Tribological Behavior of Carbon Nanotube and Polytetrafluoroethylene Filled Polyimide Composites Under Different Lubricated Conditions

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ABSTRACT: The friction and wear behavior of polyimide (PI) composites reinforced with carbon nanotube (CNT) and polytetrafluoroethylene (PTFE) were comparatively evaluated under dry sliding, water-, oil- or alkali-lubricated condition. The wear mechanisms of the composites were also discussed. Results indicate that, when comparison with the dry friction situation, PI-based composites results lower friction coefficients and wear rates under oil- or alkali-lubricated condition. The lowest wear rate of the CNT/PTFE/PI composite is recorded as 1.2×10^{-6} mm^3/Nm during the composite sliding in alkali, which is only about 40% of the value sliding under dry friction condition. The worn surface of neat PI under dry sliding is characterized by severe adhesive wear, whereas abrasive wear is the main character for CNT/PTFE/PI composites. The worn surfaces of CNT/PTFE/PI composites sliding in oil or alkali lubricated condition are smoother than those under dry or water condition. J Appl Polym Sci 121: 1574–1578, 2011

Key words: carbon nanotube; polyimide; polytetrafluoroethylene (PTFE); composites

INTRODUCTION

Carbon nanotube (CNT) has motivated numerous interesting fields of research since Iijima[1] initially identified their structures in 1991, which is because of its excellent electrical,[2] mechanical,[3] thermal, and magnetic properties,[4] low density, high surface area, and high chemical stability.[5] So far CNT has been widely used in polymer composites, including matrices such as polyethylene,[6] poly(methyl methacrylate),[7] polyurethane,[8] and so on.

Polymers are finding wide acceptance in tribological applications because of their low friction against steel counterparts and the self-lubricating ability.[9] Polyimide (PI) is a class of polymers that are known for their stability at high temperature, which have been found applications in the composite and microelectronics industries.[10] However, high friction coefficient and wear rate of pure PI limits its wider use.[11] Thus, a lot of efforts have been made to reduce friction coefficient and wear rate of neat PI by means of incorporation of particles and fibers in the composites.[12]

CNT/PI nanocomposite received common attentions for its good properties.[13] Chen et al.[14] found that CNT significantly increased the wear resistance of polytetrafluoroethylene (PTFE) composites and decreased their coefficient of friction. Meanwhile, Cai et al.[11] found CNT could effectively enhance the friction-reduction and antiwear capacity of the nanocomposite because it increased the load capacity and mechanical strength of the CNT/PI.

Many studies about the tribological properties of PI composites were done under dry conditions.[15] However, there are numerous sliding applications in which water or oil is either deliberately introduced as a coolant, e.g., in rolling mill bearing, or present as a working fluid. In the latter case, a marine environment can pose special problems because of the abrasive contamination and the corrosiveness of sea water toward many metal materials.[16] Furthermore, almost no information is available about the friction and wear behaviors of polymers sliding in alkali solution.

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In this article, PTFE was used as a solid lubricant in PI, owning to its characteristic of low friction coefficient, to improve PI's tribological properties.[17] The purpose of this article is to investigate the tribological behavior of PI composites filled with CNT and PTFE sliding against steel under different lubricated conditions, so as to acquire some insight into the selection of appropriate matching materials for actual application under different conditions. The friction and wear mechanisms of the CNT/PTFE/PI composites under different lubricated conditions were also examined.

EXPERIMENTAL

Materials

In this work, the powder of PI (YS-20, 1–10 μm) was supplied by Shanghai Research Institute of Synthetic Resins, and PTFE was commercially obtained from Dupont (7A-J, 25 μm commercial product). The CNT used in this study was purchased from Shenzhen Nanotechnologies, China. The CNT synthesized by the chemical vapor deposition method. Its diameter and length was about 50 nm and 20 μm, respectively. Deionized water, liquid paraffin, and sodium hydroxide were purchased from commercial sources and used as received.

Preparation of PI composites

It was found that polymer composites with 10–20 vol % CNT exhibited excellent antiwear properties,[11,14] although too much filler content will make the composite become fragile. As a result, the fraction of CNT was set as 10 vol %. The fraction of PTFE was set as 15 vol %.

