Tribological investigations of polyetherimide composite

U. S. TEWARI, J. BIJWE
Industrial Tribology Machine Dynamics and Maintenance Engineering Centre (ITMMEC), Indian Institute of Technology, New Delhi 110016, India

Polyetherimide (PEI), commercially known as ULTEM and manufactured by GEC (USA), is one of the newest high-performance thermoplastics. Its graphite and short-glass-fibre (GF) filled composition was evaluated for friction and wear properties. Tribological studies of the material sliding against mild steel, under different loads, counterface roughnesses and sliding distances were performed on a pin and disc configuration. It was observed that this composite displayed very good wear resistance due to glass-fibre reinforcement and low friction due to the solid lubricant graphite. The wear mechanism was studied with scanning electron microscopy by observing the worn pin and disc surfaces. Fatigue was observed to be the main factor in wear, along with adhesive and abrasive modes.

1. Introduction

Polyimides are promising tribo-materials in terms of friction and wear characteristics, thermal stability, mechanical and physical properties which are retained even at high temperatures [1–7]. Despite the advantages associated with thermoplastics such as easy processability, applicability and possibility of recycling and repair, most efforts have been devoted to tribo-investigations of thermosetting polyimides [1–7]. It was therefore considered useful to evaluate a thermoplastic polyimide as a tribomaterial.

In view of this, polyetherimide (PEI), a thermoplastic polyimide having an excellent property profile [8] was selected for the present studies. Our earlier work on polyetherimide [9], PEI + PTFE [10] and short-glass-fibre-reinforced PEI (20%) [11] revealed the following features.

1. Bulk PEI displayed low friction and good resistance to fatigue wear. However, after a critical number of cycles (characteristics of load and history of pin-surface preparation) wear due to fatigue suddenly started and was very high.
2. PEI + PTFE while showing lower friction did not show substantial improvement in wear.
3. PEI + glass fibres (GF) showed promise in terms of improved wear behaviour ($K_0 = 4.5 \times 10^{-15}$ m^3 Nm^{-1}) but not in friction ($\mu \simeq 0.35$).

Hence it was of interest to incorporate another solid lubricant which could reduce friction and/or wear in the composite. Among the potential solid lubricants it is already reported that MoS_2 oxidizes at above 400 °C, PTFE softens cold flows under low loads and moderate temperature while graphite loses water in vacuum and becomes abrasive at high temperatures [12]. However, graphite has a reinforcing effect, with the added advantage of self-lubrication and enhanced thermal conductivity, the latter being important for friction [12] Graphite was therefore considered to be an appropriate filler for reinforced PEI composites.

In tribo-science, filler effects, cannot be predicted *a priori*. It is reported that PTFE generally reduces friction, but not necessarily wear [13]. Graphite has been reported in some cases to have an adverse effect on friction and/or wear [14], but useful in others [12]. Hence the properties of the polymer composites have to be tested in the tribo system. The objective of the present study was to obtain an insight into the wear performance of short-glass-fibre-reinforced (16 wt %, average length 400 μm, diameter 10 μm) PEI composite filled with 20% graphite powder. It is also important to learn about dominating wear mechanisms in the sliding wear of this composite under various experimental conditions.

2. Experimental procedure

2.1. Characterization of the composite

The general structure of PEI has been given elsewhere [9]. The composite under study was prepared by incorporating the requisite amount of graphite in the glass-fibre-reinforced composite (ULTEM 2200). TGA and DTG studies of ULTEM 2200 and its composite with graphite are shown in Fig. 1. Details of this analysis are given elsewhere [15]. By comparing these two curves, the composition of the composite was confirmed to be PEI 64%, glass fibre 16% and graphite 20%. Pins of the composites were fabricated by injection moulding.

2.2. Materials and their preparation

Mild steel was selected as a counterface (Rockwell hardness RB 80). Details of the pin fabrication, preparation of disc surfaces with required roughness are given elsewhere [11]. The total apparent sliding area

