Journal of Ceramic Processing Research. Vol. 10, No. 3, pp. 290~295 (2009)

J O U R N A L O F
Ceramic Processing Research

Ultra precision polishing of Si_3N_4 ceramics using magnetorheological fluids and diamond abrasives

Dong-Woo Kim[a], Jung-Won Lee[a], Myeong-Woo Cho[a], Young-Jae Shin[b], Jingwen Xu[c], Ki-Ju Lee[c], Seung-Yong Shin[d] and Won-Seung Cho[c,*]

[a]*Department of Mechanical Engineering, Inha University, Incheon 402-751 Korea*
[b]*Nano-Mechanical System Technology Division, KIMM, Daejeon 305-343 Korea*
[c]*School of Materials Science and Engineering, Inha University, Incheon 402-751 Korea*
[d]*Advanced Functional Material Team, KITECH, Incheon 406-840 Korea*

Si_3N_4 ceramics were prepared by hot-pressing at 1800 °C for 2 h. Firstly, the microstructure and mechanical properties of Si_3N_4 ceramics with sintering aids of 8 mol% Y_2O_3 + 6 mol% Al_2O_3 were investigated. The Si_3N_4 ceramics showed excellent mechanical properties. Grain bridging and pullout were observed during the Vickers indentation crack propagation, implying that they are possible toughening mechanisms for Si_3N_4 ceramics. The secondary phase was identified as $Y_{10}Al_2Si_3O_{18}N_4$ (YAlSiON), which was formed by the reaction with the sintering aids, Si_3N_4, and SiO_2. Secondly, ultra precision polishing experiments for Si_3N_4 ceramics were performed using magnetorheological fluids (MR fluids) and diamond slurries. A series of experiments were performed under various polishing conditions by changing the current intensity and spindle speed for a given time period of 10 minutes. The machined surfaces were observed using a SEM and surface profiler to investigate the surface integrity changes. As a result, a very fine surface roughness of Ra = 1.012 nm was obtained within 10 minutes when the electric current was 2 A and the wheel speed was 300 rpm. Also, it was observed that the MR polishing method can provide excellent surface roughness compared with the existing lapping methods.

Key words: Si_3N_4, Ultra precision polishing, Magnetorheological fluid, Diamond slurry, Surface roughness.

Introduction

Silicon nitride (Si_3N_4) ceramics have features such as low density, high strength, wear resistance, etc.. Moreover, Si_3N_4 has been given much attention as a new structural material because it has excellent fracture strength, fracture toughness, and thermal shock resistance compared with other fine ceramics. Due to their superior mechanical and thermal properties, Si_3N_4 ceramics have been increasingly applied in high temperature and high-stress situations, such as in automotive engine components, wear resistant parts, cutting tools, advanced ceramic bearings, etc..

Although Si_3N_4 has excellent properties when it is applied as a structural ceramic, it is a very difficult-to-grind ceramic with a high efficiency resulting from its high hardness [1, 2]. Furthermore, Si_3N_4 ceramics are sensitive to defects resulting from the grinding and polishing processes due to their inherent brittleness. Failure begins in regions of surface irregularities, such as scratches, pits, and microcracks. Hence, it is important to fabricate Si_3N_4 ceramics with a superior quality and finish with minimum defects in order to obtain reliability in performance.

*Corresponding author:
Tel : +82-32-860-7528
Fax: +82-32-862-5546
E-mail: wscho@inha.ac.kr

In industry, ceramics are finished using conventional grinding followed by diamond polishing or lapping. Conventional polishing uses high loads, and diamond abrasives. The use of diamond abrasives under heavy loads can result in polishing pad wear, scratches, pits, and microcracks on the surface and subsurface of the polished ceramics. These surface defects can act as nucleation sites for cracks resulting in catastrophic failure. Thus, highly accurate mirror finishing with little mechanical damage is required when using precision components.

Recently, to solve the problem of polishing pressure, pad wear, subsurface damage, and microcracks during the polishing process, polishing technologies such as electrorheological fluids [3] or magnetorheological (MR) fluids [4] have been proposed. MR fluids are known as controllable smart materials since their flow properties, such as viscosity and stiffness, can be easily changed and controlled using externally-imposed magnetic fields.

