The effect of the starting Si powder on the sinterability and thermomechanical properties of sintered reaction-bonded silicon nitride

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The effects of the size of Si powder on the thermal conductivity and strength of sintered reaction-bonded silicon nitride (SRBSN) were investigated. Si powders with various sizes were prepared by controlling the high-energy milling duration of the starting Si powder, where the median Si size varied from 3.97 to 0.62 μm, corresponding to an oxygen content of 0.54 to 2.57 wt %. The thermal conductivity increased gradually up to the 4 h milling (D₉₀ = 1.10 μm or 1.60 wt % oxygen) owing to a gain in the relative density, but decreased by 25% for the 12 h milling (D₉₀ = 0.62 μm or 2.57 wt % oxygen). On the other hand, the flexural strength showed almost no change until the 4 h milling, but increased by 20% for 12 h milling. This opposite tendency was elucidated by the difference in grain size distribution and thickness of intergranular grain boundary phase (IGP); Clear bimodal size distribution of grains contributed to high strength, and the nearly doubled thickness of IGP was observed for SRBSN from the finest Si powder (D₉₀ = 0.62 μm or 2.57 wt % oxygen). In order to meet the required thermal properties for the substrate usings by sintering at 1900°C, the size of silicon powder of D₉₀ = 1.10 μm (1.60 wt % oxygen) is allowed for 6 h sintering, but that of D₉₀ = 0.62 μm (2.57 wt % oxygen) needs prolonged sintering time over 6 h (12 h in the current experiment) to reach required grain growth.

Key-words : Silicon nitride, Reaction bonded, Gas pressure sintering, Thermal conductivity, Flexural strength

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1. Introduction

Two typical processing routes to fabricate silicon nitride ceramics with high thermal conductivity are known: (1) conventional sintering known as sintered silicon nitride starting from Si₃N₄ powder, and (2) reaction sintering termed sintered reaction-bonded silicon nitride (SRBSN) starting from Si powder. It is considered that the reaction sintering process is more promising given such merits of higher thermal conductivity due to the low oxygen content, near-net characteristics, and cost effectiveness.¹⁻⁶ Since the mid-1990s, many research papers have been published on SRBSN focusing on variables including Si powders, sintering additives, nitriding and post-sintering processes, microstructural analyses, mechanical and thermal properties, and other facets.

As is well known, the size of Si powder is critical when seeking to control both mechanical and thermal properties. However, fact is that most of previous works only mention the size of the starting Si prior to milling process, so no information on the actual Si size (or oxygen content) used for the specimens is specified.⁷⁻¹⁰ It should be noted that the size of the Si powder can be changed in a wide range by milling depending on the energy imposed during the milling process.

In this study, the effect of the Si size (or the oxygen content) on the mechanical and thermal properties of SRBSN were investigated in a systematic manner. That is, a wide range of the size of Si powders was prepared by controlling the pulverizing time, after which the oxygen contents of specimens before and after a nitriding reaction were measured. The acquisition of data on the Si size and corresponding oxygen content for each step is expected to shed light on how the size specification of Si powders affects the microstructure and properties of SRBSN. It is stated beforehand that the effect of starting Si particle size is exactly equivalent to that of oxygen content for the analysis on the thermal conductivity in this paper.

2. Experimental procedure

Commercial Si powder (grade 4NE, 99.99%, Vesta Ceramics, Ljungaverk, Sweden) and sintering additives of yttria (Y₂O₃, grade C, H.C. Starck, Goslar, Germany) and magnesia (MgO, metal basis 99.99%, Sigma Aldrich) were used to fabricate the SRBSN ceramics. The molar ratio of

