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Preliminary effectiveness of ultrasonic nanocrystallline surface modification on the microstructure and tribological properties of sintered alumina

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This study reports the improvement in microstructural and tribological characteristics of sintered alumina (Al_2O_3) ceramic subjected to ultrasonic nanocrystalline surface modification (UNSM) technique. The surface microstructure of the untreated and UNSM-treated specimens was examined by scanning electron microscopy (SEM), laser scanning microscopy (LSM) and X-ray diffraction (XRD). The tribological properties of the specimens were assessed using a ball-on-disk tribometer against a bearing steel (SAE52100) ball. It was confirmed by SEM that the UNSM-treated specimen had much denser microstructure than that of the untreated specimen. The surface roughness of the UNSM-treated specimen obtained by AFM was found to be smoother compared to that of the untreated specimen, which may be attributed to the decrease in porosity. The tribological test results showed that the UNSM-treated specimens exhibited better tribological properties compared to those of the untreated specimens. It was also found that the UNSM technique was able to modify the surface in the topmost surface layer. The results of this study are expected to make sintered Al_2O_3 ceramic more attractive for a numerous applications in various industries.

Key words: Alumina, Friction, Wear, Surface engineering.

Introduction

Sintered alumina (Al_2O_3) is one of the most versatile of refractory ceramic oxides that has been attracting much attention because of its excellent properties and promising applications such as seal rings, drawn cones, guides and bearing parts [1]. Furthermore, because of its chemical inertness and biocompatibility, sintered Al_2O_3 is used for medical applications such as dental crowns, hip and knee prostheses, heart valves and bone implants [2]. The use of Al_2O_3 in contact-mechanical and tribological applications, where excellent mechanical and enhanced wear properties are required, has been continued growth. Favorable properties of Al_2O_3 for these applications are high stiffness and high hardness, temperature stability, corrosion and wear properties. However, monolithic Al_2O_3 suffers severe problems under mechanical and tribological loads due to its inherent brittleness, lack of defect tolerance and porosity, which can result in low resistance to wear and relatively high friction under dry sliding conditions. It has been well known that pores in most of the situations are inevitable in sintered materials due to the gas removal from interior (chamber) while sintering, even after the densification with the proper addition of the sintering additives [3]. Porosity results in detrimental influence on the microstructural, mechanical and tribological properties, and applications of sintered materials. Also, the strength of sintered materials depends basically on the presence of defects in the microstructure such as pores, agglomerates and cracks. Lindstedt et al. have reported earlier that the relatively high stress concentration at pores and pore clusters is responsible for localized slip leading to crack initiation [4]. It has been reported earlier that the tribological properties of Al_2O_3 are characterized by a transition from mild to severe wear, which occurs due to the change in mechanism from predominantly tribochemical reactions to surface damage [5]. A surface modification can result in improving the tribological performance of Al_2O_3 since microstructural parameters are significantly effect on the tribological properties. The objective of present work is to investigate the effectiveness of ultrasonic nanocrystalline surface modification (UNSM) technique on the microstructural and tribological properties of sintered Al_2O_3.

This surface modification technique is one of the newly developed surface modification technique, which is a simple, effective and economical approach for producing a nanocrystalline layer in metallic materials by reducing grain size of few layers from the surface into nanometer scale depending upon the treatment conditions. In this UNSM process, not only the static load, but also the dynamic load is exerted. The treatment is conducted striking a workpiece surface up to 2×10^4 or more times sec^{-1} with shots of an attached ball to the horn in the range of 10^3-10^5 mm^{-2}. The strikes, which can be described as cold-forging, introduce severe plastic

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deformation (SPD) and induce compressive residual stress to the surface layers with the aim of increasing the strength of material. The UNSM technique is characterized by its excellent performance and short cycle times, resulting in lower operating costs and reduced time requirements and thus, provides the users with significant competitive advantages. This simple surface modification technique is in full compliance with the today`s modern requirements. The accuracy and repeatability are guaranteed by special, fully-automatic process control. Further more details of UNSM technique can be found in our previous publications [6, 7].

Material and Methods

Specimen preparation

In this study, a series of Al_2O_3 disk specimens, with dimensions of 24 mm in diameter and 7.9 mm in thickness, were prepared using a powder metallurgy (P/M) process. The Al_2O_3 (A-480S) specimens with chemical composition of 92.13% O and 7.87% Al in wt.% were supplied by Kyocera Co., Ltd., Japan. Some properties of the specimens used in this study are listed in Table. 1. The original sintered specimens contain the micro-pores on the surface, which significantly affect the surface properties. Therefore, some of them were further subjected to UNSM technique in order to decrease the quantity of pores and to increase the strength.