In the present study, Si_3N_4 ceramics were prepared by hot-pressing at 1800 °C for 2 h. Firstly, the microstructure and mechanical properties of the Si_3N_4 ceramics with sintering aids of 8 mol% Y_2O_3 + 6 mol% Al_2O_3 were investigated. Secondly, ultra precision polishing experiments for the Si_3N_4 ceramics were performed using magneto-rheological fluids and diamond slurries. A series of experiments were performed under various polishing conditions by changing the current intensity and spindle speed.

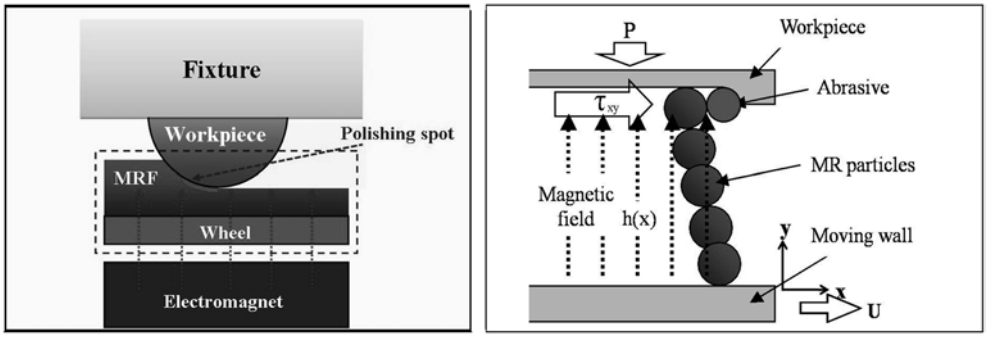

Fig. 1. Imposed magnetic field and shear direction in MR polishing.

Experimental

Preparation of Si_3N_4 monolith ceramics

The starting powders were Si_3N_4 (α crystal phase > 95%; average particle size: 0.17 μm; Ube Industries, Ltd., Japan), Y_2O_3 (> 99.9% purity; < 0.5 μm; Shinestu Chemical Co. Ltd., Japan), and Al_2O_3 (> 99.99% purity; 0.39 μm; Sumitomo Chemical Co. Ltd., Japan). In order to fabricate the Si_3N_4 ceramics, 8 mol% of Y_2O_3 and 6 mol% Al_2O_3 were added to the Si_3N_4 as sintering aids. The powders were mixed for 72 h by wet ball milling using an Al_2O_3 jar, Si_3N_4 balls, and ethanol. The mixed wet powders were dried on a hot plate using a rotating stirrer to avoid gravity-induced segregation. To obtain fully dense compacts, the dried and sieved (16 mesh) powders were hot-pressed at 1800 °C for 2 h under a pressure of 30 MPa in N_2 atmosphere. Rectangular specimens (3 × 4 × 36 mm) were prepared by cutting, grinding, and polishing the hot-pressed compact.

The sintered density was measured using the Archimedes' method, and the theoretical density of the specimens was calculated according to the rule of mixtures. Flexural strength tests were performed at room temperature on five specimens for each condition using a three-point method with a span of 30 mm. The specimens were loaded at a constant crosshead speed of 0.5 mm·minute^{-1}. The fracture toughness was estimated by measuring the crack lengths generated by a Vickers indenter with a load of 98 N [5].

The fracture surfaces and polished/etched surfaces of the specimens were investigated using a scanning electron microscope (SEM). X-ray diffraction using Cu Kα radiation was also performed for the specimens. The Si_3N_4 grain size was measured from the SEM images of the surfaces etched for 1 h using a mixed solution of HF + HNO_3 at 80 °C. The microstructure was investigated via a TEM equipped with an EDX.

Polishing principle using MR fluids

MR fluids are phase-controllable suspensions comprised of a mixture of micro-sized magnetic particles and non-magnetic fluids such as mineral oils or water. MR fluids are known as smart materials since their flow properties can be easily changed by the intensity of the imposed magnetic field. The MR fluid can form chain-like structures via magnetization of the particles in the fluid; this phenomenon is called the "MR effect" and it can reversibly transform the fluid from a fluid-like state to a solid-like state within milliseconds [3, 4]. As a result of this transformation, the flow properties of the fluid (such as viscosity and stiffness) increase due to the increased resistance to the shear stress according to the imposed magnetic field strength. Owing to such characteristics, MR fluids can be applied to dampers, clutches, and polishing systems [3, 4].