[Received April 12, 2018; Accepted December 19, 2018]
the sintering additives was 2 mol.% Y2O3 and 5 mol.% MgO, referred to as 2YSi, under the assumption that the Si powder in the green body were completely transformed into reaction-bonded silicon nitride (RBSN) without residual Si. In order to analyze the effects of the Si powder size on the microstructure and properties, high-energy milling for Si powder only (without mixing the sintering additives) was conducted using a zirconia jar and zirconia balls at 320 rpm for 0, 1, 2, 4, and 12 h. The size of the Si powder for varied milling times was measured using a dispersive laser particle size analyzer (Beckman Coulter LS-13320, USA). For the Si powder mixture (Si powder plus sintering additives), the same milling procedure was employed for the fabrication of the SRBSN specimens. The names of the Si powders, RBSN, and SRBSN specimens in this experiment were designated as m0h, m1h, m2h, m4h, and m12h after the milling times of the Si powder used. The high-energy milled powder mixtures were dried in a rotary evaporator and sieved through a polyethylene sieve of <150 μm. The green bodies were uniaxially pressing at a cylindrical metal mold of ϕ = 15 mm and subsequently cold isostatic pressing (CIP) at 200 MPa for 5 min. The green compacts were then placed in an alumina tube furnace to fabricate nitrided bodies (RBSN) by flowing a 95% N2–5% H2 mixed gas and heating up to 1450°C. The phase identification was conducted by X-ray diffractometer (D/Max 2200, Rigaku, Japan) using Cu Kα radiation at 40 kV and 100 mA. The nitridation rate was calculated by the mass change after nitridation.11) The oxygen contents for both the Si powder only and the RBSN specimens were measured by an O/N analyzer (Model: 631-800-100, LECO, USA). The nitrided bodies (RBSN) for post-sintering were embedded in a BN crucible with a 1:1 powder mixture of high-purity silicon nitride powder (grade E10, d50 = 0.3 μm, Ube) and BN powder (GP grade, Denki Kagaku Kogyo Co., Tokyo, Japan). Post-sintering was carried out in a graphite resistance furnace at 1900°C for 6 h under static N2 pressure of 0.9 MPa. For selected specimens, shortened 3 h sintering and prolonged 12 h sintering was conducted at the same temperature.

The fracture strength was measured on bar samples (2 mm × 1.5 mm × 25 mm) in a three-point bending fixture with a span of 20 mm and a crosshead speed of 0.5 mm·min⁻¹ (Instron 2406). The surfaces of the bars were machined and then polished with diamond paste down to 1 μm. In order to observe the microstructures of the RBSN and SRBSN specimens, plasma etching (SNTEK, Korea) was performed with CF4:O2 mixed gas at a ratio of 46:4. The microstructures were observed using a scanning electron microscope (SEM; JSM-5800, JEOL, Tokyo, Japan). For field emission transmission electron microscopy (FETEM, field emission gun transmission electron microscopy, 200 kV, JEM-2100F, JEOL, Japan), the specimens were machined to a thickness sufficient to allow electron transmission by ion milling. The thermal conductivity (K) at room temperature was calculated from the measured thermal diffusivity (α) using a micro-flash machine (MicroFlash, LFA437, Germany) together with the measured density (ρ) and known heat capacity (Cp = 0.68 J·g⁻¹·K⁻¹).12)

3. Result and discussion

The data of the powder size and oxygen content of the Si powders for varied milling times are summarized in Table 1. In order to focus on the effect of the Si powders, size reduction by high-energy milling was conducted for Si powders only without mixing the sintering additives. The median diameter of the Si powder decreased from 3.97 (as-received powder, m0h) to 0.62 μm (m12h) with an increase in the milling time, while the oxygen content increased from 0.54 to 2.57 wt% accompanied by an increased specific surface area of the Si powders. As reported in the literature, the effect of a size reduction of Si powder is directly related to the increased oxygen content driven by surface oxidation.

The microstructures of the RBSN specimens are illustrated in Fig. 1, where the RD numbers in the figure denote the relative density. Based on the measured weight change after nitridation, all of the RBSN specimens exhibited nitridation rates exceeding 90%. Further, no residual Si was detected by XRD analysis on the pulverized RBSN specimens and β-Si3N4 fraction calculated by the equation of Gazzara et al. was nearly the same for all the specimens ranging 9–11% for the current experiment as illustrated in Fig. 2.13) Although the relative density levels of all specimens were similar in the range of 67–71%, the size and shape of pore was directly dependent on the size of the Si powder. Heinrich et al. reported that both the pore size and pore shape of RBSN resembles those of the starting Si powders: equiaxed smaller pores by fine Si (<10 μm) and equiaxed larger pores by coarse Si (37–63 μm). It should be

