Effects of the Combination of Solid Lubricants and Short Carbon Fibers on the Sliding Performance of Poly(ether imide) Matrix Composites

Guijun Xian, Zhong Zhang

Institute for Composite Materials, University of Kaiserslautern, 67663 Kaiserslautern, Germany

ABSTRACT: Solid lubricants, that is, graphite flakes and poly(tetrafluoroethylene) powders, were incorporated with short carbon fibers into a poly(ether imide) matrix to improve the tribological performance. Wear tests were performed with a polymer pin against a mild steel counterpart at a constant sliding speed of 1 m/s under various temperatures and contact pressures. Composites filled with equilibrium contents of solid lubricants and short carbon fibers, that is, 10 vol % of each filler, exhibited the lowest wear rate and friction coefficient. The relatively lower concentration of solid lubricants adversely affected the wear resistance, whereas the friction coefficient did not vary significantly in comparison with the friction coefficient of the composites filled with only short carbon fibers. The improved tribological behavior was attributed to more continuous and effective friction films formed on the material pairs during sliding. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 94: 1428–1434, 2004

Key words: fibers; polytetrafluoroethylene (PTFE); films; composites

INTRODUCTION

One of the traditional concepts for the improvement of the friction and wear behavior of polymeric materials is to reduce their adhesion to the counterpart material and to enhance their hardness, stiffness, and compressive strength.[1,2] The incorporation of solid lubricants [graphite flakes, poly(tetrafluoroethylene) (PTFE), etc.], which help in forming transfer films between the subsurfaces of both sliding components, has been widely used for the first aim. Carbon, glass, or aramid fibers are frequently applied to enhance the mechanical profiles. Short carbon fibers (SCFs) possessing multiple functions (e.g., lubrication, reinforcement, and heat conductivity) seem more favorable for tribological applications.[1] Fiber reinforcements normally tend to disrupt the transfer films formed on the counterpart,[3] and this process deteriorates the tribological behavior.[4,5] Therefore, it is necessary to optimize the fractions of lubricants and reinforcements to optimize tribological behavior. There are several successful examples of improved tribological behavior of polymer materials through the addition of solid lubricants and fiber reinforcements.[1,2,6–9] For a poly(ether ether ketone) (PEEK) matrix, an optimum composition with 10 vol % PTFE, 10 vol % graphite flakes, and 10 vol % SCFs exhibited an overall good friction and wear performance within broad pressure–velocity (*pv*) and testing temperature ranges.[6,7] Moreover, the addition of short glass fibers and PTFE or MoS_2 together enhanced the wear resistance of poly(ether sulfone) remarkably.[8] Poly(ether imide) (PEI) modified with short glass fibers (SGF) (30 wt %) and PTFE (15 wt %) showed tribological behavior superior to that of PEI filled with only SGF at both room temperature and 150°C.[10]

PEI is a high-performance amorphous thermoplastic possessing excellent mechanical properties even at elevated temperatures because of its high glass-transition temperature (ca. 217°C). The outstanding features of PEI also make it a favored potential candidate for tribological applications, for which a high service temperature is a critical issue.[10–14] Neat PEI exhibits a relatively low coefficient of friction, but the wear resistance is less favorable.[12] Some efforts[10,13,14] have been made to modify PEI wear behavior through the incorporation of glass fibers with or without solid lubricants. It is evident that a combination of glass fibers and solid lubricants can significantly improve PEI sliding wear resistance, in comparison with that of composites filled only with glass fibers.[9] In this study, both solid lubricants (PTFE powders and graphite flakes) and SCFs were applied to a PEI matrix. The tribological behavior was assessed with a block-on-ring or pin-on-disc testing rig at various *pv* and temperature conditions. The aim was to investigate the combined influence of solid lubricants and SCFs on

Correspondence to: Z. Zhang (zhong.zhang@ivw.uni-kl.de).

TABLE I
Material Compositions

Designation	PEI (vol %)	PTFE (vol %)	Graphite (vol %)	SCF (vol %)
A	70	10	10	10
B	85	5	5	5
C	75	5	5	15
D	80	5	0	15
E	90	5	5	0
F	90	0	0	10

the tribological behavior of PEI and to study the corresponding wear mechanisms.

