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The influence of a solid lubricant dispersion on tribological behavior of Si_3N_4 based composites under water lubrication

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The tribological behavior of monolithic Si_3N_4 and solid lubricants dispersed Si_3N_4-based composites has been assessed under a high load and low speeds in an aqueous environment. The results showed that the friction coefficient of the Si_3N_4 was not significantly reduced when compared to dry sliding and this was attributed to the failure to maintain a lubricating layer between the solid-solid surfaces. In the case of the composites, the initial high friction coefficient was reduced shortly after the beginning of the wear test and maintained a low value (approximately 0.03) throughout. This was attributed to the solid lubricating effect of the composite resulting in a lower stress at the contact asperities preventing the removal of the lubricating layer. The solid lubricant content did not affect the value of the friction coefficient under hydrodynamic type lubrication, but the running in distance decreased with the solid lubricant dispersion. The type and amount of solid lubricant affected the distance required to reach a low friction regime, but all types of solid lubricant showed decreasing friction with increasing amount. In Stribeck analysis, the addition of the solid lubricant resulted in a highly graphitic transfer layer on the Si_3N_4 material, and shifted the transition points from hydrodynamic to mixed and from mixed to boundary lubrication regimes to more severe conditions. It also reduced the friction coefficient in the boundary lubrication regime.

Key words: Tribology, Friction, Solid lubricant, Silicon nitride, Composites.

Introduction

Silicon nitride is one of the most widely-employed engineering ceramics due to the fact that the properties can be tailored greatly with the microstructure [1, 2, 3]. Recently, Si_3N_4 ceramics have been used in tribological applications such as bearings for hard disks and mechanical seals in corrosive environments. In addition, many attempts at improving the tribological properties, both the friction coefficient and wear resistance, of silicon nitride ceramics have been carried out [4, 5].

In addition to an improvement of the wear resistance, a number of studies have focused on the development of fiber reinforced ceramic matrix composites in the search for improvements in the mechanical properties, in particular improving their brittle nature [6]. Recently, we have developed Si_3N_4/carbon fiber composites with excellent tribological properties under dry sliding conditions [7, 8].

Tomizawa and Fischer [9], and other researchers [10, 11], have confirmed that the sliding of Si_3N_4 in water can result in friction coefficients as low as 0.01 under certain conditions. In their reports, a decrease in friction coefficient was observed with increasing sliding velocity under a constant normal load. However, no reduction in friction coefficient was seen for low speed conditions. Chen *et al.* also reported the tribological behavior of a Si_3N_4-Si_3N_4 self-mated pair under various conditions of load and speed and showed that there was no reduction in friction under high loads and low speeds [12]. The friction behavior of Si_3N_4 ceramics in aqueous sliding conditions shows a similar behavior to that which produces the typical Stribeck curve under oil-lubricating conditions.

This means that the region of the low friction coefficient of Si_3N_4 ceramics depends on the sliding speed and normal load. In the case of high normal load and low speeds, such that the conditions represent the boundary lubrication regime, the friction coefficient of self-mated Si_3N_4 indicates relatively higher values in the range of 0.3 to 0.8.

On the other hand, Hirai and Kita reported that a Si_3N_4/Mo-Fe composite indicated good friction behavior in the boundary lubricating region under oil lubrication [13]. In this material dispersed molybdenum silicide was produced by an oxidizing heat treatment, and it was claimed that this was effective in acting as a solid lubricant under the oil lubrication environment. It is also expected that under boundary-lubricating conditions in an aqueous environment solid lubricants can improve the friction

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behavior since solid-solid contacts occur in these severe wear condition regions. In a previous study the friction behavior of Si_3N_4/carbon short fiber composites under high load and low speed conditions in an aqueous solution has been reported [14]. Also, the tribological properties of Si_3N_4/carbon fiber composites under water lubrication were clarified in detail, and the friction behavior of Si_3N_4/carbon fiber composites under various sliding conditions and with various carbon fiber contents was researched [15]. However, the effects of another solid lubricant addition to silicon nitride ceramics on their tribological properties under water lubrication were not clarified in detail. In this study, various solid lubricant-dispersed silicon nitride based composites were fabricated and the effect of the solid lubricant on tribological properties under water lubrication was researched.

