CORRELATION OF MICROSTRUCTURE WITH GRAIN SIZE AND HARDNESS OF SINTERED REACTION BONDED SILICON NITRIDE

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***Abstract*— Sintered reaction bonded silicon nitride ceramic have high hardness, high wear resistance, good thermal insulator, electrical insulator, nonmagnetic, oxidation resistant, prone to thermal shock, chemically stable and high dielectric properties due to these properties sintered reaction bonded. We have two types of sintered reaction bonded silicon nitride. One type has as-received Si powder (4.3µm) and other one has planetary ball milled powder (0.43). We fix three sintering temperature 1850, 1900, 1950 °C in both type. We will determine the grain size and effect of sintering on grain size of bimodal microstructure from different methods but Inspector Matrox 2.1 software give most suitable grain size. Grain size decreases on increasing sintering temperature. We use Vickers microhardness at 200g load for 15 sec., and hardness decreases on increasing grain size for SA and SP both.**

***KeyWords*- SRBSN, Bimodal micrtostructure, Grain Size, Image Analyzer, Inspector Matrox 2.1 software, Vicker Microhardness.**

1. INTRODUCTION

Silicon nitride is one of the most promising ceramic materials for use in gas turbine engines other high temperature structural applications because of its high temperature strength, thermal shock resistance, chemical stability and excellent creep resistance. Silicon nitride based materials have potentiality for many tribological applications because of their superior thermal and mechanical properties and corrosion resistance. Si3N4 materials have lower density and thermal expansion than metals, and their mechanical stability, corrosion and oxide resistance over a wide range of temperature are superior to those of several other high-strength ceramic materials [1, 2, 3, 4, and 5]. There are three kinds of silicon nitride ceramics.

In the present work, an attempt was made to correlate the properties and microstructure of SRBSN materials. Nitridation of high purity Si powders (Si as received as well planetary ball milled) was carried out at 1450oC for 2.5 h. Post sintering of nitridation samples were carried out using gas pressure sintering (GPS) at diferent temperatures (1850, 1900 and 1950oC) for 3 h. The SRBSN samples which were processed as mentioned above (provided by Korea Institute of Materials Science, South Korea) were used in the current work

Silicon nitride exists in two major crystallographic modifications, α and β, both hexagonal, with the c dimension of α approximately twice that of β. Different processing routes that are used to prepare Si_3N_4 are as follows [1].

The material is prepared by heating powdered silicon between 1300°C and 1400°C in an atmosphere of nitrogen:

$$3Si + 2N_2 \longrightarrow Si_3N_4$$

Carbothermal reduction of silicon dioxide in nitrogen atmosphere at 1400–1450°C has also been examined

$$3SiO_2 + 6C + 2N_2 \rightarrow Si_3N_4 + 6\,CO$$

The Carbothermal reduction was the earliest used method for silicon nitride production and is now considered as the most-cost-effective industrial route to high-purity silicon nitride powders.

Silicon nitride is difficult to produce as a bulk material. It cannot be heated over 1850°C, which is well below its melting point, due to dissociation to silicon and nitrogen. Therefore, application of conventional hot press sintering techniques is problematic. Bonding of silicon nitride powders can be achieved at lower temperatures through adding additional materials (sintering aids or "binders") which commonly induce a degree of liquid phase sintering. A cleaner alternative is to use spark plasma sintering where heating is conducted very rapidly (seconds) by passing pulses of electric current through the compacted powder. Dense silicon nitride compacts have been obtained by this technique at temperatures 1500–1700°C. The nitridation produces only a small volume change, which means that RBSN components do not need to be machined after fabrication and complex near net shapes can be produced in a single process stage [1, 2]. However, the nitridation results only a maximum of 70-75% densification of the samples. Hence Sintered Reaction Bonded Silicon Nitride (SRBSN) is preferred which involves nitridation and post sintering in order to improve density.

It has a Bimodal Microstructure, In bimodal microstructure, material presented a coarser microstructure with elongated grains Si_3N_4 and interlocked β-Si_3N_4 originating the so called “self reinforced” microstructure. Fig. 2.5 shows the bimodal microstructure.