A schematic diagram of the MR polishing mechanism is shown in Fig. 1. As shown in the figure, the MR fluid is supplied to the gap between a workpiece and a moving wall to polish the workpiece. When a proper magnetic field is applied to the MR fluid, the viscosity and stiffness of the fluid increase by more than several tens of times within milliseconds. Thus, the MR fluid can rotate continuously as long as it adheres to the wheel surface resulting from the applied magnetic field [6-8]. For polishing purposes, a suitable abrasive slurry (a mixture of DI water and abrasive particles, which are generally of non-magnetic materials) is incorporated into the fluid, which is supplied to the narrow gap between the wheel and the workpiece.

MR polishing conditions

To prepare the required fluids for MR polishing, carbonyl iron (CI) powder, which is sensitive to magnetic fields, was used. The fluids consist of approximately 50 wt% magnetic particles (CI powders, 2 μm), abrasive diamond particles (0.1-2000 μm), and DI water.

A dispersion stabilizer (glycerin) was added to the aforementioned materials as it enhances the cohesion of the magnetic fluids and facilitates proper mixing of the polishing slurry and magnetic particles. However, excessive use of a stabilizer may deteriorate the finishing quality for certain materials [4]. The compositions of the MR fluid and slurry used for the experiments are listed in Tables 1 and 2.

The experiments were performed by changing the rotating speed of the polishing wheel and the strength of the applied magnetic fields. The applied experimental conditions are listed in Table 3.

Table 1. Composition of the MR fluids

CI powder	DI water	Na_2CO_3	Glycerin
50 wt%	48 wt%	1 wt%	1 wt%

Table 2. Composition of the Diamond slurry

Diamond powder	DI water
9 wt%	91 wt%

Table 3. Experimental conditions for MR polishing

Parameter	No.	Wheel Speed (rpm)	Electric current (A)	Magnetic field (A/m)	Polishing timec (minutes)	Gap size (mm)
Rotating speed of the polishing wheel	1	50	2.0	5,500	10	0.5
	2	100				
	3	150				
	4	200				
	5	250				
	6	300				
Magnetic field	7	150	0.5	3,800	10	0.5
	8		1.0	4,700		
	9		1.5	5,200		
	10		2.0	5,500		
	11		2.5	5,700		
	12		3.0	6,100		

Table 4. Relative density and mechanical properties of Si_3N_4-8 mol% Y_2O_3-6 mol% Al_2O_3

Specimen	Relative density (g/cm^3)	Flexural strength (MPa)	Hardness (kg/mm^2)	Fracture toughness ($MPa \cdot m^{1/2}$)
Si_3N_4-8mol% Y_2O_3-6mol% Al_2O_3	99.7 ± 0.1	1,050 ± 80	1,610 ± 50	7.0 ± 0.5

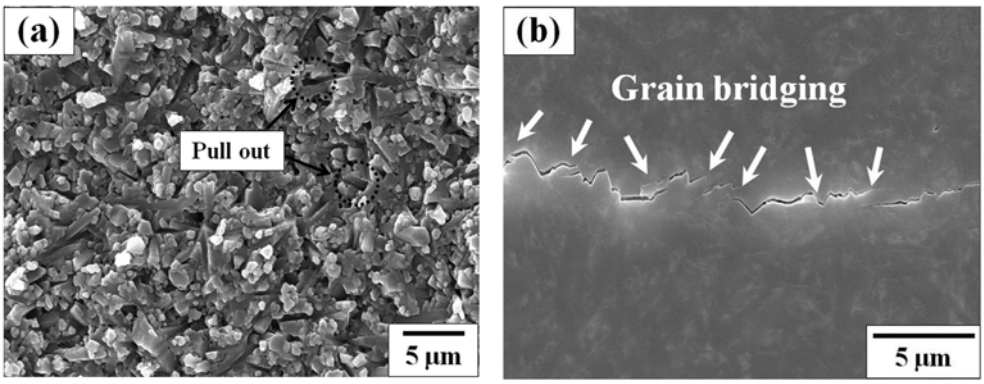

Fig. 2. SEM micrographs of (a) the fracture surface and (b) the crack path [9].