0022–2461/92 $03.00 + .12

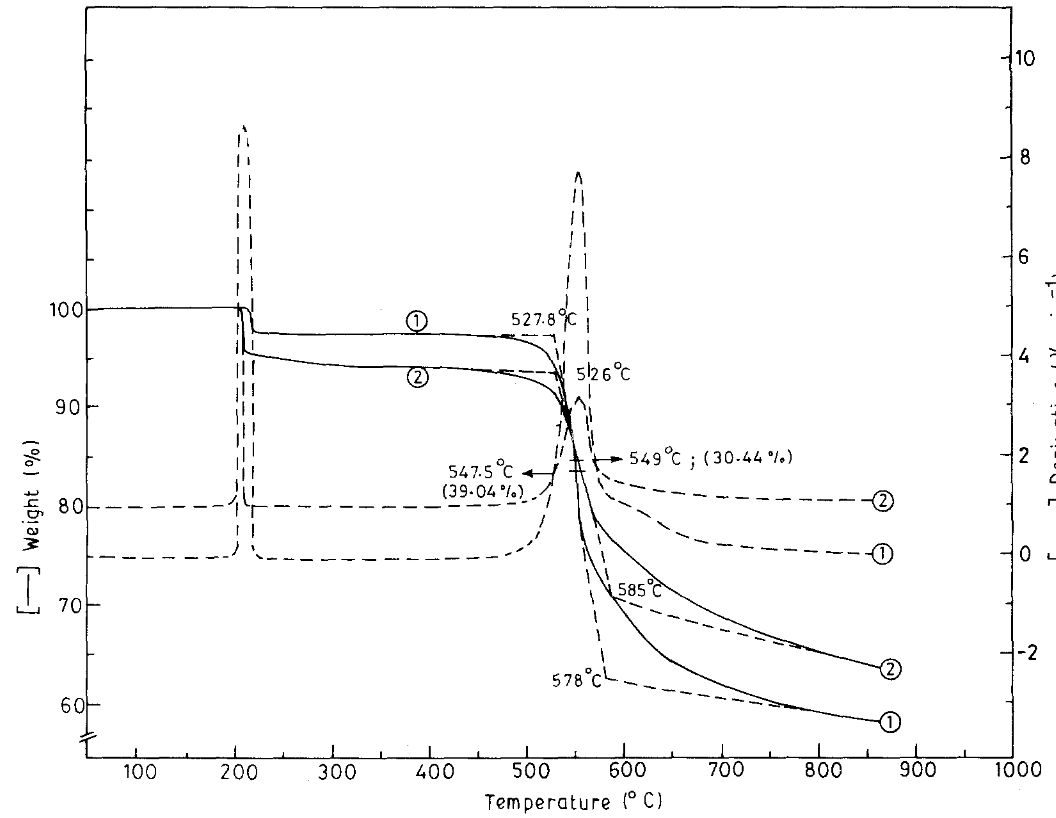


Figure 1 Thermogravimetric analysis (TGA) and DTG curves for (1) PEI + GF (20%) and (2) PEI + GF (16%) + graphite (20%) in N_2 atmosphere, heating rate 10 °C min^{-1}.

of the pins was 1.4 cm^2 and mould-filled direction (MFD) was perpendicular to sliding direction.

2.3. Friction and wear studies

Experiments were carried out using tripins on disc configuration on a friction and wear testing machine. The sliding system consisted of a ground mild-steel disc rotating against three polymeric pins. The total load varied from 43 to 132 N. All experiments were carried out at ambient temperature. All measurements were preceded by a running-in period with the same P and V values as were used in the test, on a different disc with same R_a value where P is load applied in N and V is the sliding velocity and R_a is the roughness parameter [9]. This prerubbing allowed the counterface to wear the specimen until it had full contact with the counterface.

3. Results and discussion

Friction and wear data of the composite are plotted in Figs 2–6. Scanning electron micrographs (SEM Philips 515) are shown in Figs 7–11. Polymer pins were silver sputtered to make the surface conducting.

It was observed that at lower loads the pins did not show measurable wear, nor was a wear track initiated. At an increased load of 43 N, pins started initiating a wear track and a thin graphite layer was immediately transferred, thereby reducing the friction coefficient. The friction data obtained in similar sliding conditions with and without a solid lubricant in the composite are shown in Fig. 2 for comparison.

Peaking in friction was observed in both the composites, and is thought to be due to a change in surface

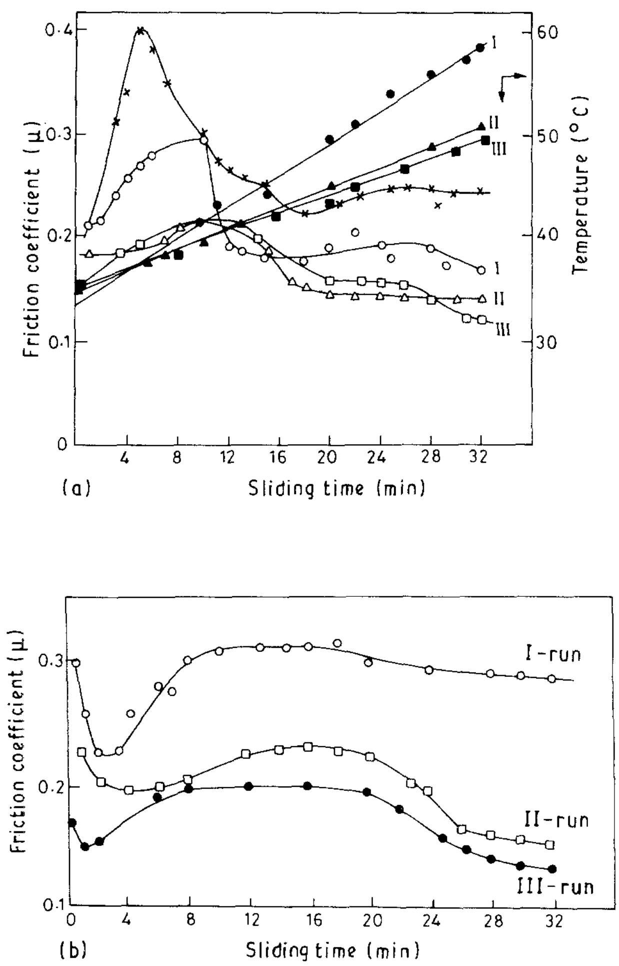


Figure 2 Friction coefficient and pin surface temperature of PEI + GF + graphite (-●-, -■-, -▲-) and PEI + GF (-×-) composites as a function of sliding time; load 43 N, speed 2.1 m s^{-1} counterface (R_a) (a) 0.15 and (b) 0.05 μm.

topography [11]. It may be noted that a reduced peak value in the friction-against-time plot appeared in the case of the graphite-filled composite (Fig. 2a), as compared to the composite without graphite. From Fig. 2a it is clear that under identical conditions of load, speed and counterface roughness, the friction coefficient is substantially reduced from 0.25 to 0.10 due to graphite inclusion. Curves I, II and III are for the friction displayed in successive runs. Because the surface is already covered with graphite layer, the friction coefficient is reduced in these runs, reaching a fairly low value ($\simeq 0.12$) in the first run. The temperature of the pin subsurface (thermocouples were inserted in the pins 3 mm away from the sliding surface) in these cycles was also measured as a function of sliding time and is plotted in Fig. 2a. Such temperature measurements are indicative of changes in actual sliding surfaces. Since friction decreases in consecutive runs, the temperature of the subsurface also decreases.

