

# Friction and Wear Behaviours of Polyimide Composites Against Steel Ball Under Rolling-Sliding Compound Motion

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## Research Article

**Keywords:** Rolling-sliding motion, high frictional temperature, tribochemical wear, black products, polyimide composites

**Posted Date:** May 17th, 2021

**DOI:** <https://doi.org/10.21203/rs.3.rs-517711/v1>

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# Abstract

Wear of polyimide composites cage due to lubrication failure has a significant impact on the lifetime of bearing, but the related mechanisms are still far from understanding due to the complex movements of bearing parts, such as rolling, sliding et al, in the real operations. Here, the tribological behaviors of polyimide composites against steel ball were studied under a motion compounded with rolling and sliding in comparison with pure sliding. The rolling-sliding motion has a strong influence on the tribological properties of polyimide composites depending on lubrication conditions. Especially under PAO4 oil lubrication, the rolling-sliding motion can facilitate the degradation of polyimide composites, resulting in severe surface wear and abundant black products forming. The analysis based on XPS and SEM measurements and the compared results from heating sliding test and thermal treatments indicates that the formation of black products is mainly attributed to the tribochemical reactions accompanying with the rupture of C-O and C=O bonds due to high frictional temperature. This process is further facilitated when the PAO4 oil and the metal from counterface participate in the tribochemical reactions.

## Highlights

- Rolling-sliding compound motion was designed to study tribological performance
- Tribological behaviors of polyimide composites against steel depends on motion mode
- R-S induces severe tribochemical wear of polyimide with black products forming
- Polyimide degradation is facilitated in R-S motion due to high friction temperature
- PAO4 oil and steel ball also play the significant roles in the tribochemical wear

## 1. Introduction

Due to its high thermal and chemical stability, excellent wear resistance, and good self-lubricity, polymer materials has widely used as the structure materials in human joints, bearings and other industrial fields [1–4]. Among them, especially polyimide and the related composite materials have received extensive attention because of their extra superior mechanical properties and high temperature resistance [5–8].

The self-lubricating properties of polymer materials normally originate from the transfer layer between the pair of grinding pairs formed due to the viscous flow of the polymer part [9]. For instance, an extreme low friction and wear state can be maintained for a long running time between the polyimide (or polyimide composites) and metal pairs due to the generation of a lubricating film combining with polyimide and metal wear debris [10, 11]. When the surrounding temperature want up to a range of 100–200°C, the densification of polymer materials improved their mechanical properties (i.e., higher rigidity, elastic modulus and hardness) and then enhanced the abrasion wear resistance [12, 13]. However, the polyimide matrix suddenly softened when friction heating induced the high flash temperature exceeding a critical value, then facilitating the surface wear [14, 15]. Several kinds of materials, such as organic aramid fibre, carbon reinforced polymer composite fibre, glass fibre, basalt fibre, polytetrafluoroethylene and MoS<sub>2</sub>, are

added in the polyimide substrates, which have presented a significant improvement of tribological performance in pure sliding motion [4, 15–20]. However, the real applications of polyimide materials in tribological components (i.e., cup in total hip arthroplasty and cage of bearings [21–24]) normally involve complicated movements such as sliding, rolling, rotating et al [25–28], so the corresponding failure mechanism in tribological study may be hindered in the pure sliding tests.

Here, we designed a compound motion with rolling and sliding and studied the tribological behaviors of polyimide composites against steel balls in comparison with the pure sliding motion. The compound motion strongly affected the tribological properties of polyimide composites depending on the lubrication conditions. The characterizations of wear tracks using scanning electronic microscope and X-ray photoelectron spectroscopy indicated the key role of frictional temperature in the wear of polyimide composites under these two motion modes, which was further proved by the thermal treatments and the heating sliding test. The results in this study provide critical insights needed for better understanding of the failure mechanism of polyimide components used in bearings.

## 2. Experimental Details

### 2.1 Tribological test set-up and test parameters

The friction and wear tests of polyimide composites against steel balls lubricated with and without PAO4 oil were carried out by a high-precision universal tribotester (HV-TRB, Anton Paar, Austria). As shown in Fig. 1, the steel ball is fixed inside the holder pressed by a screw (upper-middle inset in Fig. 1), which applies load; the polyimide composites specimen is installed on the sample table, which provides reciprocating sliding. When a gap is kept between the screw and the bearing ball (upper-right inset in Fig. 1), rolling of the steel ball would occur during the sliding process. Under this condition, the relative sliding of ball/polyimide pairs should also exist due to the interactions between the ball/screw and ball/holder contact interfaces. Then, this compound movement can be denoted as “rolling-sliding” motion. Under this condition, the critical tangential forces corresponding to the initial movement of a standard glass sample sliding against the un-fixed steel balls were kept as around 0.35 N under a applied load of 4 N to maintain a constant slide-to-roll-ratio in all compound motion studies.

In all the friction tests, the applied load was 4 N, the reciprocating amplitude was 6 mm, the moving velocity of the sample table was 8.5 cm/s. The surrounding temperature and relative humidity were controlled at  $23 \pm 2^\circ\text{C}$  and  $30 \pm 3\%$ . After the friction tests, morphologies of the wear tracks were pictured by white light interferometer (WLI, Chotest SuperView W1, Chotest Technology Inc., China), scanning electronic microscope (SEM, Inspect F50, Thermo Fisher, USA) and optical microscope (Axio Lab.A1, Zeiss, Germany). The chemical compositions of the wear region and wear debris were characterized by X-ray photoelectron spectroscopy (XPS, Escalab Xi+, Thermo Fisher, USA). To detect the degradation of the polyimide composites, the thermal treatments was carried out in room air using a tube furnace (OTF-1200X-II; Hefei kejing materials technology co., Ltd., Anhui, China).

## 2.2 Test samples

The used substrates are the thermoplastic polyimide composites containing 8 wt% polytetrafluoroethylene (PTFE). Figure 2 illustrates the chemical formulas of the polyimide substrate and PTFE, respectively. Before the friction tests, all polyimide composites surfaces were polished by sandpaper to uniform the root-mean-square surface roughness as  $100 \pm 5$  nm. The bearing balls (purchased from Jiangsu Sak Bearing Co., LTD) with a diameter of  $\sim 6$  mm are made of 440C (more detail shown in Table 1). The surface roughness is around 6 nm. The lubricating oil is PAO4 (purchased from DowPol Company). Table 2 shows the physical properties of the PAO4 oil. Before each experiment, both the bearing ball and polyimide substrate pairs were ultrasonic in alcohol and deionized water for 3 minutes, then dried by nitrogen, and finally kept in vacuum condition. As a comparison, the tribological behaviors of polyimide composites against  $\text{Al}_2\text{O}_3$  balls with a diameter of  $\sim 6$  mm under pure sliding and rolling-sliding motions with PAO4 oil lubrication.

Table 1  
Chemical composition of the used bearing ball.

Element	Fe	Cr	C	O	Mn	Si	Al
Wt %	83	12.1	3.3	0.8	0.4	0.4	0.1

Table 2  
Physical properties of the PAO4 oil.