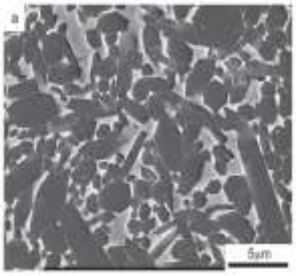

Fig. 1 Bimodal Microstructure

The mechanical properties of ceramics are a complex function of the microstructure. Almost all properties of ceramics are dependent on composition and porosity; strength and Fracture behavior apart from these depend on the grain size [13, 15]. Generally, the tensile strength is low and compressive strength is high. Hardness is important, since it is related to a number of other important properties or performance aspects of ceramics including compressive strength.

2. EXPERIMENTAL PROCEDURE

2.1 Characterization for bimodal Microstructure

Characterization of the microstructure is very important in every material for correlating the different mechanical properties like hardness, toughness, young’s modulus etc. Characterization of microstructural features plays a key-role in Materials Engineering. For characterization, different microscopes such as scanning electron microscopy, transmission electron microscopy, and atomic force microscopy can be used. In bimodal microstructure, the material presented a coarser microstructure with elongated grains Si_3N_4 and interlocked β- Si_3N_4 originating the so called ‘‘self-reinforced’’ microstructure [6].

We started with SEM microstructure of Si_3N_4–3.5%Y2O3 1.5%MgO SRBSN samples using silicon as received and planetary ball milled powder after sintering at 1850, 1900, 1950 °C respectively at 5000X SEM microstructure of Si_3N_4–3.5%Y2O3 1.5%MgO SRBSN samples using silicon as received and planetary ball milled powder after sintering at 1850, 1900, 1950 °C respectively at 5000X [2]. By using microstrucure we can measure grain size.

Grain size is one of the very important aspects of microstructure, as it affects the properties of a material. SRBSN samples have a bimodal microstructure. It has to be noted that while measuring grain size at least 1000 grains were considered (by taking number of micrographs at different locations of each sample) for the SRBSN samples. The following methods has been used for the measurement of grain size of SRBSN samples.

2.2 Image analyzer

In this method, the diameter of each grain was evaluated by the shortest grain diagonal in two-dimensional images with magnifications of 10k and/or 5k. In order to count coarse and elongated grains (major axis>8μm) which occasionally appeared in the samples sintered at higher temperatures, the micrographs with the lower magnification of 1k were also used. Aspect ratios of the normal grains were estimated from the mean value of the 10% highest observed aspect ratios [20]. Aspect ratios of the coarse grains selected from the micrographs at a Magnification of 1k were calculated by the same procedure as mentioned above.

2.3 An image analyzer Matrox Inspector 2.1

It can be also used for measurement of average grain size. In this software, grain boundary of micrograph has to be properly made by using paint. Care should be taken while making grain boundary, it should be properly join with each other. Calibration need to be done for scale. From this the maximum, minimum and mean grain diameter of grains can be measured. This software is very useful to determine size of each grain.

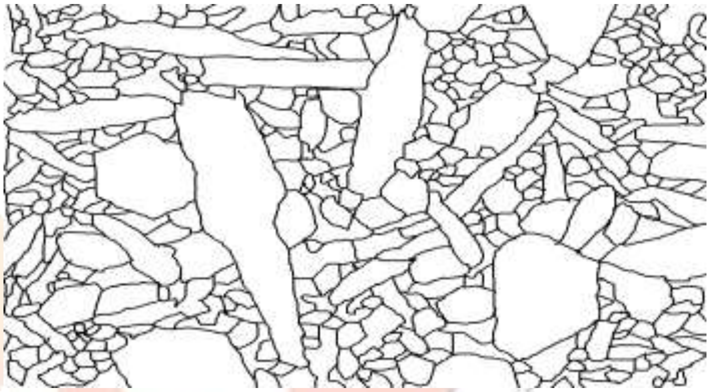

Fig. 2 Bimodal Microstructure

2.4 Hardness Test

Hardness of the samples was measured using Vickers microhardness tester (HMV-2000 SHIMAZDU). A photograph of Vickers microhardness tester is shown in **Fig. 4**. For each sample a load 200 gm and dwell time of 15 seconds was selected for Vickers microhardness measurements. To determine the average hard ness at least five readings for each sample has been taken. In this microhardness tester, initially all the parameter were set and then the length of the indentation (digonals) was selected and vickers microhardness could be read directly from the instrument.

