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Novel Method for Direct Observation of Friction Interfaces between SUJ2 Ball and Si₃N₄ Thin Film Using Scanning Electron Microscopy

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Abstract

In this study, a novel method was developed for *in situ* scanning electron microscopy (SEM) observations of friction interfaces, from a top view, using a Si₃N₄ thin film (SiN film), which has high electron transmission ability, and a microtribometer. Nanodiamond (ND) aggregates were adsorbed on the back surface of the SiN film. A JIS-SUS304 ball, on which surface ND aggregates were adsorbed and tribofilms were already formed, contacted and slid with the back surface of the SiN film. An SEM electron beam went through the SiN film, and generated secondary electrons from the ball surface contacting the back surface of the SiN film and the adsorbed ND aggregates. Thereafter, the generated secondary electrons from the ball surface penetrated again through the SiN film and reached the SEM electron detector. In other words, the contacting ball surface and the adsorbed ND aggregates were successfully imaged through the SiN film. Energy dispersive X-ray spectroscopy analyses of the friction interfaces were also accomplished. Moreover, *in situ* SEM observations of friction interfaces under boundary lubrication, using poly-alpha olefin (PAO) oil with graphene oxide aggregates and lithium grease with MoS₂ particles, were successfully accomplished. The PAO oil and lithium grease had electron transmission ability, and the friction interfaces were imaged by SEM as seen with an optical microscope.

Keywords

friction interfaces, boundary lubrication, scanning electron microscopy, *in situ* observation, contact surfaces

1 Introduction

Friction interfaces are those in which two surfaces contact and slide against each other. Complex nanosized or/and micro-sized dynamic phenomena, such as wear [1–4], tribofilm formations [5,6], and chemical reactions [7–10], occur in friction interfaces. However, it is difficult to understand the details of the complex friction phenomena in friction interfaces, because *in situ* direct observations and analyses of the friction interfaces with submicron order have not been performed. Observation of frictional interfaces has been carried out by optical microscopy [11]. However, optical microscopy does not have enough resolution for investigating the nanosized/micro-sized friction phenomena. Transmission electron microscopy (TEM) can be used to observe friction interfaces with nano/micro levels. However, TEM obtains only side views of friction interfaces [12,13]. Because friction interfaces are originally two-dimensional, the side-view observations are not enough to elucidate the friction phenomenon occurring in friction interfaces. Observations of friction interfaces using

high-resolution electron microscopy, from the top view, are necessary. *In situ* observations of friction tests have been carried out by scanning electron microscopy (SEM) [14]. However, because SEM obtains only surface images, current observation methods using SEM cannot obtain friction interfaces from a top view.

In this study, a novel method for *in situ* SEM observations of friction interfaces, from a top view, was developed using an electron transparent film and a microtribometer. A steel ball contacted the back surface of the electron transparent film. Nanodiamond (ND) aggregates were adsorbed on both the ball surface and back surface. *In situ* observations of the friction interface between the ball surface and back surface were carried out through the electron transparent film using SEM. Analyses by energy dispersive X-ray spectroscopy (EDS) were also conducted through the electron transparent film. Moreover, observations of friction interfaces under boundary lubrications were attempted, using poly-alpha olefin (PAO) oil with graphene oxide aggregates and lithium grease with MoS₂ particles.

Fig. 1 Concept of the *in situ* SEM observation of friction interfaces

2 Microtribometer in SEM

Si₃N₄ thin films (SiN film) with thicknesses of 50 nm (SN100-A50 Q05, EM JAPAN, Japan) and 200 nm (4120SN-BA, ALLIANCE Biosystems, Japan), and a window size of 0.5 × 0.5 mm were used as the electron transparent films. Surface roughnesses (R_{max}) of the SiN films were measured to be approximately 2.5 nm by atomic force microscopy. JIS-SUJ2 bearing balls (G20 grade, Surface roughness (R_{max}) of less than 40 nm) with diameters of 0.5 and 1 mm were used as the sliding balls. Wear of the SiN films and SUJ2 ball was not observed in this study, which would come from a little of friction cycles in this study. Figure 1 shows the concept of the *in situ* SEM observations of friction interfaces, from the top view, using a SiN film and a microtribometer. The sliding ball contacted the SiN film. A load was applied by a microtribometer in micro- and milli-newton levels. An SEM electron beam went through the SiN film and collided with the ball surface. Thereafter, secondary electrons generated from the ball surface, penetrated the SiN film and reached the SEM electron detector. Thus, the ball surface, which contacted the SiN film surface, was imaged. In other words, the SEM image of a friction interface between the back surface of the SiN film and the ball surface could be obtained. On the ball surface, not only secondary electrons but also characteristic X-ray were generated. In addition, the incident electrons were scattered on the ball surface. The characteristic X-ray and backscattered electrons also penetrated the SiN film and were detected. In this study, secondary electrons were detected for imaging.