EXPERIMENTAL

Materials and compounding

PEI (1000, Ultem, General Electric Plastics GmbH, Russelsheim, Germany) was used as the matrix material. Graphite flake (9039, superior, Chicago, USA), PTFE (9205, Dyneon GmbH & Co. KG, Burgkirchen, Germany), and SCF (M2007s, Kreca Chop, Tokyo, Japan) were chosen as the fillers. A twin-screw extruder was used to blend each composition (Table I). The screw temperature was set at 385°C, and the rotation speed was set at 10 rpm.

The blend was then molded into a rectangular plate ($80 \times 100 \times 4$ mm^3) via hot-press molding at 340°C and 10 MPa for 10 min and was cooled to room temperature within the machine at a cooling speed of about 10°C/min.

Wear testing

The wear measurements were performed with a pin-on-disc or block-on-ring rig unit. For the pin-on-disc tribometer [Fig. 1(a)], a polymer pin was pressed against a rotating steel disk (German Standard 100 Cr 6, HRC 62) with a pneumatic cylinder. During the wear testing, each specimen's height was recorded

with a Kistler displacement transducer (Ostfildern, Germany), and the torque was measured with a load cell, by which the friction coefficient (μ) was calculated. The rotating velocity of the disc was fixed at 1 m/s, and 2 MPa of pressure was applied. Additionally, the disc could be heated from the backside, and the temperature of the disc could be adjusted from room temperature up to 250°C. Three measurements were provided for each composition. The average μ value in the steady state is presented. The specific wear rate (w_s) and the time-related depth wear rate (w_t) were determined as described in the following. Also, the average results are given.

w_s was calculated as follows:

$$w_s = \frac{\Delta m}{\rho F_N L} \; (mm^3/Nm) \qquad (1)$$

where Δm is the mass loss during sliding, L is the sliding distance, ρ is the density of the composite, and F_N is the normal load. An alternative is w_t:

$$w_t = \frac{\Delta h}{t} \; (\mu m/h) \qquad (2)$$

where Δh is the reduction in height and t is the sliding time.

For the block-on-ring tribometer [Fig. 1(b)], a polymer pin was pressed on a rotating steel cylinder (German Standard 100 Cr 6, HRC 62) with a predetermined pressure. The test was carried out at room temperature (ca. 25°C). The rotating velocity of the cylinder was 1 or 3 m/s, and the pressure applied to the sample pin varied from 1 to 3 MPa. Because the neat PEI showed excessive wear under these conditions, only lower pressures and short sliding times were applied. The block-on-ring machine did not allow the friction force to be measured, and so only the wear rate is provided, which was determined as described previously. An average wear rate value of six measurements for each material is reported.
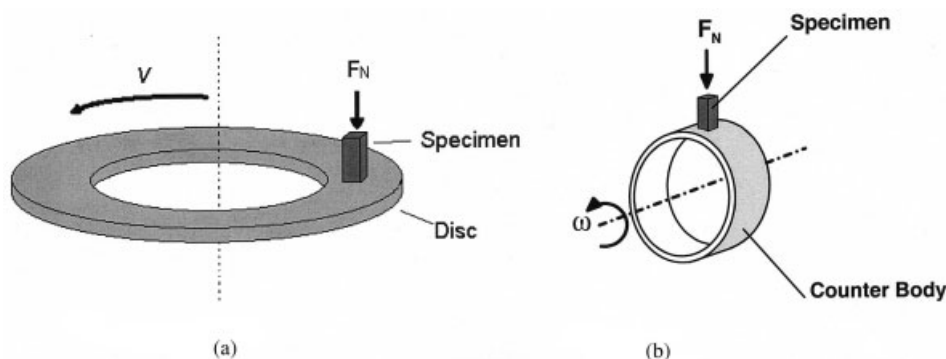

Figure 1 (a) Block-on-ring testing arrangement and (b) pin-on-disc testing arrangement.

Optical microscopy and scanning electron microscopy (SEM)

Micrographs were taken with an SEM instrument (JSM-T330A, Jeol, USA, Inc. Peabody, MA) at the worn surface of the samples after wear testing. The sliding tracks of the steel counterpart were also characterized with optical microscopy (Aristomet, Leitz, Cottonwood, CA).