Experimental Procedure

A monolithic silicon nitride was produced by mixing yttrium oxide (RU-P grade, Shin-etsu chem. Co. Ltd., Japan) and aluminum oxide powders (AKP-50, Sumitomo Kagaku Co. Ltd., Japan) as sintering additives to silicon nitride powder (over 95% α-phase content, ESP grade, Ube Industries Ltd., Japan). The composition of the starting powders was 93 mass% Si_3N_4, 2 mass% Al_2O_3 and 5 mass% Y_2O_3. The raw powder was homogeneously mixed by ball milling and then dried using a vacuum evaporator. The powders were hot-pressed at 1950 °C for 2 h at 30 MPa pressure in a 900 kPa nitrogen atmosphere.

For the silicon nitride/carbon fiber composites, the vacuum evaporated and sieved powders were mixed with high graphitized carbon fiber using planetary ball milling. The composite powders were sintered under the same conditions, and the detailed procedure for sample preparation has been reported in detail previously [7], Si_3N_4/WB [16] and Si_3N_4/BN composites were fabricated in a similar fashion (Fig. 1).

The friction behavior was evaluated by ball-on-disk experiments against polished commercial Si_3N_4 balls with

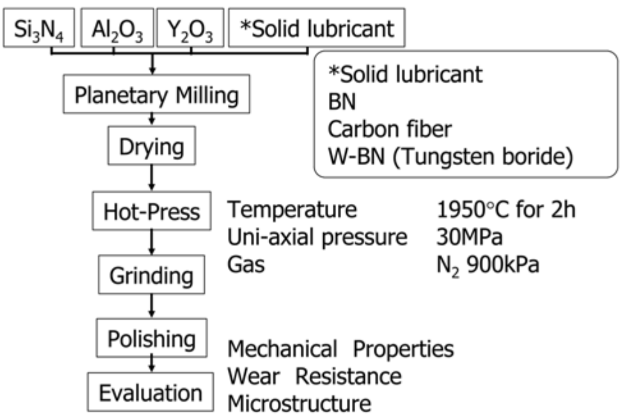

Fig. 1. Fabrication procedure for Si_3N_4-solid lubricant composites preparation.

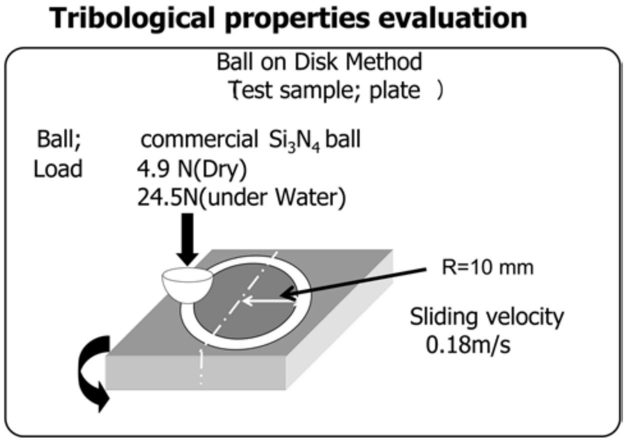

Fig. 2. Schematic image of the tribological evaluation.

diameters of 9.525 mm (Nihon Ceratec Co. Ltd., Miyagi, Japan). The samples were machined into 30 × 27 × 5 mm as the disk specimens. The samples were tested on the plane normal to the hot-press direction. Wear tests were carried out under unlubricated and distilled water at room temperature (25 ± 3 °C). Sliding conditions in dry sliding condition were set at 4.9 N normal load, 20 mm sliding diameter, 0.18 m s^{-1} sliding speed and sliding distances of 108 m. Sliding conditions under distilled water were set at 24.5 N normal load, 20 mm sliding diameter, 0.18 m s^{-1} sliding speed and sliding distances of 10000 m. The friction force and applied force were measured continuously during each test (Fig. 2).