Results and Discussion

Microstructures and mechanical properties of Si_3N_4 ceramics

Table 4 shows the relative density and mechanical properties of Si_3N_4-8 mol% Y_2O_3-6 mol% Al_2O_3 ceramics. The relative density was 99.7%, i.e. the specimen was fully densified. The flexural strength and fracture toughness of the Si_3N_4 showed excellent values of 1050 MPa, and 7.0 $MPam^{1/2}$, respectively.

Fig. 2 shows the fractured surfaces, broken parallel to the hot-pressed direction, of the Si_3N_4 + 8 mol% Y_2O_3 + 6 mol% Al_2O_3 specimen. In the specimen, evidence for pullouts of the Si_3N_4 grains was observed. Based on our previous work [9], the crack paths, obtained by indentation, are shown in Fig. 2(b) for the same specimen. The well-polished surface was indented along the vertical direction toward the hot-pressed direction. The crack paths in the specimen were sinusoidal due to the grain bridging during the crack propagation. It is well-known that grain bridging and pullout resulted in a crack resistance behavior (R-curve behavior) as the

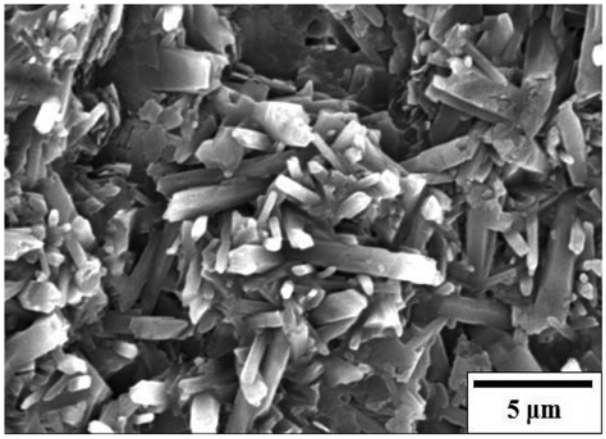

Fig. 3. SEM micrograph of the etched surface for the Si_3N_4.

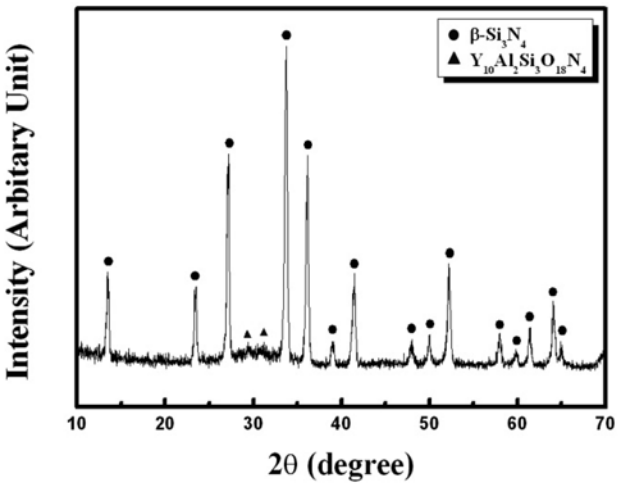

Fig. 4. XRD pattern for the Si_3N_4.

fracture toughness increased with the crack growth. Based on these microstructural observations, the grain bridging and pullout are possible toughening mechanisms for Si_3N_4 ceramics [9].

Fig. 3 shows a SEM micrograph of the etched surface for the Si_3N_4. The microstructure revealed needle-like fine β-Si_3N_4 grains. The average grain diameter was approximately 0.47 μm.

Fig. 4 shows the XRD pattern for the Si_3N_4. From the XRD analysis for the Si_3N_4 ceramic, the phase transition of α- to b-Si_3N_4 was completed. The secondary phase was identified as $Y_{10}Al_2Si_3O_{18}N_4$ (YAlSiON), which was apparently formed by the reaction with the sintering aids (Y_2O_3 and Al_2O_3), Si_3N_4, and with the SiO_2 that existed on the surface of the Si_3N_4 powders.

