LUBRICATION AT 750 °C IN VACUUM BY A TRANSFER FILM FROM SELF-LUBRICATING COMPOSITES FOR ROLL/SLIDE CONTACT OF Si₃N₄

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SUMMARY
Roll/slide friction tests were carried out at a temperature of 750 °C in vacuum. The disk specimens were made of Si₃N₄ with or without a sputtered MoS₂ film. A pin specimen was rubbed against one disk to supply a lubricating transfer film. When the pin was a MoS₂-based composite, friction coefficient was around 0.3 and almost no wear of the disks was observed after 24 hours operation at a load of 50 N, a rotating speed of 0.5 m/s, and a slip ratio was 10 %. Transferred patchy MoS₂ films were observed on the friction track. When the pin was changed to a Ni-based composite containing BN and graphite, friction increased from 0.2 to 0.7 at a test time of about 8 hours and severe disk wear was found. In an additional test using Si₃N₄ disks with a sputtered MoS₂ film without a pin, friction coefficient was around 0.3, and no wear of the disks was found after 24 hours operation. The appearance of friction track was similar with that in the test using the MoS₂-based composite pin. It seems that the sputtered MoS₂ film wore, but worn particles reattached on the friction path to develop an effective lubricating film. These results demonstrate the effectiveness of transfer film lubrication for a long-term operation in high temperature vacuum, and superior capability of MoS₂ to develop effective transfer film.

Keywords: High temperature, Vacuum, Transfer film lubrication, MoS₂-based composite, Si₃N₄

1 INTRODUCTION
One of the key technologies to realize a high-performance re-usable spacecraft is lubrication at high temperatures. Re-usable spacecrafts will experience very high temperature during re-entry to the earth due to aerodynamic heating. Some mechanical components, such as hinges of aerodynamic control surfaces (flaps), are required to operate at high temperatures in low vacuum for maneuvering spacecrafts. In "hot structure" design concept, i.e. without cooling, the operating temperature was estimated to be as high as 1600 °C.

Woydt et al. [1] assessed some candidate lubrication mechanisms for tribosystems operating at high temperature, and proposed promising substrate/coating systems for hot hinge joints of re-usable vehicles. The development activities of hot hinge joints based on the assessment have been reported [2,3], and poor wear-resistance was pointed out as a problem, although the description about the tribo-coatings and their tribological performance was very limited.

As for lubrication at high temperatures in ultra-high vacuum, Obara & Suzuki reported that long-term operation of Si₃N₄ ball bearings, more than 1400 hours, at 650 °C in vacuum was possible by using a MoS₂-based composite retainer [4]. They thought that the transfer film from the retainer was responsible to the excellent performance.

In this paper, effectiveness of transfer-film lubrication from self-lubricating composites is demonstrated in roll/slide friction configuration at 750 °C in vacuum. Feasibility of applying transfer film lubrication to hot hinge joints of re-usable vehicles is discussed.

2 EXPERIMENTAL DETAILS

2.1 Test Apparatus
A schematic view of a two-disk, roll/slide friction tester used in this study is shown in Fig.1. Figure 2 shows a schematic view of roll/slide test configuration with a pin, and Figure 3 shows a photo of the test section, where the upper furnace wall was not attached.

Two disk specimens were mounted on different shafts, and a pin specimen was rubbed against one disk for transfer film formation. Each shaft is supported by ball bearings incorporated in a magnetic fluid seal, and driven through timing belt by separate stepping motor placed outside the vacuum chamber. One rotating shaft assembly (right side in Fig.1) can rotate freely in the loading direction around support bearings. Vacuum sealing of the shaft assembly was achieved by using a bellows connector. The load between the disks was applied by hanging a dead weight on the shaft assembly at the ambient atmosphere side, as shown in Fig.1. The pin-holder-arm is attached to the vacuum chamber in the same way, and pin/disk load was also applied by a dead weight.

Frictional torque was measured using a torque transducer placed between the motor and the drive shaft of the other rotating shaft assembly (left side in Fig.1). The measured torque was the sum of friction torque of (1) disk/disk contact, (2) pin/disk contact, (3) magnetic fluid seal, (4) rotary-joint for supplying cooling water to the rotating shaft (not shown in the figure), and (5) belt-pulley system and pillow blocks. Friction torque of (1) and (2) were estimated assuming that (3) (4) (5) were constant.
This configuration for the measurement of friction torque of the disk/disk and pin/disk contacts was selected because of necessity of water-cooling of the rotating shafts, although some errors are expected. A furnace with two-piece structure was attached to surround the test section. Tantalum (Ta) wires housed in quartz tubes were used as a heater, and the quartz tubes were mounted on the inside wall of the furnace as shown in Fig.3. The quartz tubes were adopted not only as electrical insulator but also as protection against outgassing contamination from the heater. The furnace wall had three-layer structure made of 304 stainless steel. Temperature was measured using a thermocouple inserted into a 304 steel rod placed at the middle and below of two disks as shown in the lower figure of Fig.1.

