder (Ube, SN-E10 grade,  $\beta$ -phase: <5%, total oxygen content: 1.2 mass%, purity: >95%) was used as a starting material. The  $\alpha\text{-Si}_3\text{N}_4$  particles had a spherical shape, and the particle size was approximately 170 nm, as shown in Fig. 1(a), which could be confirmed by the specific surface area ( $10.9\text{ m}^2/\text{g}$ ) measured using the Brunauer-Emmett-Teller (BET) method.  $\text{Y}_2\text{O}_3$  (BB-type, Shin-Etsu Chemical Co., Ltd., Japan, purity: 99.9%) and MgO (500A, Ube Industries, Japan, purity: 99.9%) powders were used as sintering aids. The  $\text{Y}_2\text{O}_3$  particles were present in

the form of needle-like aggregates, as shown in Fig. 1(b), and the primary particle size estimated from the specific surface area ( $31.9\text{ m}^2/\text{g}$ ) was approximately 37 nm. The MgO particles had spherical shapes and were also very aggregated (Fig. 1(c)). The BET specific surface area was  $39\text{ m}^2/\text{g}$ , and the particle size estimated from the specific surface area was approximately 43 nm. The batch composition was  $\text{Si}_3\text{N}_4\text{:Y}_2\text{O}_3\text{:MgO} = 93\text{:}5\text{:}2$  (weight ratio). These starting powders were mechanically treated using a powder composer (NOB-130, Hosokawa Micron Co., Japan), in which high shear and compressive stresses were applied to the powders. First, the  $\text{Si}_3\text{N}_4$  powders were placed in a chamber and premixed at approximately 1000 rpm for 1 min. After premixing, the  $\text{Y}_2\text{O}_3$  and MgO powders were also placed in the chamber and premixed again under the same conditions. The total quantity of the raw powders was about 172 g. After that, they were mechanically treated by applying power at 5 kW for 10 min. For comparison, these raw powders were mixed by ball-milling at 110 rpm for 24 h with an added dispersant (Celuna E-503, Chukyo Yushi Co., Ltd., Japan) in ethanol. The specific surface areas of the powder mixtures were measured using the BET method (BELSORP II, BEL Japan Inc., Japan).

Approximately 1.8 g of the powder mixtures were placed in graphite dies with an inner diameter of 15 mm. They were sintered using an SPS furnace (Model SPS-2040, Sumitomo Coal Mining Co., Ltd., Tokyo, Japan) at temperatures of 1600 °C for a holding time of 0 min in an  $\text{N}_2$  atmosphere under a uniaxial pressure of 30 MPa. The SPS experiments were carried out at heating rates of 106, 475 and 950 °C/min to a temperature of 1550 °C. Then, the samples were heated to 1600 °C at a heating rate of only 50 °C/min to prevent an overshoot. Finally, they were furnace-cooled at a cooling rate of about 10 °C/s. The temperature was controlled during the sintering using a pyrometer focused at the surface of the graphite die. The linear shrinkage of the powder compacts was continuously recorded during the SPS as the displacement of the punch rod. The relative density of the fabricated  $\text{Si}_3\text{N}_4$  ceramics was measured using the Archimedes method. The phases present in the powder mixtures and  $\text{Si}_3\text{N}_4$  ceramics were identified using the X-ray diffraction (XRD) method (Multiflex, Rigaku Co., Japan). The present ratio of the  $\alpha$ - and  $\beta\text{-Si}_3\text{N}_4$  phases in the sintered  $\text{Si}_3\text{N}_4$  body when the heating rate of 950 °C was confirmed using

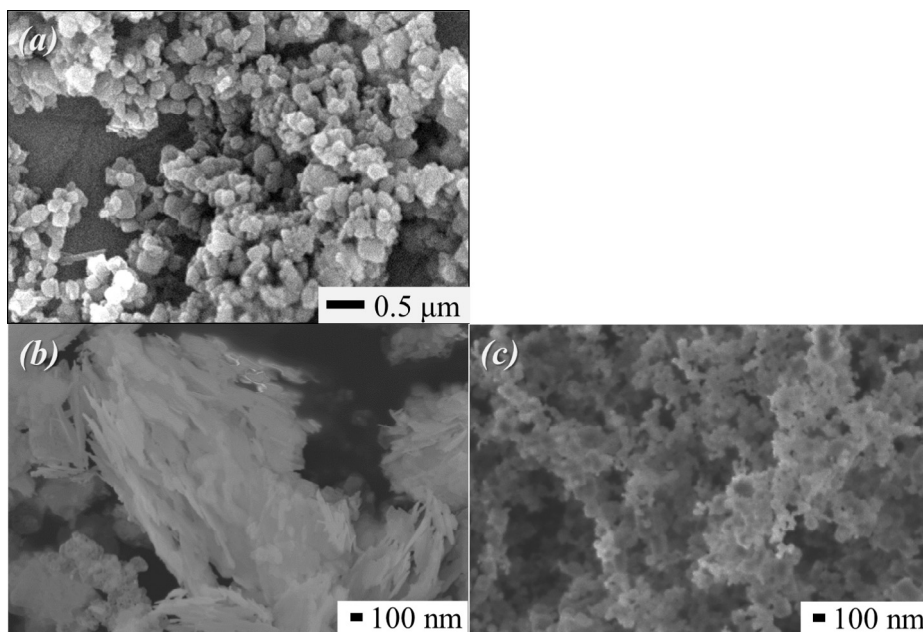


Fig. 1. SEM photographs of raw powders: (a)  $\text{Si}_3\text{N}_4$ , (b)  $\text{Y}_2\text{O}_3$ , and (c) MgO.

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