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Wear characteristic of epoxy resin filled with crushed-silica particles

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ABSTRACT

The dry-sliding wear of epoxy resin filled with 68.5 wt.% of crushed-silica particles (20–40 μm) were evaluated using pin-on-disk tests. At low sliding speeds, the bulk temperatures at contacts were below glass-transition temperature ($<T_g$); the wear process was time dependence. The volume loss increased with increasing sliding speed. Wear mechanisms were either abrasive wear or abrasive wear and particle detachment with epoxy–matrix deformation at low temperature. The wear process became temperature dependence at high sliding speeds ($>T_g$), i.e. the volume loss decreased with increasing sliding speed. Wear mechanisms were abrasive wear and particle detachment with epoxy–matrix deformation at high temperature.

1. Introduction

Polymers have simple manufacturing processes, however they have low thermal resistance and strength. On the other hand, ceramics have good thermal resistance, wear resistance, and strength, but they have low fracture toughness and complicated manufacturing process. The composites of these two engineering materials, i.e. polymer–matrix composites with small amount and size of ceramic reinforcements, could improve strength and fracture toughness comparing to those of polymers. Moreover, they could be easily manufactured and used in various applications, e.g. automobiles, ships, sport, and aerospace [1].

During service, the contact and sliding between two solids are possible, which result in the failure of their surfaces or wear. This failure is one of the main disadvantages of polymer–matrix composites. The process of material removal in dry sliding of polymer composites involves not only polymer–matrix wear but also the reinforcement wear, reinforcement fracture, and interfacial debonding. The reinforcement fracture and interfacial debonding are likely to occur sequentially, and can be considered as a combined process [2]. Cirino et al. [3] found that this combined process occurred in polymer–matrix composites with glass reinforcement, and caused an additional matrix wear or third-body abrasive wear. Thus, the polymers reinforced with fiberglass had higher wear rate than the same polymers reinforced with lower hardness material, e.g. carbon fiber.

Until present, the polymer–matrix composites filled with low contents of ceramic particles (<20 wt.%) have attacked attention from material scientists and engineers, and their wear properties have been extensively studied. Xing and Li [4] have studied the wear behavior of epoxy–matrix composites filled with uniform sized sub-micron spherical silica particles (120 nm and 510 nm in diameter). The spherical silica particles could improve the wear resistance of the epoxy–matrix, even though the content of the fillers was at a relatively low level (0.5–4.0 wt.%). Shi et al. [5] have studied the dry sliding wear of epoxy-based composites reinforced with nanometer silicon nitride particles. The significant improvement of tribological performance and mechanical properties was observed at rather low filler content (typically less than 1 vol.%).

As alternative engineering materials, the polymer–matrix composites filled with high contents (>20 wt.%) and large sizes (>20 μm) of ceramic particles have good fracture toughness and simple manufacturing process, while maintain the advantages of ceramics, i.e. high strength and hardness. Nagata et al. [6] successfully developed an epoxy resin filled with high content of crushed-silica particles (>50 wt.%), which had a fracture toughness of 2.5 MPa $m^{1/2}$. This magnitude of fracture toughness was significantly higher than those of neat epoxy resin (0.5 MPa $m^{1/2}$) and epoxy–matrix composites filled with 10 vol.% of 13 nm Al_2O_3 particles (1.15 MPa $m^{1/2}$) [7]. Unfortunately, the understanding of wear behavior and mechanisms of these composites is far from complete. It is therefore the objective of the present work to study the dry-sliding wear behavior and mechanisms of epoxy resin filled with 68.5 wt.% crushed-silica particles (20–40 μm). The effects of contact load and sliding speed on wear behavior were evaluated using pin-on-disk tests. The wear mechanisms at each tribological condition were then discussed.

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2. Materials and experimental procedures

2.1. Material preparation

Typically, the silica has a crystal structure. By melting crystallized silica and then rapid cooling, the silica with an amorphous structure can be created. The mechanical strength of amorphous silica is less than that of crystallized silica, however its thermal expansion coefficient is lower. Therefore, amorphous silica is not only a reinforced material but also an effective material for decreasing the thermal expansion of epoxy resin.