Viscosity ( $\times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ )		Density	Flash point ( $^{\circ}\text{C}$ )	Pour point ( $^{\circ}\text{C}$ )	Main composition
40 $^{\circ}\text{C}$	100 $^{\circ}\text{C}$				
16.8	3.9	0.189	222	69	Poly Alpha Olefin

## 3. Experimental Results

Here, the effect of motion mode on the tribological behaviors of polyimide composites against steel balls was studied under the contact interface lubricated with and without PAO4 oil. Under dry condition, the friction coefficient gradually increases from  $\sim 0.13$  to a stable value of  $\sim 0.36$  at a critical sliding time around 1000 s in pure sliding, and maintains a low level around 0.12 during the entire friction test in rolling-sliding motion. It is acceptable that the rolling motion can significantly weaken the tangential stress generated due to relative sliding movement of the two contact interfaces. It is intriguing that, under oil lubrication, the friction coefficient in sliding-rolling motion is larger than that in pure sliding motion during initial running process. After testing 1500s, the values are close (around 0.05) under these two motions (Fig. 3b). Compared to that in sliding-rolling motion, the decrease amplitude of friction coefficient due to the addition of PAO4 oil is much larger in pure sliding motion. It indicates that, although the oil is capable of lubricating the polyimide and steel contact interface under these two motions due to

hydrodynamic pressure effect [29–32], the effective lubricating oil film formed between the sliding interfaces may be to some extent suppressed in the sliding-rolling motion.

Not only friction but also surface wear of the polyimide composites strongly depends on the motion modes and lubrication conditions. In pure sliding motion, the surface wear of polyimide composites occurs in the entire contact region under dry condition (upper-left inset in Fig. 3a), and behaviors the formation of multiple furrow-like scratches under PAO4 oil lubrication (upper-left inset in Fig. 3b). Differently, the extra black substance are formed inside the wear tracks in rolling-sliding motion and this phenomenon is more remarkable under oil lubrication (upper-right insets in Figs. 2a and b). The formation of new products implies the occurrence of stress-associated chemical reaction (or denoted as tribochemical reaction [33]) between the polyimide/steel contact interface [34].

Figure 4 compares the wear volume of polyimide composites after running 2700 s under the different experimental conditions. Upper insets show the WLI images of the wear tracks, which is consistent of topographical results characterized by optical microscope shown in Fig. 3. In general, the PAO4 oil lubrication can significantly reduce the interfacial friction but increase the surface damage of polyimide composites under these two motions. The most possible reason is that the addition of PAO4 oil may directly participate the tribochemical reaction and then facilitate the tribochemical removal of polyimide composites (more discussion can be found in the next section). The reaction products are capable of lubricating the tribological interface but are vulnerable to mechanical removal. Similar result was observed between the polyimide/steel sliding interfaces lubricated with water. The tribochemical reaction of polyimide with water resulted in a lower friction force but a more polyimide damage [35]. This phenomenon has been broadly reported for the diamond-like carbon (DLC) coatings. For instance, although the deposition of hydrogenated DLC coating can even reduce the friction coefficient to be lower than 0.01 (denoted as superlubricity), the severe material damage is still insurmountable [36]. It is surprised that the wear volume of polyimide composites in sliding-rolling motion is slightly smaller under air condition, but becomes much larger under PAO4 oil lubrication compared to pure sliding motion.

## 4. Discussion

### 4.1 Chemical and physical characterizations of the worn polyimide composites

The possible change of chemical components inside the worn tracks formed under the different experimental conditions were characterized using XPS. Figure 5 compares the C1s XPS spectra. On a pristine polyimide composites surface, we can observed four C1s peaks in the range of bond energy from 282 eV to 295 eV, which are respectively assigned to C-C, C-H at 284.8 eV, imide carboxyl-type bonding (C-O, C-N) at 286.1 eV, C = O at 288.5 eV and C-F at 292.2 eV [37–40]. After the friction tests, the C-F peaks almost completely disappear on the worn surfaces under all experimental conditions. Table 3 display the area fractions of all components estimated from the XPS spectra in Fig. 5. Besides C-F component, both the peaks of C-O (C-N) bonds and C = O bonds weaken correspondingly compared to the pristine

polyimide composites surface. In general, the decrease amplitude of these two peak areas in rolling-sliding condition is larger than that in pure sliding motion. The addition of PAO4 oil can further reduces the area fractions of these two peaks under pure sliding and rolling-sliding motions. It is consistent with the wear behaviors shown in Fig. 4 that the lubricating oil can facilitate tribochemical reaction and then induce more severe wear of polyimide composites.

Table 3

Parameters of C 1s components of the pristine polyimide composites surface and inside the worn tracks formed under the different experimental conditions.

C 1s component		C-C, C-H	C-O, C-N	C = O	C-F
Binding Energy (eV)		284.8	286.1	288.9	292.2
Area Fraction (%)	Pristine polyimide	40.2	7.5	6.2	46.1
	Rolling-sliding without oil	88.4	6.1	5.5	0
	Sliding without oil	84.5	7.1	6.1	2.3
	Rolling-sliding with oil	93.4	4.5	2.1	0
	Sliding with oil	88.8	6.3	4.9	0

Fig. 6 shows the O1s XPS spectra of the worn regions compared to that of a pristine polyimide composites surface. On a pristine surface, the O1s peaks locating at 531.9 eV and 533.7 eV represent C=O bond and C-O bond, respectively [41-43]. Besides that, a new O1s peak at 529.9 eV that highly corresponds to metal oxide [43, 44] can be observed inside the wear tracks formed in rolling-sliding motions but is absent in pure sliding. It is impossible that the oxidation of metal is easier to be activated by rolling motion than sliding motion since the former should corresponds to a weaker shear interaction between the contact interfaces. In rolling-sliding motion, the relative sliding between ball/holder contact interfaces and ball/screw contact interfaces can induce high frictional temperature, which may result in the metal oxidation. This mechanism can also explain the much lower area fraction of C-O peaks under rolling-sliding conditions (Table 4). The high frictional temperature aggravates the rupture of C-O bond and the decomposition of the imide carboxyl bond [39]. The decrease of  $I_{C-O}/I_{C=O}$  ratio (area of C-O peak divides by area of C=O peak) estimated from O1s XPS spectra (Table 4) indicates that the C-O bonds in polyimide composites are preferentially broken in the tribochemical reactions.

Table 4  
Parameters of O 1s components on a pristine polyimide surface and in the worn regions formed under different experimental conditions.

O 1s component		Metallic oxide	C = O, C-OH*	C-O
Binding Energy (eV)		529.9	531.9	533.7
Area Fraction (%)	Pristine polyimide	0	59.5	40.5
	Sliding-rolling without oil	0	74.2	18.5
	Sliding without oil	7.3	62.3	37.7
	Rolling-sliding with oil	13.8	76.7	9.5
	Sliding with oil	0	66.8	33.2

\* This binding energy may be affected by C-OH bond [45, 46].