Stribeck curve behavior was investigated for self-mated pairs using the pin-on-disk apparatus as described above. The disk specimen had the same dimensions as above, but the counter material in this case was a 3 mm diameter pin which was fabricated from the material of the same composition by hot pressing. Prior to collecting data for the Stribeck curve analysis, the pin and disk were subjected to sliding at a 24.5 N load and 0.011 m s^{-1} sliding speed for more than 12 h in order to obtain real face-to-face contact. The sliding speed was then changed to 1.08 m s^{-1} and friction measurements were commenced. Friction measurements were recorded continuously throughout the test at different sliding speeds. For each condition, the friction coefficient was measured for 1 h of sliding, and the average value was used for Stribeck analysis.

Results and Discussions

Fig. 3 shows the typical microstructures of the fabricated composites and the Si_3N_4 ceramics (plasma etched surfaces). The matrix silicon nitride microstructure of all the specimens was bimodal, and no significant difference in the grain size and aspect ratio of the elongated grains was observed. Each solid lubricant was dispersed in the matrix homogeneously.

Fig. 4 shows the typical friction behaviors of Si_3N_4 and Si_3N_4 with solid lubricant dispersed-composites under dry friction. In the silicon nitride/carbon fiber composite,

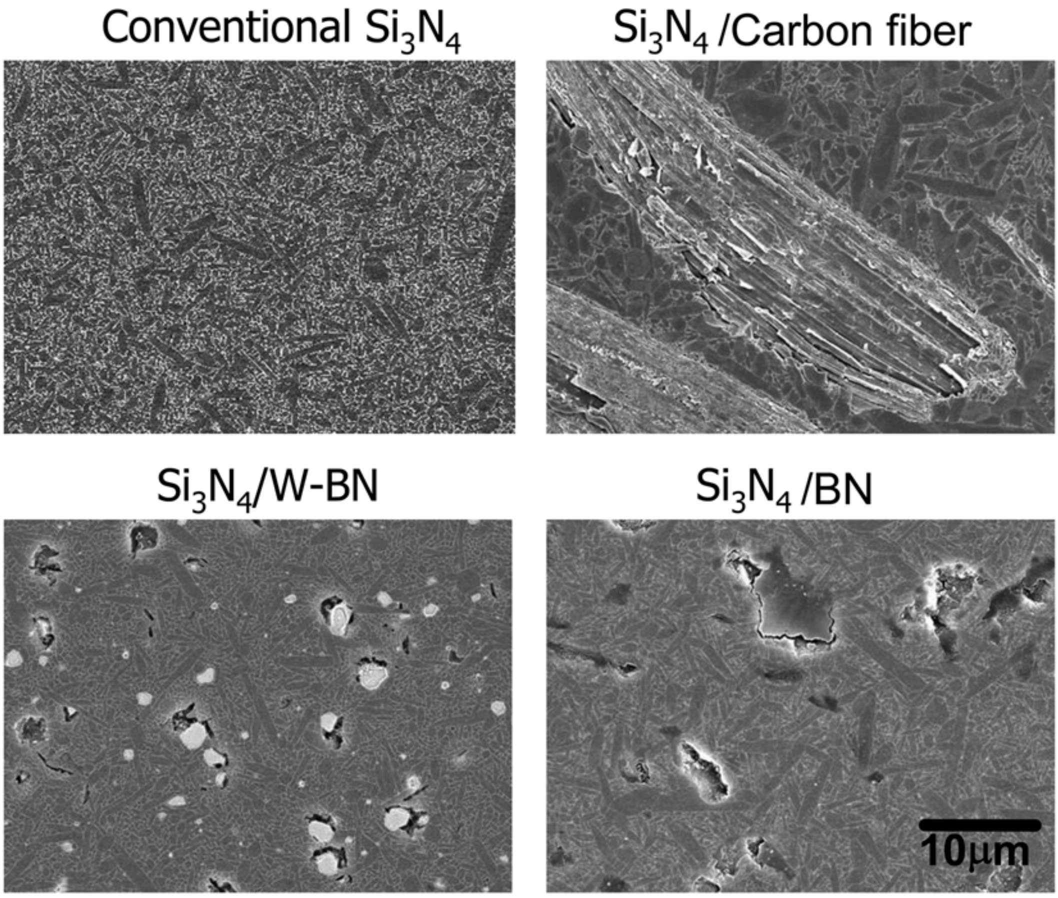

Fig. 3. Typical microstructures of the fabricated composites and the Si_3N_4 ceramics (plasma etched surface).