Fig. 2a shows friction data generated on the surface with $R_a = 0.15$ μm [9]. Friction data in Fig. 2b are for a smooth, polished disc surface with $R_a = 0.05$ μm. For such smooth counterfaces, the frictional behaviour of the composite was quite different. Friction coefficient (μ) against sliding time showed a sharp minimum after 7 kcycles of the traversals. Beyond this, the friction coefficient in each run increased slowly, reaching a steady-state value. Both the friction coefficient at minimum and at the steady state decreased in successive runs. A comparison of findings regarding the friction behaviour of the composite for two different roughnesses of the counterface is shown in Fig. 2a and b.

Fig. 3 shows the friction data generated under two extreme load conditions (43 and 132 N). Steady-state friction reduced in the second run in both cases. The peaks and steady-state value of the friction coefficient are lower in cases of high load (132 N) than for low load (43 N). The friction coefficient generally reduces with the increase in load, and has been explained by Suh [16]. Steady-state wear curves of PEI + GF and PEI + GF + graphite are shown in Fig. 4. The wear loss in each interval is plotted as a function of the distance slid in that interval, and is not cumulative wear. From this curve it is clear that the specific wear coefficient (K_0) of the glass-fibre-reinforced composite is reduced from 4.5×10^{-15} m^3 Nm^{-1} to 1.2×10^{-15} m^3 Nm^{-1}. Thus inclusion of graphite led to a reduction in friction by reducing the abrasiveness of the glass fibre with a simultaneous increase in wear resistance.

The parameters which influence the friction and wear properties of a solid polymer metal pair include applied load; speed; temperature; environment; counterface material; topography; and geometrical arrangement of the two interacting surfaces. The possible variations of these parameters are so large as to render friction and wear comparisons of different materials, in widely different applications and situations, an extremely difficult task. In such cases, only order of magnitude can be given [17]. Lancaster [18] observed that the specific wear rate ranged over 5×10^{-16} to 10^{-13} m^3 Nm^{-1}, for filled and unfilled thermoplastics during sliding against steel at room temperature on a pin and disc machine. The present composite thus seems to have fairly good wear resistance in the order of magnitude 10^{-15} m^3 Nm^{-1}. In Fig. 5, wear and specific wear rates under various loading conditions, and the corresponding temperature rise in the pin subsurface, are shown. In the wear against load relationship, specific wear rate seems to increase with load. According to Tsukizoe and Ohmae [19] such behaviour is due to the deterioration of the fibre matrix adhesion at increased load caused by higher frictional heating. As in the delamination theory [20] fibre–matrix debonding takes place due to repetitive mechanical stresses which in turn depend on fibre orientation and load. SEM observations of worn

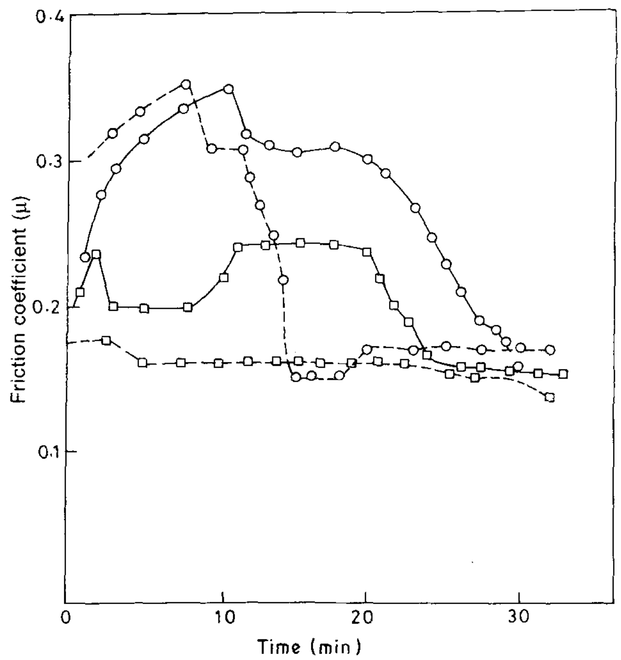


Figure 3 Friction coefficient as a function of sliding time at two extreme loads 43 and 132 N (speed 2.1 m sec^{-1} and counterface (R_a) 0.25 μm). (——) Run I; (- - -) run II, -○- L-43 N; -□- L-132 N.