Figure 2 shows a schematic drawing and photos of the developed microtribometer operating in SEM. The microtribometer used a cantilever and an optical lever system to measure the adhesion and friction forces. This was the same force measurement system as that of conventional atomic force microscopy. A laser diode and quadrant photodiode were located outside the SEM vacuum chamber to enable the easy adjustment of the laser line. The sliding ball was attached to a cantilever, which was moved to three axes by amplified piezoelectric actuators (PZT). A SEM (JSM-IT100, JEOL, Japan), which had a thermionic-emission gun using a hairpin style tungsten wire, was used in this study. A vacuum pressure in the SEM was approximately 10⁻⁵ Pa and stable during friction tests.

(a) Schematic drawing

(b) Photos

Fig. 2 Schematic drawing and photos of the developed microtribometer operating in SEM

3 Results and discussion

3.1 Observation of friction interface under dry lubrication

NDs have friction and wear reduction properties [15]. A SiN film with a thickness of 50 nm was used. An ND water dispersion was dropped on the back surface of the SiN film, followed by drying in air. The NDs were aggregated on the surface. Thus, the SiN film with the ND aggregates was prepared. A sliding ball with the diameter of 1 mm was used. The ball was already used for several friction tests using the same SiN film, with the same frictional condition. During the prior friction tests, on the ball surface, ND aggregates were adsorbed and tribofilms were formed from NDs. Figure 3(a) shows the SEM image before the SUJ2 ball contacted the SiN film. White dots shown in the SEM image before the ball contact

are the ND aggregates adsorbed on the back surface of the SiN film, which indicates that ND aggregates were imaged through the SiN film. Because the ball surface was approximately 200 μm away from the back surface of the SiN film at this time, the ball surface was not imaged. Representative white dots in Fig. 3(a) are surrounded by the thin-line circles. The SEM image after the ball contacted with the SiN film and before frictions is shown in Fig. 3(b). As mentioned before, on the ball surface, already, ND aggregates were adsorbed and tribofilms were formed. The applied load was 9 μN , and the adhesion force was measured to be 0.2 μN . Thus, the maximum Hertzian contact pressure was calculated to be approximately 50 MPa. A calculated circular contact area diameter was 0.6 μm . In addition to the ND aggregates adsorbed on the back surface of the SiN film, ND aggregates adsorbed on the ball surface (except for the white dots with the thin-line circles) were imaged. ND aggregates adsorbed on the ball surface were imaged though the SiN film. It could be seen whether ND aggregates were adsorbed to the back surface of the SiN film or the ball surface by comparing the

SEM images before and after contact. Moreover, SEM movies helped to find out where ND aggregates were adsorbed. White dots moving with the ball surface were adsorbed on the ball surface. In addition to the white dots, black features were also observed, and they were tribofilms formed on the ball surface. Figure 3(c) shows the SEM image after the frictions of 85 cycles. The sliding width was approximately 200 μm , and the sliding frequency was 10 s. The friction force was measured to be approximately 3 μN and stable during the frictions. The colors of the circles are explained later.

On comparison of the SEM images before (Fig. 3 (b)) and after (Fig. 3 (c)) the frictions, three types of variation of the white dots in the SEM images were recognized. One was that white dots existing before the friction disappeared after the frictions. Representative white dots were surrounded by thin- and thick-line red circles in the SEM image before the frictions, and the red circles in the SEM image after the frictions were also drawn in the same position as in the SEM image before the frictions. This type of white dot was seen on both the back surface of the

Fig. 3 SEM images of the friction interface between the ball surface and the back surface of the SiN film: (a) before the SUJ2 ball contacted the SiN film, (b) after the ball contacted the SiN film and before frictions, and (c) after the frictions of 85 cycles

SiN film and ball surface, and the ND aggregates were swept by the frictions. The second was that white dots appeared after the frictions. Thick-line yellow circles in the SEM image after the frictions surrounded this type of white spot, and the yellow circles in the SEM image before the friction were also shown in the same position as in the SEM image after the frictions. The second type of white dot (surrounded by the thick-line yellow circles) existed only on the ball surface. The third was that the positions and shapes of white dots were changed after the frictions. These white dots were surrounded by thick-line green circles. The third type of white dot was ND aggregates, which were affected by the frictions. The second and third types of white dots were ND aggregates which were transferred and transformed, respectively, and they were only observed on the ND aggregates adsorbed on the ball surface.