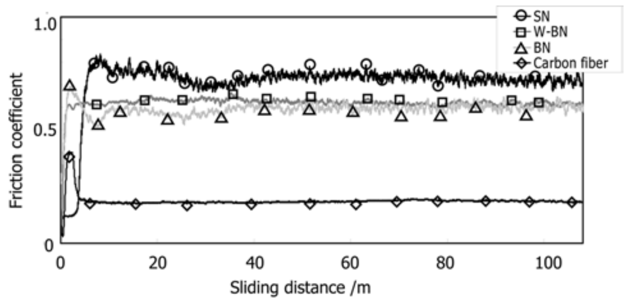

Fig. 4. Typical friction behavior of Si_3N_4 and Si_3N_4 with various solid lubricant dispersions under dry friction.

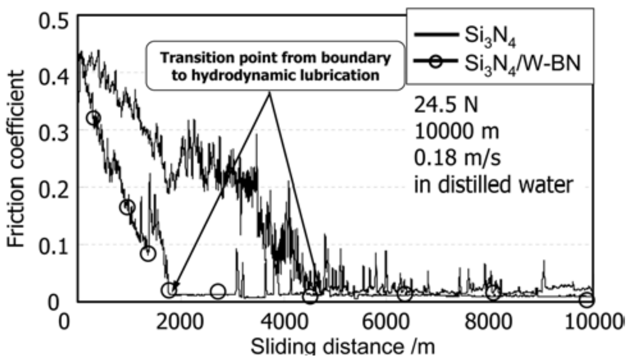

Fig. 5. Typical friction behavior of Si_3N_4 and Si_3N_4 with a solid lubricant (W-BN) dispersion under water lubrication.

the friction coefficient indicated a lower value than the other specimens. On the other hand, in BN and W-BN dispersed composites, the friction coefficient was also reduced. However, the reducing rate was not so high compared with silicon nitride/carbon fiber composite. This result indicates the mechanism of reduction in dry friction is different between the silicon nitride/carbon fiber composite and other composites. The solid lubricant effect and the mechanism of reduction of the dry friction of the fabricated composites has been reported in detail [8, 17].

Fig. 5 shows the typical friction behavior of Si_3N_4 and Si_3N_4 with a solid lubricant (W-BN) dispersion. For both materials, the friction coefficient indicated a high value in the initial stage of the sliding test, and then decreased with an increase in the sliding distance due to a decrease in the apparent normal pressure due to wear of the plate and ball (hereafter the running in region). The friction coefficient then maintained a low value (steady state region). This behavior was also reported in our previous research in the case of the Si_3N_4/carbon fiber composite in detail [15]. This tendency was the same regardless of the type of solid lubricant.

Fig. 6 shows the running in distance until steady state friction of the Si_3N_4 and the various Si_3N_4/ solid lubricant composites. The running in distance in every solid lubricant dispersed composite was reduced compared with the conventional silicon nitride ceramics (solid lubricant content

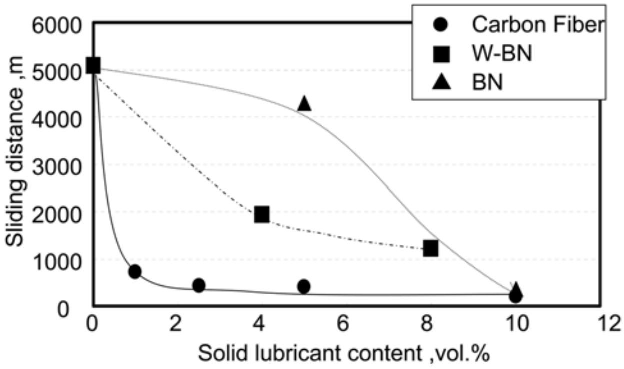

Fig. 6. The running in distance until steady state friction of the Si_3N_4 and the various Si_3N_4/solid lubricant composites.

