Role of Internal Additives in the Friction and Wear of Carbon-Fiber-Reinforced Polyimide

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ABSTRACT: Polyimide composites should function in sliding contacts under high temperatures, but the interference of carbon fibers with sliding mechanisms is difficult to predict: they often increase the coefficients of friction and act abrasively but show lubricating properties under other conditions. The friction and wear behavior of thermoplastic polyimides reinforced with short carbon fibers and filled with solid internal lubricant (polytetrafluoroethylene) or silicon oil was investigated in this study with a reciprocating cylinder-on-plate tester under 50 N at 0.3 m/s with steel counterfaces that were heated at 23–260°C. We concluded that polytetrafluoroethylene additives effectively reduced the coefficients of friction over the entire temperature range, especially under thermally controlled sliding conditions at 120°C, whereas the internal silicon oil increased the coefficients of friction. The wear rates of the fiber-reinforced polyimide significantly decreased with respect to those of the thermoplastic polyimide, whereas additional fillers slightly increased the wear rates. We further analyzed the role of internal additives by considering the deformation and maximum polymer surface temperature during sliding. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 116: 1146–1156, 2010

Key words: additives; fibers; mechanical properties

INTRODUCTION

Thermoplastic polyimide (TP) belongs to a polymer family that can be processed by regular molding techniques and offers excellent mechanical, chemical, and thermal stability. It has attracted attention as a potential candidate for use in tribological systems under high normal loads and/or high temperatures.[1–3] Although TP has an intrinsic self-lubricating ability, two problems are generally encountered: (1) it shows relatively high coefficients of friction because of the highly aromatic molecular structure and consequently difficult formation of a smoothly oriented transfer film, and (2) it often shows high wear rates because of brittleness.[4]

Efforts have been made by tribologists worldwide to tune the sliding performance of polyimide composites by reinforcing them with glass fibers[5,6] or aramid fibers.[7] Carbon fibers (CFs) have been more frequently considered, as they combine a high specific strength, specific modulus, damping capacity, good thermal conductivity, low expansion, and potentially lubricating properties.[8] After preimpregnation, the short CFs easily flow with the molten matrix resin so that heat-molding techniques remain suitable for the fabrication of composite parts.[9] Important research on the tribological behavior of carbon-fiber-reinforced thermoplastic polyimide (CF–TP) has focused on the fiber–matrix interface,[10] fiber morphology,[11] and fiber percentage.[12] However, the incorporation of a secondary phase strongly interferes with the physical and mechanical bulk characteristics: the CFs may either decrease or increase the coefficients of friction, whereas the wear rates are not necessarily lower than those of pure TP.[13] Zhang et al.[14] recently reported that it was difficult to develop CF–TP composites with low friction and low wear rates with only short CFs. To increase the applicability of CF–TP, sliding under external oil lubrication was studied.[15] Other research was devoted to the addition of solid lubricants, such as molybdenum disulfide (MoS_2), under dry and water lubrication conditions.[16] Jia et al.[17] demonstrated that graphite improved the antiwear properties and reduced friction more successfully than MoS_2 under dry sliding on stainless steel. Other fillers that effectively improved the friction-reducing and antiwear abilities of CF–TP composites include graphite and microsilica particles[18] and carbon nanotubes.[19,20] The performance of such additives is very sensitive to the operating conditions: additives functioning in one typical testing regimen may not equally perform in another situation. It was recently shown that the

TABLE I
Physical and Mechanical Properties of the TP Composites

Property	TP	CF–TP	CF–TP–PTFE	CF–TP–oil
Specific density (ASTM D 792; g/cm^3)	1.33	1.42	1.44	1.38
Modulus of elasticity (ASTM D 882; MPa)	2,760	16,800	6,300	10,800
Tensile strength (ASTM D 638; MPa)	118	230	150	190
Compressive strength (JIS K-7208; MPa)	122	207	165	202
Elongation (ASTM D 638; %)	90	2	5	3.5
Glass-transition temperature (DSC; °C)	250	248	245	244
Heat deflection temperature (ASTM D 648; °C)	238	248	240	246
Melting temperature (DSC; °C)	385	385	385	385
Thermal conductivity [ASTM C 177; W/(mK)]	0.17	MD: 0.49 TD: 0.22	MD: 0.49 TD: 0.22	MD: 0.47 TD: 0.21

DSC = differential scanning calorimetry; MD = molding direction; TD = transverse direction.

performance of graphite in sintered polyimides varied strongly with normal loads and sliding velocities because of tribochemical interactions in the interface.[21]

Although polyimide composites should function preferentially under high temperatures, most of the available test results in the literature cover a wide range of normal loads and sliding velocities. The effect of elevated temperatures on the performance of CF in polyimide composites was noticed by Xian and Zhang,[22] but interference with solid lubricants at high temperatures is relatively unknown. In this study, we aimed to further contribute to the optimization of tribological CF–TP grades by considering the incorporation of polytetrafluoroethylene (PTFE) as a solid lubricant and silicon oil as an internal lubricant; these were explicitly tested at high temperatures under a constant normal load and sliding velocity.

EXPERIMENTAL

TP was obtained by the reaction of pyromellitic dianhydride and 4,4′-bis(3-amino-phenoxy)biphenyl. The polyacrylonitrile-based high-strength CFs were 200 μm in length and about 8 μm in diameter, with a density of 1.8 g/cm^3. Four industrially available composites (Dupont de Nemours, Mechelen, Belgium) were tested, including: (1) pure polyimide as a reference material (TP), (2) polyimide filled with 30 wt % CFs (CF–TP), (3) polyimide filled with 30 wt % CFs and 15 wt % PTFE (CF–TP–PTFE), and (4) polyimide filled with 30 wt % CFs and 15 wt % silicon oil (CF–TP–oil). Although the effect of various filler concentrations was not explicitly considered, a 15 wt % concentration is often applied as a compromise between good tribological and mechanical properties.[22,23] The resin was blended with the respective fillers/lubricants in desired percentages and was then mixed in a tumbling mixer. The mixture was extruded at 420°C with a twin-screw screw extruder with a revolving speed of 80 rpm, a screw diameter of 37 mm, and a length/diameter ratio of 32.