The UNSM treatment parameters that were used to treat the specimens under strictly controlled conditions are listed in Table 2. Prior to UNSM treatment, the specimens were cleaned in acetone $(CH_3)_2CO$ and petroleum benzene (C_6H_6) mixture (ratio 1 : 1) for 10 min each using an ultrasonic bath to remove the impurities and particles from the surface.

Tribological and hardness tests

The tribological properties of the untreated and UNSM-treated specimens were investigated using a schwingung reibung verschleiss (SRV4, Germany) ball-on-disk tribometer at temperatures of 25 and 200 °C under dry and oil-lubricated conditions. All the tests were performed at a normal load of 20 N, frequency of 10 Hz with a stroke of 1 mm for 60 min. A bearing steel ball with a diameter of 10 mm was used as a counter surface specimen, which was selected because of its various applications. Each specimen was tested at least three times due to the data scattering. Poly-alpha-olefin (PAO) oil was used as a lubricant and its physical properties can be found in our previous study [7].

The Vickers hardness measurement of the specimens was performed using the Vickers hardness tester (Akashi Corp. AAV-4M, Japan) with a diamond pyramid Vickers indenter at a load of 10 N (1000 gf) and constant indenter dwell time of 30 s.

Surface analysis

The surface morphology and wear track that formed after tribological tests on the surface of the specimens were investigated using an atomic force microscope (AFM; SPA-400, Seiko, Japan) and a laser scanning microscopy (LSM; LEXT3500, Olympus, Japan), respectively. The chemical composition of the specimens was investigated using an energy dispersive X-ray spectroscope (EDS) system installed in a field emission scanning electron microscopy (FE-SEM; SUPRA40, Zeiss, Germany). X-ray diffraction (XRD) was carried out using a Bruker D8-Discover Advance and X3000 (X stress 3000) X-ray diffractometer with Cu Ká radiation in the range of 2θ= 40-80 ° with a step of 0.5 ° at a voltage of 35 kV and current of 40 mA to reveal information about the microstructure and phase identification of the specimens at the top surface. The average surface roughness and two-dimensional (2D) cross-sectional surface profiles of the specimens were obtained using an AFM and a surface profilometer (Surfcom 1500 SD3, Accretech, Japan), respectively. The cross-sectional profiles of the wear tracks were measured at three different points to quantify the wear volume. The specific wear rate and the corresponding standard deviation were quantified as the ratio of wear volume loss over the normal load multiplied by total reciprocating sliding distance.

Results and Discussion

Microstructure

Fig. 1 shows the comparison of surface microstructure for the untreated and UNSM-treated specimens. It is

Table 1. Some properties of Al_2O_3 used in this study.

Material	Method	Young's modulus, GPa	Poisson's ratio	Density, g/cm^3	Hardness, GPa
Al_2O_3	Sintering	380	0.23	3.9	17.2

Table 2. UNSM treatment parameters.

Frequency, kHz	Amplitude, μm	Horn speed, mm/min	Impact load, N	Interval, mm	Ball diameter, mm	Ball material
20	30	3000	60	0.07	2.38	WC

evident that the significant change in microstructure and reduction in surface roughness on the surface of the UNSM-treated specimen were observed. The untreated specimen consists of an irregular structure with some interconnected pores. The high peaks and valleys on the untreated specimen surface are also observed. The strikes of a WC tip deformed the peaks and partially eliminated pores during the UNSM treatment and result in the formation of a high-dense microstructure as shown in Fig. 1. However, some pores are still available after UNSM treatment as shown in Fig. 1(b), which represent the shallow valleys and a little remnant of pores. Fig. 1(c) shows the surface roughness profiles for the untreated and UNSM-treated specimens obtained from Fig. 1(d), which shows the comparison of microstructure for the untreated and UNSM-treated specimens. It can be seen that the deepest pore depth on the untreated specimen was about 2 μm, while on the UNSM-treated specimen was about 0.4 μm. The average surface roughness results measured over a scan area of 100 μm × 100 μm by AFM were found to be 47 and 17 nm for the untreated and UNSM-treated specimens, respectively. It was revealed that the microhardness of the untreated and UNSM-treated specimens were about 1750 and 1920 HV, respectively. The 2D AFM image of the indenter on the UNSM-treated specimen showed that the maximum penetration depth was about 2.8 μm. Moreover, the EDS results revealed that the UNSM technique has no effect on the chemical composition of sintered Al_2O_3 ceramic. Thus, UNSM technique is regarded as an effective method to increase the hardness, reduce the surface roughness and decrease the quantity of pores, which are beneficial for improving the mechanical and tribological properties of sintered ceramics. It is well known that the optimization of the surface microstructure of materials is of great concern since most failures such as fatigue, wear, cracks etc. occur on the surface.