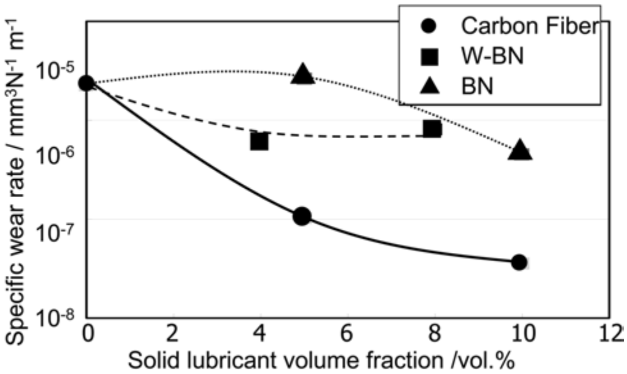

Fig. 7. Specific wear rate of the composites under water lubrication (Total wear of the ball and disk).

0vol.%). This result indicates that the addition of a solid lubricant is an effective method for reducing the running in distance. On the other hand, reducing the effect of the running in distance was different from the solid lubricant species.

Fig. 7 shows the specific wear rate of the composites under water lubrication (Total wear of the ball and disk). The specific wear rate of the specimens shows the same trend of the running in distance because solid-solid contacts, which indicate a high wear rate, are occurring in the running-in regime.

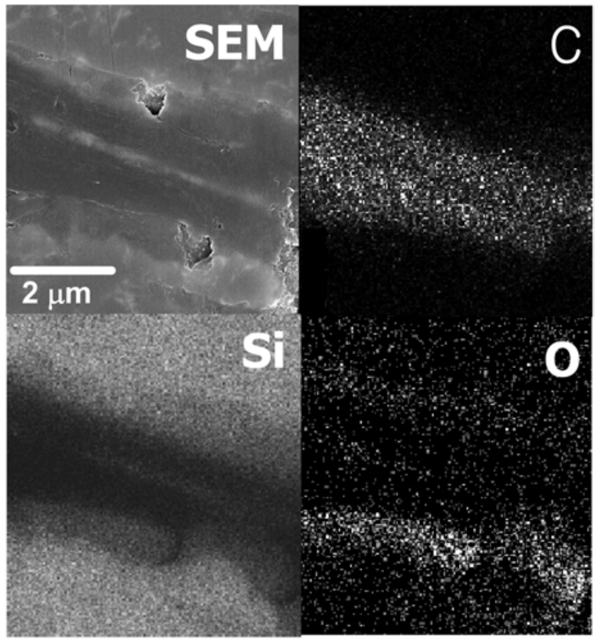

Fig. 9. EDS analysis result of a carbon short fiber on the composite surface after a water lubrication test.

Fig. 8 shows SEM images of the worn surfaces after tribological tests of Si_3N_4 and Si_3N_4 with solid lubricant dispersed composites under water lubrication. In low magnification images, the width of the worn tracks of the Si_3N_4 ceramics was obviously greater than those of the composites. In addition, many scratch traces were observed inside the worn track for the Si_3N_4 sample. These scratch traces indicate that abrasive wear occurred under this sliding condition. Because of this abrasive wear, silicon nitride indicates the high wear rate under a severe test condition. On the other hand, In the case of the specimens dispersed solid-lubricant, any grain pullout and any scratch traces were not observed at the high magnification images. However, the characteristics of the worn surfaces were slightly different.