RESULTS AND DISCUSSION

Influence of the sliding temperature

Figures 2 and 3 present w_s and μ of selected composites tested with a pin-on-disc testing rig at room temperature (ca. 25°C) and 150°C, respectively. With the exception of composition E (without SCFs), the incorporation of solid lubricants had a small effect on w_s at room temperature, as shown in Figure 2(a). Among them, composition A exhibited the best wear resistance.

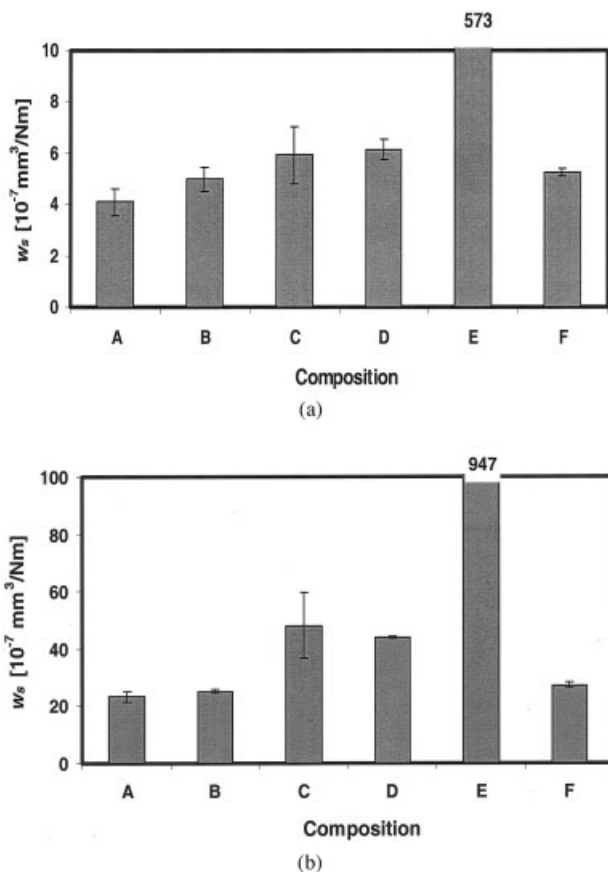At 150°C, all the composites showed remarkable increases in the wear rate, which may be due to the
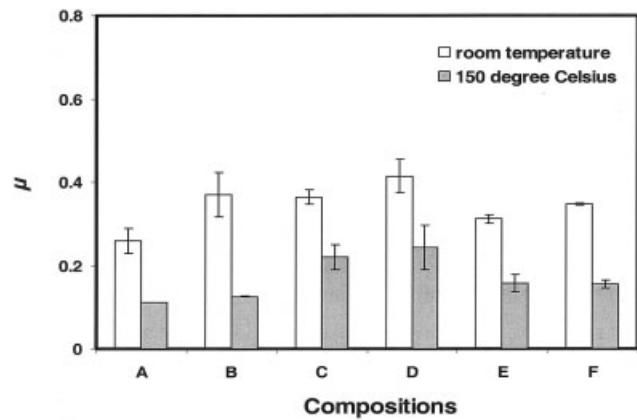
Figure 3 μ of selected composites at room temperatures or at 150°C (sliding pressure = 2 MPa, sliding velocity = 1 m/s, and sliding time = 20 h on the pin-on-disc testing rig).

deterioration of the mechanical properties.[1,6,8] Compositions A and B showed the best wear resistance, which was still comparable to that of composites filled with only SCFs (composition F). However, compositions C and D, filled with lower amounts of solid lubricants (in comparison with the SCF content) showed poorer wear resistance than the former two compositions. Composition E, filled only with lubricants, possessed the highest wear rate tested at both sliding temperatures, and this was assigned to inferior mechanical properties because of the lack of SCF reinforcement. It is clear that SCFs had an essential effect on the improvement of the wear resistance of PEI.