Furthermore, the materials were injection-molded at mold temperatures of 180–210°C and a pressure profile of 75–240 MPa for a molding cycle of 30 s. The composite properties are listed in Table I.

Composite cylinders with a diameter of $2R = 5$ mm and a width of $2b = 15$ mm were reciprocatingly slid in a dry line contact against a fixed steel counterface on a PLINT TE 77 high-frequency tribotester. The counterfaces were made of 40 CrMnNiMo8-4-6 steel (DIN 1.2738) polished with GRID 600 abrasive paper to an average roughness of 0.05 μm. The counterfaces were heated with electrical resistance cartridges to bulk temperatures of 60, 80, 100, 120, 140, 180, 220, and 260°C and controlled by PID feedback. The polyimide composite surface temperature is discussed further. A fixed normal load (F_n) of 50 N was applied on top of the polymer cylinder sliding at 0.3 m/s over a stroke of 15 mm and with a total sliding distance of 15 km. The test environment had controlled climate conditions (environmental temperature = 23°C, relative humidity = 60%). The friction force (F_f) was measured with a piezoelectric transducer in contact with the counterface, and F_n was recorded with a load cell coupled to a spring-and-lever bridge onto the polymer. The coefficient of friction is defined as the ratio of F_f to F_n (Coefficient of friction = F_f/F_n). The volumetric wear rates were calculated from the wear volumes according to the weight loss (V_w) or diameter reduction (V_d) immediately after sliding and 1 day after testing to determine the degree of deformation:

$$V_w = \Delta m/\rho \tag{1}$$

$$V_d = 2b\left(R^2 \sin^{-1}\frac{\sqrt{R^2 - (R - \Delta h)^2}}{R} - (R - \Delta h)\sqrt{R^2 - (R - \Delta h)^2}\right) \tag{2}$$

where Δm is the mass loss, ρ is the specific density, b is the semiwidth perpendicular to the sliding
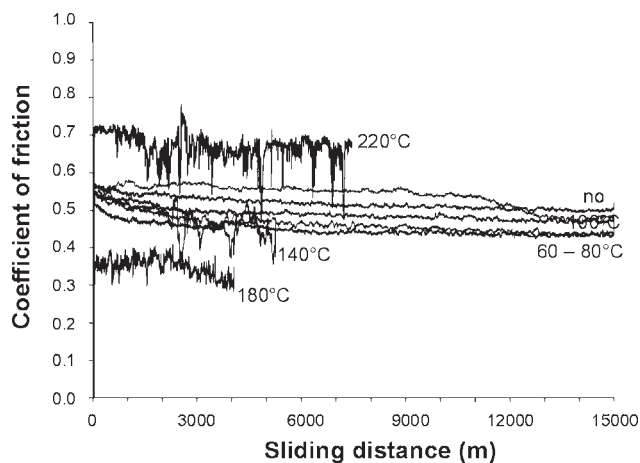
Figure 1 Online-measured coefficients of friction as a function of the sliding distance at different bulk temperatures for TP.

direction, R is the radius, and Δh is the diameter reduction.

RESULTS

Friction behavior

The coefficients of friction are plotted in Figures 1–4 as a function of the sliding distance. The steady-state coefficients of friction are given as a function of the bulk temperatures in Figure 5. Every test was repeated three times with a statistical variation of about ±7% for TP and about ±5% for the CF–TP composites.

For TP, the coefficients of friction were most constant over the sliding distance without significant differences between the running-in and steady-state conditions. In agreement with its thermoplastic properties, softening and deformation contributed to
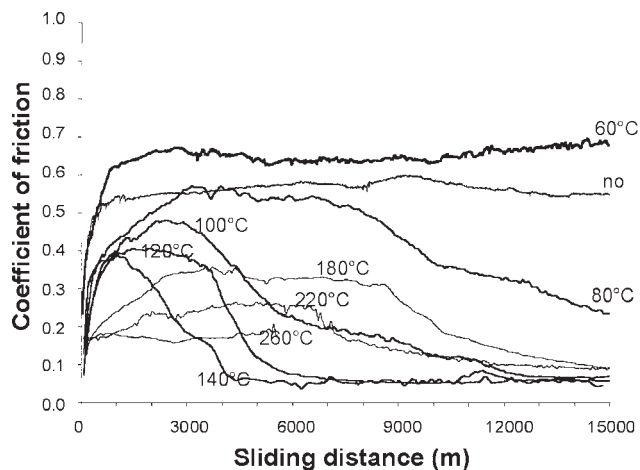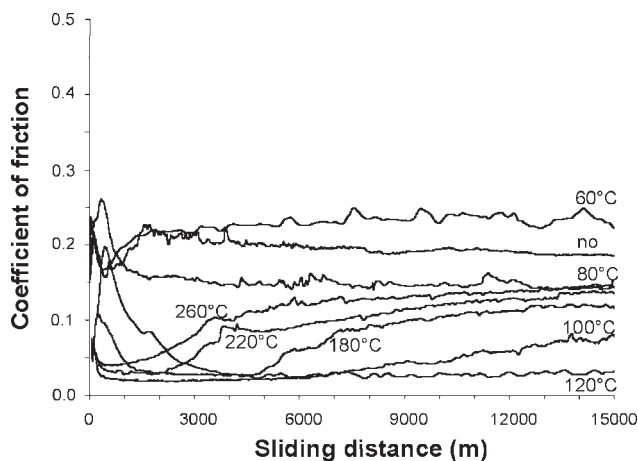

Figure 3 Online-measured coefficients of friction as a function of the sliding distance at different bulk temperatures for CF–TP–PTFE.

the formation of a lubricating transfer film, which stabilized the sliding process. The coefficients of friction increased at 80–120°C and decreased toward a minimum at 180°C. Overloads at 180–220°C were due to thermal softening and a loss in mechanical strength. The test at 260°C could not be done because of immediate melting.