Fig. 6 compares topographies of the wear tracks on polyimide composites surfaces formed in pure sliding and rolling-sliding motions under oil lubrication. In pure sliding test, the furrow-like scratches in the wear track are characterized using SEM (Fig. 6a), consistent with the WLI image shown in Fig. 4. In rolling-sliding test, the worn surface is relatively smooth and melting products can be observed at the end of the wear track. This result also supports that the high frictional temperature should play a significant role in the serious tribochemical wear of polyimide composites during the rolling-sliding motion. [38]

## 4.2 Generation of black products in pure sliding at high temperature

The optical images of the wear tracks on polyimide surfaces (Fig. 3) show that the black products are only formed in rolling-sliding motion under room temperature. The results in Figs. 6 and 7 imply that this process may be strongly related to the high friction temperature induced by the compound motion. To prove this hypothesis, a pure sliding test of polyimide composites against steel ball was conducted under a heating environment. In this experiment, the sample platform was firstly heated to 220 degrees and the temperature transferred to the polyimide composites surface decreased to around 120 degrees (estimated by finite element analysis). After that, the pure sliding tests started after addition of the PAO4 oil on the heated sample surface (upper-left inset in Fig. 8a). Different from the pure sliding wear in room temperature (upper-left inset in Fig. 3b), the black products can be observed in the relatively smooth worn region under high temperature (upper-right inset in Fig. 8a). This behavior combining with the results in Fig. 3 confirm that the black products can be formed at high temperature (heating or friction-induced).

Under the heating condition, the friction coefficient of polyimide composites sliding against steel ball under PAO4 oil lubrication is around 0.07, which is larger than that at room temperature (~ 0.05 in Fig. 3b). There are two possible reasons. One is due to the degeneration of the lubricating property along the black products forming. Another is due to the reduction of the elastic modulus of substrate at high

temperature, which corresponds to a larger contact area [12]. Figure 8b shows the O 1s XPS spectrum measured in the worn region formed in pure sliding at high temperature, which is more close to that formed in rolling-sliding at room temperature (Fig. 6). Besides the C = O bond at 533.7 eV and the C-O bond at 531.9 eV, the peak at 529.9 eV corresponding to metal oxide can be observed in the O 1s XPS spectrum, indicating that the steel ball should participate the tribochemical reaction at high temperature.

## 4.3 Degradation of polyimide composites induced by thermal treatment

The direct thermal treatments were operated to further detect the degradation of polyimide composites at high temperature. Using a tube furnace, the samples were heated at a high temperature of 220 degrees in room air. After heating 5 minutes, no obvious change of the topography was observed for the polyimide composites surface covered without PAO4 oil (upper inset in Fig. 9). Differently, the color of the polyimide composites covered with oil turned dark brown after the thermal treatment (bottom optical image). Figure 9 compares the C 1s XPS spectra measured on these two surfaces. It is obvious that, compared to a pristine polyimide composites surface, the relative intensities of the C-F peaks (292.2 eV) decrease significantly after thermal treatments under these two conditions. Besides that, the values of  $I_{C-O/C-N}/I_{C-C/C-H}$  (area of C-O, C-N peak divides by area of C-C, C-H peak) and  $I_{C=O}/I_{C-C/C-N}$  (area of C = O peak divides by area of C-C, C-H peak) after thermal treatments (Table 5) are much smaller than that of the pristine surface (Table 3). The results indicate that both the polyimide substrate and PTFE degrade under the high temperature of 220 degrees.

Here, we also observed that the area fractions of C-F peaks are very close after these two thermal treatments, but that of C-O, C-N peak and C = O peak are smaller under the condition with PAO4 oil. Therefore, the formation of dark brown products only under the thermal treatment with oil indicates that this process should not relate to the decomposition of PTFE, but relate to the degradation of polyimide substrate. Furthermore, these results also imply that the PAO4 oil may react with polyimide substrate that facilitates the rupture of C-O and C = O bonds. It is consistent with the more severe surface wear of polyimide composites under PAO4 oil lubrication in rolling-sliding motion (Fig. 4). Comparing the XPS results in Table 3 and Table 5, we can find that  $I_{C-F}/I_{C-C/C-H}$ ,  $I_{C-O/C-N}/I_{C-C/C-H}$  and  $I_{C=O}/I_{C-C/C-N}$  after thermal treatments decrease a lot compared to the pristine surface but are still larger than that measured in the formed wear tracks. During pure sliding and rolling-sliding, the mechanical interaction (i.e., shear stress, compress stress or impact stress) is capable of facilitating the bond break. Alternatively, the friction temperature in pure sliding or rolling-sliding tests may exceed the temperature in heating tests, resulting in a more severe degradation of polyimide composites.

Table 5  
Parameters of C 1s components on the heated polyimide composites surfaces covered without and with PAO4 oil.

C 1s component		C-C, C-H	C-O, C-N	C = O	C-F
Binding Energy (eV)		284.8	286.1	288.3	292.2
Area Fraction (%)	Without oil	71.1	8.8	7.7	12.4
	With oil	74.5	8.2	5.2	12.1

## 4.4 Traction curves without and with oil lubrications

The complex movements of the ball in the ball holder would affect the lubrication performance of the system, so we analyze the motion of the ball through an *in situ* optical observation system. To observe the contact region of the steel ball surface, the transparent glass substrate instead of polyimide composites was applied to match the requirement of the *in situ* observation system which imaged the ball surface through the glass sample. Here, we propose that the motions would be similar between the steel/glass pairs and steel/polyimide pairs due to the close friction coefficients of these two systems under the same experimental conditions (i.e., around 0.1 and 0.05 at the load of 4 N and the sliding speed of 8.5 cm/s without and with oil lubrications).

Figures 10a and b respectively show the optical images of the steel balls at various moments under the conditions without and with oil lubrications. A mark was made at the steel ball surface and its boundary was used to identify the ball motion. When the glass substrates were moving under a given load of 4 N and a sliding speed of 7 mm/s, the mark boundaries at the steel ball surfaces were found to move around 2 mm after 0.96 s without oil lubrication (Fig. 10a) and after 0.72 s with oil lubrication (Fig. 10b) due to the rolling of the steel balls during sliding process. Based on these observations, we can define the slide-to-roll-ratio of the experiment system and estimate the related traction curve based on the friction coefficients obtained at different sliding speeds. As the traction curves shown in Figs. 10c and d, the friction coefficients decrease with the increase of the slip-to-roll-ratio either without or with oil lubrication. One accepted reason is that local temperature rise at higher slip-to-roll-ratio would reduce the mechanical properties of the contacted materials and then lower the mechanical interaction between sliding/rolling interface [47, 48]. Furthermore, higher temperature might increase the viscosity of the lubricated oil that thicken the interfacial oil film [49–51]. Then, the interfacial shear stress would decrease, resulting lower friction coefficients. The results are consistent with the phenomenon of black products forming at steel/polyimide sliding/rolling interface due to the high flash temperature.