Fig. 5 shows a TEM micrograph at a relatively low magnification. According to TEM images and EDX analyses for the Si_3N_4, the yttrium-containing intergranular phases were usually located at the triple points of the Si_3N_4 grains; and at the Si_3N_4/Si_3N_4 interfaces, an intergranular phase was generally absent. Also, the

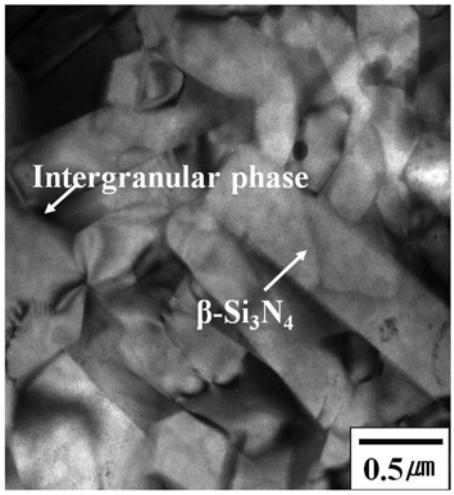

Fig. 5. TEM image for the Si_3N_4.

grain boundary regions between Si_3N_4 and the crystalline intergranular phase revealed an amorphous film of approximately 1-2 nm thickness.

Analysis results of the MR polishing process

In general, MR polishing is applied in the final finishing stage to obtain high quality surface roughness and precision due to its low material removal rate (MRR). Thus, the polishing results using MR fluids are highly dependent on the preceding machining results. In this study, lapping and conventional polishing were applied to prepare the specimens for MR polishing, and the results were compared. Fig. 6 shows the measured results using a non-contact 3D surface profiler after each method was applied. A surface roughness of Ra = 0.119 μm was obtained after the lapping process as shown in Fig. 6(a). However, several scratches were observed on the surface. In the next step, conventional sand paper polishing was used; then, a surface roughness of Ra = 54.265 nm was obtained. Even though the surface roughness improved, it was found that scratches remained on the surface of the ceramic specimens. Finally, the MR polishing method was applied to the specimen, and the results were measured as shown in Fig. 6(c). As shown in the figure, a very fine surface roughness of Ra = 1.012 nm was obtained using the MR polishing method; no serious scratches could be found on the surface of the ceramic specimens.

Fig. 7 shows the surface roughness variations according to the polishing process parameter changes. Fig. 7(a) shows the changes of the measured surface roughness with the rotational speed of the polishing wheel. The detailed experimental conditions (No. 1-6) are listed in Table 3. As can be seen from the figure, the surface roughness tends to improve with an increasing wheel speed. This is because more abrasive particles can participate in the polishing with an increasing relative velocity between the workpiece and the moving wall

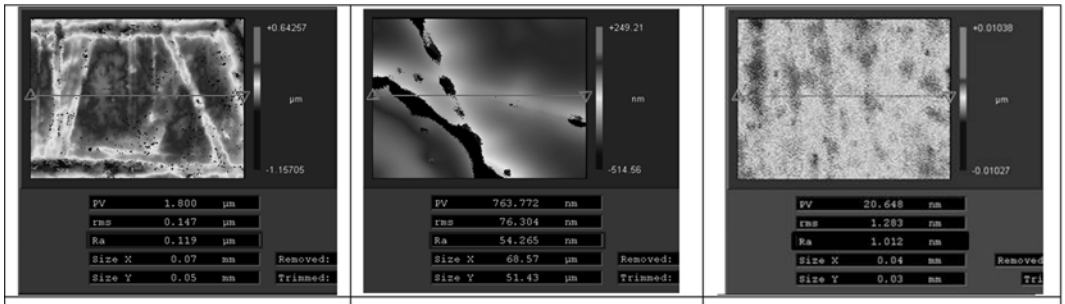

Fig. 6. Measured surface roughnesses using a 3D profiler.