For CF–TP, the coefficients of friction at 23–60°C were higher than for TP and increased with temperature. Strong transitions were noticed at higher temperatures, with significant differences between the running-in and steady-state conditions. The CFs contributed to the formation of a stable transfer film, but it developed gradually with changing properties or composition during sliding. The incorporation of CF prevented mechanical softening and provided optimum lubrication at 100–140°C. The formation of a lubricating transfer film seemed to be predominantly controlled by temperature, as transitions into
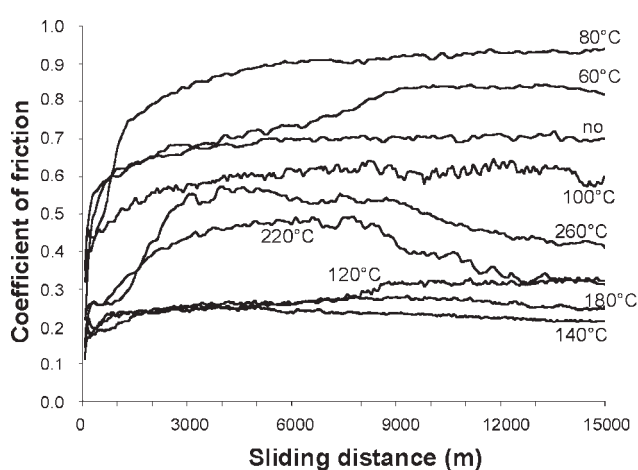

Figure 2 Online-measured coefficients of friction as a function of the sliding distance at different bulk temperatures for CF–TP.

Figure 4 Online-measured coefficients of friction as a function of the sliding distance at different bulk temperatures for CF–TP–oil.
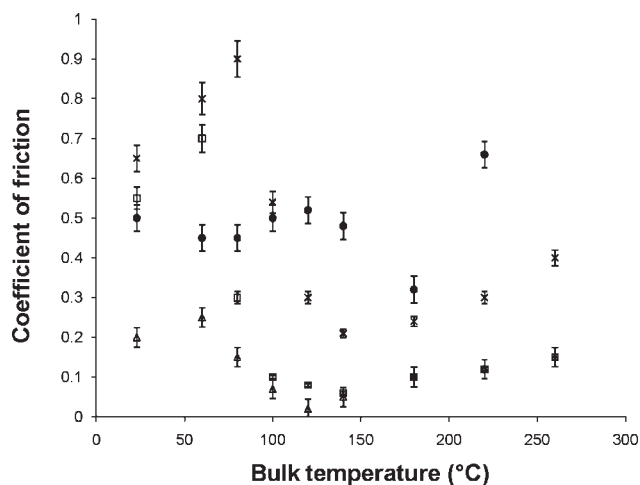
Figure 5 Steady-state coefficients of friction as a function of the bulk temperature for different polyimide composites: (●) TP, (□) CF–TP, (△) CF–TP–PTFE, and (×) CF–TP–oil.

a steady-state sliding regime occurred after shorter times at higher temperatures. At 120–140°C, the coefficients of friction stabilized and attained low values that remained constant over the entire steady-state regime. At 180–260°C, the transition into the steady state was postponed, as the thermal effects in the TP matrix interfered with the fluent formation of a lubricating transfer film. The test at 260°C had to be aborted because of melting.

For CF–TP–PTFE, the lowest coefficients of friction among all of the polyimide composites were obtained. The running-in period had a characteristic trend that first increased over short sliding distance and sharply decreased with the formation of a lubricating transfer film. The transfer was initially influenced by loose CFs; this led to a suddenly high friction. The steady-state coefficients of friction increased at 23–60°C in parallel with those of CF–TP; this indicated that the presence of CF dominated the sliding mechanisms at low temperatures. At higher temperatures, running-in progressively shortened and finally disappeared as the lubricating properties of PTFE became more efficient. The steady-state coefficients of friction significantly decreased at 60–120°C; this represented optimum sliding conditions with low and stable friction over the steady-state regime. A second transition in the steady-state coefficients of friction was noticed at 140–260°C, where the values equal those of CF–TP. The second transition in the steady-state coefficients of friction occurred more rapidly at higher temperatures; this indicated that the transfer film was modified by thermal overloads, mainly in the TP matrix.

For CF–TP–oil, the coefficients of friction were higher than for CF–TP, and the internal oil additives often negatively affected the sliding properties of

CF–TP. At 23–80°C, the coefficients of friction were highest among all of the composites, with running-in effects similar to CF–TP. The initial rise in friction at low temperatures indicated that the sliding properties were dominated by CFs without the formation of an oil lubricating transfer film. The unfavorable development of a transfer film was also observed by a transition to high friction after about 8000 m at 80°C. At 100–140°C, the coefficients of friction decreased by thermally controlled sliding conditions, whereas the running-in effects also decreased with a better lubricant supply. The main action of oil thus consisted of reducing the running-in characteristics and thereby smoothening the friction curve in a certain temperature interval. At 180–260°C, however, the internal oil became inadequate, and the friction increased again; this was coupled with significant transitions in sliding mechanisms that were attributed to TP and CFs.

Wear behavior

The volumetric wear rates from weight loss are plotted against the bulk temperature in Figure 6. The differences between the wear volumes from weight loss and diameter reduction are given as the percentage deformation immediately after testing (Fig. 7) or 1 day after testing (Fig. 8). The wear rates calculated from diameter reductions were larger, as they also accounted for viscoelastic or plastic deformation and creep. The statistical variation on weight measurements was about ±8% for TP and about ±5% for the CF–TP composites. The statistical variation on dimensional measurements was about ±10% for TP and ±7% for the CF–TP composites.
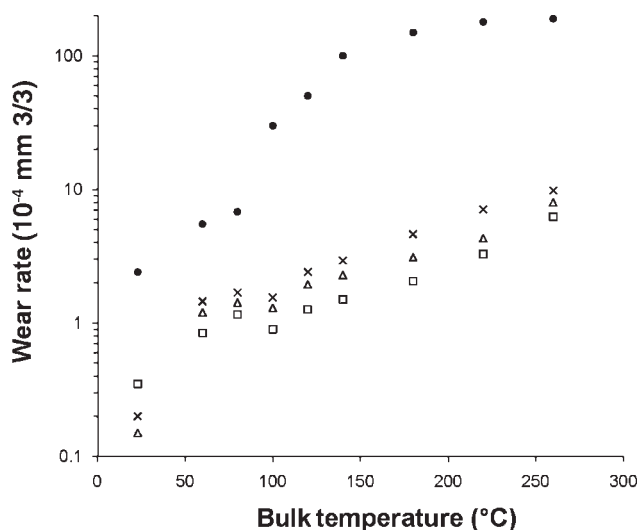