If the elongated grains Si_3N_4 and interlocked β- Si_3N_4 increases then hardness will decrease.

Fig. 4 Micro Vickers hardness tester

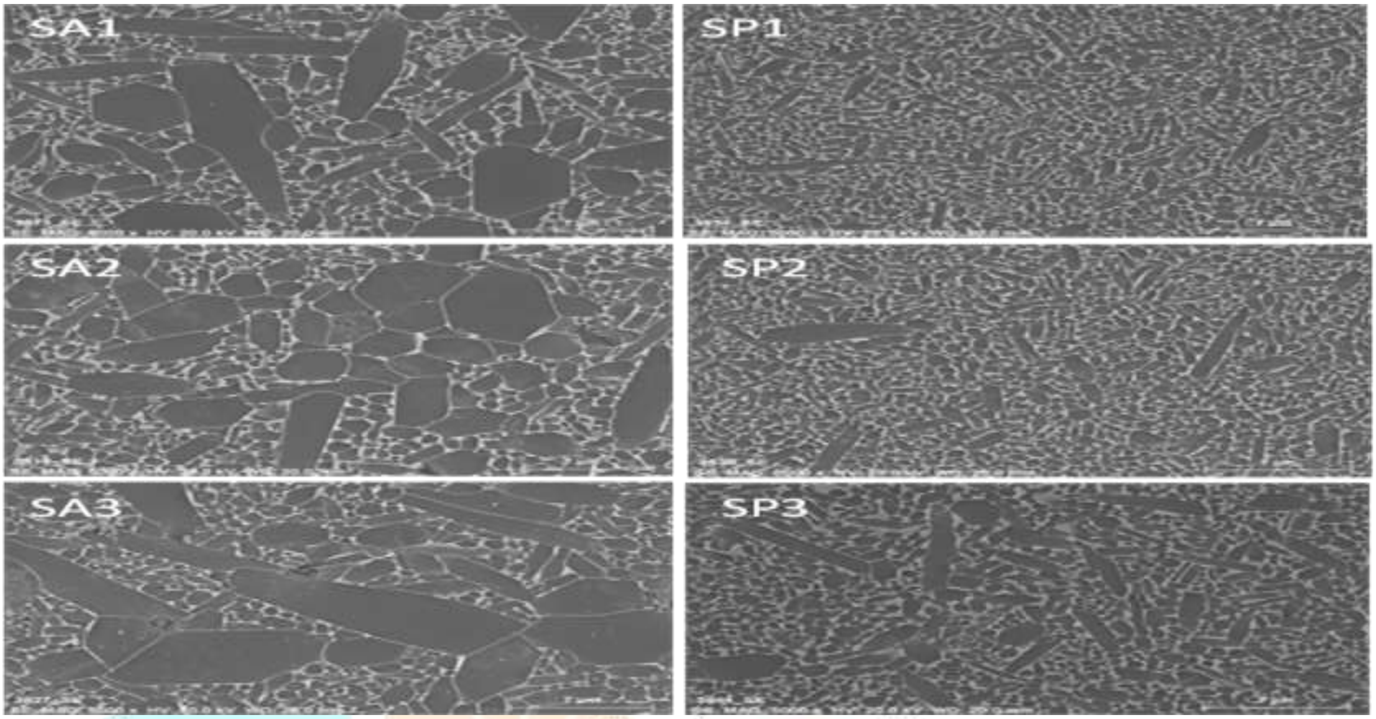

Fig. 5 SEM microstructure of Si_3N_4–3.5%Y2O3 1.5%MgO SRBSN samples using silicon as received and planetary ball milled powder after sintering at 1850, 1900, 1950 °C respectively at 5000X