2.2 Test Specimens
As shown in Fig.2, one disk specimen was barrel-shaped (rounded disk) and the other was cylindrical shaped (flat disk) to avoid edge loading and to secure the contact with a pre-determined contact pressure. The disk specimens had a diameter of 40 mm and a thickness of 10 mm. The rounded radius for the barrel-shaped specimen was 150 mm.

The tested specimen combinations are listed in Table 1.

![Fig. 1: Schematic view of high temperature, UHV roll/slide friction tester.](image1)

![Fig. 2: Roll /slide test configuration with a pin.](image2)

![Fig. 3: Photo of test section.](image3)
The disk specimens were made of Si₃N₄ with or without a sputtered MoS₂ film. The MoS₂ film was coated using a RF sputtering apparatus, and the thickness was about 0.5 µm. Two types of self-lubricating composites were tested as a pin specimen. One is a MoS₂-based composite (5S), which showed very good performance when rubbed against Si₃N₄ [5] and when used as a retainer of ball bearings in high temperature vacuum [4, 6]. The other is a commercially available Ni-based composite containing BN and graphite as a lubricant (3M1), which showed good tribological performance in high temperature air. The compositions of the composites are shown in Table 1.

<table>
<thead>
<tr>
<th>Disk/disk</th>
<th>Pin</th>
<th>Unit:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si₃N₄/Si₃N₄</td>
<td>None</td>
<td>5S Pin</td>
</tr>
<tr>
<td>Si₃N₄/Si₃N₄</td>
<td>MoS₂ based composite (5S)*</td>
<td></td>
</tr>
<tr>
<td>Si₃N₄+MoS₂/Si₃N₄+MoS₂</td>
<td>Ni-based composite (3M1)*</td>
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<tr>
<td>Si₃N₄+MoS₂/Si₃N₄+MoS₂</td>
<td>MoS₂ based composite (5S)*</td>
<td></td>
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<tr>
<td>Si₃N₄+MoS₂/Si₃N₄+MoS₂</td>
<td>None</td>
<td></td>
</tr>
</tbody>
</table>

* Compositions (wt%) of pin material
5S : 75%MoS₂+10%MoO₂+10%Nb+5%304 Steel
3M1 : 70%(15%CrB+85%Ni)+30%(8%BN+86%Gr)+6%Mica + water glass

Table 1: Tested specimen combinations

Hereafter we call the MoS₂-based composite as 5S, and the Ni-based composite as 3M1. Figure 4 shows the shapes and dimensions of the pin specimens. Both pins had a flat end face with a diameter of 8 mm, and were rubbed against the rounded disk.

2.3 Attentions for experiments in high-temperature vacuum

Material of the disk specimens (Si₃N₄) and the shaft (304 stainless steel) was different, and thus the difference of the thermal expansion ratios was a big problem to attach the specimen firmly to the shaft at high temperature. In this study, configuration with a tapered shaft and a disk specimen having the same tapered inner bore was employed, expecting that the specimens move in the axial direction when the temperature changed. To allow the movement, the disk was mounted on the shaft "softly" by means of spring loading, as shown in Figs.1 and 3. The springs used were coil springs made of Si₃N₄. A commercially available BN spray coating was applied to the shaft surface before mounting the disks to reduce the friction between the disk and the shaft at the tapered surface. Typical relative displacement between the shaft and the disk was about 0.5 mm when the temperature was changed from ambient to 750 °C. This method worked well except for one case where the disk had radial cracks, possibly because the disk could not move in the axial direction due to high friction between the shaft and the disk, causing higher hoop stress.

The disks were fastened to the shaft using a locking nut, as shown in Fig.1. To avoid possible adhesion at the thread interface in high-temperature vacuum environment, the nuts made of molybdenum were used.

2.4 Test Procedure and Conditions

After setting the specimens, the vacuum chamber was evacuated using a turbo molecular pump and a cryopump. Then the test section was heated to 750°C. The typical pressure was in the order of 10⁻⁸ Pa at ambient temperature, and in the order of 10⁻⁶ Pa at 750°C. The main residual gas species detected using a quadrupole mass spectroscopy were water vapor.

First, the pin specimen was loaded against one disk at a load of 5 N and a rotating speed of 0.5 m/s for 24 hours. Then two disk specimens were brought to contact at a load of 50 N and a slip ratio of 10 %. Slip ratio (S) is defined as S = (N₁-N₂)/N₁, where N₁ and N₂ are rotational speeds of two disks, and N₁ > N₂. The test duration was 24 hours, or until friction coefficient exceeded 0.7. The calculated maximum contact stress is 0.67 GPa for the Si₃N₄/Si₃N₄ contact.

