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Effects of the Addition of La_2O_3 on Mechanical Properties and Machinability of Si_3N_4 Ball

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Silicon nitride with adding La_2O_3 was sintered by gas pressure sintering (GPS) technique at 1950°C, in N_2 gas at 3 MPa, for 2 h. Mechanical properties such as hardness, flexural strength, and fracture toughness were determined as a function of the GPS holding time and the contents of La_2O_3 in silicon nitride. Also machinability of silicon nitride ball with various GPS holding time and amount of La_2O_3 was evaluated by magnetic fluid grinding (MFG) method. In this study it was found that machinability was influenced significantly with La_2O_3 contents. However, the different GPS holding time did not affect the machinability very much.

Key words: Silicon nitride, Lanthanum oxide, Mechanical property, Machinability, Gas Pressure Sintering (GPS)

I. Introduction

Silicon nitride ceramic materials offer many interesting properties for high speed-high temperature bearing applications. They include low specific gravity (about 40% of a conventional bearing steel) resulting in a reduced centrifugal load at high rotational speeds, high Young's modulus (about 50% more than a conventional bearing steel), non-magnetic property and low friction and high wear properties. Taking the advantages of silicon nitride, we can produce the silicon nitride ceramic ball for bearing ball.

Many studies[1-6] have been performed to improve sinterability, mechanical properties and mechanical machinability of silicon nitride ceramic ball, by adding Al_2O_3 or Y_2O_3 as additives. Y_2O_3 is well-known sintering additive for silicon nitride.[7-9] The compound of Y_2O_3 and Al_2O_3 can also improve the sinterability of silicon nitride, but thermal properties are unchanged. Many studies[7-9] have been made on the addition of Al_2O_3-Y_2O_3 to improve the densification by the liquid-phase sintering process. A serious problem associated with those additives is that the viscosity of the residual glassy phase decreases at elevated temperatures. This in turn decreases the high temperature strength of silicon nitride.[7] Utilization of other metal oxides to densify Si_3N_4 either alone or in combination, has also been attempted with success.[10] For example, a combination of Y_2O_3 and La_2O_3 was effective sintering aid for Si_3N_4.[11] The refractory $YLaO_3$, or other phases such as La_2O_3, $La_5(SiO_4)_3N_4$, and $La_2Si_6O_3N_8$ form at the grain boundaries of the sintered silicon nitride, improving the high temperature mechanical properties. Das[4] reported that three eutectic compositions of Y_2O_3, La_2O_3, and SiO_2 governs densification of Si_3N_4. In the case of using La_2O_3 as additives, in order to improve the mechanical properties by increasing the refractory nature of the residual grain-boundary phase, a higher sintering temperature was required to reach near the theoretical density. The sintering methods were gas pressure sintering (GPS), hot pressing (HP) and hot isostatic pressing (HIP) technique.

In this study, GPS was performed at 1950°C, in N_2 gas at 3 MPa, for 2 hours so that high pressure of nitrogen gas suppressed the thermal decomposition of silicon nitride. The effect of mechanical properties such as hardness, toughness and flexural strength, microstructure, and sinterability on machinability was studied in Si_3N_4 with 5 mol% of La_2O_3.

Unfortunately, silicon nitride, like the other advanced ceramics, is hard and brittle. Hence, it is rather difficult to polish by conventional methods such as grinding, lapping, polishing etc. which requires considerable machining time from several days to a few weeks to finish. In addition, expensive tools such as diamond or c-BN grinding wheels are inevitably used. Therefore, the cost of machining reaches about 30-60% of the total production cost and in some cases it reaches above 90%. In addition, the machinability of ceramics depends upon not only processing route and machining tools but also materials properties. In order to overcome some of these problems, Kato and Umehara[6, 12-14] have developed a new technique known as magnetic fluid grinding (also referred to magnetic float polishing). Magnetic fluid grinding utilizes the magnetic buoyant force of non-magnetic body in magnetic fluid effectively. Specially, the float plays an important role on the material removal rate in particular. In the early 1970s, gas pressure sintered silicon nitride was used as bearing materials of gas turbine.[15]

In this study, thereby, the optimum microstructure for silicon nitride balls exhibiting the excellent machinability was investigated by magnetic-fluid grinding (MFG) technique and an attempt was made to figure out the influence of mechanical properties on the materials removal rate of silicon nitride ball.