Figure 3 shows the steady-state μ values of selected composites tested at room temperature and 150°C. At room temperature, composite A showed the lowest μ value (ca. 0.26), whereas the other composites had comparable μ values of 0.35–0.4. As the sliding temperature increased to 150°C, μ decreased for all compositions. This kind of trend has been frequently reported for polymer composites and has been attributed to the reduction of the specific shear strength of the surfaces.[1,2,10] At an elevated temperature, composition A still had the lowest μ value (ca. 0.1), and composition B exhibited a comparable value (ca. 0.13).

Because of the higher w_s and μ values for compositions C and D, especially at elevated temperatures, in comparison with those of F (SCF/PEI), an adverse effect rather than a synergistic effect of the lubricants and SCFs was present. However, the synergistic effect was found for equilibrium contents of the solid lubricants and SCFs. With respect to PEEK, the optimal composition for sliding wear has been reported to be 10 vol % PTFE, 10 vol % graphite flakes, and 10 vol % SCFs.[6] The ranking of the compositions according to the wear resistance at both sliding temperatures was as follows: A ≈ B > F > C ≈ D ≥ E.

Figure 4 shows the worn surface appearance of compositions A and D at room temperature and

Figure 2 w_s of selected composites at (a) room temperatures and (b) at 150°C (sliding pressure = 2 MPa, sliding velocity = 1 m/s, and sliding time = 20 h on the pin-on-disc testing rig).
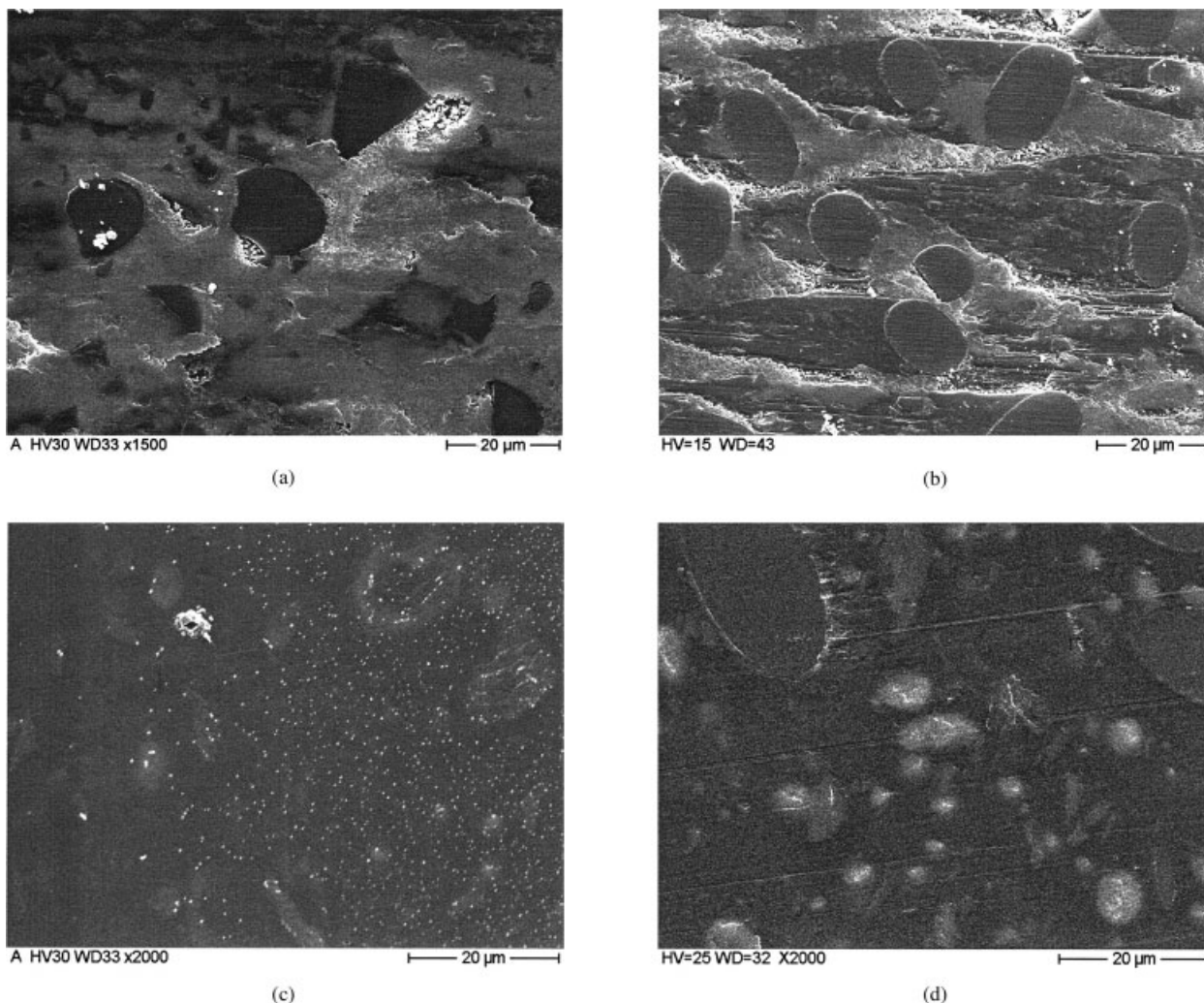
Figure 4 SEM micrographs of worn surfaces of composites (a,c) A and (b,d) D at (a,b) room temperature and (c,d) 150°C (sliding pressure = 2 MPa, sliding velocity = 1 m/s, and sliding time = 20 h).