Figure 6 Volumetric wear rates (weight measurements) as a function of the bulk temperature for different polyimide composites: (●) TP, (□) CF–TP, (△) CF–TP–PTFE, and (×) CF–TP–oil.
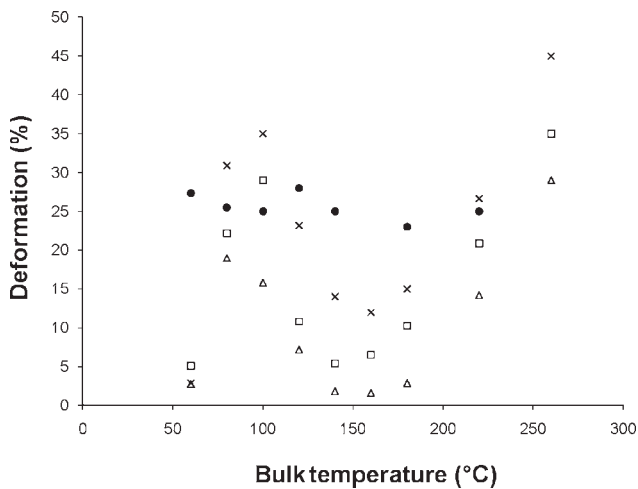
Figure 7 Deformation (%) of the test samples: a comparison of the wear rates from geometrical and weight measurements for different polyimide composites immediately after testing: (●) TP, (□) CF–TP, (△) CF–TP–PTFE, and (×) CF–TP–oil.

The volumetric wear rates of TP were the highest at all temperatures. A transition in wear for TP at 100–180°C was correlated with a regime of decreasing friction, whereas the overload conditions in wear agreed with the high friction at 180–260°C. Tewari and Bijwe[24] observed rather noncorrelated peaks in steady-state friction and wear: although the friction transition occurred at 150°C, the wear transition happened at 200°C. The wear rates of the CF–TP composites were lower than those of TP, but the wear rates of the internally lubricated CF–TP changed strongly: the internal PTFE and oil lubricants provided lower wear rates than the unfilled CF–TP at 23°C, whereas the unfilled CF–TP showed its lowest wear rates at higher bulk temperatures. The PTFE lubricants were more efficient in lowering wear than the oil lubricants over the entire testing range. The slight reduction in the wear rates for all of the CF–TP composites at 100°C coincided with the discontinuous increase in the wear rates for TP; this indicated that the composite matrix further controlled the wear rates.

The deformation strongly depended on the sliding conditions: although a fixed F_n value of 50 N was applied to all of the samples, the deformation was not constant. There was some correlation between the sample deformation immediately after testing and the coefficients of friction; this suggested that the sample deformation was not predominantly influenced by static indentation and creep but rather by mechanical shear stresses in the subcontact surface. The lower shear stresses at high temperatures prevailed over the mechanical weakening of the test samples, as only a small depth beneath the contact surface (ca. 1 μm) thermally weakened. The CF–TP–

PTFE samples had the lowest deformation, and the CF–TP–oil samples had the highest deformation, which contrasted with their relative mechanical strength but was in parallel with their low and high, respectively, coefficients of friction. After 1 day, the viscoelastic deformation was almost recovered, and some permanent deformation remained. Unfilled TP seemed to be prone to significant plastic deformation, whereas the CF–TP composites had better resistance against permanent deformation. A correlation between the permanent deformation and tensile strength of the various composites could only be drawn at 60°C; this indicated that thermally induced interactions and lubricating efficiency, rather than the mechanical properties, determined the tribological behavior.

Transfer film and polymer surface observations

The transfer films on steel and worn polymer surfaces were studied by optical microscopy for CF–TP (Fig. 9), CF–TP–PTFE (Fig. 10), and CF–TP–oil (Fig. 11). The sliding direction is indicated, and the counterface roughness grooves were perpendicular to the sliding direction.

The transfer films for CF–TP at 60–100°C contained coarse graphitized depositions accumulating in the roughness grooves with sharp edges perpendicular to the sliding direction; this, consequently, caused high friction. The polyimide was not influenced by thermal softening, and very few transfer particles were formed by mechanical shear. The composite surface was severely worn because of the abrasiveness of the CFs. The transfer films at 120–180°C were thin and oriented along the sliding
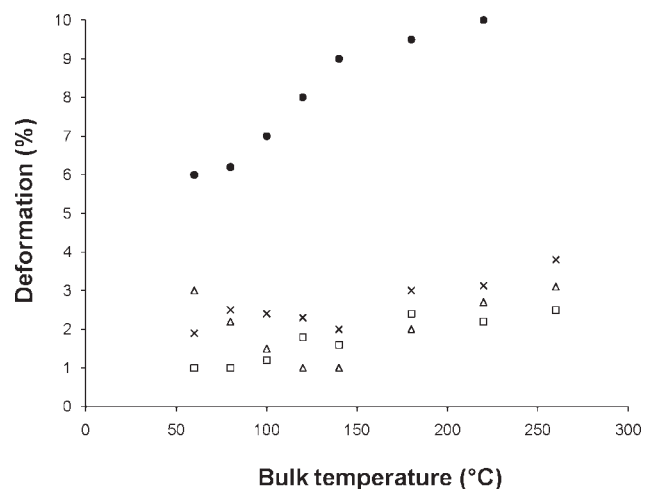