The XRD is a non-destructive analytical technique for identification and quantitative determination of the various crystalline forms and sizes. Fig. 2(a) presents the XRD patterns obtained on the surface of the untreated and UNSM-treated specimens. It is obvious from the XRD pattern of the UNSM-treated specimen that there is no significant change in full width at half maximum (FWHM) and intensity of diffraction peaks compared to that of the untreated specimen (Fig. 2(b)). It is also evident from Fig. 2(a) that the position of the diffraction peaks remained constant which indicates that the lattice parameters are unchanged by UNSM treatment and no new phases were defined. However, a small difference of the results may be attributed to the produced modified layer in the topmost surface of the UNSM-treated specimen with a thickness of about 1 μm as shown in Fig. 3. It has been previously reported that the broadening in FWHM and decrease in intensity occur as a result of small grain size of the nanocrystalline surface layer [8]. Sintered ceramics are generally hard and brittle because of bonds that hold the atoms together in the material. For this reason, producing a nanoscrystalline surface layer at the top surface is complicated and problematic than in metallic materials.

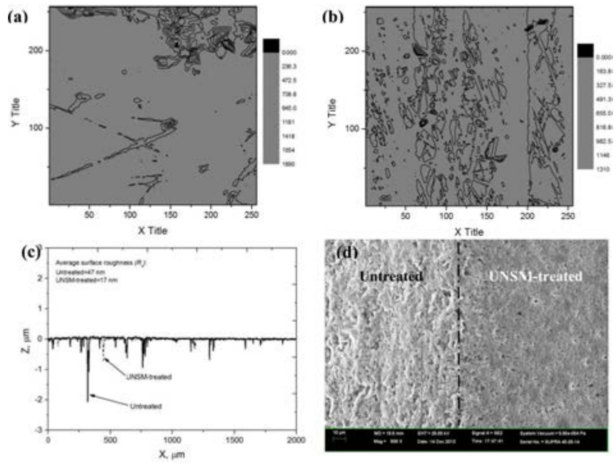

Fig. 1. Comparison of surface morphology (a, b), surface roughness profile (c) and microstructure (d) of the untreated and UNSM-treated Al_2O_3 specimens. 2D AFM scan area of 100 μm × 100 μm.

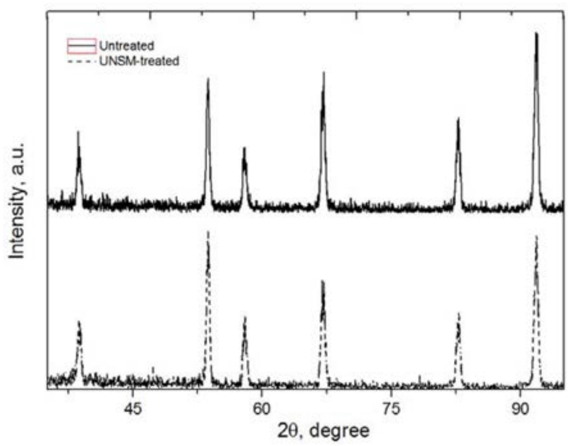

Fig. 2. X-ray diffraction patterns of the untreated and UNSM-treated specimens.

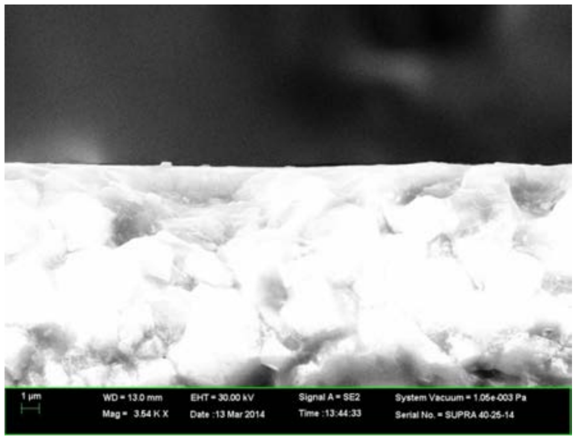

Fig. 3. Cross-sectional FE-SEM of the UNSM-treated specimen showing a modified layer in the topmost surface layer.

