Tribological properties of polyimide composites reinforced with fibers rubbing against Al₂O₃

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Abstract: Reinforcing fillers are of great importance in tribological performance and tribofilm formation of polymeric composites. In this study, the tribological properties of aramid particle (AP) and short carbon fiber (SCF) reinforced polyimide (PI) composites were added to hexagonal boron nitride (h-BN), and silica (SiO₂) nanoparticles sliding against alumina were comprehensively investigated. When sliding occurred with AP-reinforced PI composites, the tribological properties were not closely depended on the pressure × velocity \((p \times v)\) factors and the nanoparticles. The interactions between AP and its counterpart could not induce tribo-sintering of the transferred wear debris. As such, the tribofilm seemed to be in a viscous state, leading to higher friction and wear. However, the inclusion of hard SCF into the PI matrix changed the interfacial interactions with alumina. A robust tribofilm consisting of a high fraction of silica was generated when the SCF-reinforced PI was added to the SiO₂ nanoparticles. It exhibited a high load-carrying capability and was easily sheared. This caused a significant decrease in the friction and wear of the PI composite at 8 MPa·1m/s. Moreover, due to their high melting point, few h-BN nanoparticles were observed in the tribofilm of the SCF-reinforced PI when hexagonal boron nitride was added.

Keywords: reinforcing filler; polyimide composite; tribofilm; Al₂O₃

1 Introduction

Over the past decades, polymeric composites have been widely used as tribo-materials because of their high chemical stability and self-lubrication [1–6]. The fuel consumption and material loss associated with friction and wear are significant economic issues in automotive engines [7–10]. As a result, considerable efforts have been devoted to the development of lubricating polymeric composites. Formulated polyimide (PI) composite is one of the outstanding materials used in many engineering fields because of its excellent mechanical and self-lubricated properties, which has generated substantial research interest [10–13].

It has been confirmed by many reports that the addition of reinforcing fillers, such as aramid fibers, glass fibers, and short carbon fibers (SCFs), could enhance the abrasion resistance of polymer matrix by inducing high mechanical properties [14–20]. Lv et al. [21] confirmed that the incorporation of carbon fibers and aramid fibers into the PI matrix could reduce friction and wear. Moreover, the incorporation of functional nanoparticles into polymer composites significantly improved the tribological behavior.

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Chang and Friedrich [5] analyzed the effect of the high stress and temperature on the direct contact region between sliding surfaces. It was considered that the incorporated carbon fibers with a small percentage of nanoparticles had a synergistic effect on the reduction of friction and wear by forming a lubricated tribofilm, especially under high pressure and velocity conditions. In our previous studies [11, 25], we demonstrated that the addition of nano-SiO$_2$ particles into SCF-reinforced polyetheretherketone, polyphenylene sulfide, and high molecular weight polyethylene facilitated the formation of a silica-based tribofilm. This resulted in a decrease in the friction and wear of nanocomposites. Although the enhanced tribological behaviors of polymeric composites with reinforcing fillers and nanoparticles have been confirmed in many investigations [11, 25, 26], the tribological mechanisms of the reinforcing fillers have not been systematically investigated.

In addition, tribology is a system dominated by a matching rubbing pair, which plays a critical role in prolonging the lifespan and enhancing the reliability of the system. Significant attention has been focused on the development of sliding pairs, e.g., metal–metal, polymer–polymer, and polymer–metal [27–29]. In particular, polymer–metal frictional pairs are widely utilized in moving components. Nevertheless, the metallic counterpart is easily oxidized due to strong interfacial interactions, which limits its applications under extreme conditions. Ceramic materials are promising candidates that are employed in the fields of spacecraft, mechanical engineering, and the automotive industry, due to their excellent mechanical properties, oxidation resistance, and low specific density [30–33]. As reported by Song et al. [32], the sliding process could be maintained when alumina was used as the counterpart at a high temperature. Wu et al. [33] comparatively studied the tribological performance of diamond-like carbon coating films as they slid against different ceramic materials under extreme conditions. An ultralow friction coefficient and mild wear were observed due to the formation of an easy shear transfer layer. Over the last few decades, most studies have reported on the tribological performance of ceramic materials during the process of sliding against different counterparts [34, 35]. Nonetheless, the tribological performance of polymer–ceramic sliding pairs has rarely been reported. It is therefore expected that comprehensive investigation on sliding pairs can provide novel opportunities for the development of high-performance bearings.