## 4.5 Mechanism of black products formation depending on motion modes

Different from the pure sliding, the rolling-sliding motion of polyimide composites against steel ball can promote the generation of black products (Fig. 3), which weakens the tribological performance of

polyimide composites especially under PAO4 oil lubrication (Figs. 3b and 4). First, the additional sliding of ball/holder contact interfaces and ball/screw contact interfaces in rolling-sliding motion cause high frictional temperature, which facilitates the rupture of C-O and C = O bonds in polyimide substrate. This hypothesis can be clarified by the pure sliding results at high temperature where the black products are formed (Fig. 8a). As the surrounding temperature increasing, the aromatic structure of the polyimide material thermally decomposes into amide monomers, then resulting in the lower mechanical strength and more severe wear [52]. The results in heating tests (Fig. 9) prove that the black products are generated along with the C-O and C = O bonds break in polyimide material although the change of molecular structure during this reaction needs to be further detected using, for example, computational simulations.

Second, PAO4 oil should participate the tribochemical reaction and facilitate the degradation of polyimide composites. It can be proved by the XPS results in Tables 3 and 5 where the relative area fractions of C-O, C-N peak and C = O peak further decrease when the PAO4 oil is added on the polyimide composites surface in the friction tests and after the thermal treatments. At last, metal oxide may also contribute to the formation of black products in rolling-sliding motion, as predicted by the XPS results in Fig. 6. The high frictional temperature may induce not only the reaction of steel with air but also the reaction of iron with the pendent oxygen of polyimide composites during the wear process [53].

To prove this mechanism, another friction test of the polyimide composites against  $\text{Al}_2\text{O}_3$  ball was conducted under PAO4 oil lubrication. As shown in Fig. 10, the friction coefficients are 0.06 in rolling-sliding motion and 0.008 in pure sliding motion, both of which are larger than that of polyimide composites and steel pairs under the same experimental conditions (Fig. 3b). Although the black products are formed after the friction test against  $\text{Al}_2\text{O}_3$  ball (upper-right inset in Fig. 10), its amount is far less than that against the steel ball (upper-right inset in Fig. 3b). It is normally that the thermal conductivity of  $\text{Al}_2\text{O}_3$  (33 W/m·K) is higher than that of steel (80 W/m·K). The higher friction force of the polyimide composites against steel ball should correspond to a larger frictional temperature [54]. Therefore, the formation of more black products under this condition should be related to the metal oxidation.

## 5. Conclusions

A friction test compounded with rolling and sliding motions was designed to simulation the complex movements of bearing parts in real applications. The movement motions change the tribological performance of polyimide composites against steel ball strongly depending on lubrication conditions. Under dry condition, the rolling-sliding motion induces a lower friction force and slighter surface wear of polyimide composites compared to the pure sliding. Under PAO4 oil lubrication, the friction coefficients in these two motions are close but the more severe surface wear is formed in the rolling-sliding motion accompanying with the generation of black products. The XPS and SEM characterizations of the worn

surfaces and the compared results of heating sliding tests and thermal treatments reveal that the PAO4 oil and the metal counterface should participate in the tribochemical reactions under the high frictional temperature. All these concerted effects accelerate the degradation of polyimide composites. The role of high temperature in the tribochemical wear was further indicated based on the decrease of friction coefficient as the slide-to-roll-ratio increasing in traction curves under the conditions without and with oil lubrications.

## **Declarations**

### **Funding**

The authors are grateful for the financial support from the National Natural Science Foundation of China (51875486) and Sichuan Science and Technology Program (2019YFH0098 and 2021YFSY0017).

### **Conflicts of interest/Competing interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### **Availability of data and material**

The datasets used or analysed during the current study are available from the corresponding author on reasonable request.

### **Code availability**

Not applicable.

### **Authors' contributions**

Yuanyuan Jiang: Conceptualization, Investigation, Original Draft & Editing

Lei Chen: Conceptualization, Writing Review & Editing, Supervision

Chen Xiao: Resources, Investigation

Ningning Zhou: Resources

Tao Qing: Conceptualization, Methodology

Linmao Qian: Conceptualization, Writing Review & Editing

### **Ethics approval**

Not applicable.

## Consent to participate

Not applicable.

## Consent for publication

We confirm that this manuscript, or any part of it, is original and has not been published before and will not be submitted elsewhere for publication while being considered by Tribology Letters.

All authors have read through the manuscript and approved the submission to your journal.

## Acknowledgements

The authors are grateful for the financial support from the National Natural Science Foundation of China (51875486) and Sichuan Science and Technology Program (2019YFH0098 and 2021YFSY0017).

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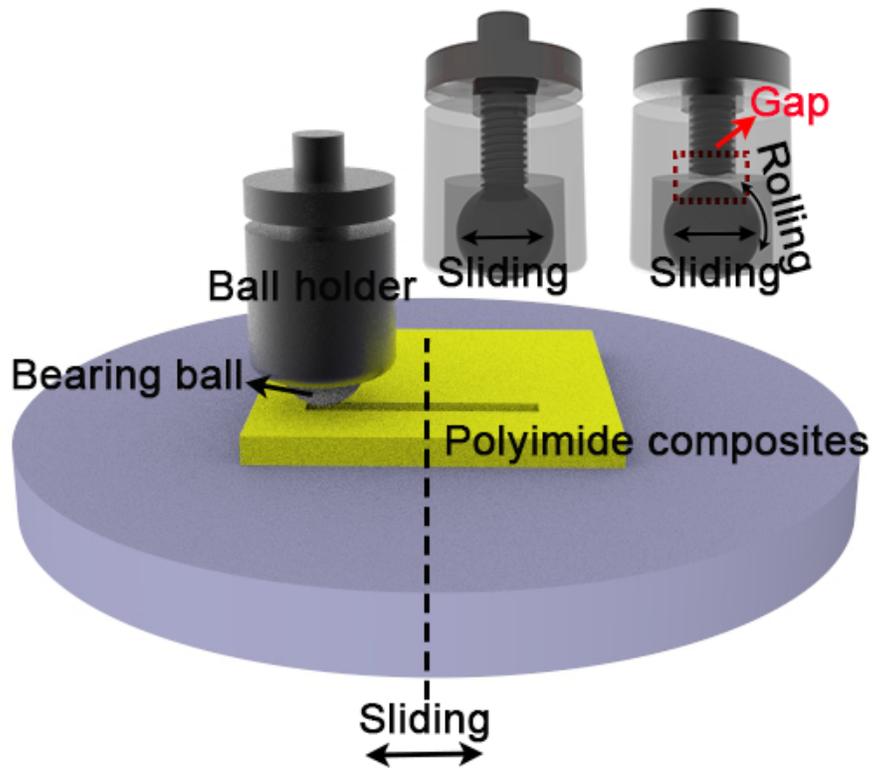
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## Figures



**Figure 1**

Schematic diagram of friction tests between steel ball and polyimide composites. The pure sliding tests were carried out when the steel ball was fixed inside the holder pressed by a screw (upper-middle inset). The sliding-rolling motion was applied as a gap was kept between the steel ball and the screw (upper-right inset).