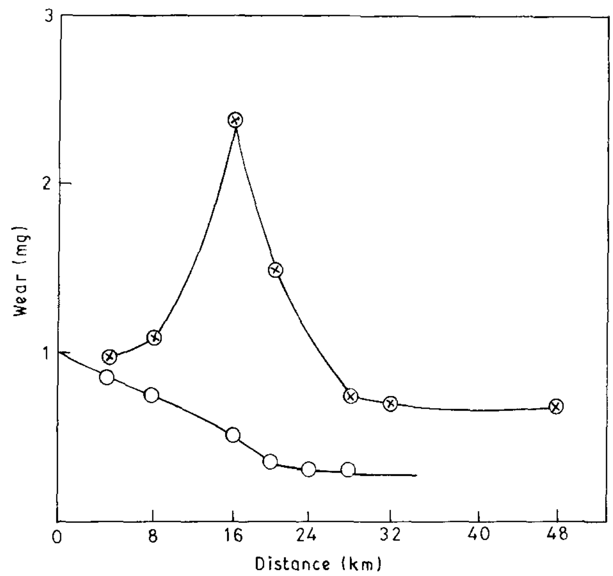


Figure 4 Steady-state wear curves for two PEI composites of PEI; (load 43 N, speed 2.1 m sec^{-1} and counterface (R_a) 0.16 μm). (-⊗-) PEI + GF; (-○-) PEI + GF + graphite.

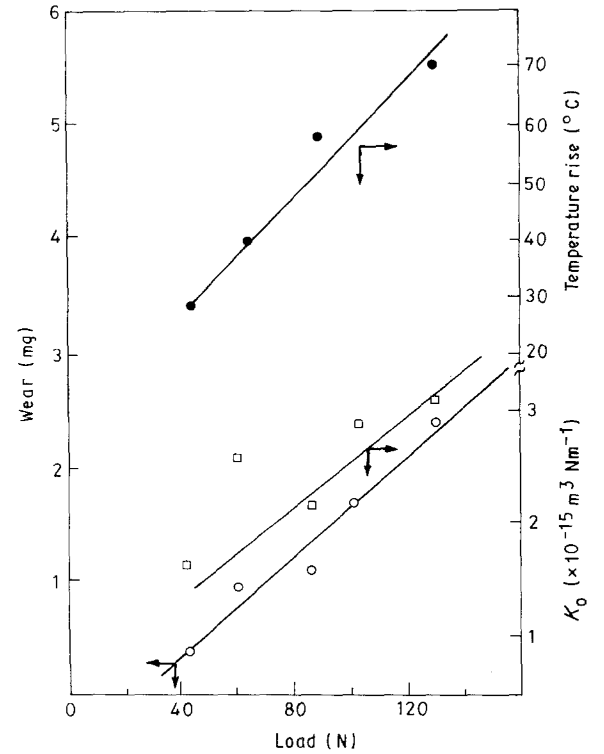


Figure 5 Wear (–○–○–); specific wear rate (–□–) and temperature rise of pin surface (–●–) as a function of load; (speed 2.1 m s^{-1}; counterface (R_a) 0.20–0.25 μm and distance slid, 4 km).

surfaces of this composite have clearly shown such debonding is due to increased load. Thus at higher loads, peeling off or pulling out of fibres or its pieces becomes easier, which in turn is reflected in a higher wear rate. In Fig. 6 the wear data of the composite are plotted as a function of sliding distance. Specific wear rates at increasing time intervals were also calculated and are plotted in the same figure. Both wear and wear rates increased with increased sliding distance. It should be noted that wear and distance in the plots are not cumulative values. Wear in each interval has been plotted as a function of distance in that interval. Wear rate as seen in Fig. 6 increases almost linearly with the increase in sliding distance; frictional heat also increases. This could influence wear mechanism in the following ways.

1. With increased temperature, lubricating efficiency of graphite decreases, as it depends on the adsorbed layer of moisture.

2. According to delamination theory [20], fibre–matrix adhesion weakens due to repetitive mechanical stresses, and fibres are easily pulled out resulting in higher wear with sliding distance.

3. Wear may increase due to fatigue induced into the sliding surface of the composite with sliding distance.

4. SEM studies

Pin and disc surfaces were observed in typical cases and are shown in Figs 7–11. The virgin pin surface at two different locations is shown in Fig. 7a, b. It is seen that most of the fibres are normal to the sliding surface (Fig. 7a). However, some fibres are in the plane of the sliding surface (Fig. 7b). Fig. 8 consists of micrographs of the pin surface and relevant EDAX studies (corresponding to Fig. 4). It is apparent that after 20 km of traversal under load 43 N and speed 2.1 m s^{-1}, cracks are generated and propagated from one fibre tip to other. If compared to a virgin surface, the worn surface is very smooth. Fibre tips normal to the surface can be seen from silicon dot mapping, as shown in Fig. 8b. It seems that the polymer surface is fully covered with the wear powder of glass fibres while sliding against mild steel. Iron-dot mapping at the same location is shown in Fig. 8c. While sliding glass fibres in the composite abrade the disc, and