II. Experimental Procedure

2.1. Sample Preparation and Mechanical Test

Starting material used was a-silicon nitride powder (E-10 grade, UBE Co., Tokyo, Japan) with an average size of 0.2 μm and higher α phase-fraction of silicon nitride. For the additives, Al_2O_3 powder from HP-DNB grade of Leynold Philadelphia, U.S.A., Y_2O_3 powder from Hermann C. Stark (Berlin, Germany) and high-purity (99.9%) La_2O_3 from High-purity Chemistry Institute Co. (Osaka, Japan) were used.

The powders were mixed in a polyethylene bottle using high purity silicon nitride balls and milled in ethanol for 24 h. The milled slurry was dried in the rotary evaporator because of big difference in specific gravities powders. The dried mixtures were passed through a 325 mesh screen, which pulverized the aggregates. Balls and plates were dry pressed under 20 MPa, prior to cold isostatic pressing (CIP) under 245 MPa. The green body was sintered at 1950°C, in N_2 gas at 3 MPa, for 2 hour. The content of La_2O_3 varied to 1, 2, 3 and 5 mol%. GPS holding time varied to 1, 2, 3 and 5 hour with 5 mol% of La_2O_3. Fig. 1 illustrates the experimental procedures for this study.

Bulk density of the GPSed specimens was measured by Archimedes method using water and the relative densities were calculated according to rule of mixture of powders. The surface of polished specimen was indented and etched by plasma etching method, and observed in SEM. The hardness and toughness were measured by indentation method. Hardness was measured under load of 196 N for 15 seconds. Hardness data were averaged using more than 10 values. The fracture toughness was calculated using the equation suggested by Evans and Charles.[16]

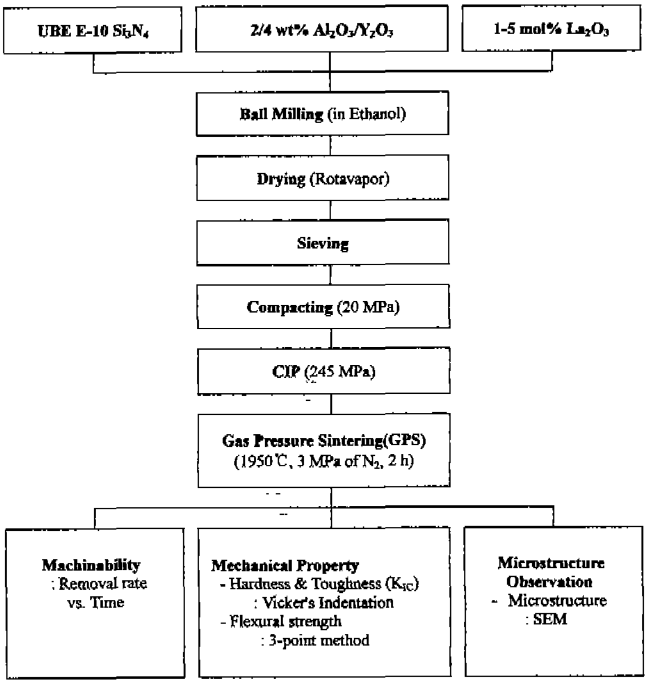

Fig. 1 Flow chart of Si_3N_4 experimental process.

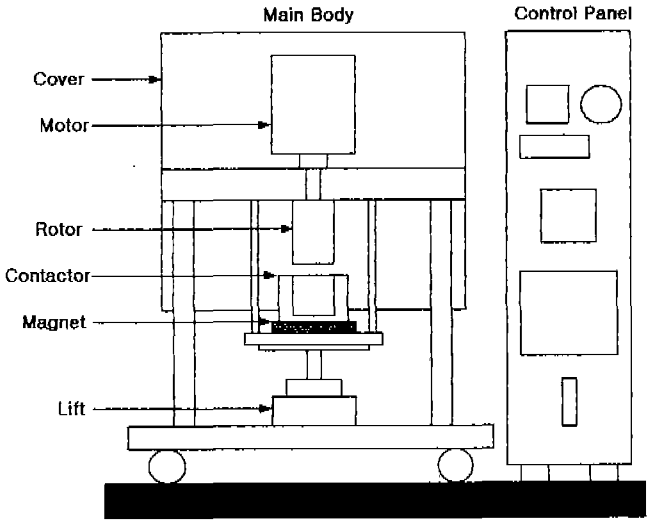

Fig. 2 Schematic diagram of magnetic fluid ball contact grinding apparatus.