150°C. The worn surfaces of A and D [Figs. 4(a,b)] tested at room temperature did not show serious fiber cracking, pulverization, or fiber/matrix interfacial separation, which are generally attributed to the wear of SCF-filled polymer composites, that is, SCF/PEEK.[13] The mild wear of compositions A and D was believed to be due to the existence of solid lubricants, which led to the formation of a transfer film on the counterface, protecting the fiber tips and matrix from the hard asperity of the counterpart.[4] Comparing the worn surfaces of A and D, it is found that the fiber tips of composition A surrounded with loose backtransferred wear debris, contrary to composition D, and this indicated relatively mild matrix wear.

At elevated temperatures, the worn surfaces of both samples became smoother, as shown in Figure 4(c,d). The fiber tips could not be easily detected by SEM on worn surfaces, especially for composition A, and this indicated the formation of a continuous and uniform friction film. This film was believed to be due to the backtransferred wear debris, which stuck together compactly because of the high temperature and normal force. However, the fiber tips of composition D could be clearly observed on the worn surface, and this implied less backtransferred wear debris. The lubricant content of compositions A and B makes it clear that more lubricant helped the formation of a compact wear debris layer on the composite surfaces.

Figure 5 shows the worn counterfaces slid by compositions A and D at both sliding temperatures. At room temperature, a thin and uniform transfer film formed on the counterface slid by composition A or D [Figs. 5(a,b)]. With an increasing sliding temperature, the transfer film became thicker, especially for composition A.