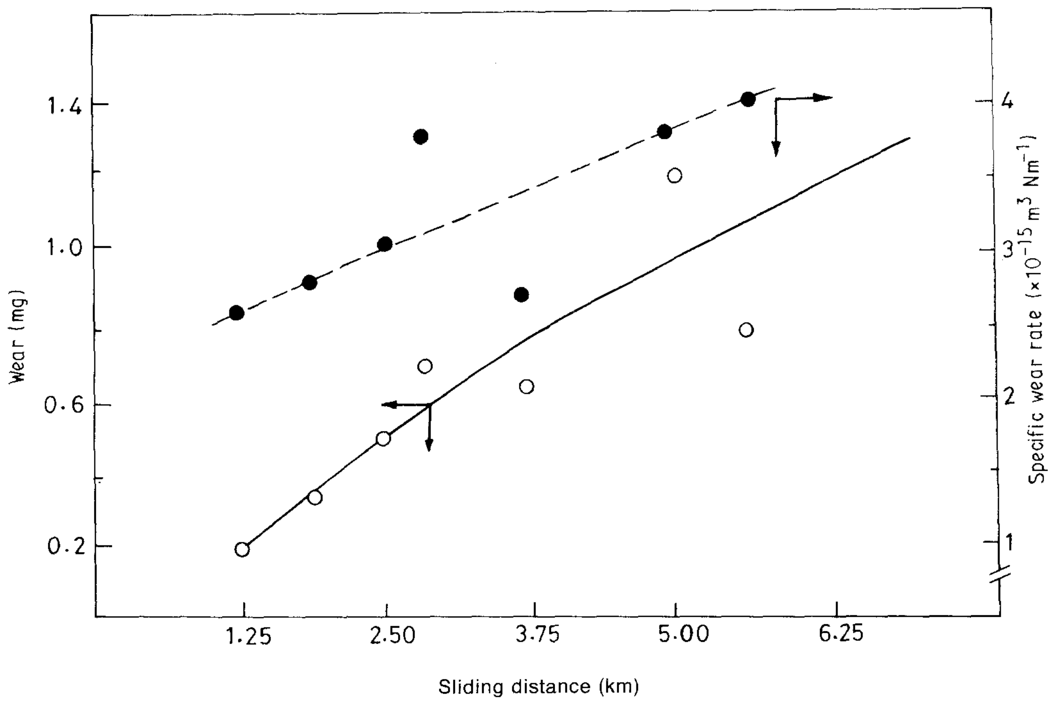


Figure 6 Wear (–○–○–) and specific wear rate (– – – –) as a function of sliding distance. (Load, 43 N; speed, 1.05 m s^{-1}; counterface (R_a) 0.25 μm).

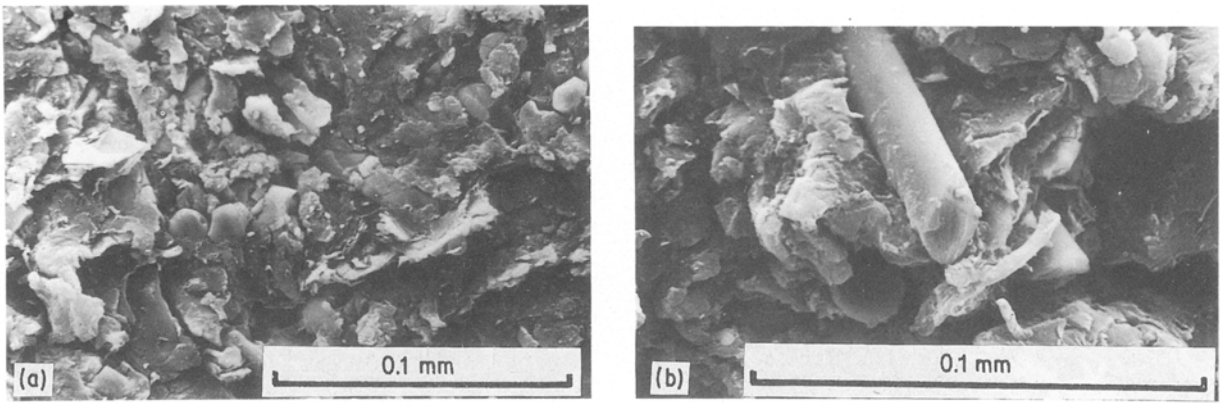
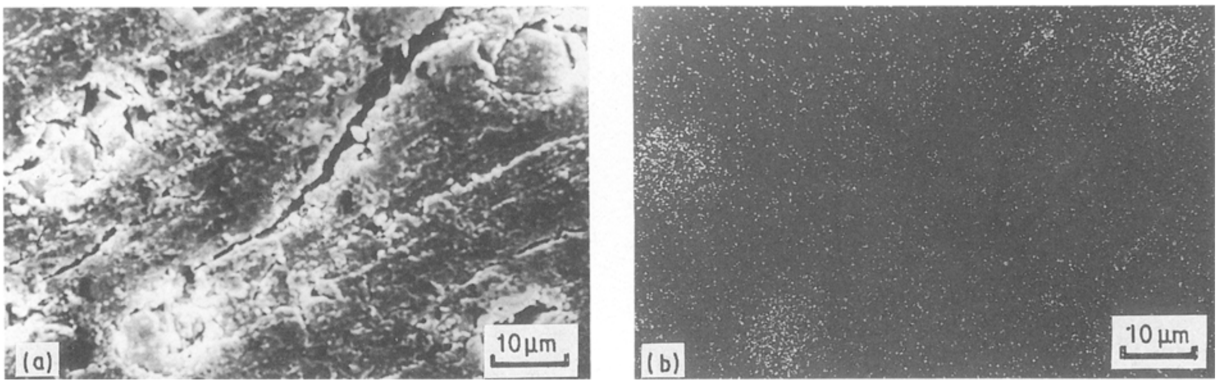


Figure 7 SEM micrograph of PEI + GF + graphite composite before wear test (a) rough surface topography and fibre tips normal to the surface; (b) fibre lying longitudinally on the surface.