Figure 8 Deformation (%) of the test samples: a comparison of the wear rates from geometrical and weight measurements for different polyimide composites 1 day after testing: (●) TP, (□) CF–TP, (△) CF–TP–PTFE, and (×) CF–TP–oil.
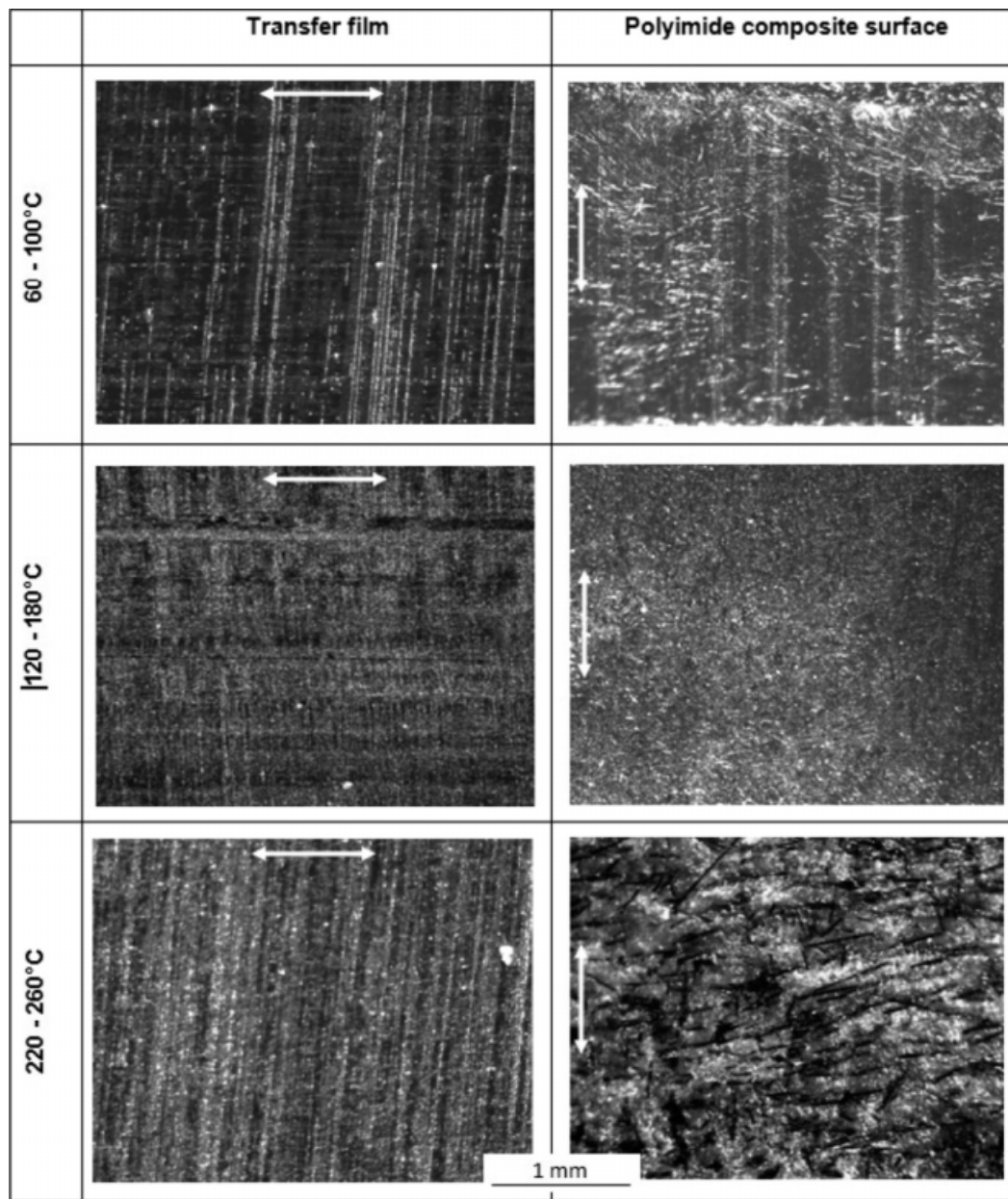
Figure 9 Microscopic observations of transfer films and worn polymer surfaces of CF–TP in different temperature regions (the sliding direction is indicated by the arrows; the same scale applies to all the micrographs).

direction. The polyimide and fillers were homogeneously mixed, so the CFs were protected by polyimide, and the CF–TP composites surfaces were smooth. The transfer film at 220–260°C was thick and continuous with polyimide depositions on top of the transfer film. Because of the better rheological properties of the transfer film at high temperatures, some flow lines were induced by shear. As the polymer matrix was preferentially worn, the CF–TP surface had unprotected CFs, which led to a high sliding resistance. Not all CFs were oriented along the molding direction; this indicated that they moved into the interface and acted as roller bearings shielded with a molten polyimide layer.