All the materials were dried at the temperature of 110°C in the air oven for 12 h before use. The preparation method was introduced as follows: at first, PTFE and PI were ground with a ball mill for ~ 1 h to reduce their particle size. After that the PI, the PTFE, and the CNT powder at selected proportions were blended mechanically. Each mixture was then dispersed in 30 mL of ethanol and sonicated in an ultrasonic bath for 30 min. Then the dispersion was partially dried under vacuum (70 mbar) at 50°C for 5 min, sonicated for another 30 min, and heated until the ethanol was completely eliminated. Subsequently, the mixed powers were compressed under the pressure of 20MPa and heated to 365°C in a mold at a heating rate of 8°C/min. The compressed composites were held at 365°C for 45 min and then cooled to ambient temperature in the mold while keeping the pressure unchanged. For friction and wear tests, the block was cut into a shape that was shown in Figure 1 (c) with 26 mm in outer diameter and 22 mm in inner diameter and 2.5–3 mm in shoulder height.

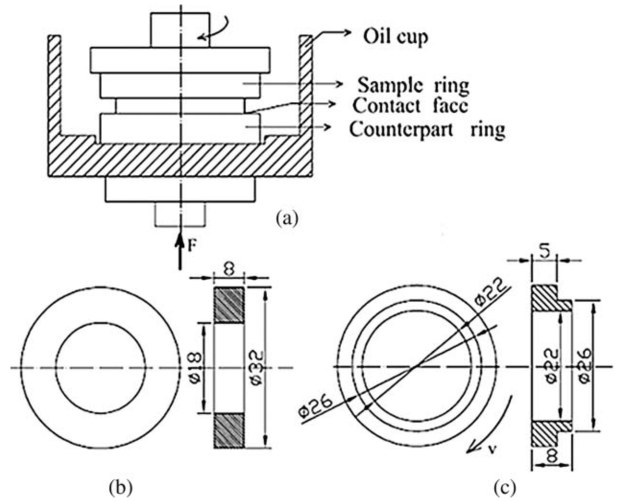

Figure 1 The schematic diagram of wear contact (a) ring on ring contact, (b) counterpart ring, and (c) sample ring.

Friction and wear tests

The friction and wear tests were conducted on a ring-on-ring model friction and wear tester. The contact schematic diagram of frictional parts is shown in Figure 1. The counter-face material was AISI1045 steel and was ring-shaped [Fig. 1(b)]. Lubricants used in this paper included deionized water, liquid paraffin, and10 wt % sodium hydroxide alkali solution. Sliding was performed under ambient dry friction conditions over a period of 1 h at sliding velocity of 1.4 m/s, normal load of 200 N. The ambient temperature was ~ 25°C and relative humidity was 50 ± 5%. The friction duration was 60 min. Before each test, the surfaces of specimen and counterpart ring were polished with 800 grit paper to a surface roughness of 0.2–0.4 μm and cleaned with acetone. The samples were put into oven at 135°C for 4 h before weighing. The friction coefficient was calculated from the frictional torque data that was recorded by a computer once a second. Results reported were the average value of the whole process. The specific wear rate, W_r (mm^3/Nm) was calculated by the following equation:

$$W_r = \frac{\Delta m}{L \cdot \rho \cdot F_N} \quad (1)$$

where Δm is the mass loss in g, L is the sliding distance in m, ρ is the density of the composite in g/mm^3, and F_N is the normal load in N.

In each experiment, the measurements were performed three times and the average of the three test results was reported. The microstructure of the worn

TABLE I
Friction Coefficient and Wear Rate of PI Composites Under Dry Friction Condition

Item	Pure PI	PTFE/PI	CNT/PTFE/PI
Friction coefficient	0.23	0.15	0.12
Wear rate ($\times 10^{-6}$ mm^3/Nm)	28.6	4.1	2.9

surface and the transfer behavior were investigated by scanning electron microscope (SEM) (QUANTA-200).