3 RESULTS AND DISCUSSIONS

3.1 Tribological performance without lubricant

First, tribological performance without lubricant was examined. A Si₃N₄/Si₃N₄ pair was tested at 750 °Cin vacuum at a load of 50 N, rotating speed of 0.5 m/s, and a slip ratio of 10 %. Figure 5 shows friction trace during the test, photos and surface profiles of the disk surfaces after the test.

Friction coefficient was around 0.7, and it was relatively stable until the test was interrupted at a test time of 15 min. Serious damage was found on both the disk surfaces in the post-test inspections. There were many worn particles on the friction tracks, and the surface was roughened and wear depth was about 5 µm.

This result indicates that the wear occurred from the beginning of the test, and progressed steadily. Worn Si₃N₄ particles or powders were seemed to always exist in the interface, and this might be responsible to the relatively stable friction behavior. This test clearly demonstrated that the Si₃N₄ pair should not be used without lubricant even for short-term operation.
3.2 Lubrication by transfer film

To examine the feasibility of transfer film lubrication in high temperature vacuum, a pin specimen made of self-lubricating composite was first rubbed against one disk to develop a transfer film, and then two disks were brought to contact. The pin was rubbed for 24 hours before the disk/disk contact started, and was continued to rub against the disk throughout the test.

Figures 6 and 7 show the results of tests when the pin was a MoS₂-based composite (5S). In Fig.6, the disks had a sputtered MoS₂ film whereas no coating was applied on the disk in Fig.7. In each figure, change in friction of the disk/disk contact during the test, photographs and surface profiles of the disks and the pin after the test were shown.

When the disks had a sputtered MoS₂ film, Fig.6, friction fluctuated from 0.2 to 0.4, and showed this relatively low value for 24 hours of operation. Post-test inspection of the disk showed that the pre-coated MoS₂ film was almost worn off, but no damage of Si₃N₄ substrate was found. Wear scar of the pin was observed at off-centered position and the wear depth was about 20 µm, as shown in lower figures in Fig.6.
Rubbing of the pin at off-centered position occurred due to mistakes in the tester design to estimate correctly the relative position of the disk and the pin at elevated temperature. Friction coefficient of the pin/disk rubbing was estimated to be 0.25-0.3, although the accuracy is not so good due to the noise of the other torque sources as explained in section 2.1. These results clearly showed that transfer film from the pin and/or the pre-coated MoS2 film worked very well to prevent direct Si3N4/Si3N4 contact, and MoS2 is effective as a lubricant even in high-temperature vacuum.

In the case of Si3N4 disk without coating, Fig.7, friction was stable around 0.3 throughout the test. Friction was much stable compared with the test using Si3N4 disks having a MoS2 film, Fig.6. In the surface profiles, no wear of Si3N4 disks was observed. Bumps were found in the peripheral of the friction track, probably the transferred material from the pin.

The appearance of the pin surface was very different from that shown in Fig.6. Several wear scars were found around the center of the pin. The shape of each wear scar coincided with the shape of the mating disk with the rounded radius of 150mm, as shown in lower figure of Fig.7. Considering that the rubbing point of the pin was off-centered as described above, this observation suggests that the pin sometimes rotated during the test changing its rubbing surface. The pin specimen was fixed to a pin holder by fastening a threaded cap made of 304 stainless steel, and the difference in thermal expansion coefficient might cause loosening of the pin from the holder. The pin seemed to be rotated by the friction force at the pin/disk interface, which worked at the off-centered position of the pin. Total wear amount of the pin estimated from the photos and surface profile was much greater than in the test of Si3N4 disks having a MoS2 film. Existence of a pre-coated sputtered MoS2 film was confirmed to be very effective to reduce the wear of the pin. Friction coefficient of the pin/disk interface was estimated to be about 0.3, almost the same as that against Si3N4 disk having a MoS2 film. The existence of a pre-coated sputtered MoS2 film had little effect on friction behavior.

This experiment demonstrated that transfer film formed from the MoS2-based composite was effective to lubricate Si3N4 disks at high-temperature vacuum. However, the transfer film was not clearly identified in the photos and surface profiles. Figure 8 shows SEM photographs of the friction track of Si3N4 disk without a pre-coated MoS2 film. White-colored "islands" elongated in the sliding direction were observed, and EPMA analysis revealed they were MoS2. Only a part of the surface was covered with the thin MoS2 films distributed like as islands. This indicates that existence of a very small amount of MoS2 at the real contact area in the roll/slide interface was enough to show good tribological performance.