| Specimen | Milling time (h) | Median diameter of Si (μm) | Oxygen content of Si (wt%) | Oxygen content of RBSN (wt%) |
|----------|-----------------|---------------------------|---------------------------|----------------------------|
| m0h      | 0               | 3.97                      | 0.54                      | 1.59                       |
| m1h      | 1               | 1.73                      | 1.18                      | 1.96                       |
| m2h      | 2               | 1.24                      | 1.32                      | 2.04                       |
| m4h      | 4               | 1.10                      | 1.60                      | 2.20                       |
| m12h     | 12              | 0.62                      | 2.57                      | 2.75                       |

*The calculated values of RBSN are obtained using the nominal compositions and measured oxygen contents of the raw materials.
noted that the dependency of the pore shape on the starting Si powder in this research is different from that in earlier work. In the referenced work, equiaxed (or roughly spherical) pores were formed on the sites of Si powders after molten Si spread through the capillaries. On the other hand, the irregularly shaped pores in the current research (Fig. 1) originated from packing defects in the green bodies, where the size of the packing defects is proportional to that of the Si powders.

Figures 3 and 4 show SEM micrographs of SRBSN specimens under low and high magnification, respectively. The RD numbers in the figures is the measured relative density, covering a range of about 92–99%. It depicts an obvious tendency in Fig. 3 in which smaller Si powders by longer milling time resulted in SRBSN with higher density. Further, the lower density resulting from the coarser Si powders by the shorter milling time is due to the difficulties in filling of large pores in the RBSN. The sequential comparison of the microstructures in Figs. 1 and 3 reveals the relationship between the size of the Si powders and the final density of the SRBSN. That is, the pore size of RBSN is dominated by the size of the Si powders, and the difficult (easy) removal of larger (smaller) pores in the RBSN leads to lower (higher) density of the SRBSN. In order to achieve >98% density, the size of Si powder must be held below 1.1 μm (m4h). The grain morphology of SRBSN samples is well recognized by the high-magnification microstructures illustrated in Fig. 4. Elongated grains with a large aspect ratio are typical for Si₃N₄ ceramics. Compared to the similar grain size distributions for the m0h, m1h, m2h, and m4h specimens, a substantially reduced grain size with an exaggerated bimodal distribution (smaller matrix grains and abnormally grown larger grains) is observed for the m12h specimen fabricated with the finest Si powders.

Figure 5 represents the flexural strength of SRBSN specimens. Strength values for m0h, m1h, m2h, and m4h have no statistical difference, because there is almost no difference in the microstructure (grain size and shape) as described above. On the other hand, the drastically enhanced strength for the m12h specimen is rationalized by both the reduced grain size and the distinctive bimodal grains. The variation in the thermal conductivity together with the relative density is plotted in Fig. 6. Monotonically increasing thermal conductivity from m0h to m4h can be explained by the increasing density considering the similar thermal diffusivities of 32.23, 31.41, 31.51 and 31.75 mm²·s⁻¹ for the m0h–m4h specimens, respectively. In spite of the highest density for m12h, the drastic decrease in thermal conductivity was caused by the lowest thermal diffusivity, 22.85 mm²·s⁻¹. Therefore, the thermal diffusivity affected by the oxygen content of Si powder mostly dominated the thermal conductivity than the density of SRBSN did. In order to investigate the effect of sintering
time on the density and thermal properties of SRBSN, the selected RBSN specimens, m4h and m12h, which had the highest and lowest thermal conductivity was post-sintered at 1900°C for 3 and 12 h. The relative density of m4h slightly decreased to 95% by shortened sintering duration of 3 h, while the thermal diffusivity of 32.16 mm²·s⁻¹ was similar to the SRBSN of 6 h sintering resulting in the similar level of thermal conductivity. For the prolonged 12 h sintering, the vigorous grain growth was observed in both m4h and m12h, so two samples had similar microstructure as shown in Fig. 7. The thermal diffusivity of m4h and m12h specimen by 12 h sintering increased to 49.71 and 51.68 mm²·s⁻¹, respectively, which implies that the effect of the Si powder size (or oxygen content) and its characteristics on the SRBSN properties became negligible due to drastic grain growth.

