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Tribological behavior of ceramic materials (Si₃N₄, SiC and Al₂O₃) in aqueous medium

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Abstract

The friction and wear behavior of self-mated Si_3N_4 , SiC and Al_2O_3 in water were investigated by varying the test conditions of applied load and sliding speed. It was found that, for self-mated Si_3N_4 and SiC ceramics, the tribochemical reaction resulted in surface smoothening with low friction coefficient at high load and high speed condition. Al_2O_3 shows high friction coefficient, but better wear rate (10^{-11} mm²/N) than other ceramic materials. © 2003 Elsevier Ltd. All rights reserved.

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1. Introduction

Advanced ceramics, being hard and chemically stable at room temperature, offer many possibilities of producing components with beneficial combinations of properties including good tribological performance. Ceramic components may serve well in various media, even aqueous media. However, there are major differences in the tribological behavior of different ceramics in water.^{1,2}

The friction and wear of Si₃N₄, SiC and Al₂O₃ under water lubrication has been studied by several research groups. Fischer and Tomizawa³ clearly showed the tribochemical wear of Si₃N₄ in water and reported a low friction coefficient of 0.002. Sasaki⁴ reported that for Si₃N₄ and SiC ceramics, the friction coefficient dropped as low as 0.01 when they slid against themselves in water. Chen et al.⁵ reported that the wear rate of both Si₃N₄ and SiC ceramics were in the same magnitude of 10⁻⁹ mm²/N. It has been reported that much shorter running-in period was taken for self-mated Si₃N₄ than for self-mated SiC, also self-mated Si₃N₄ exhibited lower friction coefficient (0.0035) after running-in in water than self-mated SiC did (0.01). Alumina wear

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rates have been reported to be in a wide range of magnitudes, from 10^{-6} mm²/N to 10^{-12} mm²/N.^{6–8} The relatively large variations observed in the wear rate of alumina have been attributed to factors such as the test configuration, test conditions and material properties.

Tribological performance of sliding pairs depends on environmental conditions, interfacial media, sliding velocity, applied load, initial surface quality, microstructure of the mated materials and other factors. ^{9–11} Though there are reports on tribological behavior of ceramic materials at the respective above said conditions, still there is a lack of knowledge about the friction and wear behavior of ceramic materials for a wide range of test conditions under a long sliding distance.

And hence, the purpose of this brief study is to evaluate the friction and wear behavior of three different types of ceramic (Si_3N_4 , SiC and Al_2O_3) materials by varying the applied load (9.8, 24.5 and 49 N) at different sliding speed (0.18, 0.54 and 1.18 m/s).

2. Experiment

2.1. Material selection

The present work is based on water-lubricated ballon-disk test with self-mated sintered ceramic specimens

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of $\mathrm{Si_3N_4}$, SiC and $\mathrm{Al_2O_3}$. All the ball specimens were supplied by Nihon Ceratec. Co. Ltd, Sendai and the disk specimens of $\mathrm{Si_3N_4}$, SiC and $\mathrm{Al_2O_3}$ were supplied by NGK Insulators Ltd., Nagoya, Ibiden Co. Ltd., Ogaki and Kyocera Corp., Kyoto, respectively. Polished balls with diameter of 9.525 mm were used in the as-received condition with a surface roughness of <0.1 μ m. The disks were ground, lapped and polished with 0.5 μ m diamond slurry, the flat test surface had a roughness of <0.02 μ m. Table 1 summarizes the commercial materials used in the present investigation together with some of their room temperature properties.

2.2. Friction and wear test

The friction and wear behavior of three different ceramics were studied by ball-on-disk tribometer in water lubrication condition. All the sliding tests were conducted at room temperature (25±3 °C). Prior to testing, the specimens were ultrasonically cleaned with acetone and dried. The sliding conditions of different sliding speed (0.18, 0.54 and 1.18 m/s) and load (9.8, 24.5 and 49 N) were applied on the self-mated ceramic materials with a sliding distance of 2000 m. The friction force and applied force were measured continuously during each test.

Wear volumes of the worn specimens were evaluated using a roughness tester and optical microscope. For the worn disk specimen, the cross-sectional area of the worn track was taken as the average of that measured at four separate locations. The worn volume of the disk (V_d) was calculated according to the following equation.

$$V_{\rm d} = 2\pi R \left(\frac{S_1 + S_2 + S_3 + S_4}{4} \right)$$

where R and S are the sliding radius and cross section area of the worn track. In all the cases, the values reported are the average of three tests.