In the present work, the amorphous silica was mechanically crushed to obtain 20–40 μm silica particles. The epoxy resin filled with crushed-silica particles was the combination between base material and hardener material at 1:1 weight ratio. Base material was the mixture between 35 wt.% of epoxy resin (bisphenol I) and 65 wt.% crushed-silica particles, while the hardener was the mixture between 28 wt.% of acid anhydride and 72 wt.% crushed-silica particles. Thus, the 68.5 wt.% (or 45 vol.%) crushed-silica particles were obtained in this composite. The liquid of epoxy resin filled with crushed-silica particles was vacuumed at 25 °C in a vacuum chamber to remove the bubbles originally trapped in liquid, poured into a mold to the form of flat plate, and cured at 85–90 °C for 12 h. SEM micrograph of the epoxy resin filled with crushed-silica particles is shown in Fig. 1. The white phases are silica particles, while the dark phase is epoxy resin. The mechanical and thermal properties of the material are given in Table 1 [6].

Table 1
Mechanical and thermal properties [6].

Tensile strength (MPa)	79.0
Modulus of elasticity (GPa)	12.5
Hardness (HRB)	114.5
Fracture toughness, K_{IC} (MPa m$^{1/2}$)	2.5
Thermal expansion coefficient (10^{-6} K)	21.0
Glass transition temperature (°C)	108
Density (g/cm^3)	1.73

2.2. Dry-sliding wear testing

Dry-sliding wear tests were performed using a pin-on-disk wear machine (Phoenix Tribology: TE-88). The geometry of specimen, and experimental procedure followed the ASTM recommendation [8]. The geometries of pin specimen and disk specimen are shown in Fig. 2. Using a diamond-cutting machine, pin specimens were cut from a plate of composite, and polished by SiC papers (80, 400 and 800 grit) to obtain a bar of 8 × 8 × 20 mm. The disk specimens were cut from a rod of wrought 304 stainless steel to obtain a diameter of 75 mm, and a thickness of 10 mm. It is known that wear resistance depends on the properties of contact surfaces. Thus, contact surfaces of pin specimen and disk specimen were manually polishing with 1000-grit SiC papers to obtain a surface roughness of 0.2 μm Ra. Before wear test, the hardness at contact area of both pin specimen and disk specimen were measured, i.e. 114.5 HRB for pin specimen and 83 HRB for disk specimen.

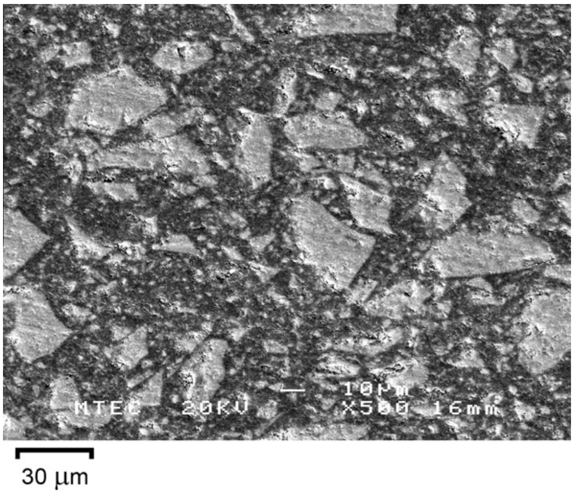

Fig. 1. SEM micrograph of epoxy resin filled with crushed-silica particles.

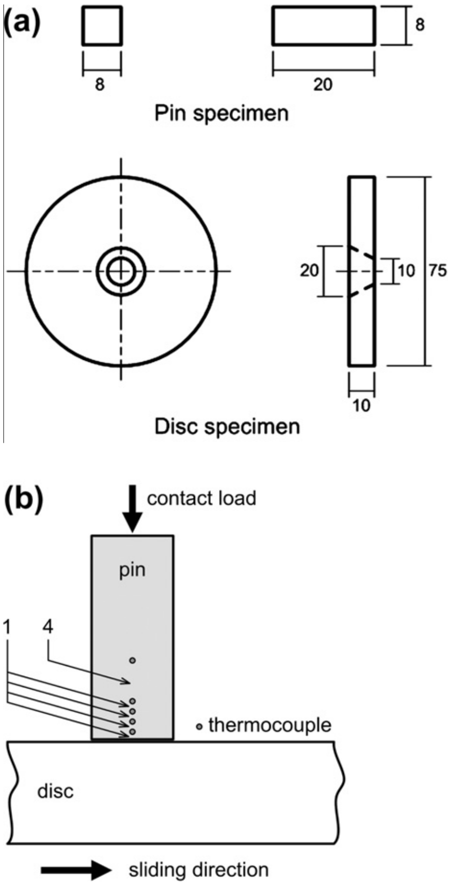

Fig. 2. (a) Geometries of specimens, and (b) schematic illustration of wear test (dimension in mm).