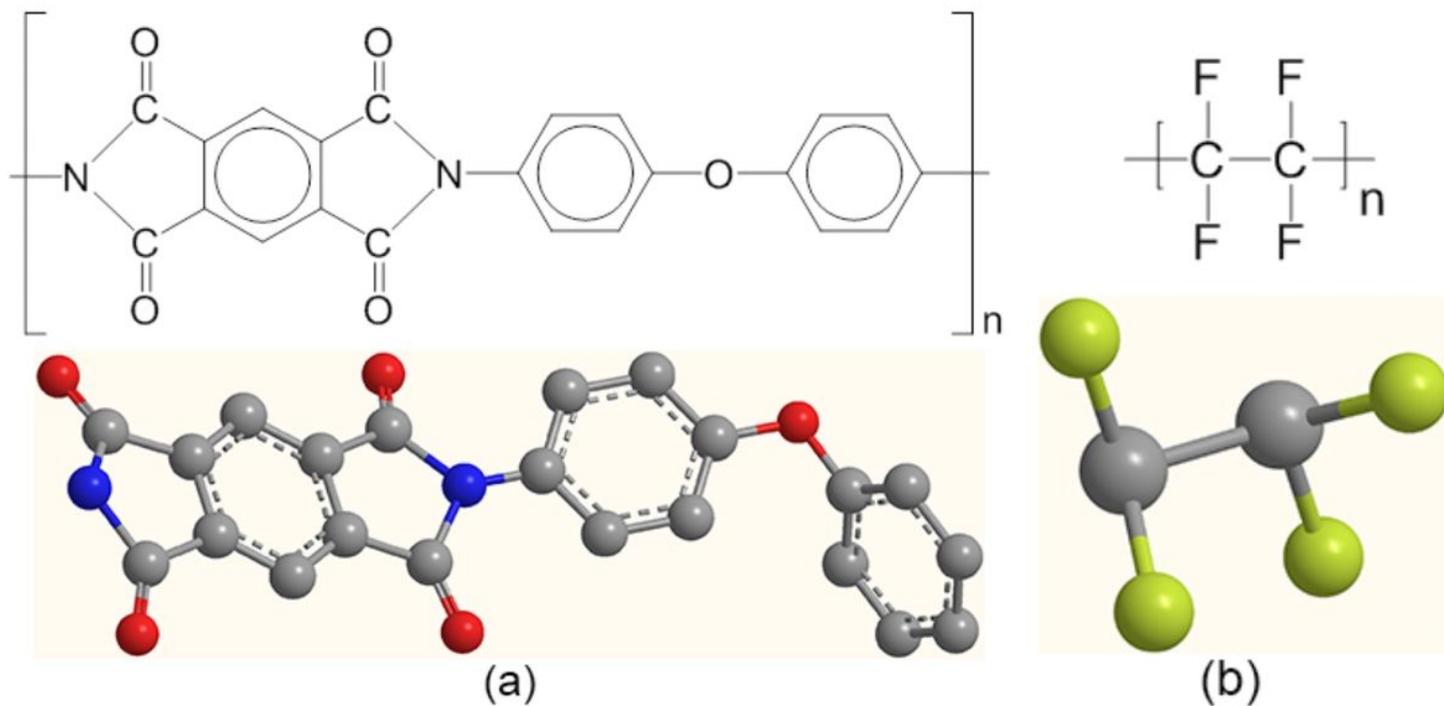


Figure 2

Chemical formulas of polyimide substrate (a) and PTFE (b)

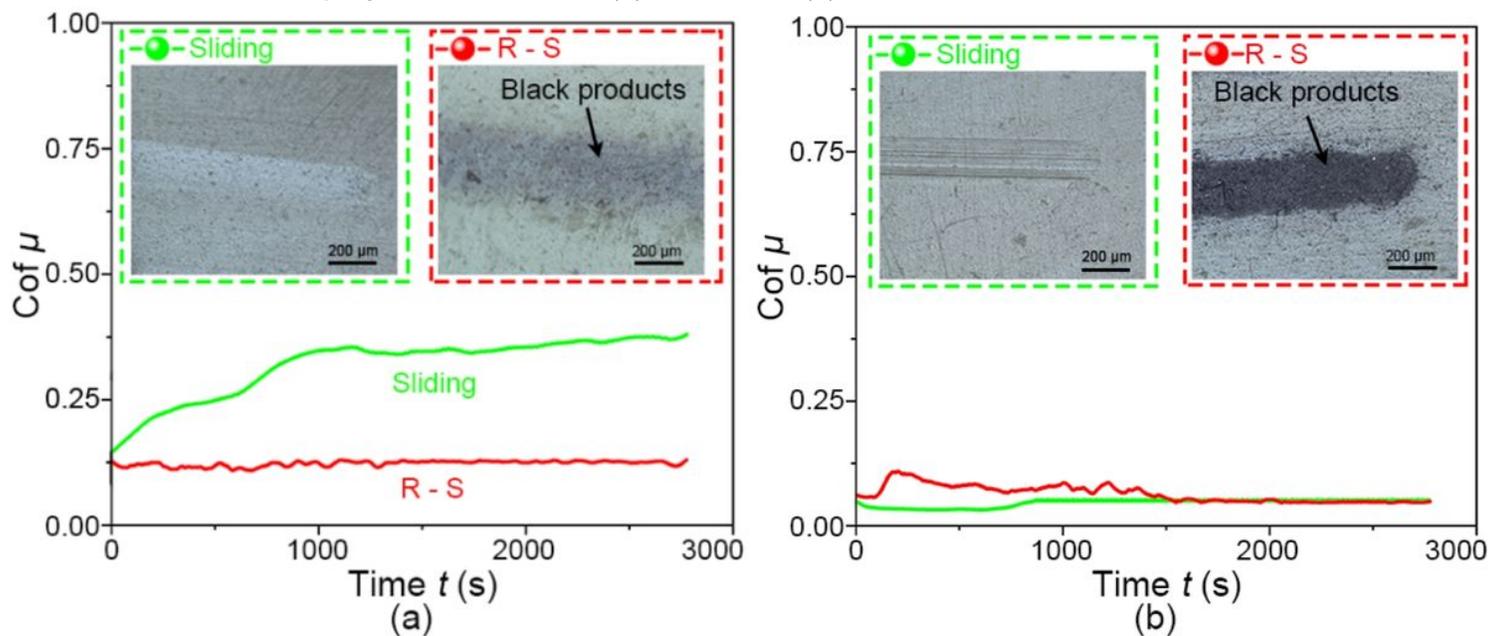
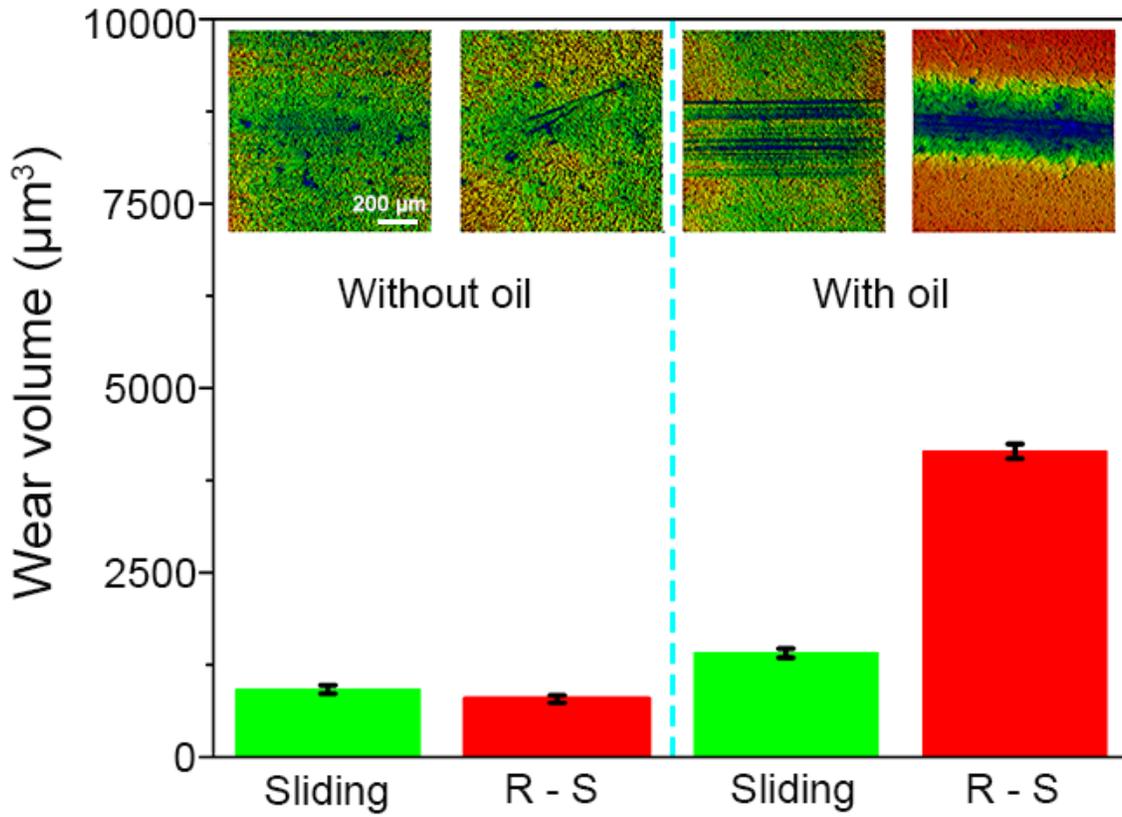