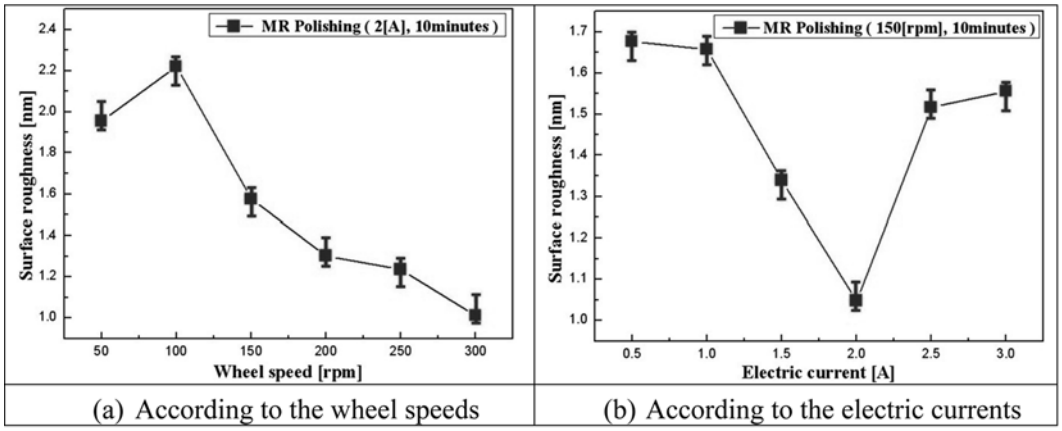

Fig. 7. Surface roughness variations according to the process parameters.

(in this study, the rotating wheel). However, continuous increases of the polishing speed do not guarantee an improvement of the surface roughness. It was observed that the improvement rate slowed as the wheel speed exceeded certain values, which varied according to the imposed process conditions. This is because a wheel rotation speed that is too fast can cause insufficient slurry flow at the polishing region.

Fig. 7(b) shows the changes of the measured surface roughness according to the electric current values. These experimental conditions correspond to No. 7-12 in Table 3. As can be seen from the figure, the best results were obtained when the applied electric current was 2 A. Since an electromagnet was used to generate the magnetic fields required for MR polishing in this study, an increase in the electric current means an increase in the applied magnetic intensity.

Also, the viscosity and stiffness of the MR fluids increase with an increasing magnetic field intensity as previously explained. Thus, Fig. 7(b) can be regarded as due to the stiffness changes of the MR fluids used. The surface roughness tends to improve as the electric currents increase, since a stiffer polishing medium (MR fluids + diamond abrasives) can remove more material from the surface within a given time frame (10 minutes). However, polishing media that are too stiff can result in scratches and/or deterioration on the surface. Thus, if the applied electric current exceeds certain values, the surface roughness becomes worse, as shown in the figure. Fig. 7 shows the results of two different sets of experiments, and it can be observed that different results were obtained with the same experimental conditions (150 rpm, 2A and 10 minutes in (a) and (b)). Since the MR polishing results are highly dependent on the preceding polishing results, different results are generally obtained for the same polishing time (10 minutes in this research).

Finally, after repetitive experiments, very fine polishing results of Ra = 1.012 nm were obtained when the electric current was 2 A and the wheel speed was 300 rpm; the results are shown in Fig. 8.

Conclusions

The microstructure, mechanical properties, and MR fluid polishing of Si_3N_4 ceramics was investigated. The results are summarized as follows.

1. Si_3N_4 ceramics showed excellent fracture strength and fracture toughness. Grain bridging and pullout were observed during the crack propagation, implying that these phenomena are possible toughening mechanisms for Si_3N_4 ceramics. The secondary phase was identified as $Y_{10}Al_2Si_3O_{18}N_4$ (YAlSiON), which was formed by the