For evaluating the worn volume of the ball specimen, the diameter of the worn area of the ball was measured at four equidistant locations. The worn volume of the

Table 1 Material properties

Material	Al_2O_3		SiC		Si ₃ N ₄	
	Ball	Disk	Ball	Disk	Ball	Disk
Density, ρ (g/cm ³) Vickers hardness, H_{ν} (GPa) Flexural strength, σ_f (MPa) Young's modulus, E (GPa)	18 450 ^a	17	3.08 22.1 500 ^b 408	3.15 22.7 500 ^b 420	13.8	3.2 17 1100 ^b 310

^a Represents the 3-pt bend strength measurement.

ball $V_{\rm b}$ was calculated according to the following equation 12

$$V_{\rm b} = \frac{\pi A^3 B}{32D}$$

where A and B are the longer and shorter diameters of the worn area of the top of the ball, and D is the ball diameter. In all the cases, the values reported are the average of three tests the same as for the case of $V_{\rm disk}$. The wear rate $(W_{\rm s})$ was determined from wear volume as.

$$W_{\rm s} = \frac{V}{Pd}$$

where P and d are the normal load and the sliding distance, respectively.

The surface of the specimens (ball and disk) after wear tests was observed using optical microscope to elucidate the wear behavior.

3. Results

3.1. Friction coefficient

The friction traces of three different ceramics at high load (49N) and high speed (1.18 m/s) condition are shown in Fig. 1. The early rise in friction coefficient over the minimum sliding distance of 100 m for all the three kinds of ceramic materials was observed and referred to initial running-in period. ¹³ After running-in, the friction coefficient of Al₂O₃ followed a gradual decrease with sliding distance, and then meandered about a mean value of 0.28. The friction coefficient decreased sharply for Si₃N₄, but for SiC, it decreased gradually and then remained constant with sliding distance. The coefficient of friction for SiC started at 0.16 and followed by a

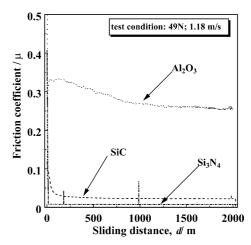


Fig. 1. Variation of friction coefficient with sliding distance.

^b Represents the 4-pt bend strength measurement.

gradual decrease and remained constant about an average value of 0.02. In the case of Si_3N_4 , the friction trace was suddenly decreased from 0.35 and attained a steady-state friction of mean value around 0.009. Few fluctuations were observed in the friction trace of Si_3N_4 , which evolved due to the formation and removal of tribochemical layer.

Different wear modes were generated on test pieces by varying the applied load and sliding speed. The average friction coefficient (μ_{av}) value determined from the friction trace for different load and speed conditions are shown in Fig. 2. The average friction coefficient value was determined from the friction trace of sliding distance between 0 and 2000 m. Initially, when the sliding speed was increased from 0.18 to 0.54 m/s, a sudden decrease in μ_{av} was noticed for Si_3N_4 ceramics from 0.18 to 0.05 and a moderate decrease from 0.31 to 0.28 in Al_2O_3 ceramics. But, μ_{av} of SiC ceramics showed a gradual or no change with sliding speed. Later, during high sliding speed (>0.54 m/s), irrespective of the ceramic materials, the friction coefficient was found to decrease faintly rather remained constant with increasing sliding speed. The coefficient of friction, had minimum effect due to applied load for all the three ceramic materials.

3.2. Wear rate

Figs. 3 and 4 show the specific wear rate of ball and disk specimens, respectively as a function of applied load and sliding speed for three different ceramic materials. The difference in the scale of *Y*-axis (Wear rate, W_s) for the respective ceramic material is to be noted. It shows that the specific wear rate of alumina ball (10^{-11} mm²/N) is lower than that of SiC (10^{-10} mm²/N) and Si₃N₄ (10^{-10} mm²/N) of one order of magnitude at any sliding condition. Similarly, alumina disks also show low wear rate of magnitude 10^{-11} mm²/N, smaller than

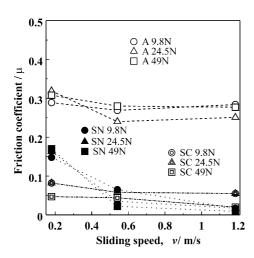


Fig. 2. Variation of friction coefficient with sliding speed and applied load.

SiC $(10^{-10} \text{ mm}^2/\text{N})$ and Si₃N₄ $(10^{-9} \text{ mm}^2/\text{N})$ of one and two orders of magnitude, respectively. For all the ceramic materials, there is no discrepancy in the trend of wear rate evaluated at different test conditions between the ball and disk specimens. Whereas, the magnitude of wear rate determined between ball and disk specimens of the respective ceramic materials are different.