It was demonstrated that thermal conductivity of Si₃N₄ ceramics strongly depends on the amount of lattice oxygen causing phonon scattering.²⁶,²⁷ Owing to the difficult pulverization of the dense SRBSN and contamination during the treatment process, the total amount of oxygen in the RBSN specimen was measured and its relationship with the starting Si powders was compared in Table 1. The calculated oxygen content of RBSN is induced under the assumption of perfect nitridation, the measured oxygen content of the Si powder, and the composition of the sintering additives. The rightmost two columns in Table 1 indicate that the oxygen content of the RBSN specimens is
nearly equal to the calculated value. Because the calculation and measurement of the oxygen content for the RBSN specimens matches well for widely varied Si powder sizes, it is safe to conclude that the total amount of oxygen content can be determined by the starting Si powders. The increased oxygen content in the specimen is postulated to be in the form of silicon dioxide, which eventually changes both the amount and composition of the liquid phase at an elevated temperature. Therefore, the acquisition of the lowest thermal diffusivity (or conductivity) in the m12h specimen is postulated as following: As the first point, the m12h specimen made from the finest Si powders with the highest total amount of oxygen resulted in the lowest thermal conductivity due to the smallest grain growth. According to Ye et al.’s report, the bulk thermal conductivity was lower for the fine-grained microstructure because of the effect of grain boundaries in spite of higher conductivity within individual grains. As the second point, the smallest grain growth for the m12h specimen gains the least benefit from the self-purification of lattice oxygen during the course of the sluggish ‘solution-precipitation’ process. As the third point, the thickness of the grain boundaries has a significant influence on the thermal conductivity in the manner that the thicker intergranular grain boundary phase (IGP) leads to the lower thermal conductivity. Figure 8 presents TEM micrographs of the SRBSN for the m4h specimen (a, b) and the m12h specimen (c, d). Except for the difference in the IGP thickness, the TEM analysis in Figs. 8(a) and 8(c) reveals nearly identical microstructural configurations consisting of crystalline grains and an amorphous triple point. In most Si₃N₄ ceramics, a glassy phase exists at the grain boundaries, and it has a thickness of about 1 nm in a thin film form. Moreover, many studies have reported that its thickness varies depending on the sintering temperature, additive composition, and impurities in the starting powder. The distinctly different microstructural evolution in the m12h specimen in contrast to the m0h–m4h specimens is related to compositional changes caused by heavy refinement of the Si size (or the highest oxygen content). It was observed that the IGP thickness of the m4h specimen was 0.9 nm, whereas it was doubled to 1.8 nm for the m12h specimen [Figs. 8(b) and 8(d)].

Lastly, we attempted an alternative interpretation in terms of oxygen content. The weight loss during sintering of silicon nitride is mainly due to the following reaction.

$$3\text{SiO}_2(s) + \text{Si}_3\text{N}_4(s) = 6\text{SiO}(g) + 2\text{N}_2(g) \quad (1)$$

$$3\text{MgO}(s) + \text{Si}_3\text{N}_4(s) = 3\text{SiO}(g) + 3\text{Mg}(g) + 2\text{N}_2(g) \quad (2)$$

According to the reference, it is reported that the reaction (2) takes place after the SiO₂ is first exhausted by the reaction (1). Therefore, the amount of SiO₂ in the Si green bodies was calculated from the initial oxygen amount of Si powder with different milling duration, and the amount of eliminated SiO₂ was estimated from the weight loss rate before and after sintering. As shown in
Table 2. Calculation of estimated remained SiO2 due to weight loss after post-sintering for 6 h at 1900°C