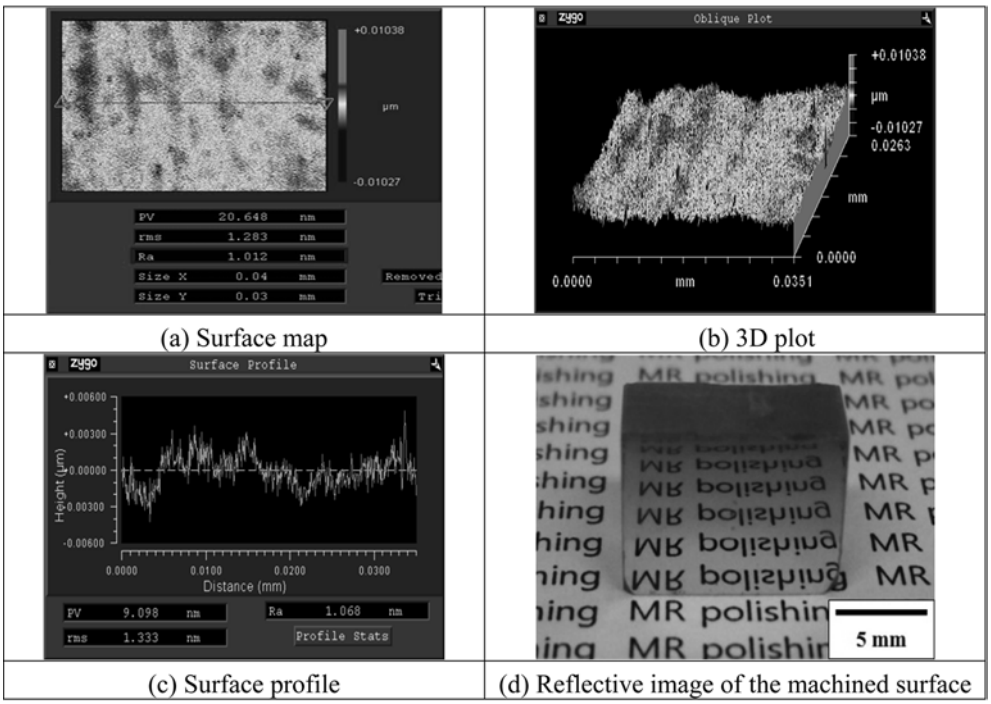

Fig. 8. MR polishing results of the Si_3N_4 ceramics.

reaction with the sintering aids, Si_3N_4, and SiO_2. Yttrium-containing intergranular phases were usually located at the triple points of the Si_3N_4 grains. The grain boundary regions revealed an amorphous film of approximately 1-2 nm thickness.

2. The purpose of this study was to obtain an ultra precision surface for Si_3N_4 ceramics using MR fluids and diamond abrasives. Lapping and conventional sand paper polishing were performed as preceding processes; then, the MR polishing method, a newly-developed polishing technique, was applied to the prepared specimens. A series of experiments were performed, and the results were measured and observed. The process parameters including wheel rotational speed and electric current were changed. Finally, a very fine surface roughness of Ra = 1.012 nm was obtained within 10 minutes when the electric current was 2 A and the wheel speed was 300 rpm. From the results of this study, it can be seen that the MR polishing method applied can be a suitable polishing technique to obtain ultra precision surface qualities of ceramics such as Si_3N_4.

Acknowledgements

This work has been supported by Ministry of Knowledge Economy in Korea through Strategic Technology (Development Project Development of Micro Functional Precision Components Manufacturing Technology).

References

1. H. Kawamurs, Key Eng. Mater. 89 (1994) 713-718.
2. R. Raj, J. Am. Ceram. Soc. 76 (1993) 2147-2174.
3. Y. Akami, K. Asari, B. Jeyadevan, and T. Fujita, J. Int. Mat. Sys. Struc. 9 (1998) 672-675.
4. W. Kordonski and D. Golini, Int. J. Mod. Phys. B 13[14-16] (1999) 2205-2212.
5. K. Niihara, Bull. Ceram. Soc. Jpn. 20 (1988) 12-18.
6. K. Kim, W. Sim, D. Jeon, and B. Choi, J. KSPE 17[8] (2000) 41-45.
7. C. Kormann, H.M. Laun, and H.J. Richer, Int. J. Mod. Phys. B 10 (1996) 3167-3172.
8. D. W. Kim, M. W. Cho, T. I. Seo, and Y. J. Shin, Sensors 8 (2008) 222-235.
9. Y.S. Yoon, S.W. Na, J. Lee, M.W. Cho, E.S. Lee, and W.S. Cho, J. Am. Ceram. Soc. 87[7] (2004) 1374-1377.