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Short Communication

Water Lubrication of Polysiloxane-Containing Polyimide Coatings on Stainless Steel Substrates

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Abstract

This study investigated the water-lubricated tribological properties of coatings made of a novel polysiloxane-containing polyimide (si-PI) material that was recently developed for the aerospace industry and can be diluted with the harmless and environmentally friendly ethanol or water. The si-PI coatings were deposited on stainless steel (JIS SUS304) substrates at curing temperatures ranging from 160°C to 275°C. Their water lubrication properties were measured by rubbing the coatings against each other in water at room temperature. The coatings exhibited lower friction than conventional polyimide materials, with a minimum friction coefficient of 0.04, which was lower than that of polytetrafluoroethylene (PTFE) measured under the same sliding conditions. Unlike the conventional polyimide, the coatings did not exhibit any obvious wear or damage. The results demonstrate that the si-PI coating is a promising low-friction and highly durable coating for water lubrication.

Keywords

polyimide, polysiloxane, resin coating, water lubrication, wear resistance

1 Introduction

Many commercial lubricants are petroleum-based oils that help reduce friction and protect metal surfaces from tribological failures. However, as the use of electric vehicles increases to reduce CO_2 emissions, the production of gasoline and petroleum-based lubricants will decrease accordingly. In addition, petroleum-based oils can cause environmental pollution when leakage occurs [1]. Therefore, it is necessary to switch to environmentally friendly non-petroleum-based lubricants.

Water has a low impact on the environment and is harmless to the human body. Therefore, it is expected to be an alternative lubricant. However, the use of water as a lubricant has several disadvantages. Because of its low viscosity, the water film between the two friction surfaces is easily destroyed, leading to a rapid increase in friction and wear and premature machine failure. In addition, water corrodes metal surfaces [2]. Therefore, measures are required to maintain the water lubricity and increase the water corrosion resistance of metal surfaces.

Resin coatings are suitable solutions for this purpose. Polytetrafluoroethylene (PTFE) coatings are widely used, for example, for bearings in submersible pumps. Although the PTFE shows a low friction coefficient of approximately 0.04 in water [3], its low wear resistance limits its long-term durability under high wear conditions [4]. The trade-off between low friction and low wear resistance is a common problem in many water-lubricated PTFE-based coatings. In addition, Europe has recently proposed restricting the use of per- and polyfluoroalkyl substances (PFAS) because their degradation products persist in the environment for longer than any other synthetic chemical, raising concerns about bioaccumulation and ecotoxicity [5]. Therefore, the development of alternative materials is desirable.

Polyimide (PI) is a resin material with excellent mechanical, heat resistance, and insulating properties. Polyimide varnishes are widely used to fabricate insulating films in semiconductor devices. However, its relatively high moisture absorption [6] and friction coefficient (approximately 0.3) [7] make it unsuitable as a coating material for water lubrication. Polysiloxane-containing polyimide (si-PI) films have recently been developed in the electronics and aerospace industries to reduce moisture absorption and exploit their excellent mechanical, thermal, and electrical properties. si-PI materials have advantages such as a curing temperature below 200°C, low moisture absorption, and strong adhesion to substrates [6]. These properties suggest that the si-PI film can be used not only for electronics and aerospace applications but also as a coating for water lubrication. However, no studies have reported the tribological properties of si-PI coatings under water lubrication.

Organic solvents, such as N-methyl-2-pyrrolidone (NMP), are usually used to dilute and coat PI-based varnishes. However, in recent years, concerns have been raised regarding their harmfulness, and they have been designated as restricted substances under EU REACH regulations since 2020 [8]. Therefore, the authors focused on a novel si-PI material that can be diluted with the harmless and environmentally friendly ethanol or water, which was recently developed for use in the aerospace industry. In this study, the tribological properties of si-PI coatings cured at various temperatures were investigated under water lubrication. The superiority of the si-PI coating was evaluated by comparing its tribological properties with those of conventional PI materials.