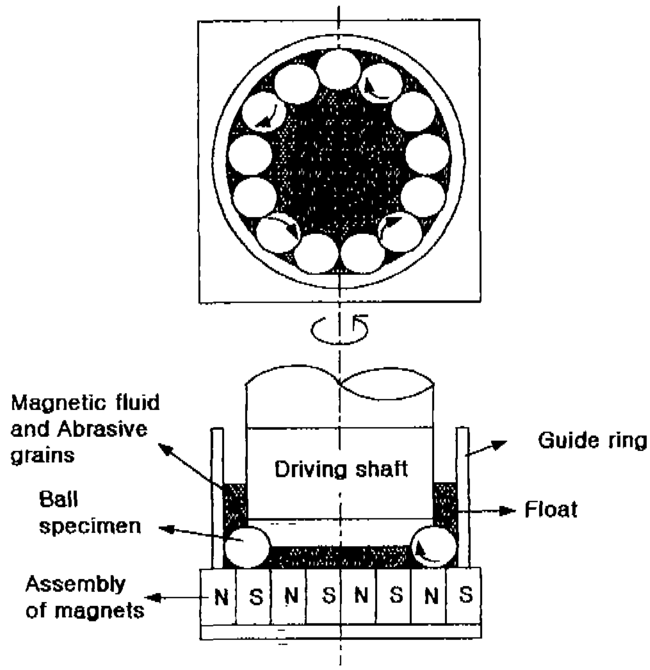

Fig. 3 Schematic diagram of the area for ball grinding.

For flexural strength test, test pieces of 3 mm×4 mm×30 in dimension were cut from samples and polished up to 0.3 μm using alumina paste. The edges of the tensile side of the flexural strength bars were chamfered at 45° using diamond wheel. All data points were the average values of at least 10 specimens fractured under crosshead speed of 0.5 μm/min and spans of 20 mm.

2.2. Measurement of Machinability

To evaluate machinability of silicon nitride balls, the sintered silicon nitride balls were ground by magnetic fluid grinding (MFG) method under grinding speed of 10,000 rpm, 20 μm SiC powder in 40 vol% magnetic fluid, for 6 hours. Following are the procedure of MFG process for silicon nitride ball. When diamagnetic abrasive powder was mixed with magnetic fluid under magnetic field, a workpiece in abrasive powder was freely rotated inside the holder. If magnetic fluid was on the top of magnetic field, the buoyancy occurred. In this study, the balls were machined in abrasive powder. In order to perform MFG, ceramic machining was consisted of rotor, holder, and magnet fluid. Fig. 2 shows a schematic diagram of MFG and the contact area for ball grinding is shown in Fig. 3.

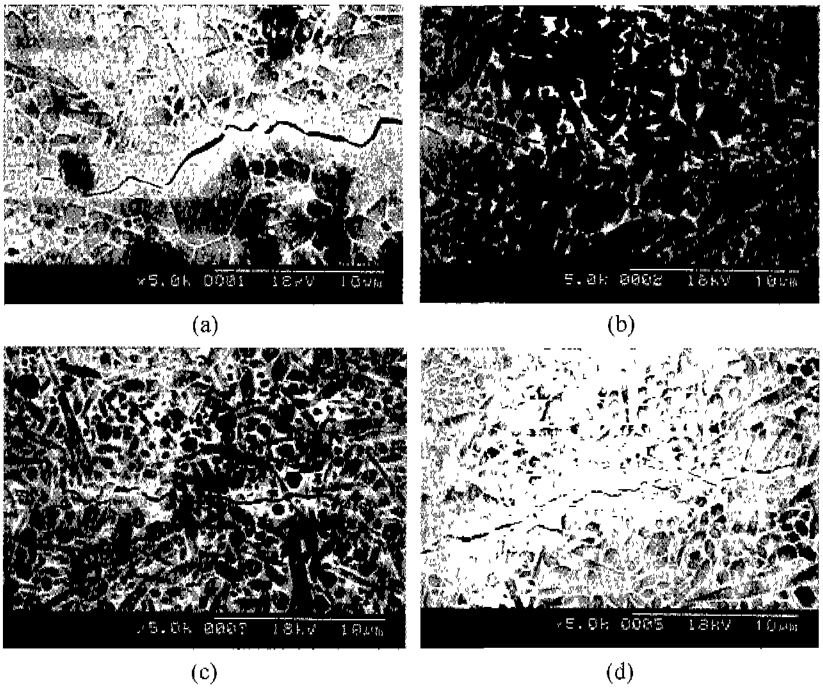

Fig. 4 SEM micrographs of the etched Si_3N_4 with various content of La_2O_3; 1 mol%, (b) 2 mol%, (c) 3 mol% and (d) 5 mol%.

