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Tribophysical and tribochemical effects of a thermoplastic polyimide

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The sliding friction and wear characteristics of a thermoplastic polyimide (PI) were investigated under different PV conditions using a pin-on-disc wear tester. The tribological characteristics of the PI were divided into three wear regions on the basis of the wear degree (slight wear, medium wear and severe wear). The friction coefficient of the PI in slight wear was stable. In the medium wear and severe regions, the friction coefficients of the PI varied over a wide range, and was unstable. The reason for the variations of friction and wear in the different wear regions was attributed to the rising of PI surface temperature, which led to the variations of the physical state of PI friction surface layer. XPS analysis indicated that carbonation occurred on the friction surface during sliding. It was proved that the reaction product may reduce the friction coefficient. *© 2000 Kluwer Academic Publishers*

1. Introduction

Polyimide (PI) and its composites attract extensive concern by tribological scientists world-wide because of their high mechanical strength, good heat resistance, high stability under vacuum, good anti-radiation, and good solvent resistant [1, 2]. Most of the research work done in this field is focused on how different fillers affected tribological properties of PIs and their application. Little study has been made of the friction and wear characteristics of PI [3–7]. The authors believe that a study of the friction and wear characteristics of PI will help provide an important scientific basis for further improving their properties. Therefore, the friction and wear behavior of a thermoplastic PI under varying loads and speeds are investigated systematically in this paper. The mechanism of the impact of load and speed on the friction and wear behavior of PI is also discussed.

2. Experimental details

PI, commercially produced as YS-20 by the Shanghai Institute of Resin, China, is a thermoplastic polymer. Its chemical structure is shown in Fig. 1. Table I lists its main physical and mechanical properties. The polymer used for the tests was formed into shape at high temperature and then processed into pin samples with a diameter of 5 mm and a length of 18 mm.

A stainless steel (1Cr18Ni9Ti) disc with a diameter of 45 mm and a thickness of 8 mm was chosen as one of the mating materials of the sliding system. The surface roughness (Ra) of the disc was 0.45 μm.

The friction and wear tests were performed on the Pin-on-disc wear tester described in Fig. 2, which consists of a single pin sample of the PI and a stainless steel disc rotating with selected speed against this pin. Frictional track diameter of the pin on the disc was 25 mm. The pin was abraded with No. 800 water-abrasive paper. Both the pin and disc surface were cleaned with cotton dipped in acetone and dried before the tests. The total sliding time was 120 min. Friction coefficients were obtained by calculating the frictional torque measured by a load cell sensor. The wear rate was measured by the loss in weight of the PI.

X-ray photograph spectra were obtained using a PHI-5702 Multi-technique Small Area X-ray Photoelectron Spectrometer (XPS). Curve fitting was carried out using fitting program PHI PC-Access 6.0. The C1s binding energy of the C-H peak in phenyl group was taken as 284.6 eV for calibration purpose.

3. Results and discussion

3.1. Tribological characteristics of PI

Figs 3 and 4 were the charts of the wear and friction coefficients of PI under different sliding speeds and specific loads concluded from the tests. Based on the variation of wear rates, three regions as shown in the figures were divided. From Fig. 3, within different PV values ($\leq$0.26 MPa $\cdot$ ms^{-1}; 0.26–1.04 MPa $\cdot$ ms^{-1} and $\geq$1.04 MPa $\cdot$ ms^{-1}), the wear rates of the PI were 10^{-14} m^3/m, 10^{-13} m^3/m and 10^{-12} m^3/m (Region I, Region II, Region III) respectively, corresponding to slight wear, medium wear and severe wear of the PI. What needs to be explained is that the PI pin became softened and deformed when the specific load was more than 8×10^6 Nm^{-2} so that the tests had to be terminated.