2 Experimental

2.1 Preparation of the si-PI coatings

The substrate was a stainless steel (JIS SUS304) plate of 15 mm × 15 mm × 1 mm with a mirror-polished surface having a surface roughness of 0.016 μmRa. The mirror-polished surface was shot-blasted using white alumina particles with an average diameter of 45–53 μm to increase the surface roughness to 0.45 μmRa and improve the adhesion of the coating. si-PI coatings were fabricated using a dip-coating method, as shown in Fig. 1. A raw si-PI solution (SIL-1400, Starfire System Inc., USA) was mixed with 99.5% ethanol in a 1:1 mass ratio and used as the precursor solution. The substrate was ultrasonically cleaned first in acetone and then in ethanol. To measure the coating thickness, part of the substrate was covered with masking tape before coating. The substrate was then immersed in the precursor solution for 60 s and withdrawn at a constant rate of 10 mm/s. The substrate was placed horizontally and dried at 85°C in air on a heating plate for 2 h. The coated substrate was placed in an electric furnace and cured at temperatures ranging from 160°C to 275°C. The heating rate was set at 1°C/min, and the curing temperature was maintained for 2 h.

2.2 Measurement of properties of the si-PI coating

After the coating, the masking tape on the substrate was removed. A surface profiler (FORMTRACER FTA-S4S3000, Mitutoyo, Japan) was used to measure the step difference between the uncoated and coated areas and determine the coating thickness. The surface roughness of the coating was measured simultaneously. The chemical bonding state of the coating was identified using Fourier-transform infrared spectroscopy (FT-IR, Spotlight 300, PerkinElmer, USA). The hardness of the coating was measured using a nanoindenter (ENT-1100b, Elionix, Japan).

The water-lubricated tribological properties were measured using a home-built ball-on-plate reciprocating tribometer. A schematic drawing of the tribometer was shown in Fig. 2. A si-PI-coated stainless-steel (JIS SUS304) ball with a diameter of 10 mm was used as the counter material. The counter ball was also shot-blasted and covered with a si-PI coating under the same conditions as the substrate coating. Sliding tests were carried out in purified water at an average sliding speed of 20 mm/s and a normal load of 3 N for 1 h. For comparison, the water-lubricated tribological properties of a plate and a ball made of a conventional PI material (Vespel SP-1, DuPont, USA) were also measured under the same conditions as those of the si-PI coating. The surface of the Vespel plate was polished to have a

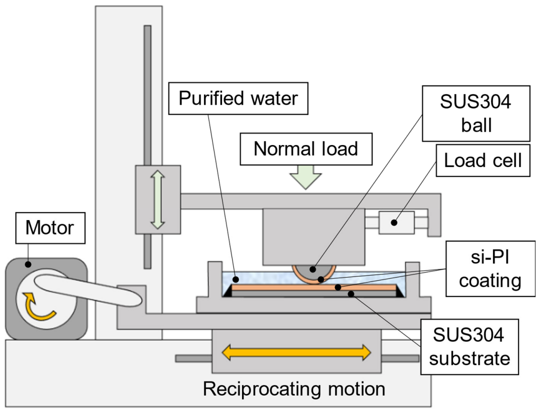

Fig. 2 A schematic drawing of a home-built tribometer used in this study

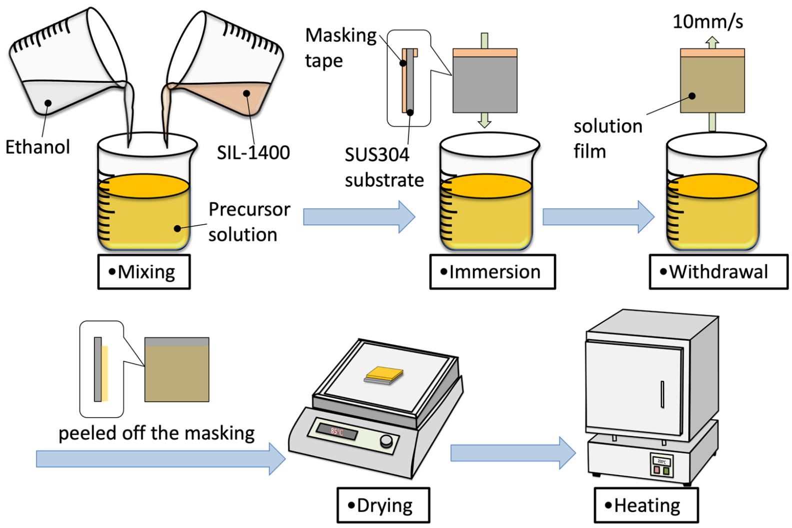

Fig. 1 Dip coating process in this study

surface roughness of 0.069 μmRa (0.092 μmRq), and the surface roughness of the Vespel ball was 0.023 μmRa (0.029 μmRq) .