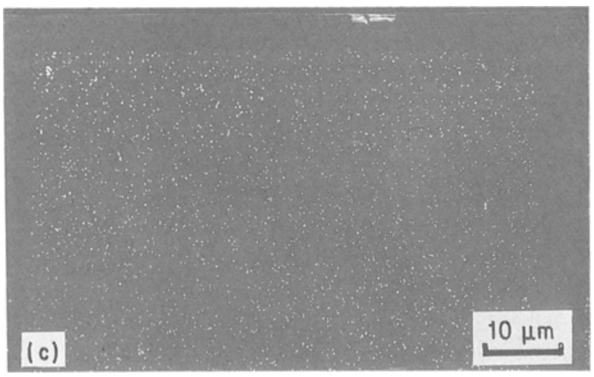


Figure 8 SEM micrograph of the pin surface worn under steady-state conditions (load, 43 N; speed, 2.1 m s^{-1}; counterspace (R_a) 0.16 µm; distance slid, 20 km). (a) crack propagating from tip to tip of a fibre; (b) and (c) corresponding EDAX analysis. (b) Silicone dot mapping and (c) iron dot mapping.

metallic wear powder (iron) generated in abrasion is back-transferred on to the soft and hot polymer surface. Such back-transfer of iron was also observed in the case of PEI + GF composites [11]. Czichos [21] has reported similar iron back-transfer in the case of PPS + GF composite sliding against mild steel.

In a brittle matrix such as epoxy, cracks generated in the matrix propagate right through the fibre when the bonding between fibre and matrix is strong. In a highly ductile matrix such as polyurethane, the cracks cannot propagate through the matrix and fibre. Therefore the fibre bends with the matrix under the asperity contact and the wear rate of this kind of composite is controlled by the wear rate of the fibre. Earlier experiments show that in such cases the ends of fibres in such composites are extremely well polished and elliptical rather than circular, although the fibres are perpendicular to the surface. This indicated that fibres are worn gradually in very small increments when these fibres are bent under sliding [16].

The SEM micrograph of a pin surface worn under high P–V values (load, 112 N; speed, 2.1 m s^{-1}) for 2 h are shown in Fig. 9. These micrographs show interesting features of the wear mode. From the micrographs in Fig. 9a–d, it seems that this composite wore in a fashion similar to highly ductile, graphite-fibre-reinforced polyurethane [16]. From Fig. 9a, the severe melt flow of the polymer in the direction of sliding due to high frictional heating under loading condition is noted. The surface appears to have an excess concentration of glass fibres, most fibres being normal to the surface. Crack propagation in the sliding direction can be distinctly seen. One fibre in the bottom seems to be pulled out and polymer material in its vicinity is lost. Multiple cracks can be observed in its vicinity. It may be surmised that as sliding commences the polymer starts wearing and is lost more readily than the fibres. Eventually, an array of protruding fibres supporting a large fraction of load is apparent from this micrograph. The fibre tips are essentially elliptical and well polished, resembling those in the polyurethane matrix [16].