Fig. 8: SEM image and EPMA analysis of disk surface after the test using a MoS2-based composite pin and Si3N4 disks without pre-coating.
Figure 9 shows the results of the test using a Ni-based composite pin containing BN and graphite as a lubricant (3M1). Friction coefficient was about 0.2 in the initial stage of the testing, which was lower than that using the MoS2-based composite. However, friction began to increase at a test time of about 200 min and finally went up to 0.7, the value was the same as Si3N4/Si3N4 combination without lubricant, suggesting that Si3N4/Si3N4 direct contact had occurred. This was confirmed by the observation of the disk surface after the test. The disks suffered serious damage, and the appearance of surface profiles was very similar with that of non-lubricated case, Fig.5.

The friction coefficient of the pin/disk interface was 0.25-0.3, higher than that of the disk/disk interface. Relatively a large amount of wear was observed on the pin surface, which had the worn shape with good conformity to the rounded disk, indicating enough amount of lubricant was supplied to the friction interface. A groove was found in the center region of the pin, as shown in Fig.9. It seems that worn Si3N4 particles entered into the pin/disk interface and wore the pin in the last stage of the test.

The more important aspect in transfer film lubrication seems to be the capability to replenish a lubricating film on the contact area where the lubricant film had just worn off. "In situ" repair or formation of a lubricating film by roll/slide action at the disk/disk interface is essential for long-term, stable operation.

3.3 Lubrication by pre-coated MoS2 film

To compare the effectiveness between lubrication by a pre-coated MoS2 film and transfer film lubrication, roll/slide friction test using Si3N4+MoS2/Si3N4+MoS2 combination without a pin was carried out. The intention of this test was to know the limitation of lubrication by a pre-coated MoS2 film, but the result was beyond the intention.

![Fig. 10: SEM image and EPMA analysis of disk surface after the test using a Ni-based composite pin and Si3N4 disks without pre-coating.](image)

In this test, transfer film failed to lubricate Si3N4/Si3N4 pair in a relatively short term, presumably because the wear rate of the transfer film was higher than the formation rate. Figure 10 shows a SEM photograph of the friction track and EPMA analysis result. White-colored material elongated in the sliding direction was observed on the wear scar as the test using the MoS2 pin. The area analysis of Ni by EPMA confirms that the white islands were transferred films from the pin. Compared with Fig.8 for the test using the MoS2 composite pin, the transfer films covered a larger area of wear scar, although it did not result in longer life.

These results indicate that supply of a larger amount of lubricant by sliding action at the pin/disk interface was not enough for the good tribological performance. The
the area other than "islands". The appearance of the surface indicates that the white-colored islands were not a part of the pre-coated MoS$_2$ film still remained, but re-attached films or transfer films.

Interesting finding was that many cracks were found in the pre-coated MoS$_2$ film in un-rubbed region, as shown in Fig.12 (c). These cracks were caused by the difference in thermal expansion ratios between MoS$_2$ and the substrate Si$_3$N$_4$, which resulted in tensile stress in the MoS$_2$ film. The cracked films seem to easily peel off, and this may provide good source of transfer film.

Friction behavior and the observations of the surfaces suggest that the pre-coated MoS$_2$ film was completely worn off in the initial stage of the test, but the worn particles became the source of the transfer film for the remaining test period. The long-term, good tribological performance seemed to be achieved by transfer film lubrication where the source of the transfer film was the worn-off or peeled-off MoS$_2$ film. Friction coefficient was about 0.2 for the pre-coated MoS$_2$ film, and it was 0.3 for transfer film lubrication. This suggests that the nature of the transfer film was different from the pre-coated sputtered MoS$_2$ film.

4 CONCLUDING REMARKS

Effectiveness of transfer film lubrication with MoS2 was demonstrated at 750 °C in vacuum. This seems to be achieved by the superior capability of MoS2 to develop effective transfer films. A Ni-based composite containing BN and graphite as a lubricant tested in this study formed a low-friction transfer film but film-replenishing ability was not good, resulting in shorter life. These results indicate that "in situ" repair or formation of a lubricating film at the friction interface, combined with enough supply of the lubricant, is essential for long-term, stable operation in transfer film lubrication.

For applications to the hot hinge joints of re-usable space vehicles, the assessment in the simulated operating environment is mandatory. The environment will range from a high-temperature, low-vacuum condition to a medium-temperature, ambient pressure condition. It is well known the adverse effect of oxidation for MoS2, and the existence of oxygen and/or water vapor in the operating environment may pose serious problems even in low vacuum condition.

However, Suzuki recently reported that the existence of a small amount of oxygen or water vapor had a good effect for extending the life of MoS2 film [7]. He thought that the long life was achieved by transfer film lubrication, and the environment affected the nature of the transfer film.

Further experimental studies are needed to examine the feasibility of transfer film lubrication for applications at high temperature and in low vacuum conditions, and to better understand mechanisms of transfer film lubrication in the environment containing a small amount of oxygen and/or water vapor.

5 REFERENCES