3 Results and discussion

3.1 Coating properties

The si-PI coatings on the SUS304 substrates cured at temperatures ranging from 160°C to 275°C were shown in Fig. 3. All coatings were uniform at the central region of the substrates where the sliding tests were conducted, and had a certain degree of transparency. Figure 4 shows the dependence of the coating thickness and surface roughness on the curing temperature. The coating thickness was approximately 6–12 μm and slightly decreased with the curing temperature. The surface roughness was 0.07–0.15 μmRa, which was smaller than that of the blasted substrate. Figure 5 shows the FT-IR spectra of the coatings. In the coating cured at 160°C, absorptions at 1700 and 1775 cm^{-1} due to C=O stretching in the imide group and absorption at 1396 cm^{-1} due to C-N stretching in the imide group [9, 10] were observed, indicating that imide bonds were formed even when cured at 160°C. In addition, absorption at 1256 cm^{-1} due to symmetric C-H deformation in Si-CH_3, absorption at 780 cm^{-1} due to stretching of Si-CH_3, and broad absorptions at 1008 cm^{-1} and 1048 cm^{-1} due to vibration of N-Si-O [11, 12] and asymmetric stretching of Si-O-Si [13, 14] were observed, indicating the incorporation of polysiloxane. Although no significant changes in FT-IR spectrum with curing temperature were observed, the relative intensities of the absorption peaks at 1008 cm^{-1} and 1048 cm^{-1} were reversed when the curing temperature was above 260°C. This change is considered to show that when the curing temperature exceeded 260°C, some Si-O-Si bonds were broken and bonded to N in imide groups, resulting in an increase in the proportion of N-Si-O bonds. Figure 6 shows the nanoindentation hardness values of the coatings. No clear correlation with the curing temperature was observed. The average hardness was 0.27 GPa, slightly higher than that of Vespel.

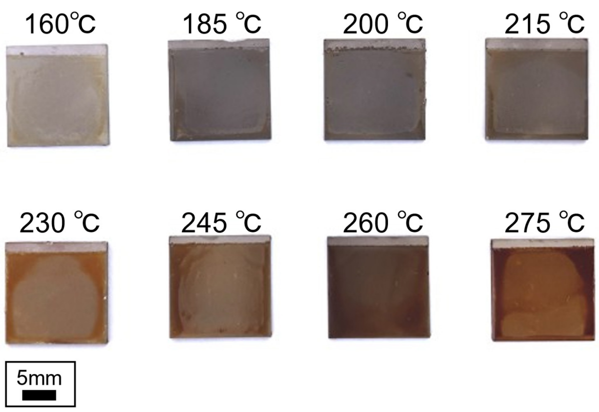

Fig. 3 Photographs of si-PI coatings on SUS304 substrates cured at temperatures ranging from 160°C to 275°C

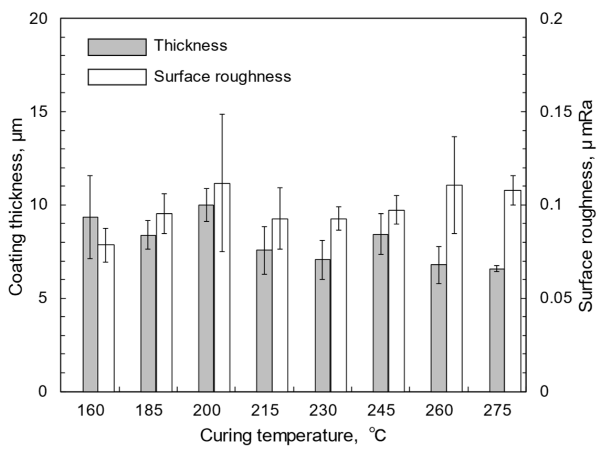

Fig. 4 Curing temperature dependence of the thickness and surface roughness of si-PI coatings