RESULTS AND DISCUSSION

Friction and wear properties of PI composites under dry condition

The friction coefficient and wear rate of neat PI and its composites filled with PTFE and CNT under dry sliding condition are shown in Table I. As seen in Table I, the friction coefficient and wear rate of PI decrease obviously after adding PTFE in the neat PI. This indicates that PTFE has good lubricating effect on PI. Furthermore, after adding CNT in the PTFE/PI composite, the friction coefficient and wear rate decrease markedly, which are only about 50% and 10% of that of neat PI, respectively. Therefore, conclusion can be drawn that CNT has outstanding ability in decreasing the friction coefficient and increasing the wear resistance of PI composites.

Friction and wear properties of PI composites under different lubricated conditions

Figures 2 and 3 show the friction coefficient and specific wear rate of PI composites under different lubrications sliding at 200 N and 1.4 m/s. It is seen from Figure 2, the friction coefficient of CNT/PTFE/PI composites is decreased when the composites slide under lubricated conditions. This is attributed to the boundary lubricating ability of the medium. Especially, the CNT/PTFE/PI composite registers the lowest friction coefficient when it slides against the steel under alkali solution lubricated condition, which indicates, for CNT/PTFE/PI composite, the effect of alkali on reducing friction force, which is better than that of water and oil. The relationship of the friction coefficient of PI composites sliding under different lubricated situations is $\mu_{alkali} < \mu_{oil} < \mu_{water} < \mu_{dry}$.

From Figure 3, it is clear that the antiwear ability of CNT/PTFE/PI composites, when compared with the value under dry friction condition, is improved when the composite slides under alkali or oil condition. Although the situation is different, when the composite slides in water, whose wear rate is about 2.2 times higher than the value under dry friction

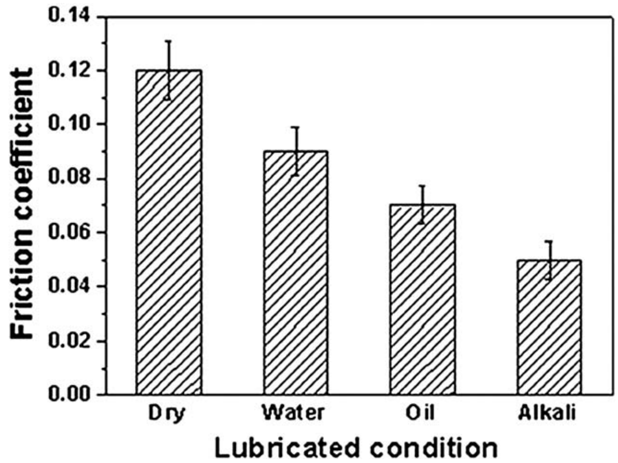

Figure 2 Friction coefficient of CNT-PTFE-PI composites under different lubricated situations.

condition, it means that water deteriorates the antiwear properties of CNT/PTFE/PI composites, which may be attributed to the reason that water destroys the bonding between filler and the matrix causing the easier separation between the filler and matrix during sliding. Moreover, the lowest wear rate is recorded as 1.2×10^{-6} mm^3/Nm during the composite slides in alkali, which is only $\sim$ 40% of the value sliding under dry friction condition.

SEM analysis of the worn surface

Figure 4 shows the scanning electron micrographs of the worn surfaces of the neat PI and CNT/PTFE/PI composites sliding under different lubricated conditions. The worn surface of neat PI under dry sliding is characterized by severe adhesive wear and microcracking [Fig. 4(a)]. After filling PTFE in PI, the adhesive wear is reduced [Fig. 4(b)]. Contrary to the above, the CNT/PTFE/PI composite is characterized

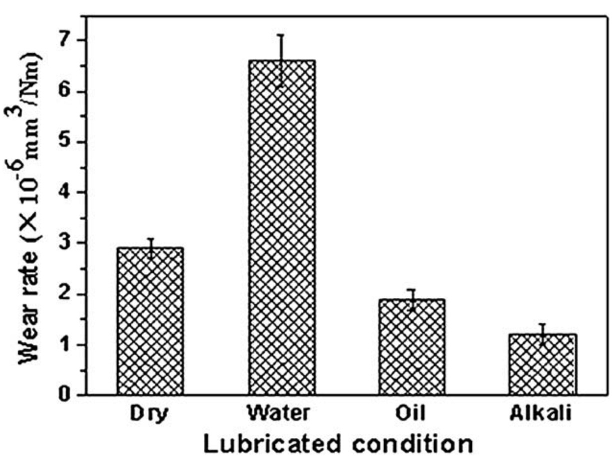

Figure 3 Wear rate of CNT-PTFE-PI composites under different lubricated situations.