The transfer films for CF–TP–PTFE at 60–100°C were inhomogeneous, with alternating graphitized CF and PTFE. PTFE was transferred by mechanical shear as thin platelets surrounding the graphitized depositions; this limited their abrasiveness. Although TP almost did not transfer at low temperatures, the transfer film for CF–TP–PTFE favorably oriented parallel to the sliding direction through PTFE additives. The polyimide composite surface was protected, and shear bands near the edges resulted from the mechanical shear of PTFE. The continuous and smooth transfer films at 120–180°C contained a homogeneous mixture of graphitized CF, polyimide, and PTFE. The lubricating properties
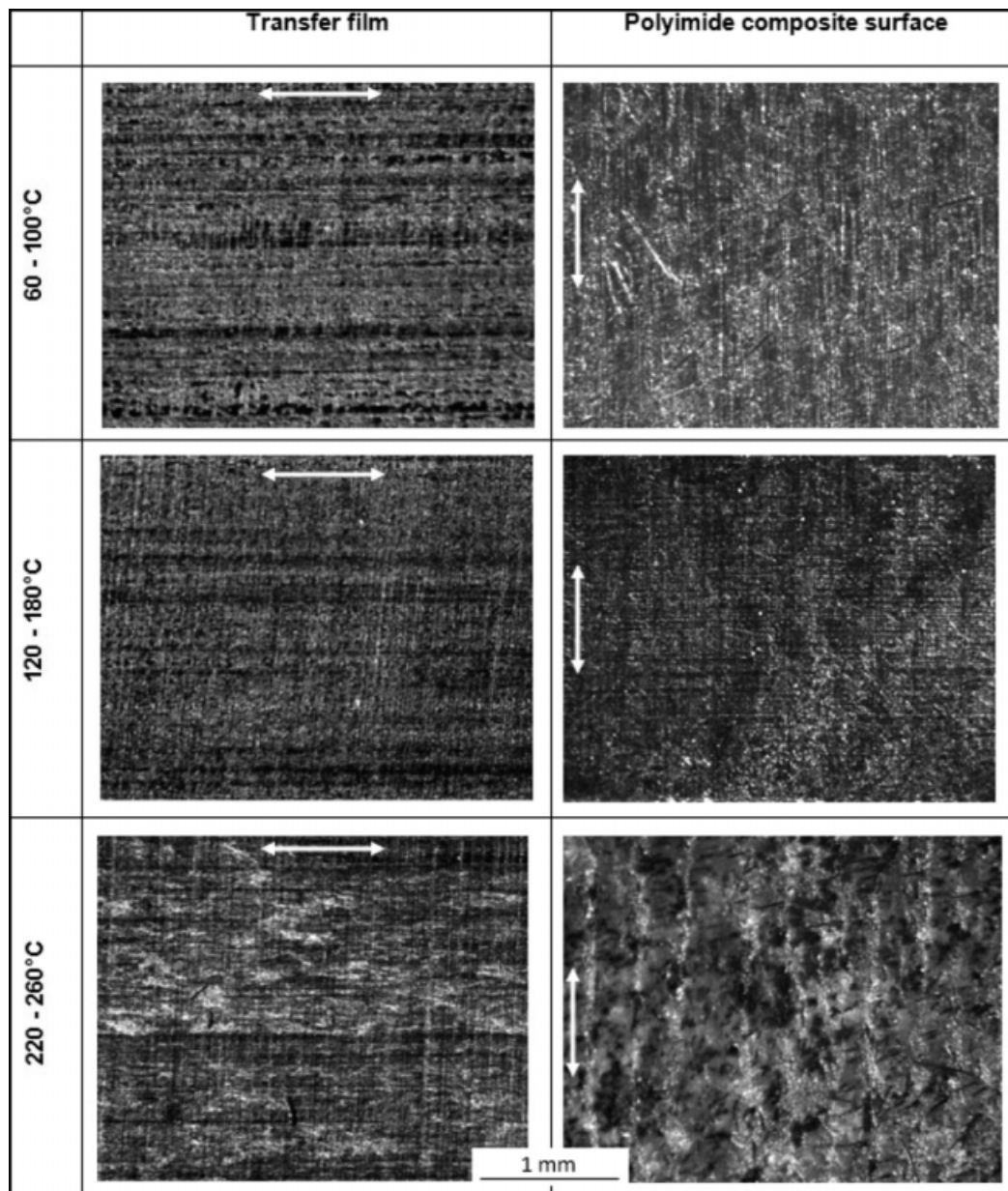
Figure 10 Microscopic observations of transfer films and worn polymer surfaces of CF–TP–PTFE in different temperature regions (the sliding direction is indicated by the arrows; the same scale applies to all the micrographs).

of PTFE were best in this temperature range. Also, the CF–TP–PTFE composite surface was completely smooth; this corresponded to the lowest coefficients of friction among the TP composites. The transfer film at 220–260°C had lumpy flakes along the sliding direction because of thermal degradation. A thick PTFE/TP layer developed on the composite surface, embedding the CFs and protecting against abrasion. The molten surface film had a similar morphology to CF–TP, and as shown by the almost similar steady-state coefficients of friction for CF–TP and CF–TP–PTFE, the lubrication of PTFE into a molten polymer film at high temperatures might have been limited.