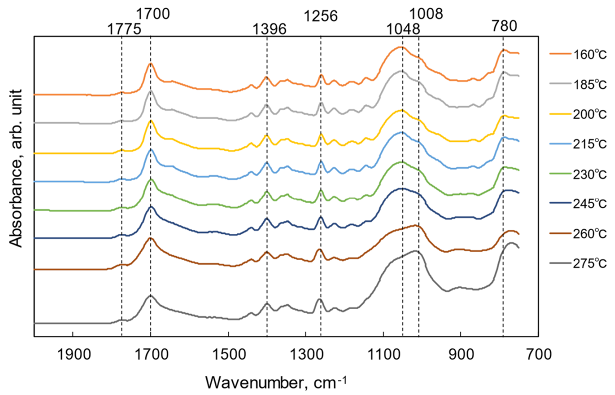

Fig. 5 FT-IR spectra of si-PI coatings cured at a temperature between 160°C and 275°C

3.2 Water lubrication properties

A typical water-lubricated friction property of the si-PI coating cured at 200°C against the si-PI-coated ball is shown in Fig. 7, together with the friction property between the Vespel ball and plate. In both cases, the friction was almost stable after a sliding distance of 30 m; however, the friction coefficient of the coating was lower. Figure 8 (a) shows the friction surfaces of the si-PI coatings on the substrate and ball, and Fig. 8 (b) exhibits the Vespel friction surfaces. The Vespel plate and ball were clearly worn, whereas the si-PI coatings were not worn or damaged, and only a slight deformation was observed on the ball side. To investigate the effect of water affinity on water lubrication properties, the water contact angles were measured

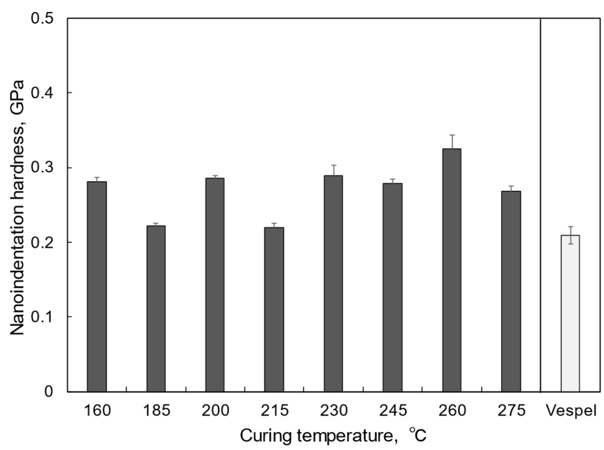

Fig. 6 Nanoindentation hardness of si-PI coatings cured at a temperature between 160°C and 275°C and a conventional PI material (Vespel)

on both the si-PI coating and Vespel plate before and after the sliding tests. Figure 9 shows measurement results. There were no significant difference of the water contact angles after the sliding test.

Figure 10 shows the sliding speed dependence of the average friction coefficient for the last 10 min of the sliding test. As the sliding speed increased, the friction coefficients of both the si-PI coating and Vespel decreased. However, the si-PI coating showed a lower friction coefficient over the entire sliding speed range, and the rate of decrease in friction coefficient with sliding speed was also larger. At a sliding speed of 40 mm/s, the friction coefficient of the si-PI coating was less than half that of the Vespel. Figures 11 (a) and (b) show the friction surfaces of the coatings on the ball and Vespel ball, respectively. Although wear was clearly observed on the Vespel ball at all sliding speeds, no obvious wear or damage were observed on the coating. The same situation was observed for the coatings on the substrate and Vespel plate. Thus, the si-PI coating not only had better friction properties but also a higher wear resistance than the conventional PI material under water lubrication. It was found from Figs. 10 and 11 that in a boundary lubrication regime and a mixed lubrication regime, the si-PI coating exhibited almost no visible wear, whereas the Vespel showed obvious wear. This suggests that adhesion between si-PI coatings with each other could be lower than that of Vespels. The si-PI material contains siloxane (–Si–O–Si–) segments. The incorporation of the siloxane segments into polymer network can reduce surface energy and adhesion, making it low adhesion compared to the Vespel. Thus, it is considered that lower adhesion nature of the si-PI coatings caused lower friction and wear than the Vespels.