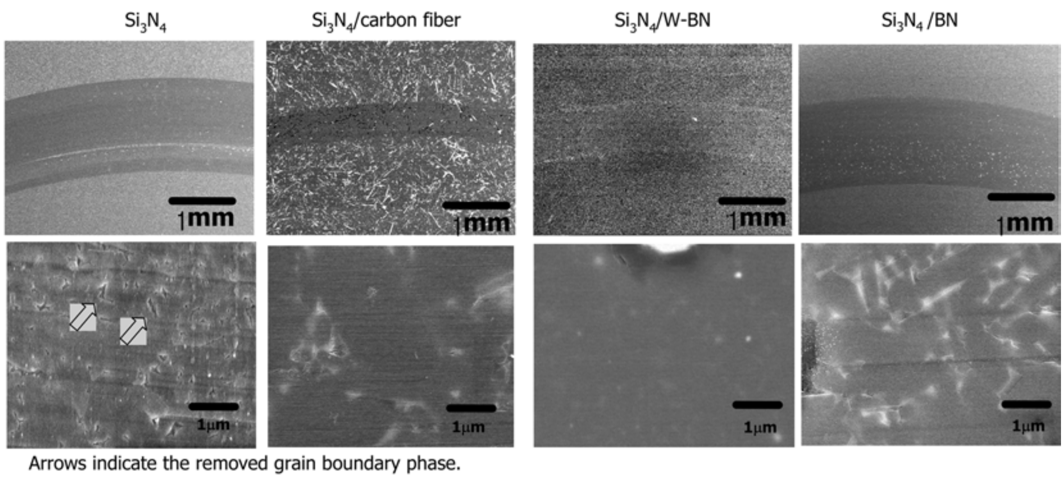

Fig. 8. SEM images of the worn surfaces after tribological tests of Si_3N_4 and Si_3N_4 with solid lubricant dispersed composites under water lubrication.

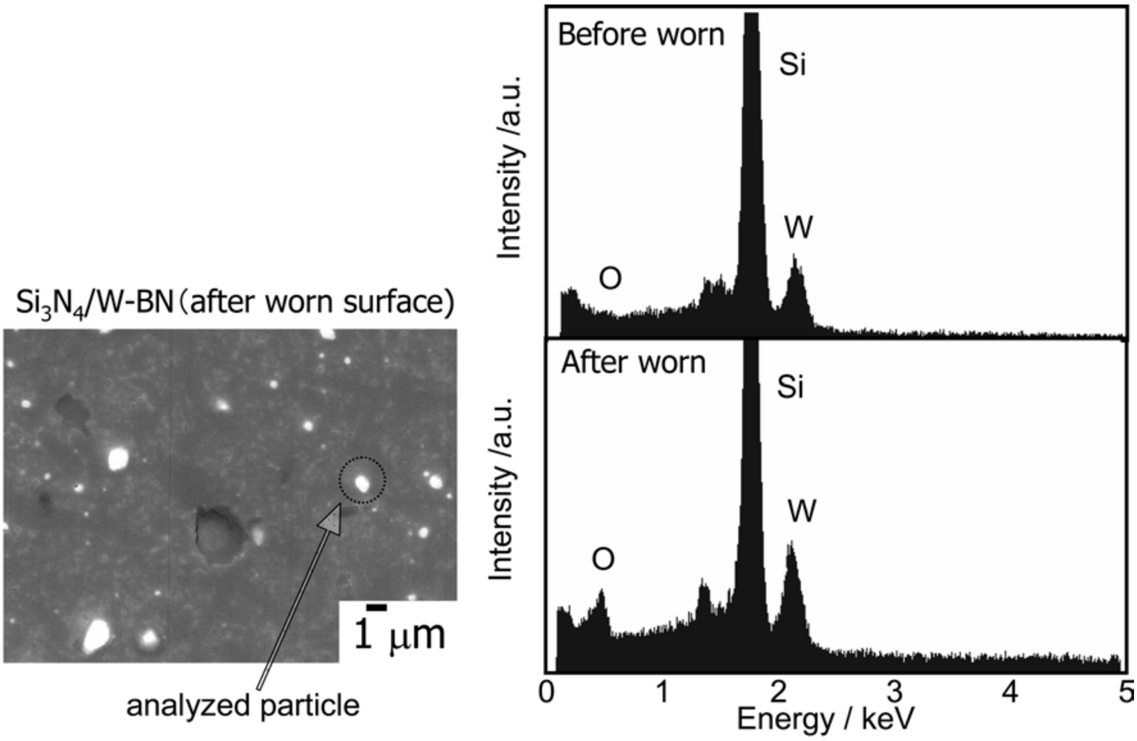

Fig. 10. EDS analysis result of the dispersed tungsten on the composite surface after a water lubrication test.

Fig. 9 shows the EDS analysis result of the Si_3N_4/carbon short fiber composite after a water lubrication test. At the surface of the carbon fiber, a white debris could be observed adhered along the long axis direction of the carbon fiber. Because the fiber used in this study was pitch-derived with highly-oriented graphite layers, this debris is considered to be located between the graphite layers. From the EDX analysis, it was confirmed that the debris consisted of Si and O elements, which was formed by a tribo-chemical reaction (Fig. 7) . This Si-O material is effective for friction reduction and is difficult to remove from the worn surface because it was located between the graphite layers.