At low sliding speed (0.18 m/s), the wear rate of $\mathrm{Si}_3\mathrm{N}_4$ ceramics was found to increase with increasing applied load. In SiC ceramics, a trivial change in wear rate with applied load was noticed. But, in the case of $\mathrm{Al}_2\mathrm{O}_3$ ceramics, the reverse trend of decreasing wear rate with increasing applied load was observed. For all the ceramic materials, when the sliding speed was increased, a noticeable decrease in wear rate was recognized. And similarly, the wear rate was found to decrease with increasing applied load.

The wear tracks of ball and disk specimens of different ceramics were analyzed after sliding over 2000 m.

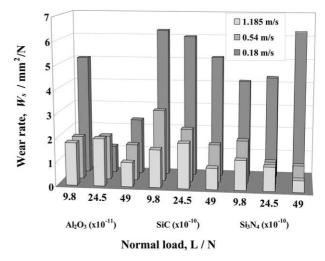


Fig. 3. Variation of wear rate of ball with applied load and sliding speed.

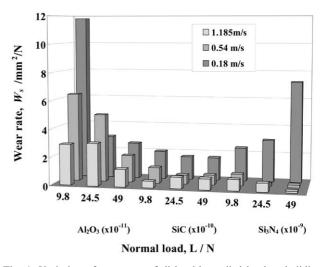


Fig. 4. Variation of wear rate of disk with applied load and sliding speed.

Optical microscopy of wear track of disks (Fig. 5) show, very flat and smooth worn surface with very few wear particles. In particular, Si₃N₄ and SiC worn tracks were smoothened like polished surfaces. Also, the difference in worn width between the ceramic materials explains the respective change in wear rate.

4. Discussion

The effects of normal load, sliding speed, temperature and test duration on the friction of ceramics are usually be interpreted in terms of changes in the tribochemical surface films and the extent of fracture in the contact region.

When self-mated Si_3N_4 and SiC ceramics slide against in water, a high friction coefficient was obtained at low sliding speed. Particularly, in Si_3N_4 ceramics, a high friction coefficient was observed at low speed (0.18 m/s) and high load (49 N) condition. Also, an increase in wear rate was noted with applied load at low sliding speed, which corresponded well with high friction coefficient. At low sliding speed, when the applied load increases, the separating lubrication medium between the sliding pairs gradually get lost. And hence, the increase in wear rate and friction are attributed to the contact of initial surface asperities with high contact stress. In the case of SiC ceramics, the friction coefficient (0.08) at low sliding speed was not as high as Si₃N₄ ceramics. Also, the change in wear rate at low sliding speed was much insignificant with increasing applied load.

When the sliding speed was increased, a sudden and a gradual decrease in friction coefficient have been noticed for Si₃N₄ and SiC ceramic materials, respectively. This decrease in friction with increasing sliding speed is

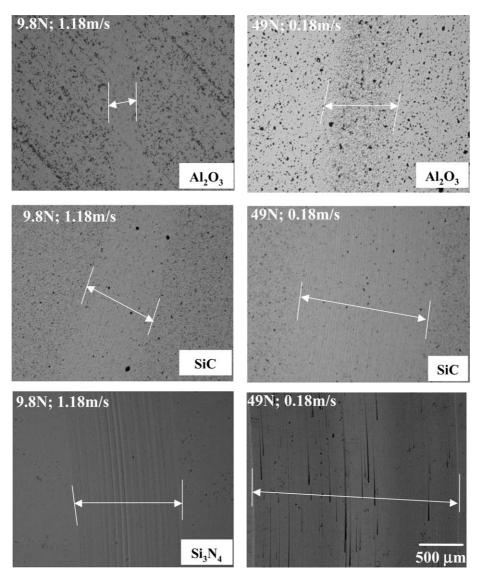


Fig. 5. Optical micrographs of the worn images of different ceramics.

associated with the high energy developed from sliding, tends to accelerate the tribochemical reaction. And this effect has been attributed to hydrodynamic lubrication after an ultra smooth surface is obtained by tribochemical wear. As the contact surfaces have better conforming by wear, tribochemical film of colloidal silica grows on wear surface and it is then removed from the surface by forming fine rolls or dissolved into water. ¹⁴ Further, at high sliding speed, the lubrication effect was expected to be very good with increasing applied load attributed to the enhance of tribochemical reaction from sliding.