Figure 12 summarizes the relationship between the curing temperature and average friction coefficient at a sliding speed of 20 mm/s. In all cases, no obvious wear or damage was observed on the coatings on the ball or substrate. However, in some cases, only deformation was observed on the coating surface on the ball as shown in Fig. 8(a). The friction coefficient had no clear

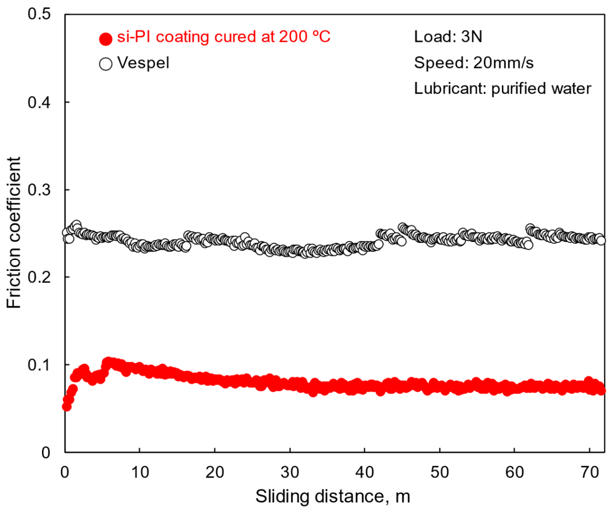

Fig. 7 Water-lubricated friction properties of a si-PI coating cured at 200°C (red filled circle) and conventional PI material (Vespel) (black open circle)

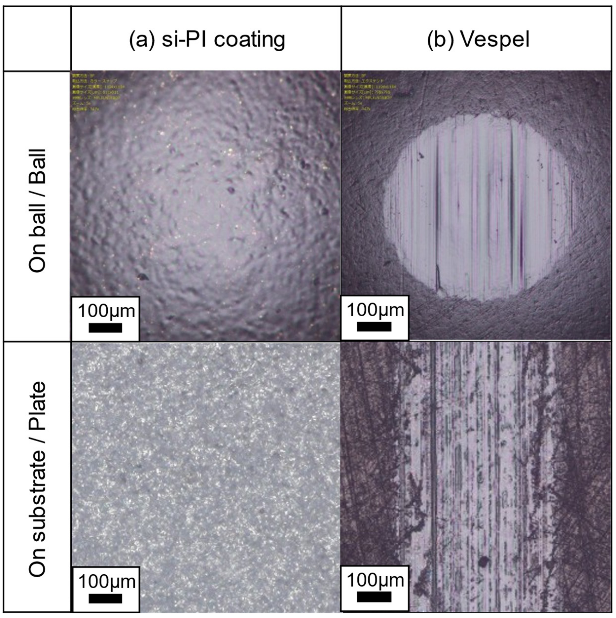

Fig. 8 Optical microscope images of the friction surfaces of si-PI coatings on a ball and substrate (a), and Vespel ball and plate (b)

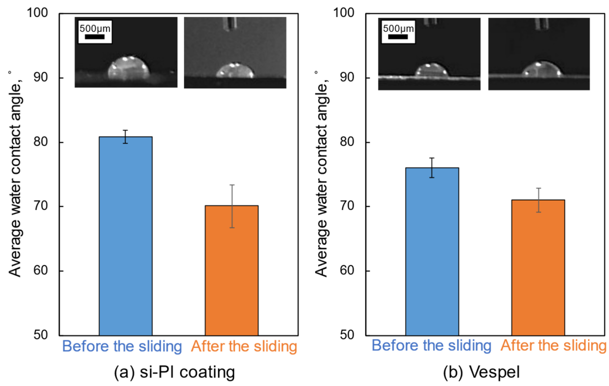

Fig. 9 Water contact angles of the si-PI coating (a) and Vespel (b) before and after the sliding test at 20 mm/s in water

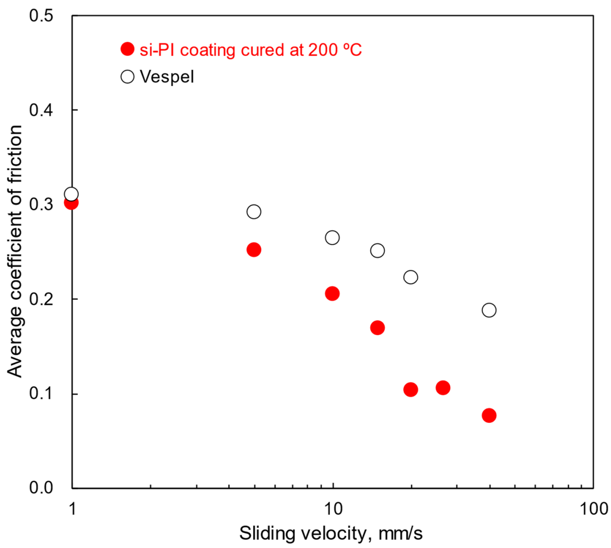

Fig. 10 Sliding speed dependence of the average friction coefficient for the last 10 min of the sliding test in purified water