As previously demonstrated, composition A was expected to form more uniform and more effective friction films on the sliding pair than composition D with less lubricant content, and this may be the main
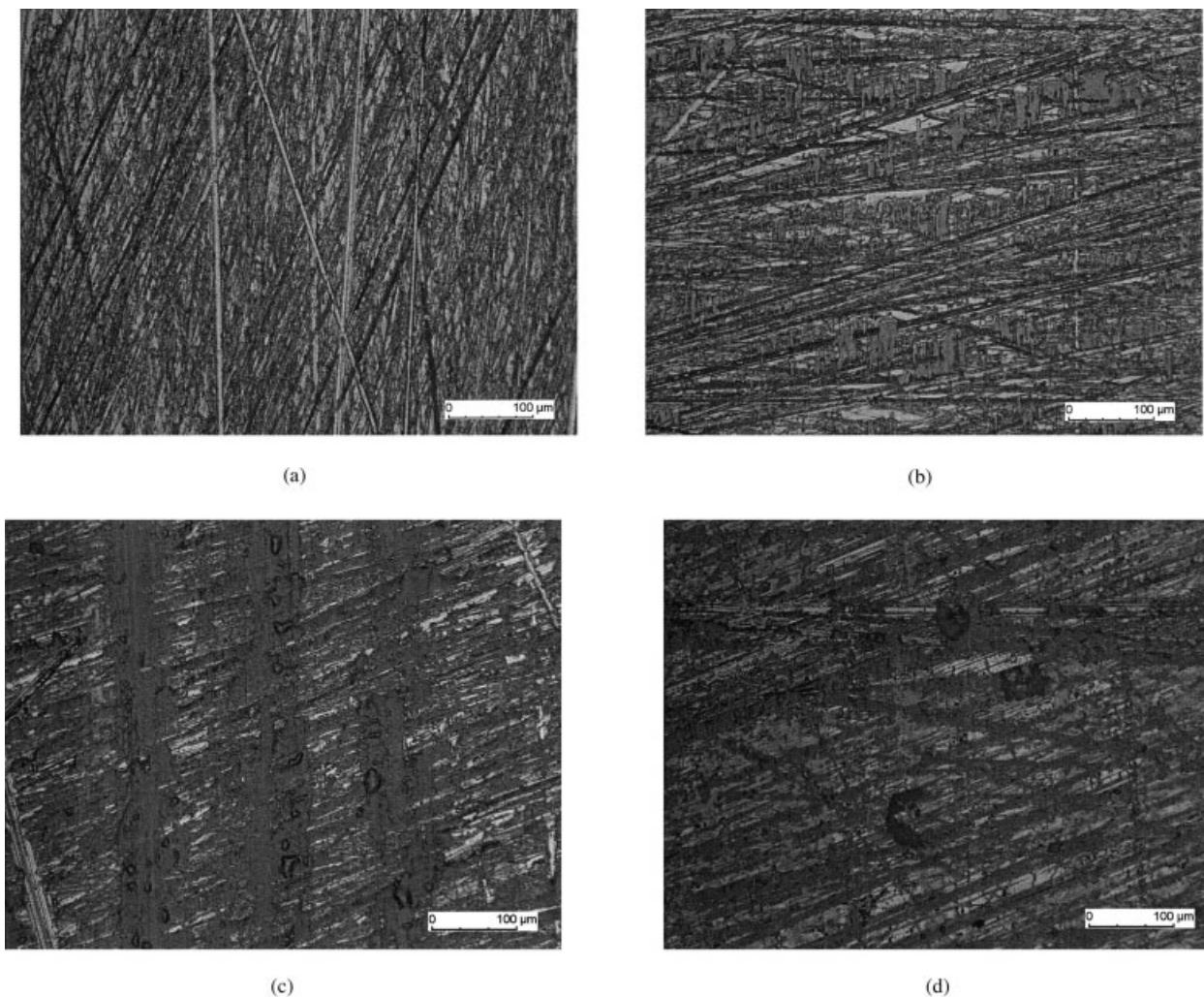
Figure 5 SEM micrographs of worn counterfaces due to composites (a,c) A and (b,d) D at (a,b) room temperature and (c,d) 150°C (sliding pressure = 2 MPa, sliding velocity = 1 m/s, and sliding time = 20 h).

reason that composition A exhibited a synergistic effect of the lubricants and SCFs on the wear behavior.

Furthermore, composition E, without an SCF component, did not show the formation of a continuous transfer film on the counter steel surface, as shown in Figure 6(a) (optical micrograph of the worn counterface). Figure 6(b) shows an SEM image of the worn surface of composition E. Significant debris was found in the shape of sheet attached to the smooth worn sample surface. This sheetlike wear particle was related to the fatigue mechanisms.[14]

Influence of the pv factor

The effect of the pv factor on the wear behavior was studied with block-on-ring tests at room temperature. Composition E was not tested because of the high wear rate shown in the previous tests. w_t against pv is presented in Figure 7. The compositions selected in this study showed almost linear