Similarly, depending on varying friction coefficients of the PI shown in Fig. 4, the tribological characteristics of the PI could be divided into Region I, Region II and Region III. In Region I, the variation of friction

TABLE I Main properties of PI(YS-20)

Density (Kg/m^3)	1380	Impact strength (kJ/m^2)	70–120
Tensile strength (MPa)	100	Brinell hardness (MPa)	371
Breaking elongation (%)	50	Glassy transition temperature (°C)	260
Flexural strength (MPa)	210	Heat decomposition temperature (°C)	530–550

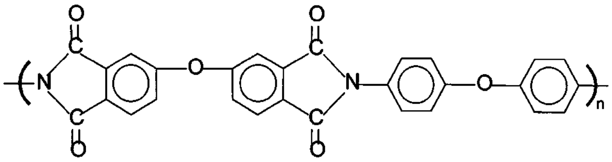


Figure 1 Chemical structure of PI(YS-20).

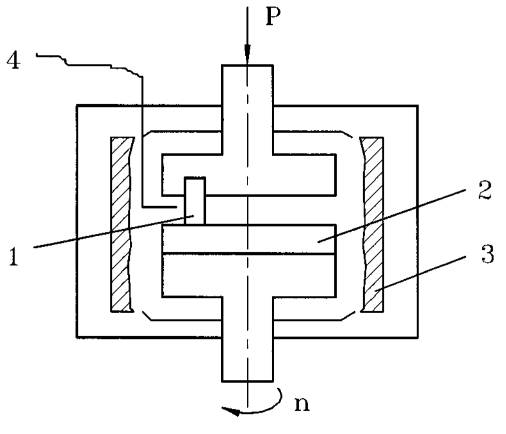


Figure 2 Schematic diagram of the pin-on-disc wear tester P-Applied load, 1-Pin specimen, 2-Disc specimen, 3-Electric furnace, 4-Thermocouple.

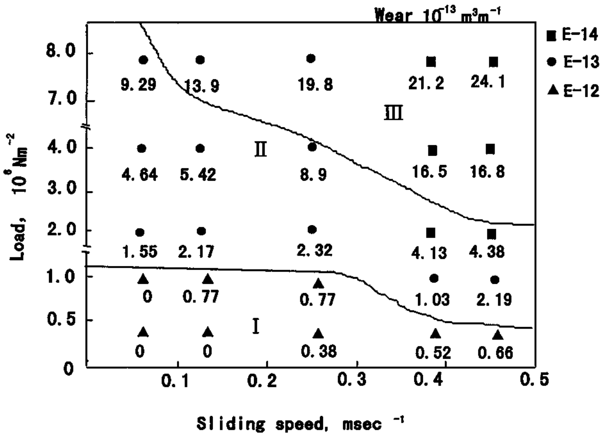


Figure 3 Wear map of PI against stainless steel.

coefficient of the PI was slight and remained about 0.30; In Region II, the variation in the friction coefficients of the PI was large and the maximum value of the friction coefficient (shown above the marks) remained 0.68 ± 0.24 and the minimum value of the friction coefficient (shown below the marks) remained 0.42 ± 0.12; In Region III, the variation in the friction coefficient was very large. The maximum value remained 0.72 ± 0.19, the lowest value remained 0.22 ± 0.02.

Typical curves of the friction coefficient of the PI with sliding time were shown in Fig. 5a–c, which were in Region I, Region II, Region III respectively. The variations in the friction coefficients of the PI with sliding time were apparent in different regions. The friction coefficient was very stable in Region I. In Region II, the friction coefficient of the PI increased slowly with sliding time extended and then decreased gradually and the higher friction coefficient remained about 60 min. In Region III, the friction coefficient of the PI increased quickly to the maximum and then decreased rapidly after sliding for 10–20 min. Finally, the friction coefficient remained stable at a value of about 0.22

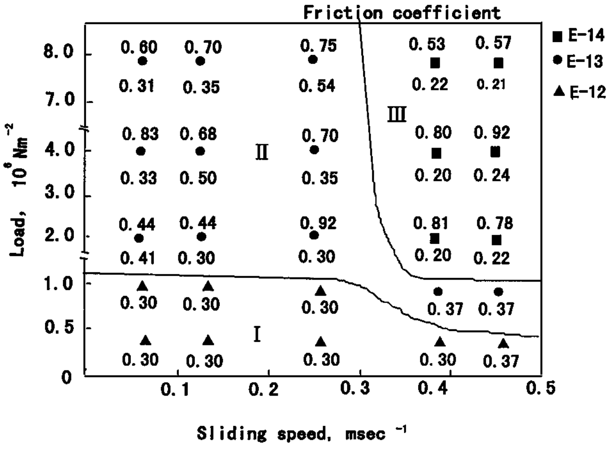


Figure 4 Friction coefficients of PI under varying PV conditions.