The wear tests were carried out under ambient environment (25 ± 2 °C and 55 ± 5% relative humidity) with a sliding speed range of 0.2–2 m/s, a contact load range of 70–200 N (approximately 1–3 MPa contact pressure), and a sliding distance of 2 km. After removing loose debris by high-pressure air, the difference in weights of pin before and after wear test (mass loss of pin) was determined. The weight measurement was performed by an analytical balance with 0.1 mg precision. Consequently, the volume loss was determined by dividing the mass loss by density. The wear rate was calculated as the rate of volume loss per contact load and sliding distance. The wear tests were repeated twice for each tribological condition, and the averages of volume losses and wear rates were determined.

During wear test, the volume loss was obtained by continuously measuring the pin height by a linear transducer. The temperature at side surface of pin during the wear test was also measured using chromel–alumel type thermocouple probes, which were attached at approximately 1–8 mm above the contact area (Fig. 2b). The epoxy adhesive was used for the attachment of thermocouples. The calibration of thermocouples was performed in accordance with the ASTM recommendation [9]. The pin height, temperature, and time were simultaneously recorded every second with personal computer-controlled data acquisition. On the other hand, the friction force was measured by a load cell equipped within the wear machine. Investigations of worn surfaces and loose debris were undertaken using a scanning electron microscope (SEM) equipped with an energy dispersive X-ray spectrometer (EDS).

3. Results and discussion

3.1. Wear behavior

For polymeric materials, the volume loss during wear test could be influenced by thermal expansion and creep deformation. Using volume loss measured by a linear transducer to assess the wear rate may not reliable. Therefore, the volume losses estimated from mass losses of pin were compared with those measured using a linear transducer. It was found that the differences in volume loss between both methods were less than 5%. The thermal expansion and creep deformation in longitudinal direction of pin were unlikely the dominant mechanism, and the volume loss obtained from both methods could be used in the present work.

Relationships between volume loss and sliding distance are shown in Fig. 3. A steady state was reached after approximately 200–300 m of running-in state. For neat polymers sliding against steel [10], it was found that the wear resistance increased with sliding distance. This increase was caused by progressive smoothing of the surface, and by the formation of a protective transfer film of polymer on the steel disk. However, no transfer film of polymer on the disk specimens was found in this work. It is likely that the contact and sliding between crushed-silica particles and surface of disk specimen may prohibit the formation of the protective transfer film.

The volume losses of pins after wear tests are shown as a function of sliding speed in Fig. 4. The volume losses at all sliding speeds increased with increasing contact load. At each contact load, there was an increase in volume losses over a range of low sliding speeds, and then the volume losses decreased after some transition sliding speeds. The transition sliding speeds were approximately 1.4 m/s for wear tests at 70 N contact load, and 0.8 m/s for wear tests at 140 and 200 N contact loads. It is noted that the volume loss of polymeric composites generally depends on sliding speed or temperature, i.e. it increases with the increase of sliding speed or temperature. However, the opposite behavior was observed here, i.e. the volume loss at high sliding speed or high temperature decreased with the increase of sliding speed or temperature. After wear tests, the volume losses of disk specimens were also determined. The wears of disk specimens at the present tribological conditions were mild, i.e. less than 0.5% of original volumes were removed from the surfaces of disk specimens.

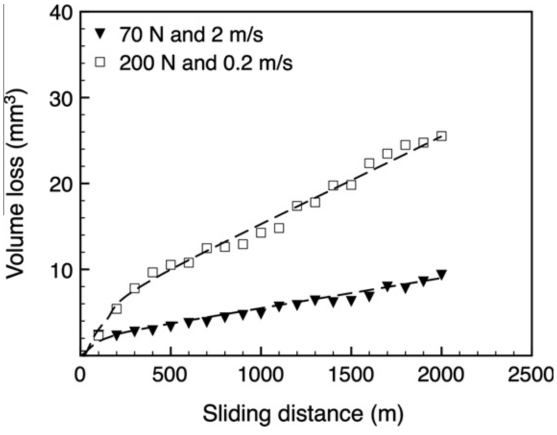

Fig. 3. Relationships between volume loss and sliding distance.