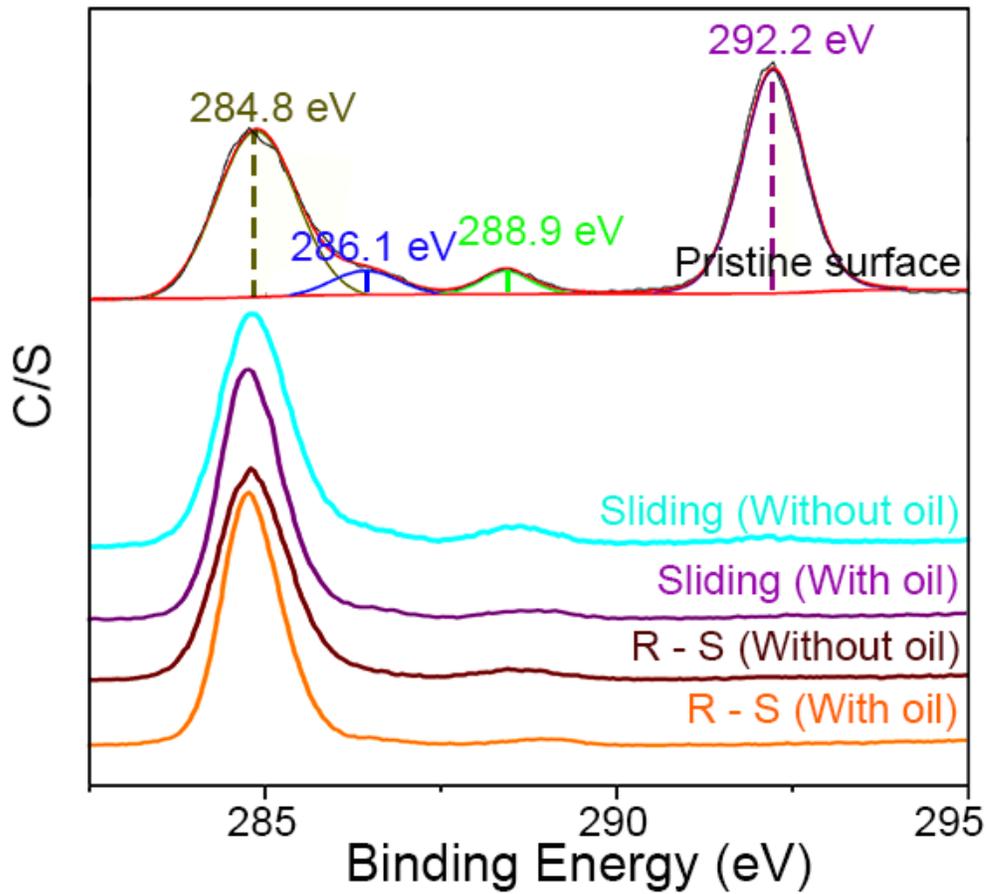
Figure 3

Friction and wear behaviors of polyimide against steel balls in pure sliding and sliding-rolling motions under the conditions without PAO4 oil lubrication (a) and with oil lubrication (b). Upper insets showing the optical images of the wear tracks formed after running 2700s under the corresponded conditions.