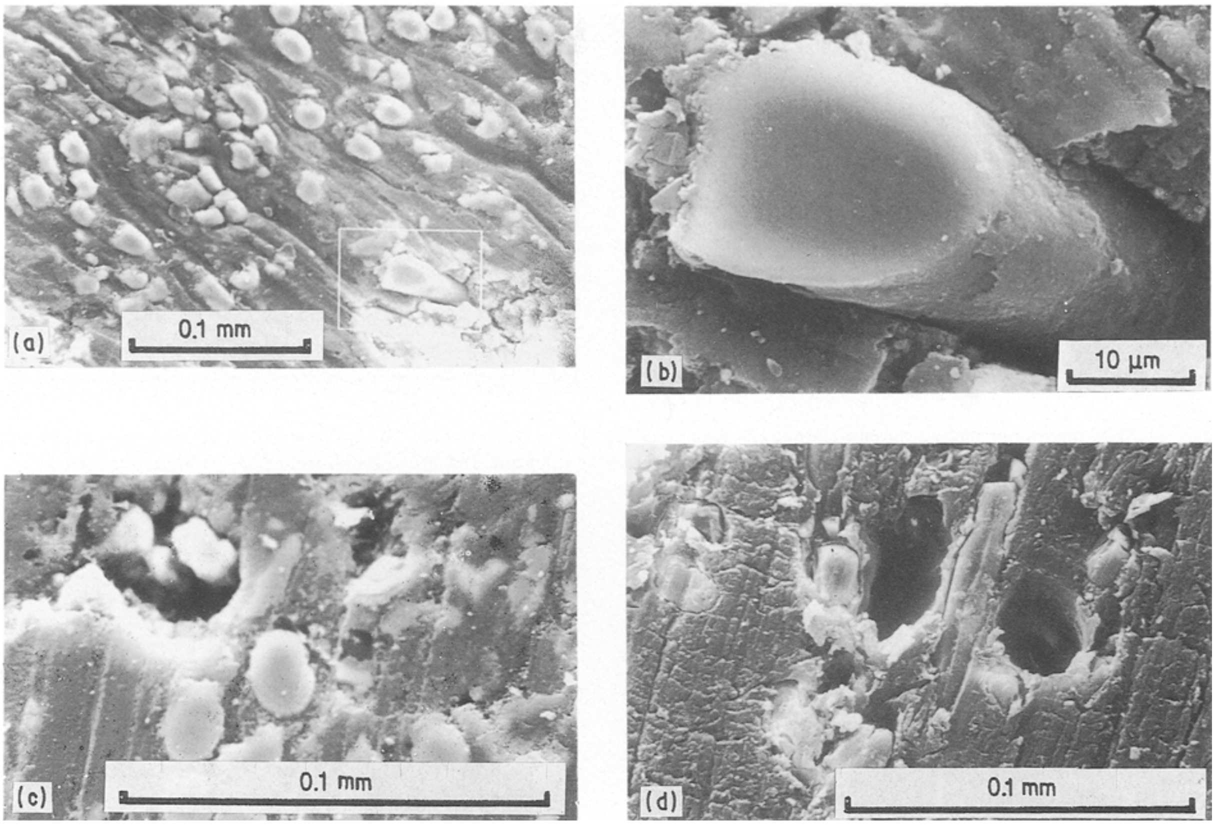


Figure 9 SEM micrograph of pin surface at different locations. (a) Severe melt flow of polymer in sliding direction, maximum fibres with elliptical tips normal to surface, cracks generated in sliding direction and a pull out of fibre. (b) Magnified view of pulled-out fibre from the matrix with elliptical and polished tip. (c) Elliptical fibre tips, crater where molten material is chipped off with array of unworn fibres (left top). (d) Multiple parallel microcracks perpendicular to the sliding direction indicating fatigue, cavities due to fibre consumption, deterioration in fibre matrix adhesion.

A magnified view of one such fibre is shown in Fig. 9b, supporting the suggestions discussed earlier. As the load and sliding time increase, fibre–matrix bonding deteriorates. In Fig. 9c, different locations on the same pin surface are shown. Many worn tracks seem to be essentially broken "cold-welded" adhesions. Some polymeric molten material has been peeled off and appears to be embedded with elliptical fibre tips. The surface where some material has been chipped off has a crater, with an array of unworn fibres with circular tips inside it. Severe plastic deformation and the local melt adhesion of the resin from some portions to the metal are the dominant features of Fig. 9c. Fig. 9d shows multiple parallel microcracks perpendicular to the sliding direction throughout the surface. Crack initiation is believed to be due to the tensile component of the principal stress in the contact zone, and these cracks grow due to repetitive sliding. Jain and Bahadur [22] observed similar cracks in the surface of filled polyamidemide (PAI) at load 57 N and speed 1 m sec^{-1}. The progressive crack growth due to repetitive sliding at asperity contacts is indicative of fatigue mode of wear [22]. A number of small pits are also seen in Fig. 9c and d which are created by the extraction of graphite particles from the matrix under the effect of frictional force. Another interesting feature of Fig. 9d is the wear thinning and fibre breakage which is embedded longitudinally on the surface. Fibre–matrix separation (debonding) along the interface due to high mechanical stresses can be seen. Some glass fibres seem to be consumed due to wear, leaving behind cavities of appropriate sizes. One worn-well polished fibre tip can be seen in the left portion of the micrograph. Severe plastic deformation associated with the removal of larger material patches and fibres from the surface is apparent in the middle portion.

In Fig. 10, the corresponding disc surface worn under similar sliding conditions to those seen in Fig. 9 (load, 112 N; speed, 2.1 m sec^{-1}) is shown. From the micrograph it is apparent that because of the high frictional heat generated at the interface under such severe sliding conditions, molten polymer was extruded from the pin surface. Because of stronger adhesion of this material to the counterface, it is readily

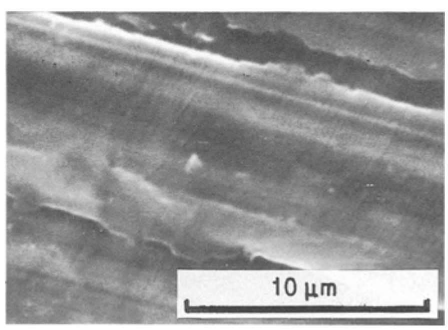


Figure 10 SEM micrograph of a counterface (mild steel disc) showing transferred layers of molten polymer (load, 112 N; speed, 2.1 m s^{-1}; sliding time 2 h).

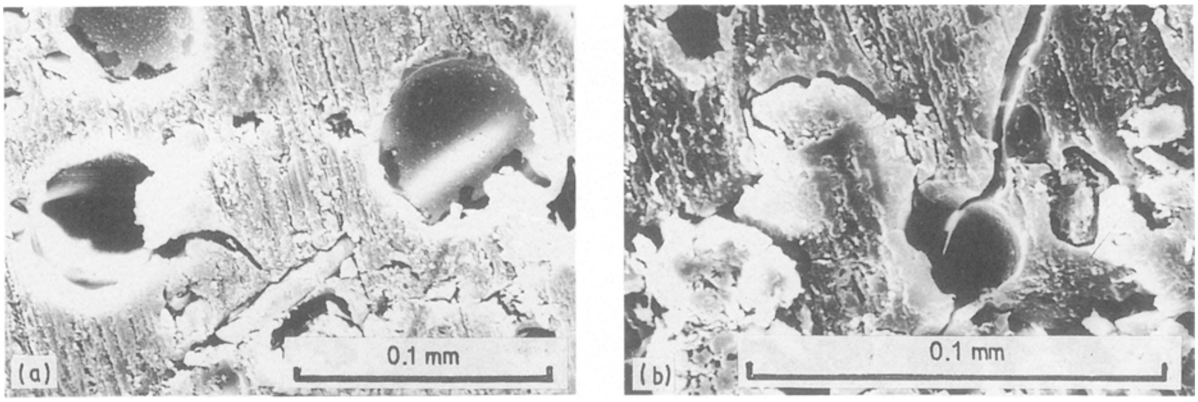


Figure 11 SEM micrographs of pin surface worn under high load (132 N) and speed (2.1 $m s^{-1}$). (a) Fibre–matrix debonding wear thinning of a longitudinal fibre, cavities due to fibre consumption. (b) Deep cracks initiating and propagating from fibre to fibre, pits formed due to graphite extraction and fibre consumption.

transferred on to the disc in the form of overlapping layers, leaving the craters behind as shown in Fig. 9c. Wedge-shaped asperities are covered in various places by such molten material.