Therefore, in this study, we mainly investigate the tribological behaviors of two types of PI composite systems as they slide against alumina ceramic (Al$_2$O$_3$) at different pressure × velocity ($p \times v$) factors. This includes PI filled with aramid particles (AP), SiO$_2$ and hexagonal boron nitride (h-BN) nanoparticles, and PI filled with SCF, SiO$_2$, and h-BN nanoparticles. The tribological performance and tribological mechanisms of PI composites filled with various reinforcing fillers are compared. The objective of the present study is to determine the effect of interfacial interactions between reinforcing fillers and counterparts on tribofilm formation. The results of this study are expected to provide guidance for the improved design of PI composites as they slide against ceramic.

2 Experimental

2.1 Material preparation

Thermoplastic PI (YS-20, particle size < 75 μm) at a softening temperature of 239 °C was obtained from Shanghai Synthetic Resin Institute (Shanghai, China). Aramid particles (Fig. 1(a)) (Twaron 5011, Japan) and polyacrylonitrile-based SCFs (Sigrafil C10M080UNS) were used to reinforce the PI matrix (Fig. 1(b)). The Mohs’ hardness and $E$-modulus of aramid particles are < 4.5 [36] and 60–120 GPa [37], respectively. With regard to SCFs, the Mohs’ hardness is 4 [36] and the $E$-modulus is 230–600 GPa [38]. The h-BN (particles with an average size of 120 nm) was provided by Shanghai St-Nano Science Technology Co., Ltd. SiO$_2$ (particle size ≈ 20 nm) was purchased from Nanjing Emperor Nano Material Co., Ltd. The Al$_2$O$_3$ counterpart with a diameter of 42 mm was supplied by Shenzhen Jin Long Da Ceramic Technology Co., Ltd., and cut into the disc with a thickness of 5 mm using Diamond Wire Saw (STX-202A Shenyang, China).

PI powders were compounded with fillers using a high-speed crusher (FW177, Tianjin Taisite Instrument Co., Ltd.) at 8,000 rpm for 30 min to ensure appropriate blending. The preparation procedure for the composites was based on the hot press molding technique as described in Ref. [11]. They were heated from 25 to
375 °C and then held at a constant pressure of 12 MPa for 2 h to allow full compression sintering. After cooling to room temperature, the specimen was cut into the sizes of 4 mm × 4 mm × 12 mm for tribo-tests. The composition of two kinds of PI composite systems and the abbreviated forms are listed in Table 1.

The distribution of various fillers in the PI matrix is shown in Fig. 2. The micro AP and SCF fillers are uniformly embedded in PI matrix (Figs. 2(a) and 2(b)), whereas the nanoparticles are mainly distributed around the AP and SCF as indicated by the arrows. We measured the shore hardness (D) of the PI composites consisting of different fillers using a hardness tester (TH210, Beijing Time High Technology Co., Ltd., China). At least five locations were tested for each specimen. The result in Fig. 3 demonstrates that the hardness of the SCF-reinforced PI composites is higher than that of the AP-reinforced composites.

### 2.2 Tribology tests

Tribology tests were conducted in an air atmosphere on a pin-on-disc test rig (POD, TRM-100, Wazau, Germany). The contact form of the friction pair and the surface morphology of the counterpart are shown in Fig. 4. A polymer pin was rotated against a ceramic disc with a contact surface of 4 mm × 4 mm. The

| Abbreviated form | PI  | AP  | SCF | SiO\(_2\) | h-BN |
|------------------|-----|-----|-----|----------|------|
| PI/AP            | 90  | 10  | —   | —        | —    |
| PI/AP/SiO\(_2\) | 87  | 10  | —   | 3        | —    |
| PI/AP/BN         | 87  | 10  | —   | —        | 3    |
| PI/SCF           | 90  | —   | 10  | —        | —    |
| PI/SCF/SiO\(_2\) | 87  | —   | 10  | 3        | —    |
| PI/SCF/BN        | 87  | —   | 10  | —        | 3    |
tribological behavior was evaluated under low, medium, and high-pressure × velocity factors, i.e., 1, 4, and 8 MPa·1m/s. Sliding was performed for 5 h and was repeated at least three times for each condition. Prior to the tribology tests, the roughness of Al₂O₃ was controlled in the range of 0.30–0.40 μm by polishing with W20 SiC grinding paper. The friction coefficient was recorded online, and the specific wear rate \( W_s \) (mm³/(N·m)) was calculated using the following equation:

\[
W_s = \frac{\Delta M}{(FL\rho)}
\]  

where \( \Delta M \) (mg) represents the mass loss of the specimen, \( F \) (N) represents the normal load as shown in Fig. 2, \( L \) (m) represents the total sliding distance, and \( \rho \) (g/cm³) represents the composite density.