behavior in the pv range tested. At low pv values (i.e., <3 MPa m/s), close w_t values were found for all the compositions, but w_t varied significantly at high pv values. PEI filled with 10 vol % PTFE, 10 vol % graphite, and 10 vol % SCFs (composition A) showed the lowest w_t value. The value increased in the order of B, C, F, and D. Composition D had worse wear resistance than composition F without any lubricants. The incorporation of PTFE (5 vol %) of composition D may be beneficial to the formation of a transfer film but detrimental to the mechanical properties. Under severe pv conditions, the mechanical properties may become the dominant factor, and so the wear resistance of composition D was reduced in comparison with that of composition F, which was filled with only SCFs. The sequence of the compositions according to the wear resistance under high pv conditions was similar to that shown at higher temperatures: A > B > C > 10 vol % SCF/PEI > D.
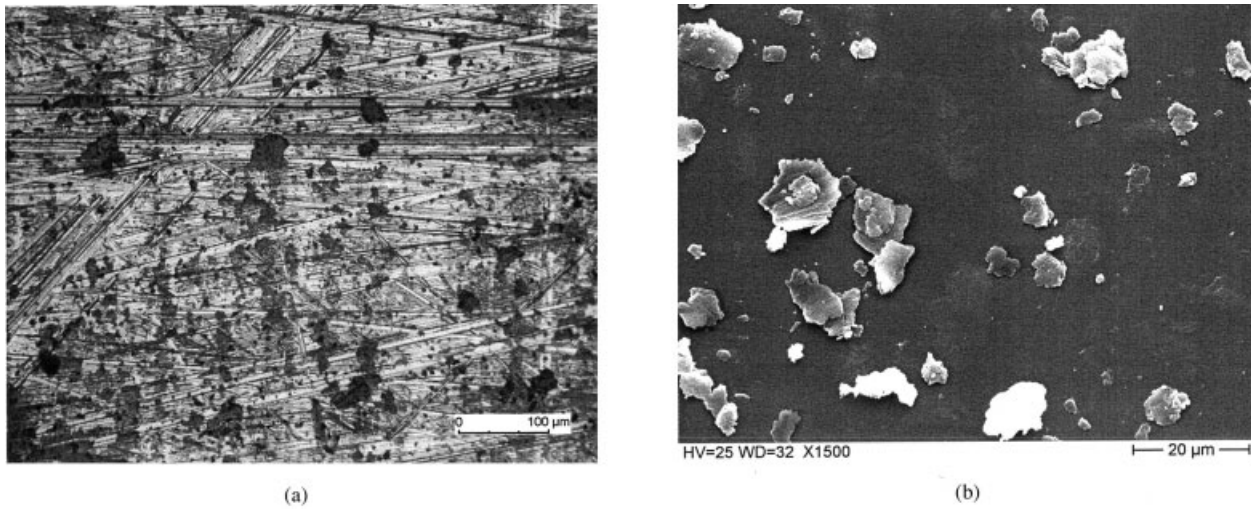
Figure 6 (a) Optical micrograph of a worn counterface slid by composite E and (b) SEM micrograph of worn composite E (sliding pressure = 2 MPa, sliding velocity = 1 m/s, and sliding time = 10 h at room temperature).

Influence of the pressure on the friction and wear of composition A

Because composition A showed the best wear behavior under the severe conditions described in this study, a further investigation into the pressure response (sliding speed = 1 m/s) was performed on the pin-on-disc rig. As shown in Figure 8, w_s initially increased steeply from 4.3×10^{-7} to 7.0×10^{-7} mm^3/Nm as the contact pressure increased from 2 to 4 MPa and then decreased to 5.0×10^{-7} mm^3/Nm slowly until the pressure of 9 MPa. This indicates that composition A could take a high load during sliding.

The evolution of μ as a function of the contact pressure is presented in Figure 8. μ initially reached the maximum value of 0.3 at the pressure of 4 MPa, and this was followed by a linear decrease to 0.17 (Fig. 8). As expected, the contact temperature increased with the increased contact pressure at a constant sliding speed, and

this could lead to a smoother transfer film, as previously described. Thus, μ decreased with high-pressure sliding.

CONCLUSIONS

The synergistic effects of solid lubricants (PTFE powders and graphite flakes) and SCFs on the reduction of both the wear rate and μ for compositions filled with 5 or 10 vol % graphite, PTFE, and SCFs were determined. The effects became more significant under severe sliding conditions, such as high sliding temperatures and high contact pressures. Microscopy observations suggested that the composition with equilibrium contents of the lubricants and SCFs helped in forming more continuous and uniform friction films on the sliding pair.