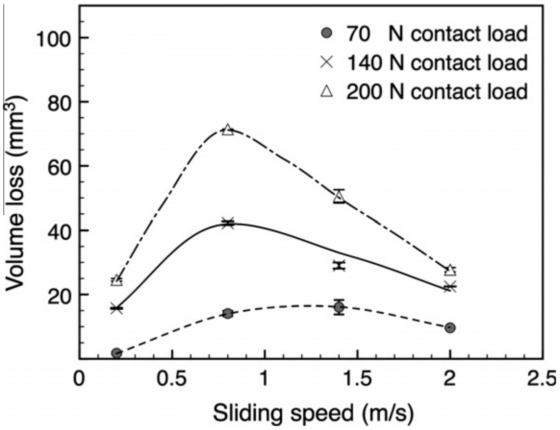

Fig. 4. Relationships between volume loss and sliding speed.

The friction force was high at the beginning of wear test, decreased with increasing sliding distance, and then became stable. At the present tribological conditions, the stable friction coefficients were in the range of 0.2–0.4. It was observed that the onsets of sliding distance for stable friction force corresponded to those for steady-state volume loss (Fig. 3).

During wear test, the friction heat could generate at the tiny contact areas (asperities of material), which results in high temperature at these areas (flash temperature). Since the crushed-silica particles are the major phase of the present composite (Fig. 1); the contact occurred mainly between surfaces of crushed-silica particles and 316 L stainless steel. For low thermal conductivity material like silica, the flush temperature was constrained around the contact surface of crushed-silica particles. This flush temperature rapidly decreased with the increase of the distance from the contact surface of pin, and became the bulk temperature. Since the size of crushed-silica particles is relatively large (20–40 μm) and the epoxy resin has a low thermal resistance; the integrity between epoxy resin and crushed-silica particles is likely to be influenced by the bulk temperature around the contact surface.

At each tribological condition, the temperatures at side surface of pin increased with increasing sliding distance, and then reached a steady state. The pin temperatures at steady state increased with decreasing distance from contact, and became almost stable at 1 mm from contact, as shown in Fig. 5. Thus, these stable temperatures were considered as the bulk temperatures at contact surfaces. The relationships between pin temperature at 1 mm from contact and sliding speed are shown in Fig. 6. For comparison, the glass-transition temperature (T_g) of this composite was added into Fig. 6. The pin temperature increased with increasing contact load and sliding speed. At 70 N contact load, the influence of sliding speed on pin temperature was weaker than those of 140 N and 200 N contact loads.

It is known that the fracture toughness of epoxy resin is time dependence, i.e. fracture toughness decreases with increasing loading rate [11], and increases with increasing temperature [12]. For wear tests at 70 N contact load, the pin temperatures at 0.2–1.4 m/s sliding speeds were lower than glass-transition temperature (T_g). Thus, the effect of loading rate on fracture toughness

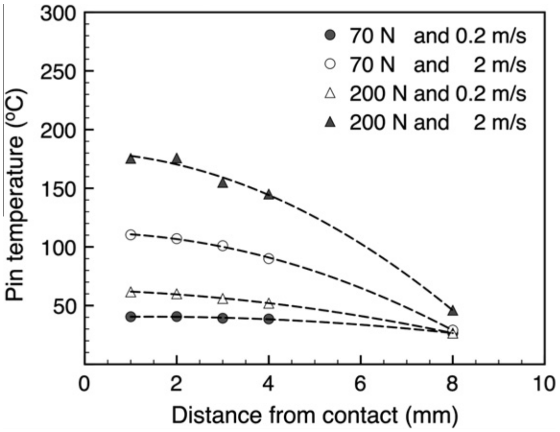

Fig. 5. Relationships between pin temperature and distance from contact.