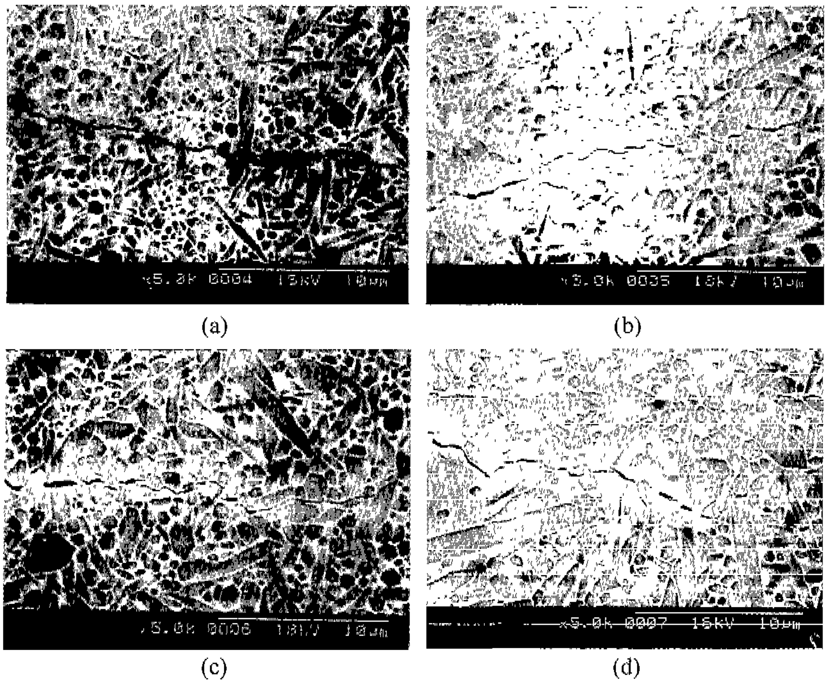

Fig. 5 SEM micrographs of the etched Si_3N_4 with various GPS holding time for 5 mol% La_2O_3 in Si_3N_4; (a) 1 hour, (b) 2 hour, (c) 3 hour and (d) 5 hour.

III. Results and Discussion

Figs 4 and 5 show the microstructure of Si_3N_4 with 5 mol% of La_2O_3 as a function of La_2O_3 content and GPS holding time of the specimen. For 1 mol% La_2O_3 in Si_3N_4-2 wt% Al_2O_3-4 wt% Y_2O_3, the grain size increased because of increasing grain growth caused by Y_2O_3.[7] For over 2 mol% of La_2O_3, the grain size was reduced from 2 to 0.85 due to the La_2O_3 rather than Y_2O_3. La_2O_3 could suppress the grain growth since the grain-boundary phases remained. The elongated grains were observed in 2 or 3 mol% La_2O_3 as shown in Fig. 4. For the two compositions, we could see the interlocking among β-phases of Si_3N_4. In addition, the aspect ratios were about from 3 to 8 in all of the cases. The grain size increased with increasing GPS holding time as shown in Fig. 5. Concurrently, the abnormal grain growth also occurred up to 5 hours.

Fig. 6 (a) and (b) show the variation of relative densities as a function of La_2O_3 content and GPS holding time, respectively. With addition of 3 mol% La_2O_3 in Si_3N_4, relative density was close to 99.9%. The 2 hours holding time could be optimum as explained in the previous work.[5] The mechanical properties such as hardness, toughness and flexural strength were shown in Fig. 7. Hardness increased with increasing the amount of La_2O_3, but toughness was decreased because of decreasing grain size, as mentioned before (Fig. 7(a)). Flexural strength was 560 MPa, for 3 mol% of La_2O_3 in Si_3N_4. When increasing the GPS holding time, hardness did not change much for a short holding time such as up to 3 hours, but decreased sharply after 3 hours. In addition, toughness did not changed significantly, but flexural strength increased (Fig. 6 (b)). The crack was more difficult to propagate along the grains with increasing the GPS holding time. Consequently, mechanical properties were optimized at 3 mol% of La_2O_3 and for 2 hours of the