Representative ND aggregates adsorbed on the back surface of the SiN film are surrounded by the thin-line circles in Figs. 3(a)-(c). The ND aggregates with the thin-line red circles were swept after the frictions, and the ND aggregates with the thin-line blue circles were stable during the frictions. Although some stable ND aggregates adsorbed on the SiN film (indicated by green arrows) existed, the ND aggregates with the blue and red circles were divided by a purple line, as shown in Figs. 3(b) and (c). It appears likely that an area to the right from the purple line was where many ND aggregates adsorbed on the SiN film were swept by the friction. The right area was much wider than the calculated circular contact area (the diameter was 0.6 μm). As discussed below (in Fig. 4), the tribofilms on the ball surface swept the ND aggregates on the SiN film surface. The tribofilms would be protruded from the ball surface. The heights of the large ND aggregates on the ball surface would be a micron-order. The ND aggregates on the SiN film surface were protruded from the SiN film surface. Therefore, it is very likely that, in the right area, the ND aggregates on the SiN film contacted with the tribofilms, the ND aggregates on the ball surface, and the ball surface, and were swept by them. In the present, contact areas are not recognized only by SEM images, and are supposed by transitions of SEM images by frictions and

contacts.

The SEM images shown in Fig. 3 were still images, and a movie was taken during the frictions. Figure 4 shows a series of capture images of the SEM movie obtained in the friction interface between the back surface of the SiN film and the ball surface. A series of capture images of SEM movies were obtained at a time interval of 1/15 s. In the SEM image at $t = 0.80$ s, a blue arrow indicates the sliding direction of the ball, a white dot in the center of a red circle was the ND aggregate adsorbed on the SiN film surface, and a black feature indicated by a red arrow was a tribofilm formed on the ball surface. At $t = 0.93$ s, the black feature was close to the white dot. Although the position of the white dot was not changed, its brightness decreased. At $t = 1.07$ s, the position of the black feature was closer to the white dot, and the brightness became darker. Finally, at $t = 1.67$ s, the black feature moved out of the image, and the white dot on the film disappeared. Because the black feature is more elevated than elsewhere, the black feature (tribofilm) swept the white dots (ND aggregates).

Figure 5 shows a series of capture images of the SEM movie obtained in the friction interface, in the same position as in Fig. 4. In the SEM movie image at $t = 9.07$ s, a white dot surrounded by a yellow circle was an ND aggregate adsorbed on the tribofilm (black feature) formed on the ball surface. This was because the white dot moved with the black feature. In other words, the relative positions of the white dot and the black feature did not change before $t = 0.80$ s. The black feature indicated by a red arrow was the same as that in Fig. 4. At $t = 9.60$ s, the brightness of the white dot decreased, and the relative position did not change. At $t = 10.07$ s, the relative position was slightly changed. Finally, at $t = 10.73$ s, the ND aggregate was transferred on the SiN film. After several frictions, the transferred ND aggregate was finally swept by the tribofilm (not shown). The adsorption force of the ND aggregate on the SiN film was weak. Thus, ND aggregates on the SiN film were swept by the frictions, as shown in Fig. 3(c).

The SEM image and EDS data of the friction interfaces are shown in Fig. 6, at a similar position as that in Fig. 3. The EDS

Fig. 4 Series of SEM movie capture images of the friction interface between the back surface of the SiN film and the ball surface. The red circle indicates the ND aggregate which was adsorbed on the SiN film surface and swept by the friction. The blue arrow indicates the sliding direction of the ball. The red arrow indicates the tribofilm formed on the ball surface.

Fig. 5 Series of the SEM movie capture images of the friction, in the same position as in Fig. 6. The yellow circle indicates the ND aggregate which was adsorbed on the tribofilm and then transferred on the SiN film surface. The blue arrow indicates the sliding direction of the ball. The red arrow indicates the tribofilm formed on the ball surface.