The transfer films for CF–TP–oil were inhomogeneous at all temperatures; this led to high friction. At 60–100°C, no transfer film was observed: the striations parallel to the sliding direction represented abrasive grooves into the counterface. At 140–180°C, the mixing of polyimide and graphitized CF was prevented, and oil drops remained visible as a separate phase on the polyimide surface. The transfer film had graphite depositions that did not mix with the polyimide and were consequently abrasive. The wear products were readily removed from the sliding interface, which was observed as a large volume of debris at the edges of the sliding area sticking together into larger agglomerates. In contrast, the
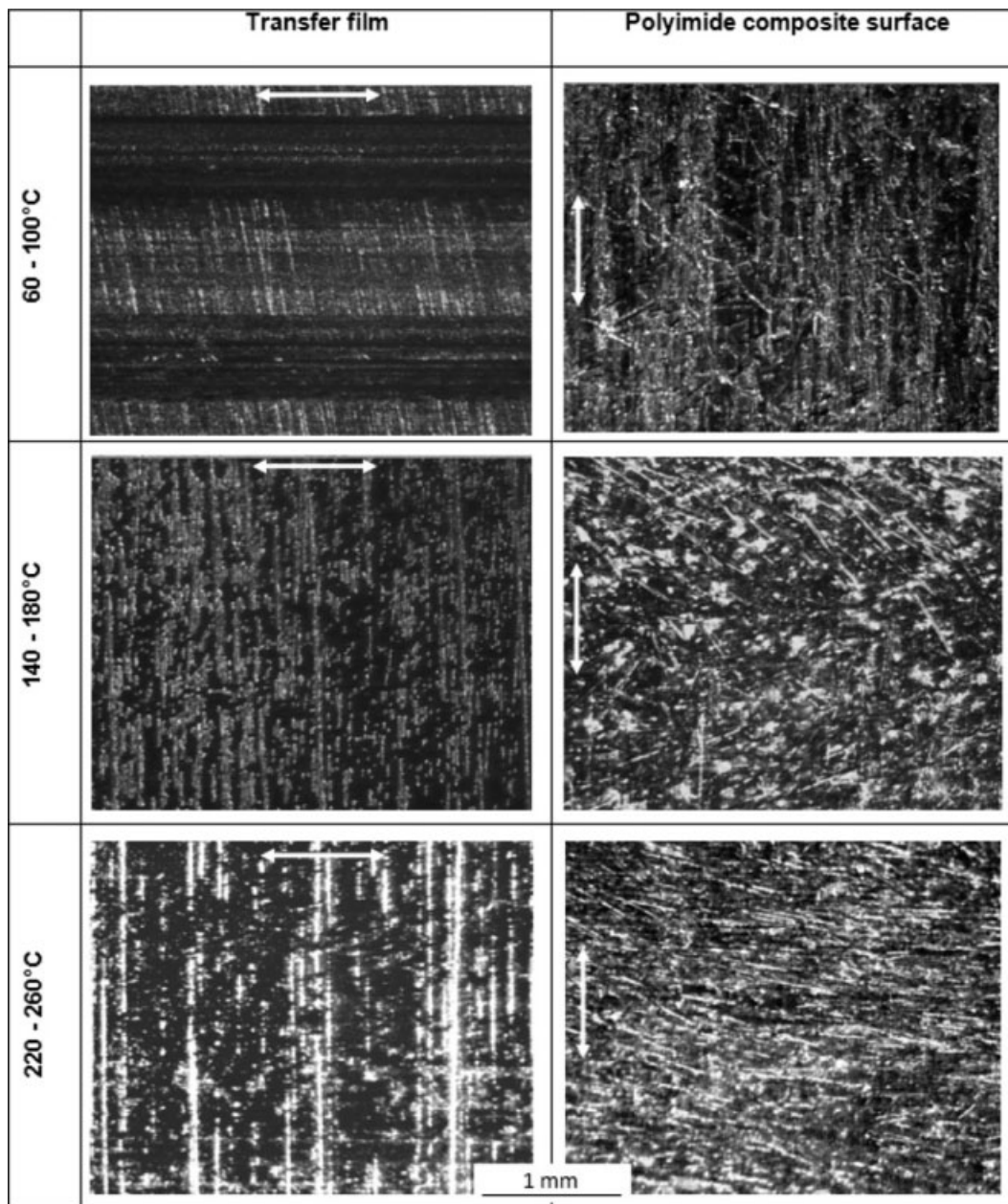
Figure 11 Microscopic observations of transfer films and worn polymer surfaces of CF–TP–oil in different temperature regions (the sliding direction is indicated by the arrows; the same scale applies to all the micrographs).

debris for CF–TP and CF–TP–PTFE was rather fine and powdery. At 220–260°C, a very thick and lumpy transfer film consisted of a viscous mixture with graphite, oil, and polyimide. As the polyimide matrix was easily worn, the CFs became free.

DISCUSSION

Effects of frictional heating

The interfacial temperature was higher than the applied bulk temperature because of frictional heating. For this sliding configuration, we verified a

model to calculate the maximum polymer surface temperature (T^*):[25]

$$T^* = T_{bulk} + 1.13\sqrt{\frac{\ell a}{v}}\frac{\mu p v}{k} \qquad (3)$$

where μ is the coefficient of friction, T_{bulk} is the bulk temperature, p is the contact pressure in the steady state, v is the sliding velocity, ℓ is the semicontact length, k is the polymer thermal conductivity (perpendicular to the fiber direction), and a is the thermal diffusivity. T^* was calculated for all of the tests (Table II). The running-in conditions were

TABLE II
Calculated T^* Values (°C) with Running-In and Steady-State Sliding for TP Composites
at Different Applied Bulk Temperatures

Bulk temperature (°C)	TP		CF–TP		CF–TP–PTFE		CF–TP–oil	
	Running-in	Steady-state	Running-in	Steady-state	Running-in	Steady-state	Running-in	Steady-state
60	150	80	210	102	100	95	202	172
80	177	98	293	97	110	103	224	213
100	190	120	285	105	128	110	248	145
120	210	152	293	124	155	122	250	153
140	232	182	300	143	172	147	283	166
180	298	192	310	185	203	189	304	203
220	350	244	340	225	232	231	324	245
260	398	292	384	266	285	266	380	295

determined by the Hertz contact pressure (90 MPa for $F_n = 50$ N). p was geometrically calculated with eq. (4) for worn cylinders with semiwidth b, radius R, and diameter reduction Δh under a normal load F_n:

$$p = \frac{F_n}{4b\sqrt{R^2 - (R - \Delta h)^2}} \quad (4)$$

The evolution of the coefficients of friction with sliding distance indicated important transitions caused by T^*. For TP, the heat-deflection temperature of 238°C was approached during running-in at 140°C bulk temperatures ($T^* = 232$°C); this caused sliding instabilities by softening. Melting was observed during running-in at 260°C ($T^* = 398$°C); this caused overloads. For CF–TP, CF–TP–PTFE, and CF–TP–oil, melting was avoided in all cases. For CF–TP, the coefficients of friction remained constant over the sliding distance at 60°C ($T^* = 210$°C), as there was no softening. The transitions at higher bulk temperatures were not only related to softening mechanisms of the polyimide bulk, but they likely resulted from a combination of chemical interactions between the polyimide matrix and additives, as reflected in the transfer film morphology. Tribochemical interactions likely occurred over a longer period of time and were rather induced by steady-state T^* values: hydrolysis of the imide moieties was observed at $T^* = 120–140$°C, whereas imidization took place at $T^* > 180$°C, as previously illustrated by Raman spectroscopy and thermal analysis.[26]