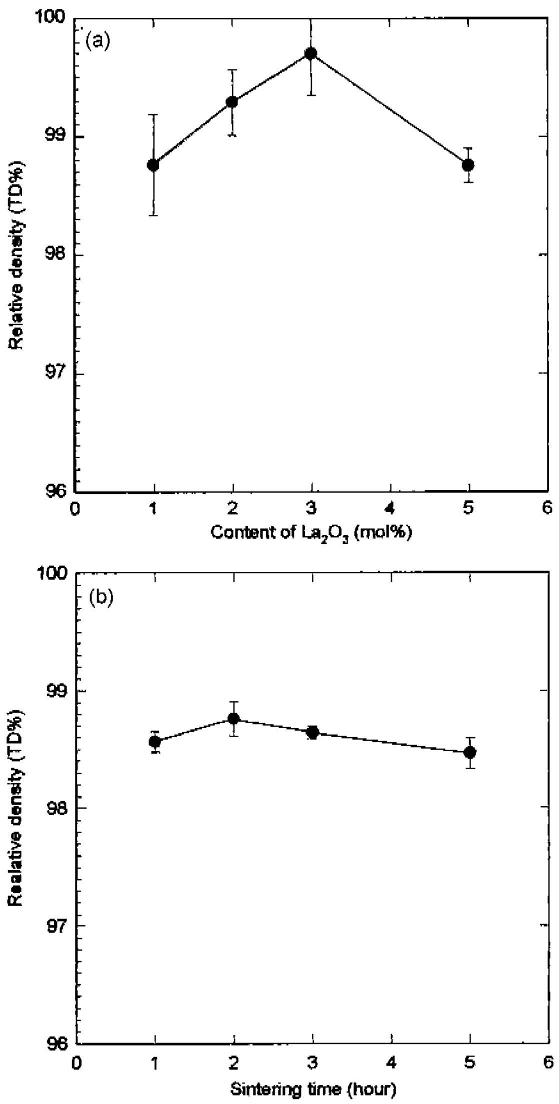

Fig. 6 Variation of relative densities of Si_3N_4 as a function of (a) the content of La_2O_3 and (b) the GPS holding time for Si_3N_4-5 mol% La_2O_3.

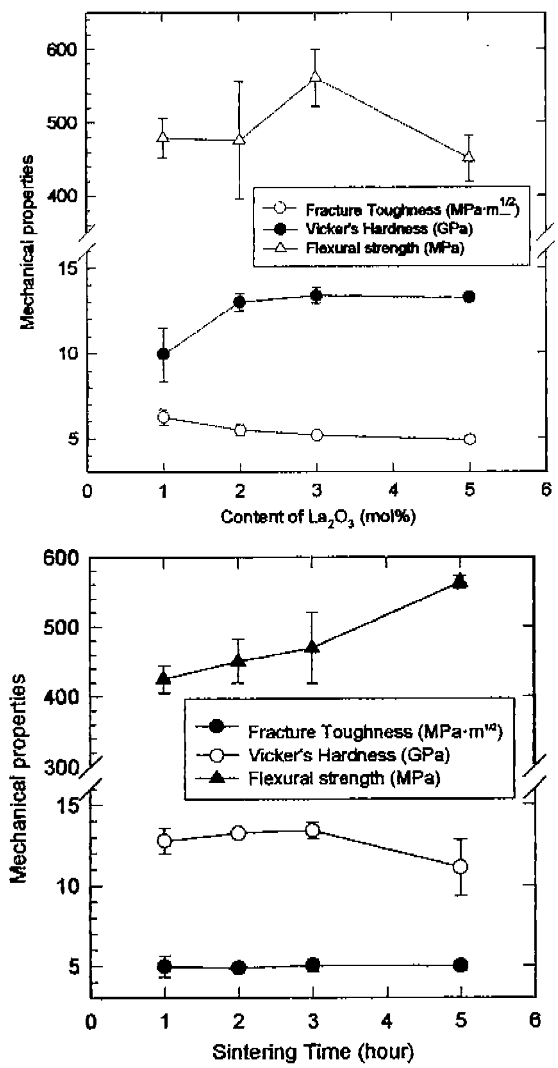

Fig. 7 Variation of mechanical properties of Si_3N_4 as a function of (a) the content of La_2O_3 and (b) the GPS holding time for Si_3N_4-5 mol% La_2O_3.

GPS holding time.