At high sliding speed, the formation of smoothened surface by tribochemical reaction with increasing applied load resulted in low wear rate for both the $\mathrm{Si}_3\mathrm{N}_4$ and SiC ceramic materials. The tribochemical reaction is thought to have the following reactions associated with Gibb's free energy, ¹⁵

$$Si_3N_4 + 6H_2O \rightarrow 3SiO_2 + 4NH_3$$
; $\Delta G = -566.5 \text{ kJ/mol}$
 $SiC + 2H_2O \rightarrow SiO_2 + CH_4$; $\Delta G = -369.1 \text{ kJ/mol}$

The free energy of both Si₃N₄ and SiC are found to be high. And hence, there exists high possibility for the formation of SiO₂ film. The difference in friction coefficient between self-mated Si₃N₄ and SiC tribopairs suggest that the lubrication mechanisms of the respective tribopairs sliding in water are different. The formation and removal of SiO₂ film evolved from Si₃N₄ sliding pairs is expected to be rapid than SiC tribopairs. And this might be attributed to more wear loss of Si₃N₄ than SiC. Probably, Si₃N₄ tribopairs are left behind with the tribochemical product even after the removal of SiO₂ film, which attributed for the low friction coefficient. Whereas, in the case of SiC/SiC sliding, the formation of SiO₂ film is not easily removed and hence resulted with low wear rate.

A high friction coefficient of about 0.3 was obtained from Al₂O₃/Al₂O₃ sliding when compared to non-oxide ceramics. When self-mated alumina slides in water, a lubricious trihydroxide layer is expected to form on the wear surface. The effectiveness of the tribochemical product decides the friction generated from the respective sliding pairs. The fluid film generated from sliding should be thinner than a certain value (<1) and hence the occurrence of high friction coefficient. Such high friction coefficient results by the occurrence of complete loss of film support. This corresponds to the lubrication regime well above the boundary lubrication, typical for dry sliding. Also, it can be noticed that the friction coefficient of alumina does not vary appreciably even for different sliding speed and applied load.

The free energy calculated from the tribochemical reaction of Al_2O_3 is very low ($\Delta G = -25.9$ and -21.6 kJ/mol for the formation of aluminium trihydroxide and aluminium hydroxide, respectively), which suggests

the very low reaction rate of Al_2O_3 with water. Since the reaction rate between alumina and water is low, the lubrication effect will be minimum driving to the regime above boundary lubrication.

Though the friction coefficient is very high for Al₂O₃/Al₂O₃ sliding in water, it results in a very low wear rate when compared to Si₃N₄ and SiC ceramics. The wear mechanism described for the self-mated Al₂O₃ sliding in water is a cooperation between mechanical and chemical interactions to bring about a series of reactions. In this tribochemical reaction, the contribution of "tribo" component namely, anisotropic shear stress and high temperature through frictional sliding are driven by the subsequent chemical reaction derived from the "chemical" component of the reaction sequence.¹⁷ And hence, the low wear rate evolved from Al₂O₃/Al₂O₃ sliding is merely due to the chemical reaction, similar like etching. The low wear rate of alumina by grain orientation and chemical etching can be noticed from the wear track (Fig. 5, upper right) developed from high load.

Inspite of applied load and sliding speed, alumina shows better wear rate when compared to other ceramic materials. At low sliding speed, trend of wear behavior with applied load was unusual and reversal to the other materials. In particular for the minimum applied load (9.8 N), a high wear rate was seen. In order to clarify the present result, a roughness profile (Fig. 6) is shown, illustrating the worn depth of Al₂O₃ ceramics at different sliding condition and has been compared with Si₃N₄ ceramics. In the case of Al₂O₃ ceramics, the mean worn depth was found to be very low of about 0.1µm and was same even when the applied load increased from 9.8 to 49 N. The sharp peaks observed in the roughness profile represent the inclusion of pores in the initial surface. At both the conditions, the wear volume was also determined to be very low of 0.02 mm³. Also, there was not much difference noted in friction coefficient of alumina at the respective above conditions. From the above wide range of test conditions, the drastic change in wear rate (Figs. 3 and 4) and negligible difference in wear volume (Fig. 6) are not comparable. And hence, it led us to suggest the limitation in experimental error while measuring the wear rate (remains at the same narrow order of magnitude). Whereas, it is undoubtedly seen from the roughness profile of Si₃N₄ that the drastic increase in worn depth is comparable with increasing wear rate at high load (49 N) and low speed (0.18 m/s) sliding condition. Therefore, the change in wear rate of alumina ceramics for various test conditions is very low and lying in the narrow order of range, explicit the experimental error. Still, if the test conditions are widened, either increasing the sliding distance or applied load may clear the present diffusive results.