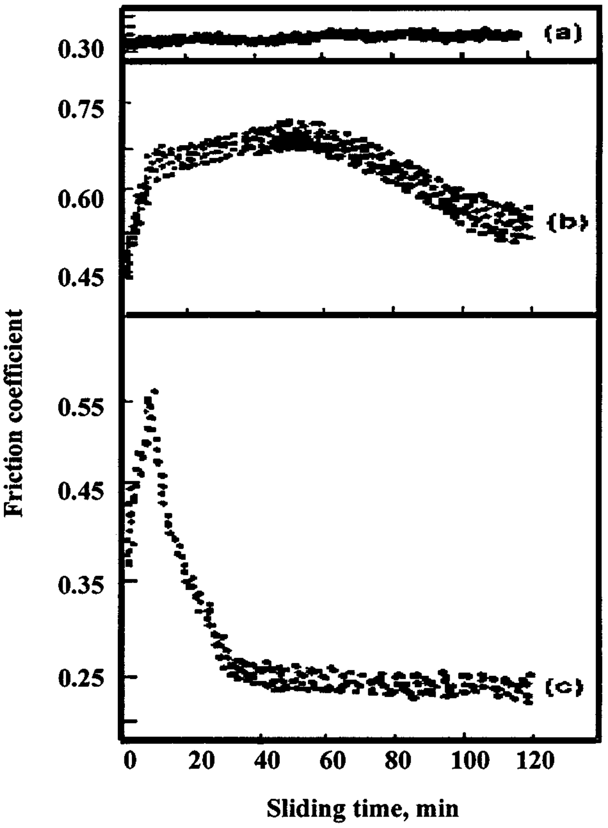


Figure 5 Typical curve variation of friction coefficients of PI with sliding time in different regions (a) Region I, (b) Region II, (c) Region III.

The division of the three regions of the wear and friction coefficients of the PI was basically identical when Figs 3–5 were compared. This indicated that a common rule was followed when the wear and friction of the PI in the three regions were under varying speed and load.

3.2. The tribophysical effect of PI

It is well known that thermoplastic polymer shows different physical states under different temperatures. In the glassy state, the molecule segment of the polymer was frozen and the deformation of the polymer under external force was little. In the viscous elastic state, the molecule segment of the polymer was likely to move and deformation was possible under external force. In the viscous flow state, the molecule main chain could move freely and the polymer displayed melting flow under external force. Under sliding friction conditions, the effect of load P and speed V led to the rising of temperature on the worn surface. The amount of heat generated was determined by the formula $Q = \mu PV$, where μ is the friction coefficient, P is the load, V is the speed. More frictional heat was accumulated on the surface of the PI under higher PV value conditions, which made the temperature increase on the surface layer, thus changing the physical state of the PI. As the adhesion and deformation in the formula of frictional force were related to the variation of the physical state of the PI, the tribological characteristics also varied.

According to the above-mentioned analyses and results from the tests in Figs 3–5, it is believed that the cause of the change of the tribological characteristics of the PI was the change of tribological characteristics of the physical state of the PI surface layer when frictional heat accumulated and reached a balance under a certain speed and load combination. It could be induced that the physical state of the PI surface could change at different regions as shown in Fig. 6. In Region I, the total amount of frictional heat was not sufficient to change the glassy state properties when the friction coefficient was relatively low and stable with the value of 0.30 (Fig. 6-I-A). But in Region II and Region III, the frictional heat was sufficient enough to form it viscous elastic state (Fig. 6-IIB and C) within a certain thickness on the surface layer and even led to the melting (Fig. 6-IIID and E) of some or part of the friction surface of the PI. As a result, the worn surface had viscous elasticity changes that matched Region II. Because of the viscous elasticity, the adhesion and deformation in the formula of frictional force played more roles and the friction coefficient increased to about 0.42. When higher frictional heat led to a large area of melted layer which matched Region III, the frictional coefficient of the PI would decrease to about 0.22 because of the reduced friction of the melted part.