2.3 Characterization of worn surfaces of PI composites and tribofilms formed on Al₂O₃ surface

To elucidate the tribological mechanism, field emission scanning electron microscopy (FE-SEM) (Mira 3, Xmu, Tescan) was used to characterize the morphology of the polymer pins after sliding against Al₂O₃, in addition to the tribofilms formed on Al₂O₃ counterparts. Energy dispersive X-ray spectroscopy (EDS) was combined with FE-SEM to detect the elemental composition of the tribofilms. In addition, elemental mapping of the worn surface of Al₂O₃ was performed using FE-SEM equipped with EDS (Zeiss Ultra55, Germany). To elucidate the tribo-chemical reactions that occur on the surface of the ceramic, the functional groups of the tribofilms that formed on the Al₂O₃ surface were identified using attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR TENSOR 27, Bruker).

3 Results and discussion

3.1 Tribological performance of PI composites reinforced by aramid particles

The average friction coefficients and wear rates of PI composites that are reinforced with aramid particle, i.e., PI/AP, PI/AP/SiO₂, and PI/AP/BN for the case of sliding against Al₂O₃ at 1, 4, and 8 MPa·1m/s are illustrated in Fig. 5. It was determined that the friction coefficients of the AP-reinforced PI composites were not dependent on the \( p \times v \) factors and the incorporated nanoparticles, whereas the corresponding wear rates exhibited distinct differences. As shown in Fig. 5(b), the increasing \( v \) value resulted in a lower wear rate. In particular, PI/AP yielded the lowest wear rate (0.91 × 10⁻⁶ mm³/(N·m)) for sliding at 8 MPa·1m/s. This represents a reduction of approximately an order of magnitude in comparison to that at 1 MPa·1m/s. The incorporated nanoparticles notably decreased the wear resistance of PI/AP/SiO₂ and PI/AP/BN. Wang et al. [39] reported that the released nanoparticles acting as a polishing agent could abrade the PI composite and increase its wear rate. However, when sliding occurred at 1 MPa·1m/s, the wear rate of PI/AP/SiO₂ was much lower than that of PI/AP/BN. It was considered that although the polishing action of the nanoparticles could enhance the wear rates of the composites, the spherical nano-silica particles probably carried more loads of composites compared to the lamellate h-BN, leading to a better wear resistance of PI/AP/SiO₂ than PI/AP/BN.

The evolution of the friction coefficients of PI/AP, PI/AP/SiO₂, and PI/AP/BN at 1, 4, and 8 MPa·1m/s are
compared in Fig. 6. At the initial stage, the friction coefficient of PI/AP decreased and then increased at approximately 0.5 h. However, the running-in duration of PI/AP at 4 MPa·1m/s was much shorter than the sliding duration at 1 and 8 MPa·1m/s. It is supposed that the replenishment and the removal of transferred wear debris rapidly reached a dynamic equilibrium at 4 MPa·1m/s [11, 22], which resulted in a stable friction coefficient of PI/AP (0.21). However, the addition of nanoparticles into the PI composites changed the evolution of the friction coefficient. Unlike the impact of the addition of nanoparticles to the SCF-reinforced polymers [11], the reduction in the friction coefficients of PI/AP/SiO₂ and PI/AP/BN was not significant. Moreover, it was determined that the friction coefficient of PI/AP/SiO₂ fluctuated dramatically at 8 MPa·1m/s in comparison to that of PI/AP/BN. It was considered that the h-BN particles released onto the sliding interface reduced the friction coefficient of PI/AP/BN due to the easy-to-shear property of its lamellar structure.