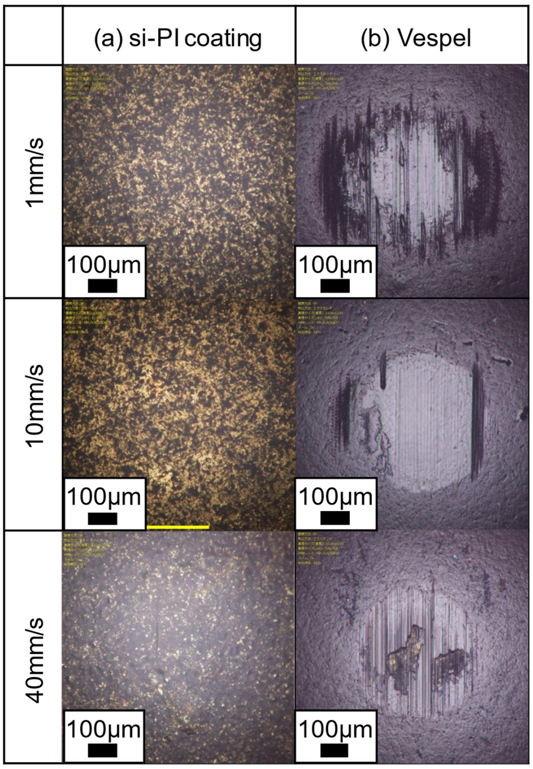

Fig. 11 Optical microscope images of frictional surfaces of the si-PI coating on a ball (a) and Vespel ball (b) after the sliding test at 1, 10, and 40 mm/s in purified water

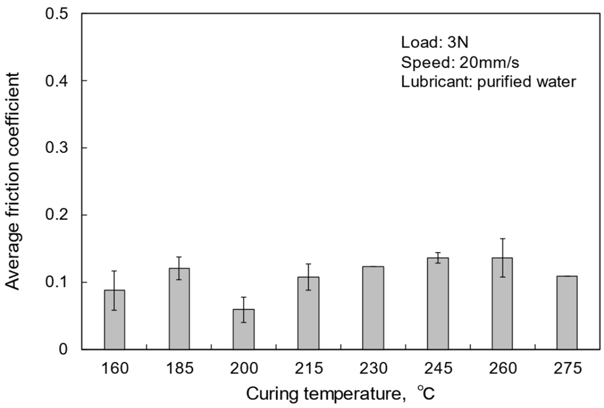

Fig. 12 The relationship between the curing temperature and the average friction coefficient for the last 10 min of the sliding test at a sliding speed of 20 mm/s in purified water

correlation with the curing temperature and exhibited a wide distribution from 0.04 to 0.16. From Fig. 10, it can be considered that the lubrication state at a sliding speed of 20 mm/s was mixed lubrication. Because the surface roughness affects the friction properties under mixed lubrication conditions [15, 16], the relationship between the composite root-mean-square roughness (RMS) and the friction coefficient was investigated. The RMS was defined as follows:

$$RMS = \sqrt{R_{q(sub)}{}^2 + R_{q(ball)}{}^2}$$

where $R_{q(sub)}$ and $R_{q(ball)}$ are the root-mean-square roughness values of the coated substrate and coated ball, respectively.