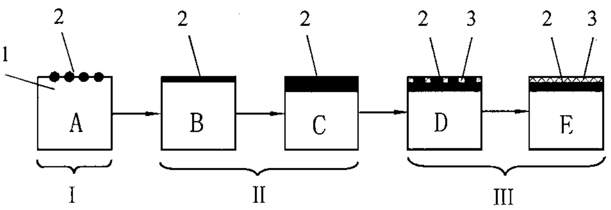


Figure 6 The schematic diagram of the change of the physical state of the friction surface layer of the PI in different regions 1. glassy state, 2. viscous elastic state, 3. viscous flow state.

As for wear, the accumulation of frictional heat led to the variation of the physical state of the PI surface and also resulted in related change [8].

It is worth pointing out that it is of great significance to further understand the role of frictional heat on the surface of polymers, whose properties show strong temperature dependency. It had been noted that under some experimental conditions, a friction coefficient peak that first increased and then decreased appeared. Whether it appeared or not and how long it lasted were quite different under different conditions. No such peak appeared in Region I. A peak value appeared both in Region II and Region III. As shown in Figs 5 and 6, some friction coefficients were hardly stable under some conditions. This showed that the change of the tribological characteristics of the polymer was attributed not only to speed and load, but also closely to the viscous elastic behavior of the PI.

In order to verify the tribophysical effect, the following test was performed. The PI pin and the stainless steel disc after the first friction test were not abraded further and were used again in the second test after they were cooled down to room temperature. The variations of the friction coefficients obtained from the two tests were compared. For polymers, the change of physical state could be reversible in the test.

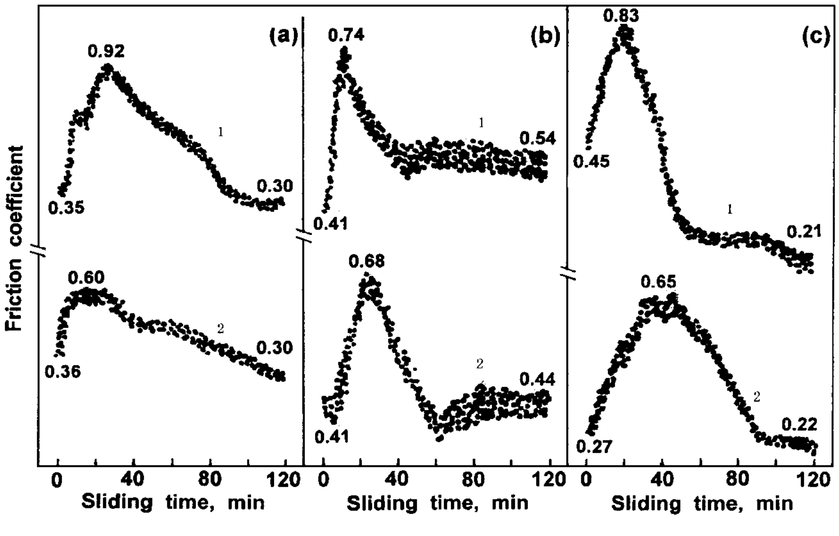


Figure 7 Variation of friction coefficients of PI with sliding time, 1-First test, 2-Second test, (a) Specific load: 2×10^6 Nm^{-2}, speed: 0.26 msec^{-1}, (b) Specific load: 8×10^6 Nm^{-2}, speed: 0.26 msec^{-1}, (c) Specific load: 4×10^6 Nm^{-2}, speed: 0.39 msec^{-1}.

The variations of friction coefficients of the PI with sliding time in the two tests under the test conditions in Region I, Region II and Region III were shown in Fig. 7. The friction coefficients of the PI first increased and then decreased with the sliding time extended in the second test, but the friction peak in the repeated test confirmed that the physical state of the PI had an important influence over the tribological characteristics. We believe that the decrease of the friction coefficient was coused by tribochemical reaction on the friction surface layer of the PI affected by heat and force during sliding and its product made the friction coefficients of the PI decrease in the repeated test.