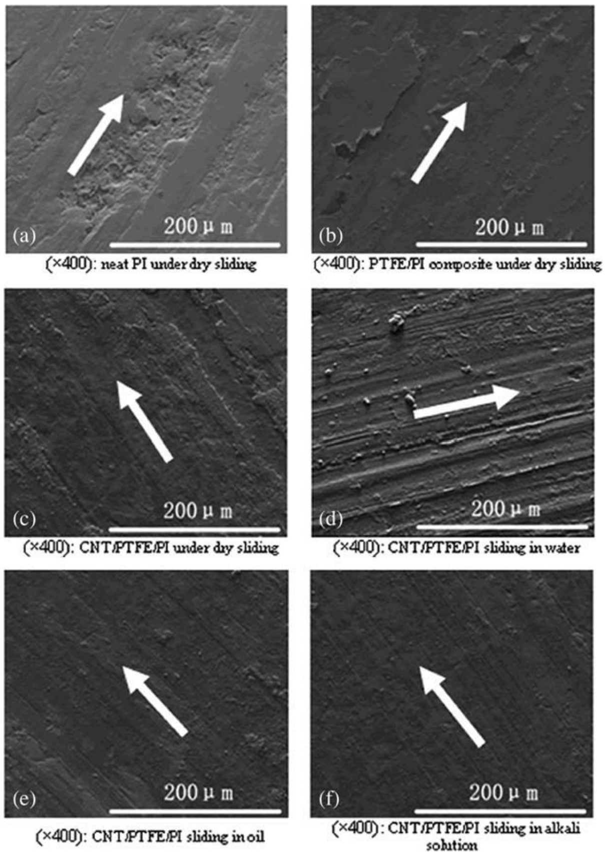

Figure 4 SEM morphologies of the worn surfaces. Arrow indicates the sliding direction.

by abrasive wear under all conditions [Fig. 4(c–f)]. The worn surfaces of CNT/PTFE/PI composites are very rough after sliding under dry friction [Fig. 4(c)] or water lubricated condition [Fig. 4(d)].

Moreover, almost no obvious scuffing is visible on the worn surface of the CNT/PTFE/PI composite under alkali-lubricated condition [Fig. 4(f)], whereas deep furrows are visible on the worn surface sliding under dry friction condition [Fig. 4(c)] or water lubricated condition [Fig. 4(d)]. The less scuffing on the worn surface of CNT/PTFE/PI composites under oil- or alkali-lubricated condition accounts for its corresponding better wear resistance among the tested composites. This observation also agrees with the different wear-resistance of the composite under dry, water, oil, or alkali lubricated condition.

SEM analysis of the transfer film

The scanning electron micrographs of the transfer films of the neat PI and CNT/PTFE/PI composites sliding in different medium are shown in Figure 5. A large amount of transferred debris can be observed on the surface of steel for neat PI or PTFE filled PI sliding under dry friction condition [Fig. 5(a,b)]. After filling CNT in PTFE/PI, the surface of the counterpart becomes smooth [Fig. 5(c)]. There are lots of obvious furrows on the counterpart surface after the CNT/PTFE/PI composite sliding in water, and no signs of PI transfer are observed in this case [Fig. 5(d)]. This means that water inhibits the transfer of PI onto the metal counterpart surface. On the contrary, a uniform transfer film is gained on the counterpart surface after the composite slides under oil or alkali lubricated condition [Fig. 5(e,f)].