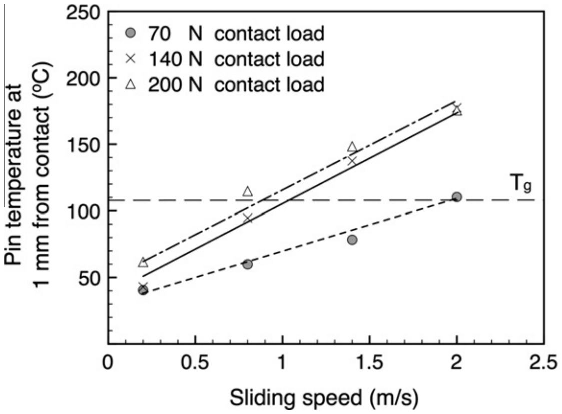

Fig. 6. Relationships between pin temperature at 1 mm from contact and sliding speed.

of epoxy–matrix was stronger than the effect of temperature, and the wear behavior under these tribological conditions was controlled by the effect of sliding speed. The volume loss then increased with increasing sliding speed until the transition sliding speed of 1.4 m/s (Fig. 4). Beyond this transition sliding speed, the pin temperature was higher than glass-transition temperature. Above glass-transition temperature, the ductility and fracture toughness of epoxy–matrix increased significantly, then the wear behavior was controlled by the effect of temperature. The volume therefore decreased with increasing sliding speed (Fig. 4). Similar wear behaviors were observed for wear tests at 140 and 200 N contact loads. However, their pin temperatures reached the glass-transition temperature at slower sliding speeds, i.e. 0.8 m/s sliding speed.

3.2. Wear mechanism

For the present composite material, it was found that the effects of sliding speed and/or temperature on fracture toughness of epoxy–matrix could influence epoxy wear as well as crushed-silica detachment. According to the analysis of worn surfaces and debris, the wear mechanisms could be divided into three categories, as follows.

3.2.1. Abrasive wear without particle detachment

SEM micrographs of worn surface and debris from wear test at 70 N contact load and 0.2 m/s sliding speed are shown in Fig. 7. The grooves dominated worn surface (Fig 7a), and the debris comprised of small particles and their agglomerations (Fig. 7b). None of large crushed-silica particle detachment was observed in debris. According to the EDS analysis (Fig. 7c), the debris was the combination between small fragments of silica and stainless steel. It is noted that EDS analysis could give only semi-quantitative results when analyzing light elements. Thus, the presence of epoxy resin in debris could not be confirmed by an EDS analysis. However, the uniform appearance of grooves (Fig. 7a) indicated that the particles of epoxy resin were removed from the pin specimen. Then, it is believed that the epoxy resin was a part of the debris. Since the hardness of disk specimen was lower than that of pin specimen; the small silica particles in debris were likely the cause of grooves on worn surface, i.e. third-body abrasive wear. Schematic illustration of this wear mechanism is shown in Fig. 8a.

Without the detachment of large crushed silica, the volume loss at 70 N contact load and 0.2 m/s sliding speed was significantly low (Fig. 4). Similar observation was reported by Durand et al. [13], who studied dry-sliding wear behavior of polymer reinforced with large ceramic particle (>20 μm) at 10 N contact load and 0.1 m/s sliding speed. They found that the ceramic particles embedded and protected the polymer matrix during wear process, which resulted in low wear rate. Unfortunately, the wear behaviors and mechanisms at high sliding speeds and high contact loads have not been evaluated in their study.

3.2.2. Abrasive wear and particle detachment with epoxy–matrix deformation at low temperature

Below glass-transition temperature, the fracture toughness of epoxy–matrix was time dependence [11], i.e. it decreased with increasing sliding speed. As sliding speed increased; the fracture toughness of epoxy–matrix became lower; and the crushed-silica particle detachment was possible. Evidences of abrasive wear with crushed-silica detachment at 200 N contact load and 0.2 m/s sliding speed are shown in Fig. 9. Large cavities and grooves were prominent on worn surface (Fig. 9a). While, the debris was the combination between plate-like, and particle-like debris (Fig. 9b). According to the EDS analysis, the plate-like debris was silica. While, the particle-like debris was the mixture of epoxy resin, silica, and stainless steel. Thus, the wear mechanism was the combination between third-body abrasive wear and particle detachment with epoxy–matrix deformation at low temperature ($<T_g$ of composite), as schematically shown in Fig. 8b. It was also observed that more crushed-silica particles were removed with increasing sliding speed. This observation corresponded to the previous finding, i.e. the volume losses at temperatures below T_g increased with increasing sliding speed (Fig. 4).