TABLE III Curve fitting results from C1s spectra of the PI before and after friction

	Original surface	Worn surface	Functional group
Binding energy eV	288.0	288.2	$\overset{\displaystyle O}{\overset{\Vert}{—C—}}$
Atomic concentration wt %	6.1	4.9	
Binding energy eV	285.7	286.0	—C—O—
Atomic concentration wt %	16.7	11.9	
Binding energy eV	284.6	284.6	⌬,
Atomic concentration wt %	77.2	83.2	Graphitised carbon

3.3. The tribochemical effect of PI

In order to understand whether chemical reaction occurred and its mechanism, the results by XPS analysis of relative atomic concentration of the PI surface before and after sliding were listed in Table II.

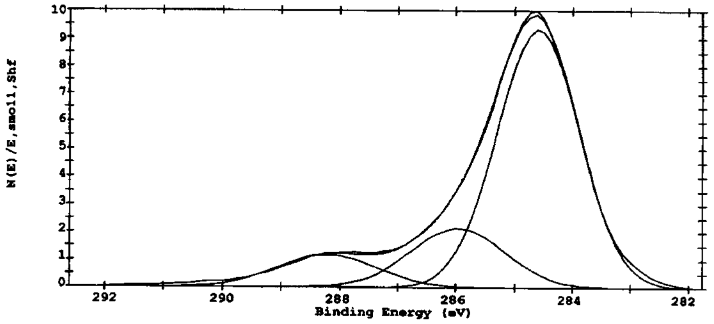
As shown in Table II, the relative atomic concentration of C on the worn surface of the PI after the test increased, where by both O and N decreased. This result proved the chemical reaction that occurred on the worn surface of the PI.

TABLE II The relative atomic concentration of the PI before and after friction test (specific load 8 × 10^6 Nm^{-2}, speed: 0.46 m/s, sliding time: 120 min)

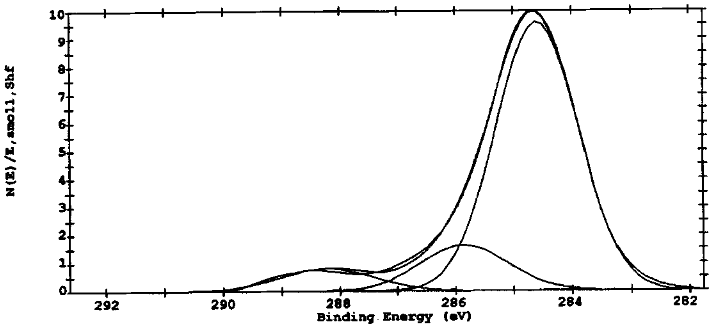
	Relative atomic concentration (wt.%)		
	C	N	O
Original surface	81.8	2.7	15.4
Worn surface	84.8	2.5	12.7

Fig. 8a and b showed the C1s spectra of the PI before and after sliding. The curve-fitting results were listed in Table III. Functionality corresponding to binding energy of C1s was also listed in Table III.

As seen from Fig. 8, the binding energy peaks of C1s before and after sliding consisted of a main peak and two chemical shifted signals. This indicated that no new functional group appeared on the surface of the PI. But it is noticed that the atomic concentration of C in the functional group around 284.6 eV that matched the worn surface of the PI increased by 6%. As the formation of benzyl needed high heat under both high temperature and high pressure, it was impossible for it to form during sliding. Therefore, the increase of C



a Original surface



b Worn surface

Figure 8 C1s spectra of the PI before and after friction 120 min.

atomic concentration on the friction surface of the PI might be caused by the generation of graphitised carbon, that is, the chemical reaction consisting of carbonisation which occurred graphitised carbon, that is, the chemical reaction consisting of carbonisation which occurred on the surface of the PI during sliding. The results further supported the inference of the mechanism in Fig. 7.

4. Conclusion

a. Wear of the PI could be divided into three regions depending on load and speed: slight wear, medium wear and severe wear.

b. The main mechanism of different friction and wear characteristics lies in the variation of physical state on the friction surface of the PI as a result of the rising of the PI surface temperature.

c. During sliding, tribochemical reactions with carbonisation dominating occurred on the PI surface and its products may reduce friction coefficients of the PI.

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