Figure 7 shows the worn surfaces of PI/AP, PI/AP/SiO₂, and PI/AP/BN for rubbing against Al₂O₃ at 4 and 8 MPa·1m/s. There were plowing marks on the worn surfaces of three kinds of composites when the sliding occurred at 4 MPa·1m/s. This is indicative of the abrasive wear of the composites. Nevertheless, the incorporated SiO₂ and h-BN nanoparticles caused a deep scratch on the surface of the PI/AP/SiO₂ and PI/AP/BN, and led to higher wear rates compared to
PI/AP. It is considered that the nanoparticles released onto the sliding interfaces were mixed with the remnant polymer and the abraded counterpart, thereby forming large nano-based wear debris. This resulted in a wide groove of the worn surface as indicated by the white arrows in Figs. 7(c) and 7(e). Increasing the load from 4 to 8 MPa resulted in obvious adhesion and plastic deformation. As indicated by the red circles in Figs. 7(b), 7(d), and 7(f), the wear debris of PI/AP/SiO₂ and PI/AP/BN exceeded that of PI/AP, which probably accounts for the high adhesion between the sliding interfaces.

To reveal the tribofilm structure and elucidate the tribological mechanism, SEM images and EDS analysis of the tribofilms formed on the Al₂O₃ surface after sliding against the AP-reinforced PI composites at 4 and 8 MPa·m/s are presented in Fig. 8. In contrast to the morphology of Al₂O₃ before the tribology tests (Fig. 4), the counterfaces exhibited significant differences after sliding against three kinds of composites. When PI/AP was considered, the transferred materials almost covered the entire Al₂O₃ surface, which resulted in an improved load-carrying capability of the sliding surface, and thereafter improved the anti-wear of PI/AP. With respect to the sliding of the counterfaces against PI/AP/SiO₂ and PI/AP/BN, EDS analysis confirmed that the transferred wear debris mixing with the counterpart materials formed carbon-based tribofilm. This is inconsistent with our previous observations of SCF-reinforced nanocomposites [11, 26]. However, the tribofilms generated from PI/AP/SiO₂ and PI/AP/BN at 4 MPa·m/s seemed to be in a viscous state compared to PI/AP. This decreased the load-carrying capability of the tribofilms and led to the higher wear rates of PI/AP/SiO₂ and PI/AP/BN compared to PI/AP. Moreover, a possible high adhesion between the tribofilms and Al₂O₃ counterface could account for the high friction coefficients of PI/AP/SiO₂ and PI/AP/BN [26]. It seemed that at 8 MPa·m/s, the wear debris of PI/AP/SiO₂ released on Al₂O₃ surface was more compact than that of PI/AP/BN, and as such, a tribofilm with a high load-carrying capability was obtained. This resulted in the superior wear resistance of PI/AP/SiO₂ compared to that of PI/AP/BN.

Figure 9 gives the elemental mappings of the tribofilms generated from PI/AP/SiO₂ and PI/AP/BN.

![Fig. 8](image_url)  
**Fig. 8** Tribofilms on Al₂O₃ surface after rubbing with (a, b) PI/AP, (c, d) PI/AP/SiO₂, and (e, f) PI/AP/BN at 4 and 8 MPa·m/s. The EDS analysis of the red squares in (c) and (e) are shown on the right.
3.2 Tribological performance of PI composites reinforced by SCFs

Figure 10 illustrates the mean friction coefficients and wear rates of SCF-reinforced polyimide composites for sliding against Al₂O₃ at different \( p \times v \) values. In contrast to the AP-reinforced PI composites, their friction and wear were closely dependent on nanoparticles and \( p \times v \) factors. For PI/SCF, the lowest friction coefficient (0.21) and wear rate \((3.3 \times 10^{-6}\) mm\(^3\)/(N·m)) were obtained at 4 MPa·m/s. However, when the \( p \times v \) factor was improved to 8 MPa·m/s, the friction and wear of PI/SCF increased significantly. Moreover, the addition of SiO₂ nanoparticles improved the tribological performance of PI/SCF/SiO₂. The friction coefficient and wear rate of PI/SCF/SiO₂ were reduced with the increase of the \( p \times v \) value, and the lowest friction coefficient and wear rate \((0.13 \times 10^{-6}\) and \(0.72 \times 10^{-6}\) mm\(^3\)/(N·m)) were obtained at 8 MPa·m/s. It could be concluded that the influence of nano-SiO₂ on the tribological behavior of SCF- and AP-reinforced PI was significantly different. In comparison, no significant reduction was observed in the friction and wear of PI/SCF/BN at 1 and 4 MPa·m/s compared to PI/SCF/SiO₂.