| Specimen | Weight loss (%) | Estimated Initial SiO2 (g) | Theoretical weight loss by SiO2 (%) | Estimated remained SiO2 (g) | Thermal conductivity (W/mK) |
|----------|-----------------|----------------------------|------------------------------------|-----------------------------|-----------------------------|
| m0h      | 0.96            | 0.00645                    | 1.21                               | 0.25                        | 0.00132                     | 70.97                      |
| m1h      | 1.71            | 0.01406                    | 2.65                               | 0.94                        | 0.00500                     | 73.01                      |
| m2h      | 2.09            | 0.01585                    | 2.96                               | 0.87                        | 0.00467                     | 72.78                      |
| m4h      | 3.13            | 0.01922                    | 3.60                               | 0.48                        | 0.00255                     | 71.04                      |
| m12h     | 4.57            | 0.03083                    | 5.81                               | 1.24                        | 0.00659                     | 59.54                      |

*Assuming that SiO2 is completely eliminated by reaction (1), the calculated value.

Table 3. Calculation of estimated remained SiO2 due to weight loss after post-sintering for 12 h at 1900°C

| Specimen | Weight loss (%) | Estimated Initial SiO2 (g) | Theoretical weight loss by SiO2 (%) | (2)Theoretical weight loss by MgO (%) | Thermal conductivity (W/mK) |
|----------|-----------------|----------------------------|------------------------------------|--------------------------------------|-----------------------------|
| m4h      | 19.17           | 0.01703                    | 3.75                               | 2.59                                 | 6.35                        | 108.12                     |
| m12h     | 22.31           | 0.02659                    | 6.00                               | 3.14                                 | 9.14                        | 113.88                     |

(1) Assuming that SiO2 is completely eliminated by reaction (1), the calculated value.
(2) Assuming that MgO is completely eliminated by reaction (2), the calculated value.

Table 2, it was confirmed that the weight loss increases as the amount of SiO2 increases. In addition, the theoretical weight loss was calculated by assuming that SiO2 was depleted by 100% through reaction (1). Calculating the amount of residual SiO2 by subtracting the actual weight loss from the theoretical value, the residual amount of SiO2 in m12h was 1.24%, which was the highest among the five samples; such large amount of residual SiO2 thickens IGP and increases the thermal resistance, hence the thermal conductivity decreases sharply. Besides, the weight loss of the specimens sintered for 12 h is shown in Table 3 together with theoretical weight loss by reaction (1). Since the actual weight loss value exceeded the theoretical weight loss by the reactions (1) and (2), there is no significant difference in the amount of remained oxygen. Therefore, it is considered that m4h and m12h exhibited similar thermal conductivity after sintering for 12 h.

Considering that the microstructural evolution of the Si3N4 system is governed by the liquid-phase sintering mechanism, the amount of in-situ added silicon dioxide for the m12h specimen is more than critical, leading to suppressed grain growth. The nearly doubled thickness of IGP hinders the material transformation, resulting in reduced grain growth with a bimodal distribution. That is, the highest strength and lowest thermal conductivity for the m12h specimen are attributed to suppressed grain growth caused by the increased diffusion distance between the grains.

4. Conclusion

In this study, the exclusive effect of the Si powder size (or oxygen content) was investigated on the properties of SRBSN by characterizing the Si powders with respect to the milling time. The size of the Si powders controls the
sinterability that it determines the pore size of RBSN. It was revealed that larger pores in RBSN specimens caused by larger Si powders were difficult to remove during the post-sintering stage. By the post-sintering at 1900°C for 6 h, the highest thermal conductivity and moderate strength was achieved for m4h specimen, while the lowest thermal conductivity and the highest strength was measured for m12h specimen. The highest oxygen content in m12h specimen resulted in the lowest thermal conductivity, since the smallest grain growth and thicker IGP caused by the compositional change in the liquid phase hinders the thermal passage. In order to meet the required thermal properties for the substrate usages by sintering at 1900°C, the size of silicon powder of D50 = 1.10 μm (1.60 wt% oxygen) is allowed for 6 h sintering, but that of D50 = 0.62 μm (2.57 wt% oxygen) needs prolonged sintering time over 6 h (12 h in the current experiment).

Acknowledgment This work was financially supported by Fundamental Research Program of Korea Institute of Materials Science (Grant no. PNK5580).

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