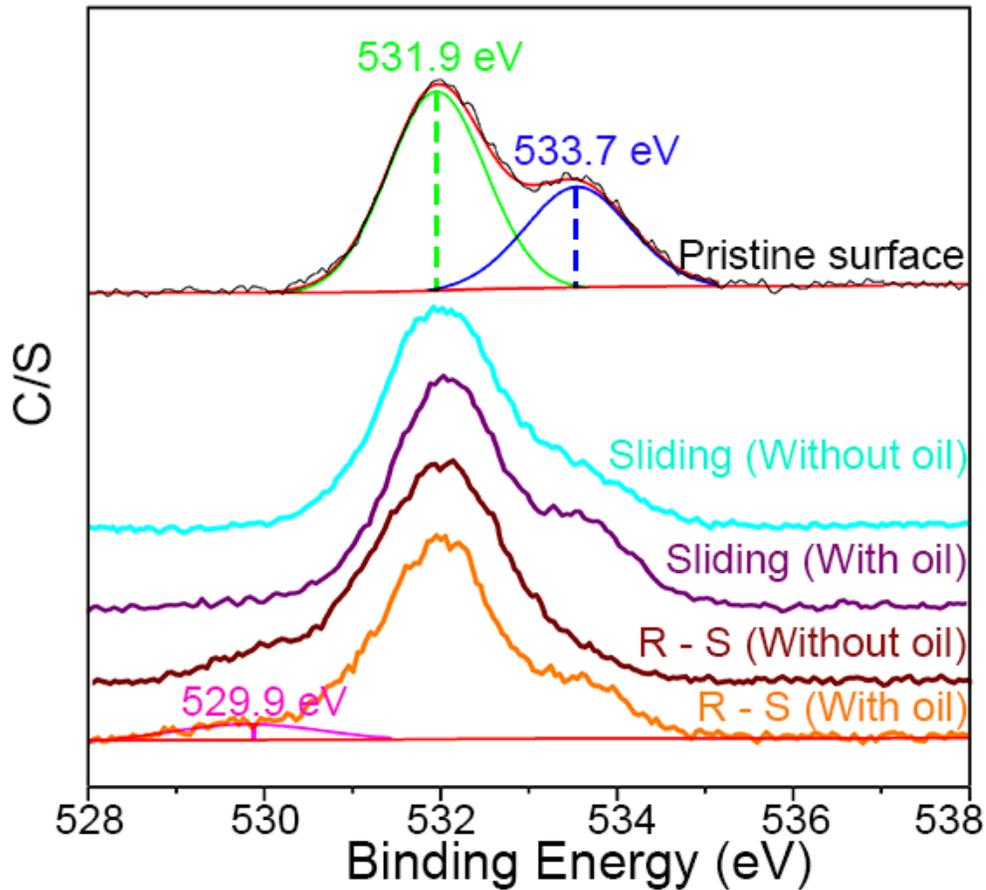
**Figure 4**

Comparison of wear volume of polyimide composites under the different experimental conditions. Upper insets showing the related WLI images of the wear tracks.



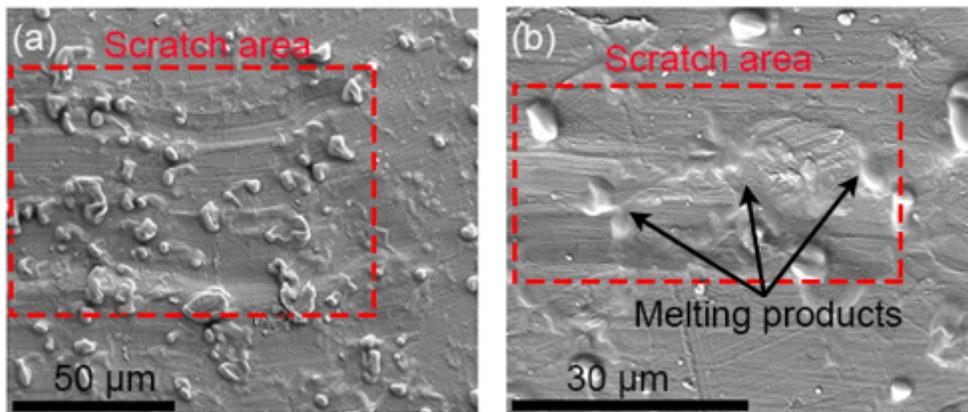
**Figure 5**

C 1s XPS spectra measured on a pristine polyimide composites surface and inside the worn tracks formed under the different experimental conditions.



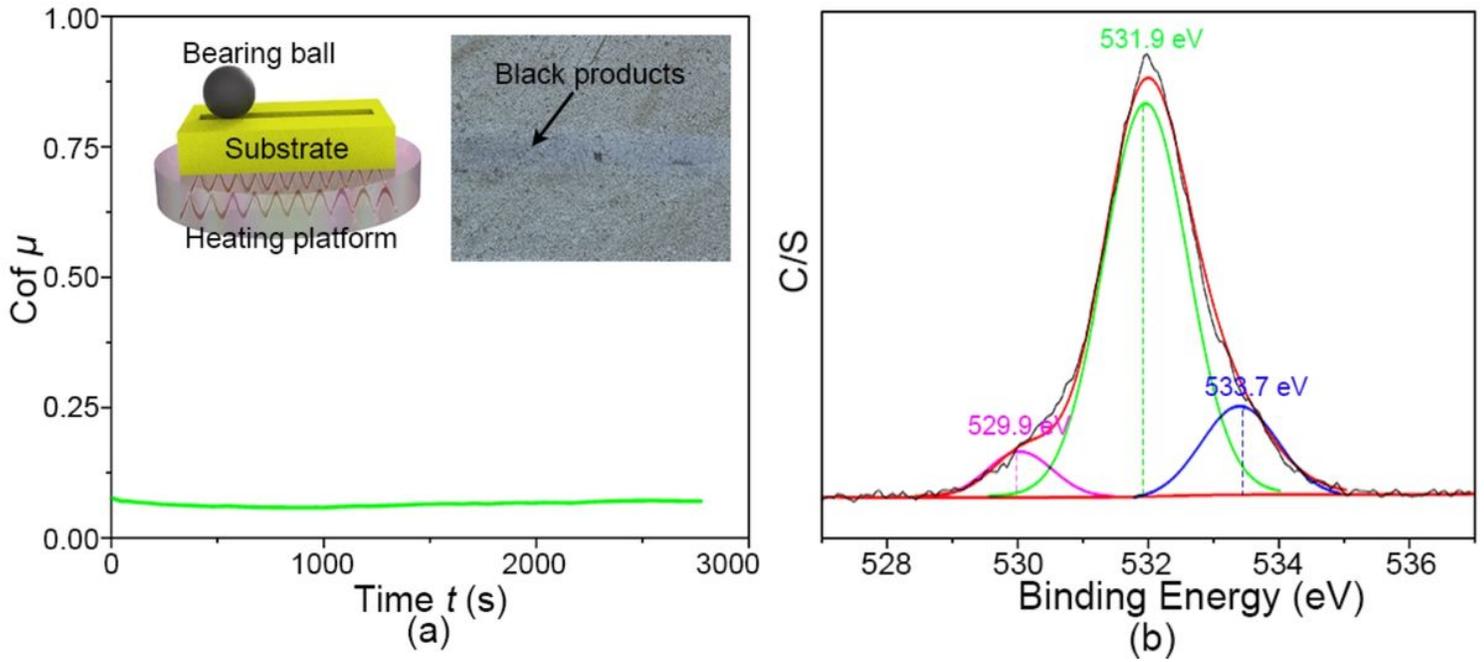
**Figure 6**

O 1s XPS spectra measured on a pristine polyimide composites surface and inside the worn tracks formed under different experimental conditions.