As shown in Fig. 11, the evolution of the friction coefficient of the SCF-reinforced PI with or without nanoparticles is different after sliding against Al₂O₃ at 1, 4, and 8 MPa·m/s. When PI/SCF was considered, the friction coefficient increased sharply at the initial stage, and the running-in duration lasted approximately 1.5 h at 1 MPa·m/s. Moreover, the friction coefficient

Fig. 9 Tribofilms and corresponding elemental mappings generated from (a) PI/AP/SiO₂ and (b) PI/AP/BN for sliding against Al₂O₃ at 4 MPa·m/s.
Fig. 10  (a) Average friction coefficients and (b) wear rates of PI/SCF, PI/SCF/SiO$_2$, and PI/SCF/BN after sliding against Al$_2$O$_3$ at 1, 4, and 8 MPa·1m/s.

Fig. 11  Evolution of friction coefficient at different sliding time when PI/SCF, PI/SCF/SiO$_2$, and PI/SCF/BN were slid against Al$_2$O$_3$ at 1, 4, and 8 MPa·1m/s.

SiO$_2$ nanoparticles in the SCF-reinforced PI played a critical role in the tribological behavior of PI/SCF/SiO$_2$. The friction coefficients of PI/SCF/SiO$_2$ decreased initially, then achieved a stable state. It was considered that material transfer initially dominated the formation of the tribofilm. Subsequently, the wear debris and the released nano-SiO$_2$ were sintered to produce a tough SiO$_2$-based tribofilm due to the strong interfacial interaction between the carbon fibers and the Al$_2$O$_3$ surface [22]. With regard to the PI/SCF/BN, the evolution of the friction coefficient at 1 MPa·1m/s fluctuated drastically. The curve subsequently became smooth and rapidly reached a stable value at 8 MPa·1m/s.

Figure 12 shows the worn surfaces of the SCF-reinforced PI composites as they rub against Al$_2$O$_3$ at 4 and 8 MPa·1m/s. Carbon fibers were exposed to the sliding surface after the sliding process, which is inconsistent with the properties of the PI composites reinforced by aramid particles. As indicated by the arrows in Figs. 12(a) and 12(b), carbon fibers protruded from the worn surfaces of PI/SCF, which induced high wear resistance in the PI/SCF. It seemed that the addition of nanoparticles changed the wear mechanism of PI/SCF became unstable when the $p \times v$ factor increased, and fluctuated dramatically at 4 and 8 MPa·1m/s. In contrast to PI/AP/SiO$_2$, the incorporated
of the SCF-reinforced PI composites. For the PI/SCF/SiO$_2$ transferred wear debris accumulated on the surface as indicated by the red circles in Figs. 12(c) and 12(d). As reported by Qi et al. [11], the nanoparticles released from the composites could be sintered into a compact piece. With regard to PI/SCF/BN, the morphology of the worn surface exhibited a significant difference between 4 and 8 MPa·1m/s. Adhesion and plastic deformation were deemed to be the wear mechanism at 4 MPa·1m/s [40]. Moreover, SCF seemed to sink into the PI matrix during the sliding process as indicated by the red rectangle in Fig. 12(e). This resulted in a decrease in the load-carrying capability of PI/SCF/BN, leading to a low wear resistance at 4 MPa·1m/s. The worn surface of PI/SCF/BN at 8 MPa·1m/s was smooth and the protruding SCF improved its wear resistance.