3 RESULT AND DISCUSSUIONS

3.1 Grain size and aspect ratio measurements by Image analyzer procedure

Table 1 Grain size and aspect ratio by Image analyzer procedure

Sample	Avg. Grain size (µm)	Aspect ratio (normal grains)	Aspect ratio (Elongated grains)
SA1	1.33	5.82	8.25
SA2	1.55	7.37	10.47
SA3	1.62	8.01	11.41
SP1	1.23	5.00	6.47
SP2	1.16	5.44	7.47
SP3	1.03	6.10	7.66

The grain size and aspect ratio of samples estimated using Image analyzer is presented in Table 1. The average grain size of both

the SA and SP samples are almost comparable despite the variation in starting powders particle size and sintering temperature. Hence image analyzer method is also not right method for the average grain size measurement of bimodal microstructure. On the other hand, aspect ratio (AR) increased considerably for SA samples and slight increment for SP samples. AR of normal grains for SA samples varied from 5.8 to 8.0 and for SP samples it lies between 5.0 to 6.1. In case of elongated grains, the AR for SA samples measured to vary from 8.2 to 11.4 and for SP samples from 6.5 to 7.6. These result shows aspect ratio increases with sintering temperature.

3.2 Grain size measurements by Inspector2.1 Matrox software

The mean grain size, mean grain diameter for matrix grains and mean grain diameter for coarse grains measured by Inspector Matrox 2.1 software is shown in the Table **2**. The average grain size measurements by planimetric method are not giving proper grain size. In case of Image analyzer procedure, although the average grain size measurements are not accurate enough the aspect ratio of the samples could be measured reliably. As Inspector Matrox 2.1 software provides mean grain size with respect to matrix and coarse grains (by defining limit) this method gives more accurate grain size than the other methods. This gives similar grain size at different magnification. Hence it can be said that Inspector Matrox 2.1 software is better method among all for grain size measurement of bimodal microstructure

Table 2. Average grain size from Inspector 2.1 Matrox software

SAMPLE	Mean grain size (µm)	Mean grain diameter for matrix grains (µm)	Mean grain diameter for coarse grains (µm)
SA1	1.336 ± 1.0489	1.219 ± 0.685	5.906 ± 2.071
SA2	1.479 ± 1.1744	1.317 ± 0.717	6.262 ± 2.128
SA3	1.531 ± 1.298	1.331 ± 0.738	6.289 ± 2.098
SP1	0.842 ± 0.383	--	--
SP2	0.964 ± 0.530	--	--
SP3	1.020 ± 0.462	--	--

3.3 Vickers microhardness of sample

The Vickers microhardness of the SRBSN samples is presented in the following Table. 3. In order to understand the effect of microstructure on hardness of the samples, the grain size and aspect ratio of the samples is also included in the Table. 3. The hardness of SA sample varied Between 14.5 to 18. 4 GPa, however, SA1 sample exhibited the maximum hardness. Similarly the hardness of SP sample varied between 15.0 to 17.1 GPa, and the SP1 sample exhibited the maximum hardness.

Table 3: Grain size and Vickers microhardness of SRBSN sample

Sample	Mean grain size (µm)	Mean grain diameter for coarse grains (µm)	Aspect ratio (Elongated grains)	HV (GPa)
SA1	1.336	5.906 ± 2.071	8.25	18.42
SA2	1.479	6.262 ± 2.128	10.47	15.93
SA3	1.531	6.289 ± 2.098	11.41	14.49
SP1	0.842	--	6.47	17.12
SP2	0.964	--	7.47	15.59
SP3	1.020	--	7.66	15.01

Hardness measurements should have been carried out at different loads to understand the indentation behavior of samples. Due to various constraints, those aspects could not be done. The hardness of the SRBSN samples is comparable or even better than that of the reported results.

4. CONCLUSION

1) Average Grain size of sample increases with increasing sintering temperature. On increasing sintering temperature more elongated and interlocked β grains formed and due to this Average Grain size of sample increases.
2) Inspector Matrox 2.1 software gives the more accurate Average Grain size for bimodal microstructure.
3) For both SA and SP samples, microhardness decreases with increasing sintering temperature and grain size.

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