Fig. 10 shows the EDS analysis result of the Si_3N_4/W-BN composite before and after a water lubrication test. From the comparison of the EDX analysis results between before and after the tribological test, the oxygen content of tungsten-rich particles which were dispersed in the Si_3N_4 matrix was increased after the wear test. It was confirmed that the dispersed W particles consisted of W and O elements, which were formed by a tribo-chemical reaction during the tribological test under water lubrication. Oxidized tungsten has a solid lubricant effect (m = 0.3) the as same as molybdenum oxide. The running in distance reduction is related to the formation of tungsten oxide.

Fig. 11 shows typical Stribeck curve behavior of solid lubricant dispersed composites under water lubricated conditions. Under the fluid lubrication regime, the friction coefficient of all specimens is almost the same. This trend is the same as that observed for the conventional friction test results when sliding against a monolithic Si_3N_4 counterbody.

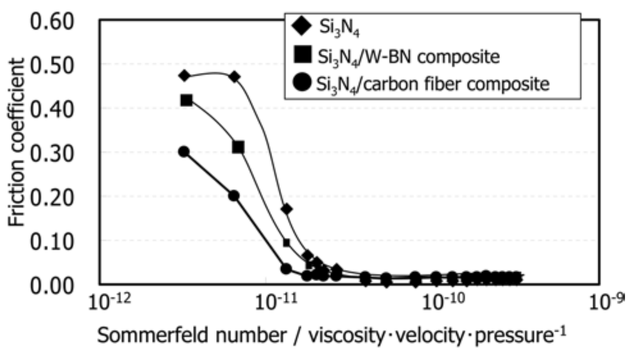

Fig. 11. Stribeck curve behavior of several solid lubricant dispersed composites and conventional Si_3N_4 ceramics under water-lubricated conditions.

However, the transition point from liquid lubrication to a boundary lubrication regime is different. The Si_3N_4/carbon fiber composite maintained a low friction coefficient to more severe wear conditions, such as low speed and high load. The Si_3N_4/W-BN composite also maintained the low friction coefficient to more severe wear conditions. However, this composite a smaller reducing effect comparied with the Si_3N_4/carbon fiber composite. The monolithic Si_3N_4 ceramics however showed an increase in friction coefficient under the same conditions. In addition, the friction coefficient of every composite in the boundary lubrication region was lower than that of the monolithic Si_3N_4 ceramic. These results indicate that the solid lubricant additions to monolithic Si_3N_4 do not reduce the friction coefficient under a hydrodynamic lubrication region, however, the friction coefficient under boundary lubricating conditions was reduced and the transition point from a state of hydrodynamic to boundary lubrication was displaced to the right on the Stribeck curve. In the boundary lubrication regime solid-solid contact partially occurred, dispersed solid lubricant reduces the contact stress due to the solid lubrication effect. Reducing the friction coefficient is effective in reducing the stress during sliding. Slicon nitride ceramics shows a low friction under water lubrication due to the formation of the tribo-reacted Si-O derived film. However, it is easy to remove this from the surface because the applied tensile stress is high during the sliding. On the other hand, the film is effective in reducing the stress to reduce the friction coefficient. Solid lubricant additions to silicon nitride ceramics restrict the removal of the low friction thin film due to the friction reduction. As a result, formed lubricant substrate formed is maintained during the sliding test, and shortens the running in distance. On the other hand, the difference of the friction coefficient in more severe sliding condition is due to the formation of lubricating substrates. In the carbon fiber composite, a lubricating substrate was formed inside the carbon fiber due to the carbon fiber having a layered structure. On the other hand, in the case of the Si_3N_4/W-BN composite, a lubricating substrate was formed on the surface of dispersed W-rich particle. From this result, the lubricating substrate formed between carbon fiber layers was difficult to remove from the surface. As a result, the low friction range is maintained in more severe conditions.

Summary

·ESolid lubricant dispersions in bulk Si_3N_4 are an effective method to reduce the running in distance due to the solid lubricant acting to reduce the friction coefficient under high normal pressures corresponding to the boundary lubrication regime.

·EThe efficiency of reducing the running in distance greatly varied with the types of solid lubricants dispersed in the Si_3N_4 matrix.

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