SEM results of the morphology of the worn surfaces of the ceramic counterparts are presented in Fig. 13. When sliding of PI/SCF, PI/SCF/SiO$_2$, and PI/SCF/BN occurred, the transferred materials almost covered the entire Al$_2$O$_3$ surface. The tribofilm of PI/SCF was very thin, and cracks were present at 4 MPa·1m/s. When the $p \times v$ factor was increased to 8 MPa·1m/s, the wear debris around the cracks was scraped away and a patch-like tribofilm was obtained (Fig. 13(b)). This resulted in the poor tribological performance of PI/SCF at 8 MPa·1m/s. With respect to the counterface after sliding against PI/SCF/SiO$_2$, a thick tribofilm was formed at 4 MPa·1m/s (Fig. 13(c)). EDS analysis confirmed that the thick tribofilm consisted of a high fraction of silica and C materials. When the $p \times v$ factor was increased to 8 MPa·1m/s, the tribofilm almost covered the entire Al$_2$O$_3$ surface (Fig. 13(d)), which could separate the direct contact of the sliding surfaces and reduce the friction and wear of PI/SCF/SiO$_2$ [11, 41]. However, in the case of PI/SCF/BN, the tribofilm’s structure was similar to that of PI/AP/BN at 4 MPa·1m/s, and appeared to be in a viscous state. Therefore, the high adhesion between the tribofilm and Al$_2$O$_3$ resulted in high friction and wear of PI/SCF/BN. Moreover, it is noted from the EDS results that almost no BN nanoparticles were detected in the tribofilm of PI/SCF/BN at 4 MPa·1m/s. Under a
high \( p \times v \) condition, compaction of wear particles dominates tribofilm formation (Fig. 13(f)). In this case, the tribofilm with a high load-carrying capability significantly reduced the friction and wear of PI/SCF/BN at 8 MPa·1m/s [42].

The elemental mapping of the tribofilms generated from PI/SCF/BN and PI/SCF/SiO\(_2\) at 8 MPa·1m/s in Fig. 14 confirmed the composition of the SCF-reinforced PI composites. It is apparent that the content of nanoparticles in the tribofilms of PI/SCF/BN and PI/SCF/SiO\(_2\) are different at 8 MPa·1m/s. Most of the nano-SiO\(_2\) covered the Al\(_2\)O\(_3\) surface during sliding with PI/SCF/SiO\(_2\). This was probably because the hydroxyl and residual unsaturated bonds in the nano-silica were prone to adsorption on the counterpart [42]. Moreover, the released nanoparticles could abrade the Al\(_2\)O\(_3\) counterpart under the strong interfacial interactions between SCF and the counterpart that was tribo-sintered to a compact layer with the abraded Al\(_2\)O\(_3\) and remnant polymer. However, the mixing of Al\(_2\)O\(_3\) with the transferred wear debris constituted the tribofilm of PI/SCF/BN. Consistent with the composition of the tribofilm at 4 MPa·1m/s, few BN nanoparticles were detected at 8 MPa·1m/s. It was proposed that h-BN nanoparticles were difficult to sinter during sliding process due to their high melting temperature [42].

### 3.3 Tribological mechanism of PI composites

Based on the aforementioned analysis, it is evident that the properties of the reinforcing fillers in the PI composites played an important role in tribo-physical and chemical behavior on the sliding surface, which determined the tribological performance and tribofilm formation. Generally, the formation of a tribofilm includes the following four procedures: (i) Formation of a carbon-based film by PI composite transfer; (ii) release of nanoparticles onto the sliding surface due to the degradation of the polymer matrix; (iii) accumulation and compaction of wear debris in the contact regions; (iv) tribo-sintering of wear particles including nanoparticles, remnant polymer, and the counterpart. Based on the formation process of a tribofilm, it was considered that the tribological mechanism and the tribofilm formation for SCF- and AP-reinforced PI composites were notably different. Schematic diagrams depicting the interfacial interactions of two kinds of PI composites with Al\(_2\)O\(_3\) and their tribofilm formation mechanisms are compared in Figs. 15(a) and 15(b).

In the case of AP-reinforced PI composites, the tribofilm formed on the Al\(_2\)O\(_3\) surface seemed to be in

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**Fig. 14** Tribofilms and corresponding elemental mappings generated from (a) PI/SCF/SiO\(_2\) and (b) PI/SCF/BN when sliding against Al\(_2\)O\(_3\) at 8 MPa·1m/s.
Fig. 15  Schematic diagrams of interfacial interactions between (a) AP-reinforced PI composites and (b) SCF-reinforced PI composites and Al$_2$O$_3$ counterpart.