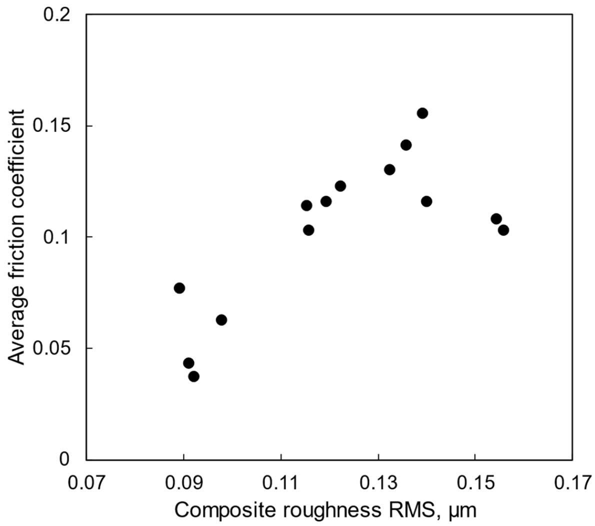

Fig. 13 Correlation of the average friction coefficient and composite root-mean-square roughness between the coatings of the substrate and ball

The results are shown in Fig. 13. A positive correlation was observed, demonstrating that the variation in the friction coefficient was caused by the variation in surface roughness. Therefore, a very low friction and highly wear-resistant coating could be achieved by minimizing the surface roughness of the si-PI coatings.

4 Conclusions

In this study, a novel si-PI coating was fabricated on a stainless-steel substrate (JIS SUS304), and its water-lubricated tribological properties were investigated. The following results were obtained:

- The coating was cured even at 160°C and then imide bonds were formed.
- The friction coefficient in water depended on the surface roughness of the coatings, and the lowest friction coefficient was 0.04, which was lower than that of conventional polyimide materials and also lower than that of PTFEs measured under the same sliding conditions.
- No obvious wear or damage was observed, even under conditions in which the conventional polyimide material showed significant wear.

Thus, the si-PI coating could be cured at temperatures even lower than the typical tempering temperatures for steel materials, showed high wear resistance, and exhibited a friction coefficient lower than that of PTFE under water lubrication. Therefore, the si-PI coating is expected to be promising for water lubrication.

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Appendix – Friction and wear properties between polytetrafluoroethylenes (PTFEs) in water under the same condition as the sliding tests between the si-PI coatings

To compare the water-lubricated tribological properties of PTFEs and si-PI coatings, sliding tests were conducted using a PTFE ball and plate under the same test conditions as used for the si-PI coatings. The surface roughnesses of the PTFE ball and plate were 0.022 μmRa (0.027 μmRq) and 0.062 μmRa (0.079 μmRq), respectively.

The friction property of the PTFEs in water is presented in Fig. A1. The average friction coefficient for the last 10 min of the sliding test was approximately 0.09, which was higher than a minimum friction coefficient of the si-PI coating. Figure A2 shows optical microscope images of friction surfaces of the PTFE plate and ball. Obvious wear was observed on both surfaces, and wear debris were observed around the wear tracks.

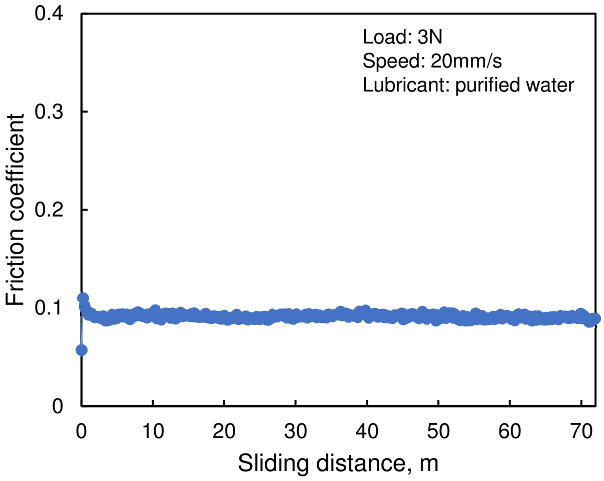

Fig. A1 Water-lubricated friction properties between a PTFE plate and ball

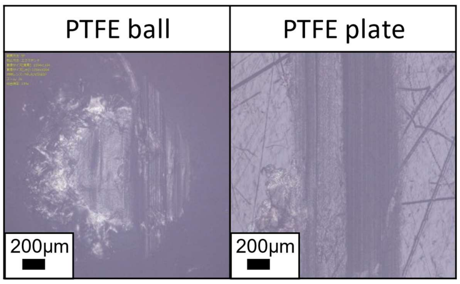

Fig. A2 Optical microscope images of the friction surfaces of the PTFE ball and plate after the sliding test in water under the same sliding condition as used for the si-PI coating