Friction and wear behavior

Fig. 4(a) shows the friction coefficient behavior as a function of reciprocating sliding time for the untreated and UNSM-treated specimens at a normal load of 20 N and temperatures of 25 and 200 ºC under dry and oil-lubricated conditions. It can be seen from Figs. 4(a) and 4(b) that the untreated specimens exhibited a high friction coefficient value of about ~ 0.76 and ~ 0.87 with high fluctuation throughout the test period at temperatures of 25 and 200 ºC, respectively. However, the UNSM-treated specimens exhibited a relatively lower friction coefficient value of ~ 0.64 and ~ 0.73 throughout the test period, respectively. As shown in Fig. 4(c), the UNSM-treated specimen showed a lower friction coefficient compared to that of the untreated specimen under oil-lubricated condition at a temperature of 25 ºC. Interestingly, the untreated specimen exhibited very stable friction behavior at the initial stage under oil-lubricated conditions at a temperature of 200 ºC, but increased abruptly after sliding time of about 6 min. However, the UNSM-treated specimen exhibited unstable friction behavior at the initial stage, but got stable after sliding time of about 7 min and then increased and decreased again. This friction behavior of the specimens at a temperature of 200 ºC may be attributed to the supplied oil evaporation, which considered as a transition friction behavior that corresponds to the interface accommodation between the disk and the counter surface ball specimens followed by stationary regime in which materials removal takes place at a constant rate [9]. In the running-in regime, the ball had low interaction with the disk specimen, once the initial contact with the disk specimen occurs on the asperities and these asperities do not experience plastic deformation. In the stabilized regime, those asperities experience plastic deformation and the wear debris take part at the sliding interface.

The wear behaviors of the specimens were further investigated using a LSM in order to better understand the influence of the UNSM technique on the wear resistance. Figs. 5 and 6 present the LSM images of wear tracks formed on the untreated and UNSM-treated specimens under dry and oil-lubricated conditions at temperatures of 25 and 200 ºC, respectively. It can be clearly seen after sliding tribological tests that the UNSM-treated specimens slid against a bearing steel ball exhibited a very small difference in wear track compared to those of the untreated specimens with evidence of severe signs of abrasion inside the wear track, which is typically caused by abrasion due to asperities of the counter surface or particles and debris generated during reciprocating sliding tribological test. Moreover, the UNSM-treated specimen under oil-lubricated conditions at a temperature of 200 ºC exhibited very mild wear track as shown in Figs. 6(b) and 6(d), which may be attributed to the oil evaporation.

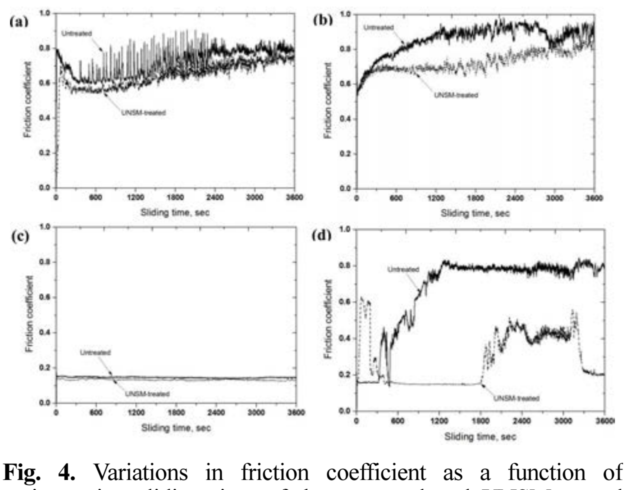

Fig. 4. Variations in friction coefficient as a function of reciprocating sliding time of the untreated and UNSM-treated Al_2O_3 specimens under dry (a, b) and oil-lubricated (c, d) conditions at temperatures of 25 and 200 ºC, respectively.

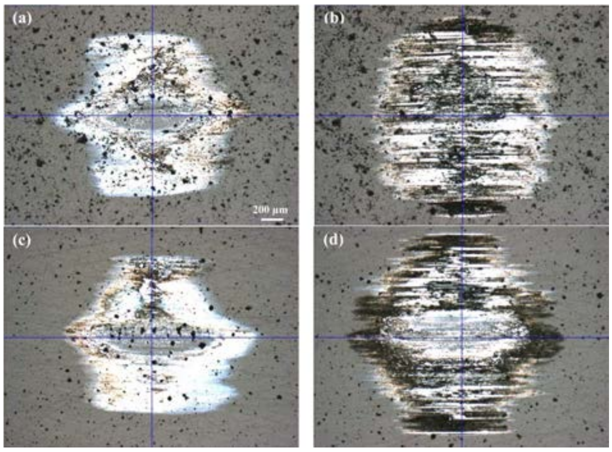

Fig. 5. LSM images of wear tracks of the untreated (a, b) and UNSM-treated (c, d) Al_2O_3 specimens under dry conditions at temperature of 25 and 200 ºC, respectively.