It is reported the polar imide radicals in the composite molecules cause the composite to be liable to adsorb water,[18] which may destroy the interaction between CNT and the matrix during the CNT/PTFE/PI composite slides in water. As a result, the wear rate of the composite in water is much higher than that under dry friction condition. Although PI is apt to melt at high pressure velocity (PV) values, the cooling effect of oil or alkali solution hinders the melting of PTFE/PI surface and inhibits the generation of wear debris on the counterpart surface. Thus, a smoother worn surface CNT/PTFE/PI composite is observed under oil or alkali lubrication, when compared with that under dry sliding.

Water and alkali solution has different effects on the friction and wear properties that may be attributed to the fact that the viscosity of 10 wt % alkali

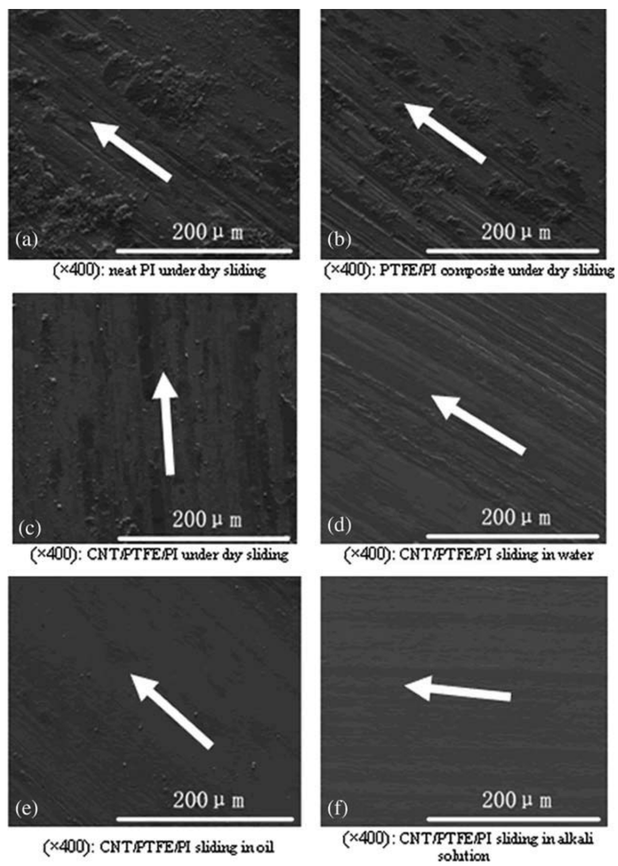

Figure 5 SEM morphologies of the transfer films. Arrow indicates the sliding direction.

solution is 1.77 times higher than that of water at the temperature of ~ 40°C during test (0.00115 Pas for10 wt % alkali solution and 0.00065 Pas for water).[19] The thickness of alkali boundary layer is bigger than that of water. So the cooling and boundary lubricating effects of alkali solution is better than that of water.

The above investigations on the worn surfaces and transfer films on the counterpart surfaces are also consistent with the wear rate data of CNT/PTFE/PI composites.

CONCLUSIONS

The friction and wear properties of PI composites reinforced with CNT and PTFE under dry, water, oil, and alkali lubricating situations were studied. Some insights into the friction and wear mechanisms under different lubricating situations were also investigated. The following conclusions can be made based on this study:

1. The incorporation of CNT and PTFE in PI contributes to greatly decrease the friction coefficient and wear rate of PI composites sliding against steel under dry friction condition. Furthermore, the friction coefficient and wear rate of CNT/PTFE/PI composite are only approximately 50% and 10% of that of neat PI, respectively.
2. In comparison with the tribological properties of CNT/PTFE/PI composites under dry friction condition, the material shows lower friction coefficient and much better antiwear ability under oil or alkali lubricated condition. Although the wear rate is increased, when it slides in water, whose wear rate is about 2.2 times higher than that under dry friction condition, the lowest wear rate is recorded as 1.2×10^{-6} mm^3/Nm during the composite slides in alkali, which is only ~ 40% of the value measured under dry friction condition.
3. The worn surface of neat PI under dry sliding is characterized by severe adhesive wear and micro cracking, whereas abrasive wear is the main character for CNT/PTFE/PI composites. The worn surfaces of CNT/PTFE/PI composites sliding under oil or alkali lubricated condition are smoother than that under dry or water lubricated condition.

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