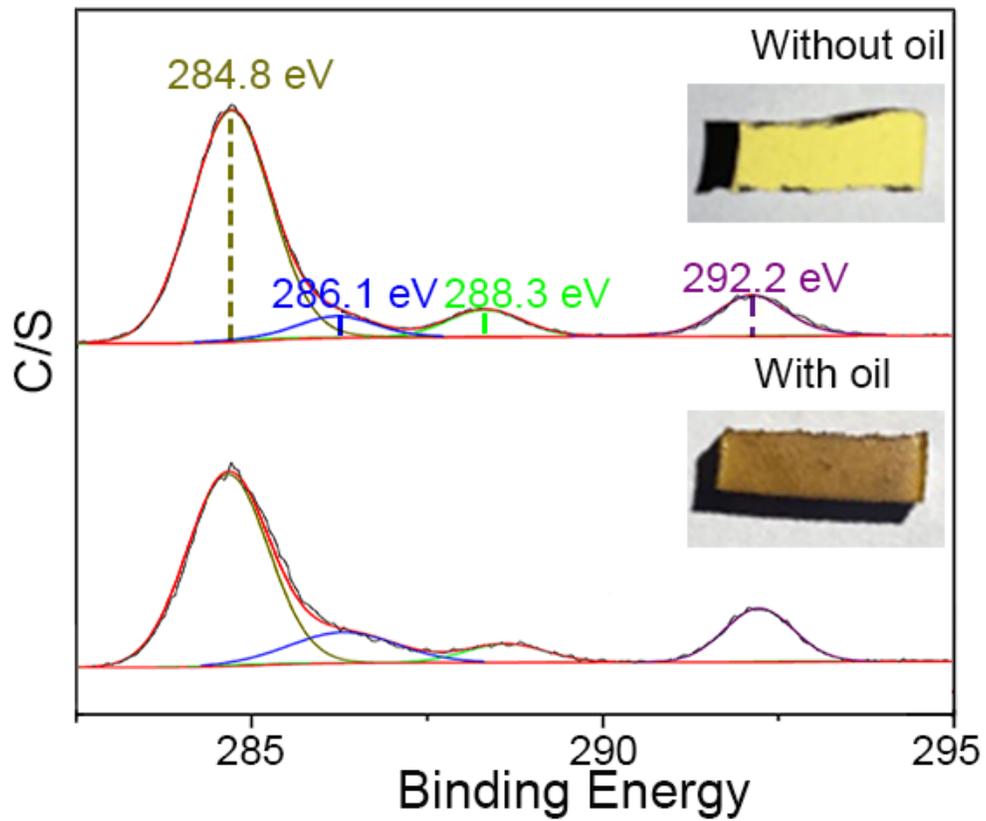
**Figure 7**

SEM images of the wear tracks on polyimide composites surfaces formed in pure sliding (a) and rolling-sliding (b) motions under PAO4 oil lubrication.



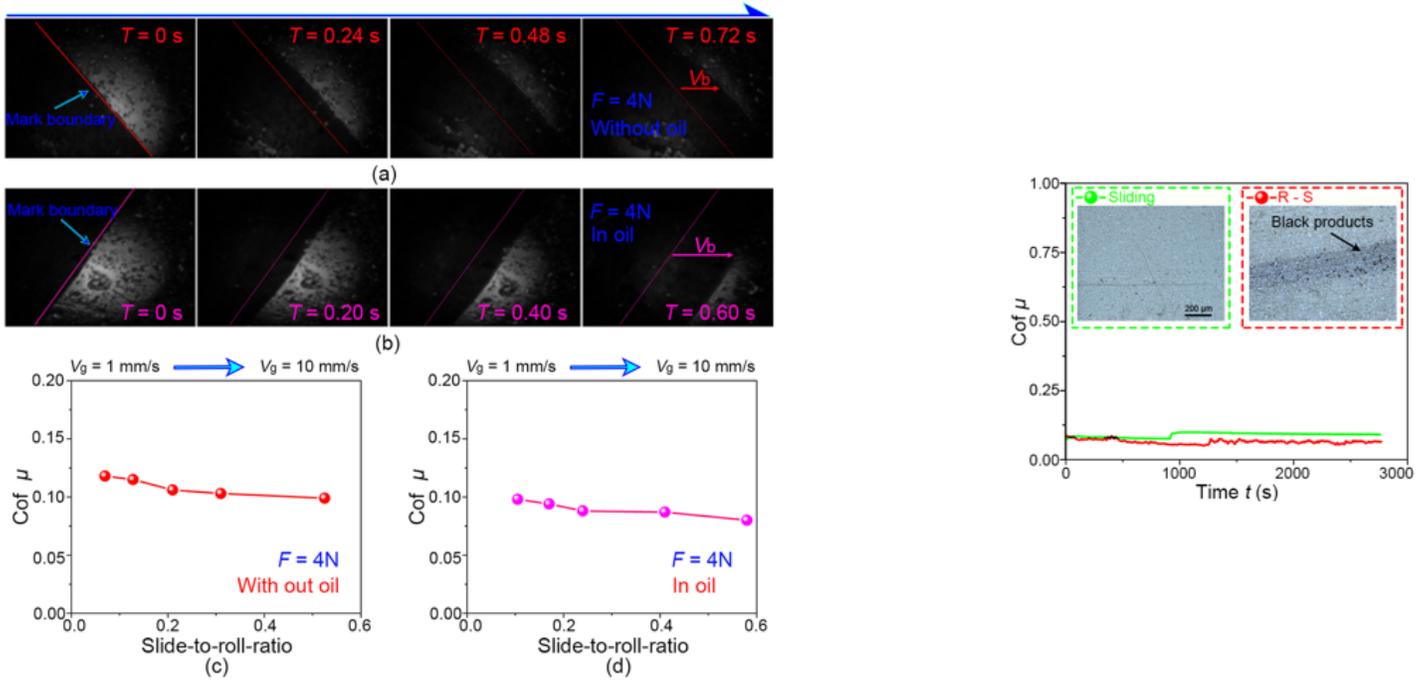
**Figure 8**

Tribological behaviors of polyimide composites sliding against steel ball under PAO4 oil lubrication at a sample surface temperature of  $\sim 120$  degrees. (a) Friction coefficient as a function of sliding time in pure sliding at high temperature. Insets respectively showing the schematic diagram of friction test at high temperature (left) and the optical image of the formed wear track (right). (b) O 1s XPS spectrum measured inside the wear track formed at high temperature.



**Figure 9**

C 1s XPS spectra measured on the heated polyimide surfaces covered without (upper) and with (bottom) PAO4 oil. Insets respectively showing the related optical images of heated polyimide samples.



**Figure 10**

In situ optical observations of the steel balls rolling during sliding against glass surfaces and the estimated traction curves. (a) and (b) respectively showing the optical images of the balls rolling at various moments without and with oil lubrications under a constant sliding speed of 7 mm/s. (c) and (d) illustrating the traction curves (friction coefficient vs. slide-to-roll-ratio) without and with oil lubrications as the sliding speed ranging from 1 mm/s to 10 mm/s. The applied load was 4 N in all tests. Friction and wear behaviors of polyimide composites against Al<sub>2</sub>O<sub>3</sub> ball under PAO4 oil lubrication. Upper insets respectively showing the optical images of wear tracks formed after running 2700 s in pure sliding and rolling-sliding motions.