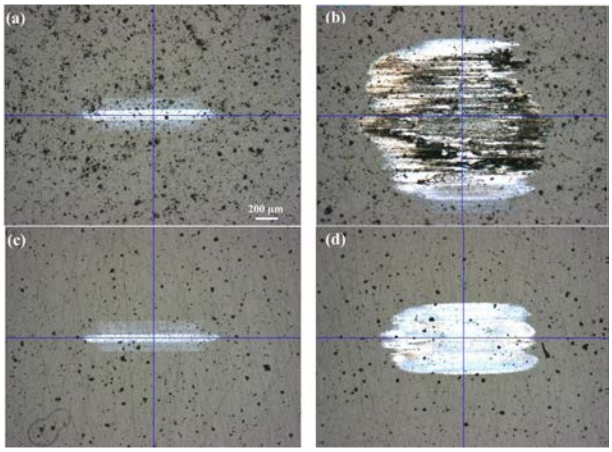

Fig. 6. LSM images of wear track of the untreated (a, b) and UNSM-treated (c, d) Al_2O_3 specimens under oil-lubricated conditions at temperature of 25 and 200 ºC, respectively.

Fig. 7 shows the quantified wear rate of the untreated and UNSM-treated specimens derived under dry and oil-

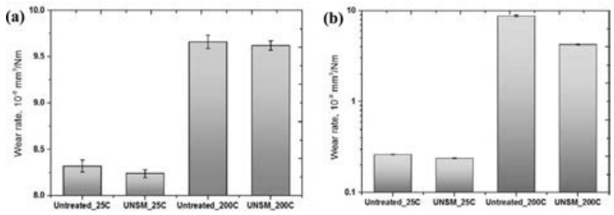

Fig. 7. Wear rate of the untreated and UNSM-treated Al_2O_3 specimens under dry and oil-lubricated conditions at temperatures of 25 and 200 °C, respectively.

lubricated conditions. It is obvious that there is no significant effects of UNSM technique on wear property of Al_2O_3 under dry sliding conditions, but a significant effectiveness was observed under oil-lubricated consitions. Despite the distinction in friction coefficient was remarkable at under dry conditions, but in wear rate it was relatively small since the modified surface layer in the topmost surface was worn out very rapidly under due to high friction. The relatively lower friction coefficient and higher resistance to wear of the UNSM-treated specimens compared to the untreated specimens may be attributed to the increase in hardness, decrease in the quantity of pores and reduction in surface roughness. The initial surface roughness also plays an important role in defining the wear rate. It is well known that the lower initial surface roughness is responsible for less wear and low friction cofficient [10]. According to Hertzian contact-based models of brittle ceramics, the wear loss largely depends on the mechanical properties such as hardness, fracture toughness etc. [11]. It was also found that the friction coefficient and wear rate of the untreated and UNSM-treated specimens increased with increasing temperature. It has been previously reported that the friction coefficient and wear rate of Al_2O_3 very much depend on the temperature, which increased with increasing the temperature [12]. Wang and Hsu [13] have studied the influence of operating parameters and environment on the wear of Al_2O_3. They have showed that wear phenomenon can be demonstrated in practically in all cases by a sudden icrease in friction coefficient and it depends on environment, operating parameters and microstructure. Moreover, Trabelsi et al. [14] have showed that the influence of temperature is predominant, because of the strong temperature dependence of toughnening mechanisms.

It is worth mentioning that the effectiveness of UNSM technique on the surface properties of metallic materials is significant rather than sintered ceramics, which may be attributed to the brittleness of sintered ceramics. It is well known that sintered ceramics undergo large plastic deformation without rupture since they are strong, but brittle because of the bonding present between the metal and non-metal components of the materials. Bonds are very strong and require a relatively strong amount of energy to break and they can stretch during plastic deformation. Therefore, an applied impact load of UNSM technique should be good enough to break the bonds completely to generate a plastically deformed surface layer at the top surface of sintered ceramics [9].

Conclusions

In this study, the effectiveness of UNSM technique on the microstructural and tribological properties of Al_2O_3 was investigated. It was found that the UNSM-treated specimens had better friction and wear properties compared to those of the untreated specimens under dry and oil-lubricated conditions at temperatures of 25 and 200 °C. The improvement may be ascribed to the increase in hardness, decrease in the quantity of pores and reduction in surface roughness. It was also found that the UNSM technique was able to modify the surface layer at the top surface having a high hardness. The results of this study are expected to make sintered Al_2O_3 ceramic more attractive for a numerous applications in various industries.

Acknowledgments

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