Effect of the CFs

CFs effectively increase the mechanical performance and thermal conductivity of a polyimide matrix, but they intrinsically have no self-lubricating properties. The thermal degradation of CF–TP in the presented temperature range was limited, as no obvious

weight loss was observed at temperatures below 490°C. The sliding properties of CFs depended on the tribochemical interactions in the sliding interface and interactions with the polyimide matrix. CF is a graphitized carbon with the hexagonal planes of its crystals aligned perpendicular to the fiber axis. The combination of mechanical shear and thermal processes exposed the CF as graphitized substances in the transfer film, which had flakelike graphite structures similar to those formed after the sliding of the graphite-filled sintered polyimides.[27] The lubricating ability of graphitized carbon should reduce the coefficients of friction and wear rates, but it is influenced by the chemical reactivity of graphite and the presence of dangling bonds at its surface.[28] Low friction and graphite lubrication only happens when the dangling bonds are deactivated through a reaction with the environment or transferred products. Graphite shows, therefore, favorable sliding properties under humid conditions. At steady-state T^* values below 100°C, the TP was inert, and it did not react with the fillers or it did not transfer. This was confirmed by a large fraction of polyimide wear debris outside the sliding interface. The active graphite sites were consequently not deactivated and strongly adhered to the counterface (this resulted in the formation of a thick transfer layer) and the sliding sample (this resulted in high friction). At steady-state T^* values of 120–180°C, hydrolysis of the polyimide moieties made them chemically active, and interactions between the polyimide debris and the graphitized CF resulted in more homogeneous transfer. Less wear debris simultaneously accumulated at the borders of the sliding stroke.

Effect of the PTFE additives

The lubricating properties of semicrystalline PTFE are associated with its planar structure and weak intermolecular bonding; these allow for the slipping of crystalline aggregates under shear and orientation.[29] Although PTFE has good thermal resistance

up to 400°C, efficient lubrication is only attained in a self-stabilizing temperature range where lamellar transfer layers provide optimum loadability and rheological properties. The thermal fluctuations in friction of PTFE were reported by Pleskachevsky and Smurugov:[30] the coefficients of friction decreased from 0.20 to 0.13 between 60 and 120°C and increased at higher temperatures. According to Yang,[31] the amount of PTFE transferred was almost constant during sliding at 130–180°C and mainly depended on the amount of amorphous PTFE. From this knowledge, there was very good agreement between the thermal dependency of the sliding properties for pure PTFE and the CF–TP–PTFE composites. The latter offered the lowest coefficients of friction and homogeneously mixed transfer films at $T^* = 120$°C, which was within the range of the thermally controlled sliding of PTFE. The running-in T^* values were higher and only governed during a short time, whereas tribochemically reactions and amorphous-to-crystalline transformations developed over longer periods. Thermally controlled sliding of PTFE only governed in a certain temperature range, as higher temperatures caused thermal weakening and a flakelike transfer film. The large amount of PTFE was beneficial for low friction but showed higher wear rates; this was in agreement with the conclusions of Hufenbach et al.[32] for poly(ether ether ketone)–PTFE composites: blends with 7.5 wt % PTFE showed the best wear behavior, whereas blends with 30 wt % PTFE exhibited the best friction performance.

Effect of the oil additives

The lubricating efficiency of oil in combination with CF–TP composites was limited at high temperatures, as the coefficients of friction and wear rates increased. The oil lubrication depended on exposure from the polymer bulk and the behavior of lubricant (mixed with polymer debris) in the sliding interface. Stable sliding requires a permanent oil supply and the formation of a thin sliding film. Means of lubricant supply were studied by Marchetti et al.[33] and were attributed to static effects through pores in the matrix (migration and capillary effects caused by the surface roughness and Maragoni effects caused by the temperature gradients) and dynamic effects (reciprocating sliding). Thermal effects in the oil supply were demonstrated with a sintered porous polyimide reservoir with good thermal resistance, and the extraction rate improved during heating at 35–100°C. The TP composites were relatively softer, and any polymer matrix deformation interfered with the oil supply mechanisms. Deformation may have favorably squeezed lubricant out of the bulk, but it may also have blocked the oil migration paths by

softening. The running-in T^* values for CF–TP–oil were significantly higher than the glass-transition temperature, and oil migration into the interface was consequently hindered through matrix deformation. Moreover, the incorporation of oil into the transfer film seemed difficult and prevented smooth transfer. The tribochemical interactions between the oil and graphitized CF were limited, whereas the oil visually adhered more likely onto the TP–composite surface. The higher compatibility between the oil and polymer resulted from the carbonaceous structure of both elements; this led to better structural adhesion. The sliding conditions under low velocities did not favor the development of a boundary-lubricated transfer film, whereas in some preliminary tests, the oil additives became more efficient in lowering the friction and wear rates at high velocities.

CONCLUSIONS

The compatibility of internal lubricants such as solids (PTFE) or liquids (silicon oil) for controlling the coefficients of friction and wear rates of thermoplastic carbon-fiber-reinforced polyimides was investigated as a function of the counterface temperatures.

The coefficients of friction for carbon-fiber-reinforced polyimide were either higher or lower than those of the TP, depending on the thermal regime. Internal lubricants such as PTFE caused the lowest coefficients of friction over the entire range of temperatures, with an optimum around bulk temperatures of 100–120°C, whereas internal silicon oil increased the coefficients of friction over the entire range of temperatures. The wear rates of the carbon-fiber-reinforced polyimide significantly decreased with respect to those of TP, whereas additional fillers slightly increased the wear rates. The latter reflected the improvements of mechanical strength for carbon-reinforced polyimide and the slight deterioration in strength with additional internal lubricants.

We obtained better insights into the sliding mechanisms of the carbon-fiber-reinforced polyimides by considering the T^* values. Mechanical overloads were determined by melting during a short sliding time at running-in, whereas thermochemical interactions took longer time and, therefore, better related to the steady-state polymer surface temperatures. The transfer film was abrasive and consisted of pure graphitized depositions at low temperatures where the polyimide was inert. The hydrolysis of the polyimide at intermediate sliding temperatures made the polyimide more active, and interactions between the polyimide debris and graphite resulted in more homogeneous transfer. The homogeneously mixed transfer in the presence of PTFE agreed with the thermal controlled sliding regime of PTFE lubricants

around 120°C, whereas the supply of oil lubricant in the sliding interface was hindered by the matrix deformation above the glass-transition temperatures during the running-in period. The deformation was related to the magnitude of friction or shear stresses in the subsurface. The interactions between the oil and graphitized depositions were limited, whereas it likely adhered onto the composite surface.

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