3.2.3. Abrasive wear and particle detachment with epoxy–matrix deformation at high temperature

At high contact loads and sliding speeds, the temperatures of pins became higher than the glass-transition temperature of composite (Fig. 6). The composite loosed its brittleness, then the crushed-silica particle detachment with deformation of epoxy–matrix was possible. Evidences of crushed-silica detachment and epoxy–matrix deformation at 200 N contact load and 2 m/s sliding speed are shown in Fig. 10. Large elliptic-shape cavities and grooves were prominent on worn surface (Fig. 10a). While, the debris was the combination between plate-like, and particle-like debris (Fig. 10b). According to the EDS analysis, the plate-like debris was silica. While, the particle-like debris was the mixture of epoxy resin, silica, and stainless steel. The wear mechanism was therefore the combination between third-body abrasive wear and particle detachment with epoxy–matrix deformation at high temperature ($>T_g$ of composite), as schematically shown in Fig. 8c.

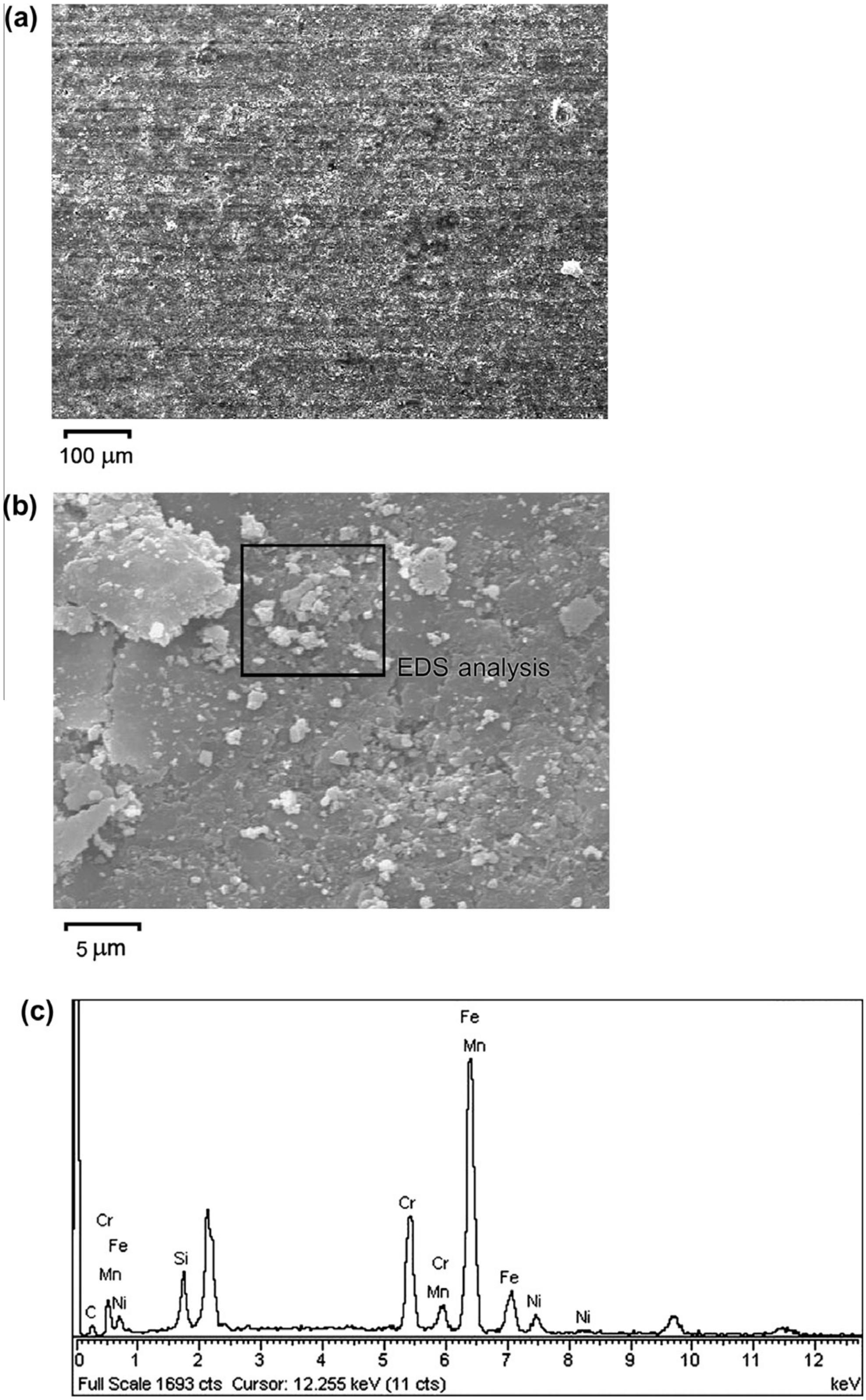

Fig. 7. Abrasive wear without particle detachment at 70 N contact load and 0.2 m/s sliding speed; (a) worn surface (sliding direction from left to right), (b) debris, and (c) EDS analysis.