Under elevated sliding temperature, all the studied compositions exhibited decreased wear resistance and μ. In the pv range tested, all the compositions showed an almost linear increase of w_t with pv. At high pv values,
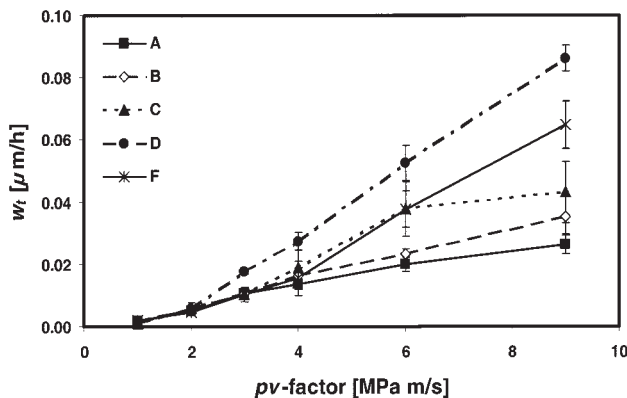

Figure 7 Dependence of w_t on the pv factor tested with a block-on-ring testing rig (sliding velocity = 1–3 m/s and contact pressure = 1–3 MPa at room temperature).
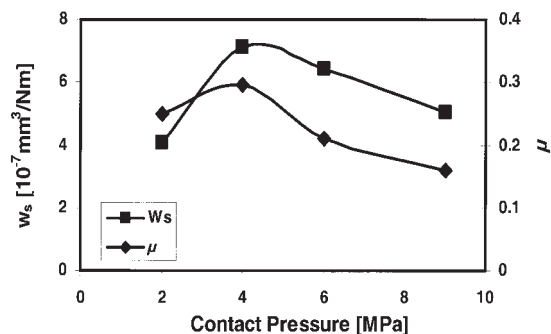

Figure 8 Variation of w_s and μ as a function of the contact pressure for composite A tested with a pin-on-disc testing rig (sliding velocity = 1 m/s at room temperature).

that is, greater than 3 MPa m/s, compositions A and B, containing 10 or 5 vol % graphite, 10 or 5 vol % PTFE, and 10 or 5 vol % SCF, respectively, showed lower wear rates. With the sliding speed kept constant at 1 m/s and with the contact pressure varied between 2 and 9 MPa, composition A showed the peak values for both the wear rate and μ at 4 MPa.

References

1. Häger, A. M.; Davies, M. In Advances in Composite Tribology; Friedrich, K., Ed.; Composite Materials Series 8; Elsevier: Amsterdam, 1993; p 107.

2. Friedrich, K.; Reinicke, R.; Zhang, Z. Proc Inst Mech Eng Part J 2002, 216, 415.

3. Khedkar, J.; Negulescu, I.; Meletis, E. I. Wear 2002, 252, 361.

4. Bahadur, S. Wear 2000, 245, 92.

5. Yu, L.; Yang, S.; Liu, W.; Xue, Q.; Bahadur, S. J Appl Polym Sci 2000, 76, 179.

6. Lu, Z. P.; Friedrich, K. Tribol Int 1995, 181–183, 624.

7. Zhang, Z.; Breidt, C.; Chang, L.; Friedrich, K. Tribol Int 2004, 37, 271.

8. Bijwe, J.; Rajesh, J. J.; Jeyakumar, A.; Ghosh, A.; Tewari, U. S. Tribol Int 2000, 33, 697.

9. Reinicke, R.; Haupert, F.; Friedrich, K. Compos A 1998, 29, 763.

10. Tewari, U. S.; Bijwe, J. In Advances in Composite Tribology; Friedrich, K., Ed.; Composite Materials Series 8; Elsevier: Amsterdam, 1993; p 159.

11. Yoo, J. H.; Eiss, N. S. Wear 1993, 162, 418.

12. Stuart, B. H. Tribol Int 1998, 31, 647.

13. Voss, H.; Friedrich, K. Wear 1987, 116, 1.

14. Czichos, H. In Advances in Composite Tribology; Friedrich, K., Ed.; Composite Materials Series 1; Elsevier: Amsterdam, 1986; p 12.