Fig. 6 SEM image and EDS spectra of the friction interface (at a near position as that in Fig. 3)

spectra were obtained in a black feature (001), white dot (002), and a place without them (003). Since the EDS spectra shown in Fig. 6 were obtained through SiN films, the EDS spectra had Si K_{α} peaks. The EDS spectrum of the black feature (001) had a carbon peak, which indicates that the tribofilm on the ball surface formed from NDs. The EDS spectrum of the white dot (002) also had a carbon peak. This supports that the white dots observed in SEM images were NDs (aggregates of NDs). There is no carbon peak in the place (003), where the black features and white dots were not existed. Since the ball was contacted with the back surface of the SiN film, Fe K_{α} peaks were observed in the all EDS spectra and Fe L_{α} peaks existed in the places (002) and (003). The EDS spectrum of the place (001) had a very small Fe L_{α} peak (0.705 keV). It is possible that since the X-ray energy of the Fe L_{α} peak is low and the tribofilm was formed on the ball surface, the X-ray intensity of the Fe L_{α} peak was disturbed and decreased by the tribofilm. The EDS spectra at friction interfaces (through SiN films) can be obtained.

3.2 Observations of friction interfaces under boundary lubrication

PAO oil with graphene oxide (GO) aggregates was used for observation of friction interface under boundary lubrication. A SiN film with a thickness of 200 nm was used. A SUJ2 ball with a diameter of 1 mm was dipped in the PAO oil and contacted with a back surface of a SiN film. Figure 7(a) shows the SEM image of a contact region between the SUJ2 ball and the back surface of the SiN film. The applied load was 0.56 mN. The adhesion force was around 1.3 mN. Thus, the real contact load was approximately 1.9 mN. Thus, the maximum Hertzian contact pressure and circular contact area diameter were calculated to be approximately 0.29 GPa and 3.5 μ m, respectively. The PAO oil was imaged by SEM as seen with an optical microscope. A meniscus of the PAO oil was formed around the contact region. The brightness of the PAO oil decreased from the contact region to the outside. The ball surface near the contact region were observed through the PAO oil. This means that the PAO oil had electron transmission ability. When the thickness of the PAO oil was thin, the electron beam and secondary electrons went through the PAO oil. When its thickness was thick, they were disturbed. GO aggregates in the PAO oil were also imaged.

The imaged GO aggregates would be existed near the back surface of the SiN film. Figure 7(b) shows the SEM image after frictions of approximately 50 cycles. The friction force was stable and measured to be 0.09 mN. A blue arrow indicates the sliding direction of the ball. After the frictions, the PAO oil was spread, the GO aggregates seen before the friction were moved. Moreover, new GO aggregates were appeared. The appeared GO aggregates would have been hidden in the thick PAO oil region (the dark region of the PAO oil) before the frictions.

Moreover, *in situ* SEM observation of friction interface in commercial lithium grease with MoS₂ particles was also

Fig. 7 SEM images of the friction interface in the PAO oil with the GO aggregates, (a) before and (b) after the frictions. The blue arrow indicates the sliding direction.

Fig. 8 SEM images of the friction interface in the lithium grease with MoS₂ particles, (a) before and (b) after the frictions. The blue arrow indicates the sliding direction.

attempted. A SiN film with a thickness of 200 nm and a SUJ2 ball with a diameter of 0.5 mm were used. A small amount of the lithium grease was put on the ball surface, and the ball surface was contacted with the back surface of the SiN film. Figure 8 shows the SEM images of the contact region, using the lithium grease, before and after frictions of approximately 100 cycles. A blue arrow indicates the sliding direction. The applied load and the adhesion force were 0.56 μ N and 0.55 mN, respectively. Thus, the real contact load was approximately 1.1 mN, and the maximum Hertzian contact pressure and circular contact area diameter were calculated to be approximately 0.39 GPa and 2.3 μ m, respectively. The friction force was stable and measured to be 0.07 mN. The lithium grease also had electron transmission ability, and a meniscus of the lithium grease was formed. Before the friction, many white particles (MoS₂ particles) were observed. In addition, EDS spectra of the white particles were measured, and S and Mo peaks were detected. The observed particles decreased, after the frictions. This would be because the MoS₂ particles were hidden in the thick grease region after the frictions. The novel method developed in this study accomplished the observations of the friction interfaces under boundary lubrication.

4 Conclusion

In this study, the *in situ* SEM observation of the friction interface was successfully performed. The SEM images and movies were obtained through the SiN film. The transfers, disappearances, and adsorptions of the ND aggregates in the friction interface were observed. In addition to the observations, analyses of the friction interfaces using EDS were also accomplished. Moreover, the friction interfaces in the PAO oil with the GO aggregates and the lithium grease with MoS₂

particles under boundary lubrication were imaged by SEM. It was found that the PAO oil and the lithium grease had electron transmission ability.

As SEM observations are more convenient than those of TEM, and the tribometer used in this study is sufficiently small to be equipped in conventional SEM. Thus, this method has potential to be widely used in tribology research. Observations of friction interfaces using this method can contribute to clarifying many friction mechanisms.

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