Fig. 8 shows a variation of removal rate of silicon nitride balls as a function of the La_2O_3 content and the GPS holding time for the 5 mol% of La_2O_3 in Si_3N_4, respectively. The removal rate of the materials was estimated by the decrease in diameter of the silicon nitride balls of grinding test. The removal rate of the materials per unit running time was calculated. The removal rate (μm/min) represents the numerical value for machinability of the materials. The previous study of machinability of the silicon nitride ball was related to physical and mechanical properties such as relative density, hardness, fracture toughness, and flexural strength, etc.[17]

As shown in Fig. 8(a), the removal rate of silicon nitride balls increased with increasing the content of La_2O_3 from 0.4 μm/min to 1.0 μm/min. With increasing the GPS holding time, the removal rate of silicon nitride ball was not changed to significantly from 0.5 μm/min. However, the rate of 1.0 μm/min occurred for the specimen of which the GPS holding time was 2 hour (Fig. 8 (b)). To optimize machinability of silicon nitride ball, a relation between mechanical properties and machinability was tried to be investigated. Consequently, it was found that mechanical properties and mechanical machinability were optimized at the content of 3 mol% La_2O_3 and for 2 hours of the GPS holding time. It was claimed that the machinability of the single crystal materials was mainly related to fracture toughness.[18] However, it is considered that machinability in the chemical-mechanical polishing of silicon nitride[19] is influenced by other parameters. Moreover, it would be required more wide studies to confirm machining parameter in order to improve the machinability.

IV. Conclusion

The Si_3N_4 with 2 wt% Al_2O_3, 4 wt% Y_2O_3 and La_2O_3 as additives was sintered by a gas pressure sintering (GPS) method at 1950°C, at 3 MPa of N_2 gas, for 2 hours. The GPSed silicon nitride with various La_2O_3 contents and the GPS holding time were investigated to determine machinability as well as mechanical properties such as hardness, flexural strength, and fracture toughness. Based on this investigation, the following conclusions can be made,

1. With increasing the content of La_2O_3, grain size was reduced since the La_2O_3 suppressed grain growth by generating the refractory grain-boundary phases. Therefore, hardness and flexural strength increased, but fracture toughness decreased.

2. With increasing the GPS holding time, fracture toughness increased, and hardness and flexural strength did not change significantly because of the longer β silicon nitride grains.

3. With increasing the content of La_2O_3, the materials removal rate of silicon nitride increased significantly.

4. With increasing the GPS holding time, the materials removal rate were changed except for 2 hours holding time.

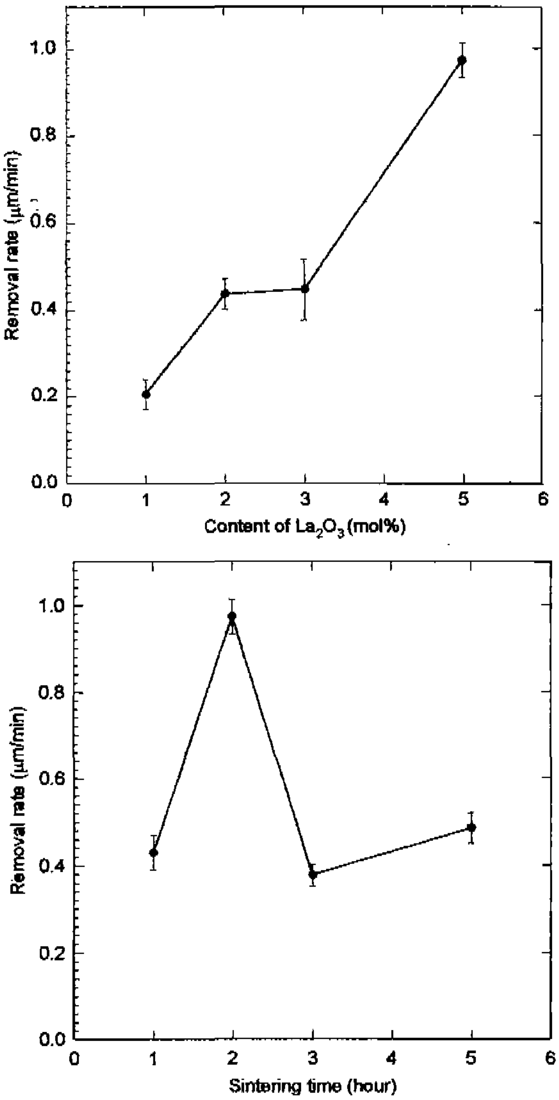

Fig. 8 Variation of machinability of Si_3N_4 as a function of the content of La_2O_3 and (b) the GPS holding time for Si_3N_4-5 mol% La_2O_3.

5. Consequently, it was found that mechanical properties and mechanical machinability were optimized at the content of 3 mol% La_2O_3 and for 2 hours of the GPS holding time.

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