a viscous state, which can be verified from the SEM images. It was concluded that the hardness and modulus of AP led to the weak interfacial interactions between the sliding surfaces. As a result, the transferred wear debris mixed with the Al$_2$O$_3$ nanoparticles was hardly tribo-sintered to a compact layer (Fig. 15(a)). Moreover, the released nanoparticles in the sliding surface could lead to abrading of PI/AP/SiO$_2$ and PI/AP/BN. In this case, the wear rates of PI/AP/SiO$_2$ and PI/AP/BN were higher than that of PI/AP. Nevertheless, the inclusion of SCF into the PI composites changed the interfacial interactions. It was considered that the strong interaction between the hard carbon fibers and Al$_2$O$_3$ could result in a special stress concentration and the high temperature of the sliding surface [5]. Hereafter, tribo-sintering of nanoparticles, remnant polymeric composites, and Al$_2$O$_3$ occurred, resulting in the formation of a nano-based tribofilm (Fig. 15(b)). It was determined that the nanoparticles in the tribofilm could bear most of the load of the sliding interface due to their high modulus. Moreover, the tribofilm prevented direct contact and reduced the adhesion of the sliding interfaces. Therefore, the high load-carrying capability and easy-to-shear property of the tribofilm led to the superior tribological performance of the SCF-reinforced PI composites compared to that of the AP-reinforced PI composites.

To further explore the tribo-physical and chemical behavior on the Al$_2$O$_3$ surface, ATR-FTIR spectra of the tribofilms of AP- and SCF-reinforced PI composites are presented in Fig. 16. After rubbing against PI/AP, the absorption peaks of tertiary amide at 1,714, 1,614, and 1,375 cm$^{-1}$, and C–O at 1,230 cm$^{-1}$ were confirmed in the PI matrix (Fig. 16(a)) [11]. This indicated that material transfer dominated tribofilm formation. However, these absorption peaks disappeared in the case of sliding against PI/AP/SiO$_2$ and PI/AP/BN. Two broad infrared absorption peaks at 1,180 and 1,010 cm$^{-1}$ were evident in the spectra obtained for the tribofilms. This corresponds to C–O in carbonaceous material, demonstrating that tribo-chemical reactions such as the breakage of polymer molecular chains and the oxidation of free radicals occurred on the sliding interface [43, 44]. However, as illustrated in Fig. 16(b), the tribo-chemical reactions were prone to occurring.

Fig. 16  ATR-FTIR spectra of tribofilms formed on Al$_2$O$_3$ surface when sliding against (a) AP-reinforced PI composites and (b) SCF-reinforced PI composites at 8 MPa·1m/s.
under the interfacial interactions between the SCF and the counterpart. Two broad infrared absorption peaks at 1,180 and 1,010 cm\(^{-1}\) were obtained in the tribofilms of SCF-reinforced PI composites. This indicated that similar tribo-chemical reactions to PI/AP/SiO\(_2\) and PI/AP/BN occurred at 8 MPa·1m/s.

4 Conclusions

In this study, the impact of aramid particles and carbon fibers on the tribological performance of PI composites with incorporated nanoparticles was comparatively investigated. The tribofilms formed on Al\(_2\)O\(_3\) counterpart were comprehensively characterized to elucidate the tribological mechanism of the AP- and SCF-reinforced PI composites. The main conclusions are as follows:

1) Reinforcing fillers play an important role in the tribological performance of PI composites. When the AP-reinforced PI composites were slid against Al\(_2\)O\(_3\), their friction coefficients were not dependent on the \(p \times v\) values and nanoparticles. However, the addition of SiO\(_2\) and BN nanoparticles decreased the wear resistance of PI/AP/SiO\(_2\) and PI/AP/BN. In the case of SCF-reinforced PI composites, the incorporation of nanoparticles into PI/SCF exerted an influence on the reduction of the friction and wear of PI/SCF/SiO\(_2\) and PI/SCF/BN, especially at 8 MPA·1m/s.

2) Reinforcing fillers are of great importance in tribofilm formation. With regard to the AP-reinforced PI composites, the incorporation of SiO\(_2\) and h-BN nanoparticles led to the formation of viscous-like tribofilms. However, after the addition of SCF into the PI matrix, tribo-sintering of remnant composites, nanoparticles, and Al\(_2\)O\(_3\) occurs due to the strong interactions between SCF and the counterpart, and a compact tribofilm could be obtained.

3) As identified in this study, tribo-chemical reactions of polymer molecules occurred under the interfacial interactions between the sliding surfaces. It was demonstrated that the reactions could not occur during the process of sliding against PI/AP due to the weak interactions between AP and Al\(_2\)O\(_3\). With regard to the SCF-reinforced PI composites, tribo-chemical reactions readily occur and robust tribofilms could be formed.

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