The observed worn tracks of three different ceramic materials selected from few specific conditions are

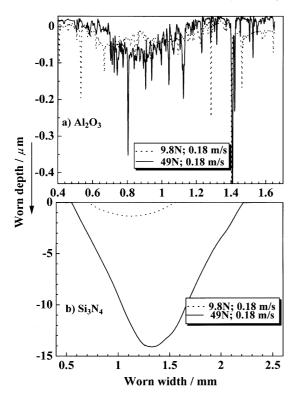


Fig. 6. Surface profiles of wear tracks of Al₂O₃ and Si₃N₄ ceramics.

shown. Both the wear tracks of SiC and $\mathrm{Si}_3\mathrm{N}_4$ disks were observed to be very smooth with no wear debris. The nature of smoothening between carbide and nitride ceramics reveals the effect of tribochemical reaction between the respective material and water. The change in worn width between the materials irrespective of the wear conditions explains the difference in wear rate evaluated for different materials. The narrow worn width observed from alumina sliding interprets the low wear rate when compared with other materials under study.

5. Conclusion

The wear properties of Si₃N₄, SiC and Al₂O₃ ceramics were studied at various test conditions. It is concluded that the wear of all ceramic materials under all test conditions was due to reaction with water and not by mechanical. The basic tribological properties confirmed from the present study are, (i) Si₃N₄ possessed very low friction coefficient with high wear rate, (ii) SiC showed low friction coefficient with medium wear rate, but

stable for any condition and, (iii) Al_2O_3 revealed high friction coefficient with low wear rate.

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References

- Tomizawa, H. and Fischer, T., Friction and wear of silicon nitride and silicon carbide in water: hydrodynamic lubrication at low sliding speed obtained by tribochemical wear. *Am. Soc. Lubr. Eng. Trans*, 1987, 30, 41–46.
- Andersson, P., Water-lubricated pin-on-disc tests with ceramics. Wear, 1992, 154, 37–47.
- Fischer, T. E. and Tomizawa, H., Interaction of tribochemistry and microfracture in the friction and wear of silicon nitride. *Wear*, 1985, 105, 29–45.
- Sasaki, S., The effects of the surrounding atmosphere on the friction and wear of alumina, zirconia, silicon nitride and silicon carbide. Wear, 1989, 134, 185 - 200.
- Chen, M., Kato, K. and Adachi, K., Friction and wear of selfmated SiC and Si₃N₄ sliding in water. Wear, 2001, 250, 246– 255.
- Jeng, M. C. and Yan, L. Y., Environmental effects on wear behavior of alumina. Wear, 1993, 161, 11–16.
- Hsu, S. M. and Shen, M. C., Ceramic wear maps. Wear, 1996, 200, 154–175.
- 8. Takadoum, J., Tribological behavior of alumina sliding on several kinds of materials. *Wear*, 1993, **170**, 285–289.
- Woydt, M., Kalffke, D., Haibig, K. H. and Czichos, H., Tribological transition phenomena of ceramic materials. *Wear*, 1990, 136, 373–381.
- Hisakado, T. and Morijiri, H., Friction and wear mechanisms of ceramics in vacuum and characteristics of normal load. *Jpn. J. Tribol*, 1995, 40, 239–250.
- Wang, H. C., Umehara, N. and Kato, K., Frictional characteristics of ceramics under water-lubricated conditions. *Tribol. Lett.*, 1998, 5, 303–308.
- Japanese Industrial Standards Committee, Testing methods for wear resistance of high performance ceramics by ball-on-disk method, JIS R 1613. Japanese Standard Association, Tokyo, Japan 1993.
- Bushan, B., Principles and Applications of Tribology. John Wiley and Sons, New York, 1999.
- Xu, J., Kato, K. and Hirayama, T., The transition of wear mode during the running-in process of silicon nitride sliding in water. Wear, 1997, 205, 55–63.
- 15. Sasaki, S., The effect of water on friction and wear of ceramics. *J. Jpn. Soc. Lub. Eng.*, 1988, **33**, 620–628.
- Khonsari, M. M. and Booser, E. R., Applied Tribology, Bearing Design and Lubrication. John Wiley and Sons, New York, 2001.
- Gates, R. S., Hsu, S. M. and Klaus, E. E., Tribochemical mechanism of alumina with water. *Trib. Trans*, 1989, 32, 357– 363.