Fig. 11a, b shows the worn pin surface under extreme load conditions (load, 132 N; speed, 2.1 $m sec^{-1}$) polymer pin surface seems to be damaged severely and extensive plastic deformation is apparent. Cavities are formed due to consumption of fibres which were perpendicular while wearing. The intersection of multiple cracks, severe melt flow of polymer and the pits formed due to extraction of graphite particles can also be seen (Fig. 11a, b). Thinning and breakage of fibres lying in the plane of the pin surface, their interfacial debonding with the resin and cracks in the direction perpendicular to sliding (Fig. 11a) are indicative of severe damage due to high loading.

5. Conclusions

Graphite inclusions in the glass-fibre-reinforced composite of PEI were found to be effective in the tribological sense. Friction coefficient was reduced to a steady, low value (from 0.25 to 0.12). Specific wear rate also showed improvement from 4.5×10^{-15} to 1.2×10^{-15} $m^3 Nm^{-1}$ which indicates good wear resistance. Glass fibres seem to improve the wear resistance of polymer composites, due to the fact that fibres exposed to the sliding surface support the applied load. Also, penetration of the steel asperities into the polymer surface was reduced due to graphite inclusions, so that micro-ploughing and micro-abrasion were no longer effective. The composite did not wear by the polymer film transfer mechanism, but by graphite transfer, and in the severe sliding condition, gross transfer of molten polymer on the counterface was observed. Fatigue was found to be dominant in wear along with abrasive and adhesive processes.

Acknowledgements

Part of this paper was presented at the sixth International Colloquium on Industrial Lubricants—Properties, Application and Disposal, Technical Academy Esslingen, Germany, January 1988.

References

1. R. L. FUSARO, *Tribol. Trans.* **31** (1988) 174.
2. U. S. TEWARI, S. K. SHARMA and P. VASUDEVAN, *Tribol. Int.* **21** (1988) 27.
3. K. TANAKA and Y. YAMADA in "Polymer Wear and Its Control", edited by L. H. Lee (ACS Symposium Series 287, Amer. Chem. Soc., Washington DC, 1985) pp. 103–128.
4. R. L. FUSARO, in "Polyimides—Synthesis, Characterization and Applications", Vol. 2, edited by K. L. Mittal (Plenum, New York, London 1984) p. 1053.
5. P. SUTOR, in "Polymer Wear and Its Control", edited by L. H. Lee (ACS Symposium Series 287, Amer. Chem. Soc., Washington DC, 1985) p. 269.
6. M. R. C. ZADEH and N. S. EISS, *Wear* **110** (1986) 259.
7. K. FRIEDRICH, *Tribol. Int.* **22** (1989) 25.
8. R. O. JOHNSON and H. S. BURLHIS, *J. Polym. Sci. Polym. Symp.* **70** (1983) 129.
9. J. BIJWE, U. S. TEWARI and P. VASUDEVAN, *J. Mater. Sci.* **25** (1990) 548.
10. J. BIJWE, U. S. TEWARI and P. VASUDEVAN, *J. Synth. Lub.* **6** (1989) 179.
11. J. BIJWE, U. S. TEWARI and P. VASUDEVAN, *Wear* **132** (1989) 247.
12. F. J. CLAUSS, "Solid Lubricants and Self Lubricating Solids" (Academic, New York, London, 1972).
13. B. BRISCOE, L. H. YAO and T. A. STOLARSKI, *Wear* **108** (1986) 357.
14. S. BAHADUR and D. TABOR, in "Polymer Wear and Its Control", edited by L. H. Lee (ACS Symposium Series 287, Amer. Chem. Soc., Washington DC, 1985) p. 253.
15. P. VASUDEVAN, J. BIJWE and U. S. TEWARI, in Proceedings of the Sixth International Colloquium TAE, Esslingen, West Germany, January 1988, p. 5.5.1.
16. N. P. SUH, "Tribophysics" (Prentice-Hall, Englewood Cliffs, New Jersey, 1986) p. 223.
17. J. P. GILTROW, *Composites* **4** (1973) 55.
18. J. K. LANCASTER, *Tribol. Int.* **5** (1972) 249.
19. T. TSUKIZOE and N. OHMAE, in "Friction and Wear of Polymer Composite", Composite Material Series 1, edited by K. Friedrich (Elsevier, Amsterdam, 1986) p. 205.
20. N. H. SUNG and N. P. SUH, *Wear* **53** (1979) 129.
21. H. CZICHOS, *ibid.* **88** (1983) 27.
22. V. K. JAIN and S. BAHADUR, *ibid.* **123** (1988) 543.

Received 31 January
and accepted 29 October 1990