Above the glass-transition temperature, the ductility and fracture toughness of epoxy–matrix were temperature dependence [12], i.e. they increased with increasing temperature or increasing sliding speed. As the fracture toughness of epoxy–matrix increased; less crushed-silica particles could be removed. From the observations of worn surfaces and debris, it was found that the detachment of crushed-silica particles at high temperature (>T_g of composite) decreased with increasing sliding speed. This

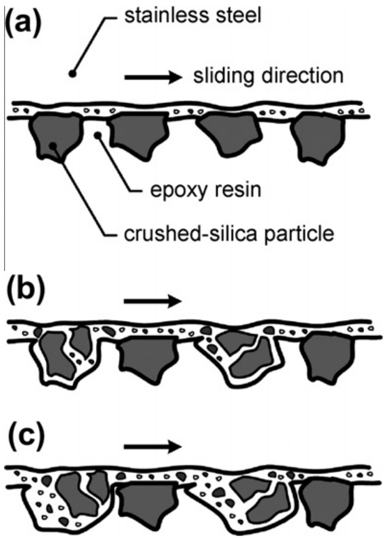

Fig. 8. Schematic illustrations of wear mechanisms; (a) abrasive wear without particle detachment, (b) abrasive wear and particle detachment with epoxy–matrix deformation at low temperature, and (c) abrasive wear and particle detachment with epoxy–matrix deformation at high temperature.

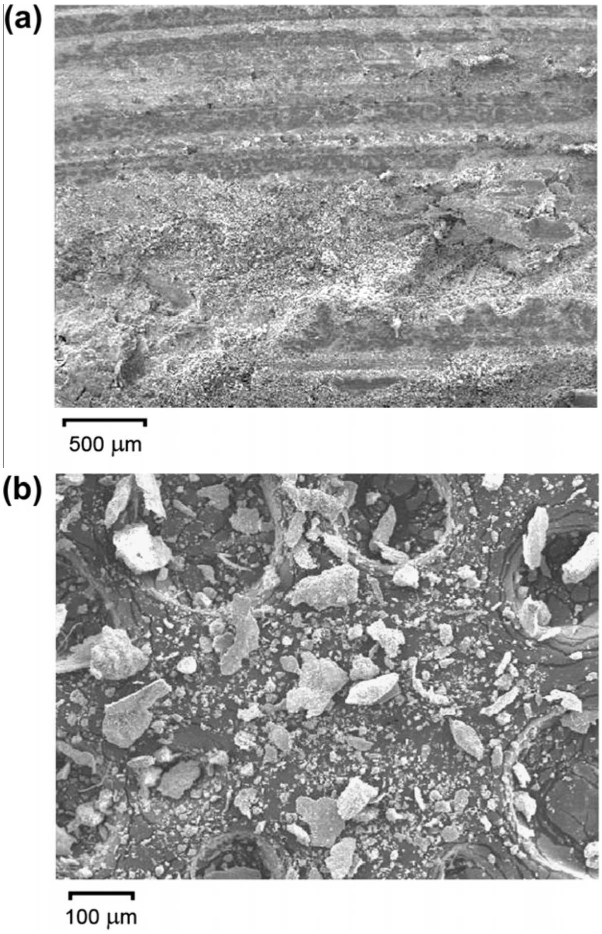

Fig. 9. Abrasive wear and particle detachment with epoxy–matrix deformation at low temperature at 200 N contact load and 0.2 m/s sliding speed; (a) worn surface (sliding direction from left to right), and (b) debris.

finding corresponded to the decrease in volume loss with increasing sliding speed (Fig. 4).

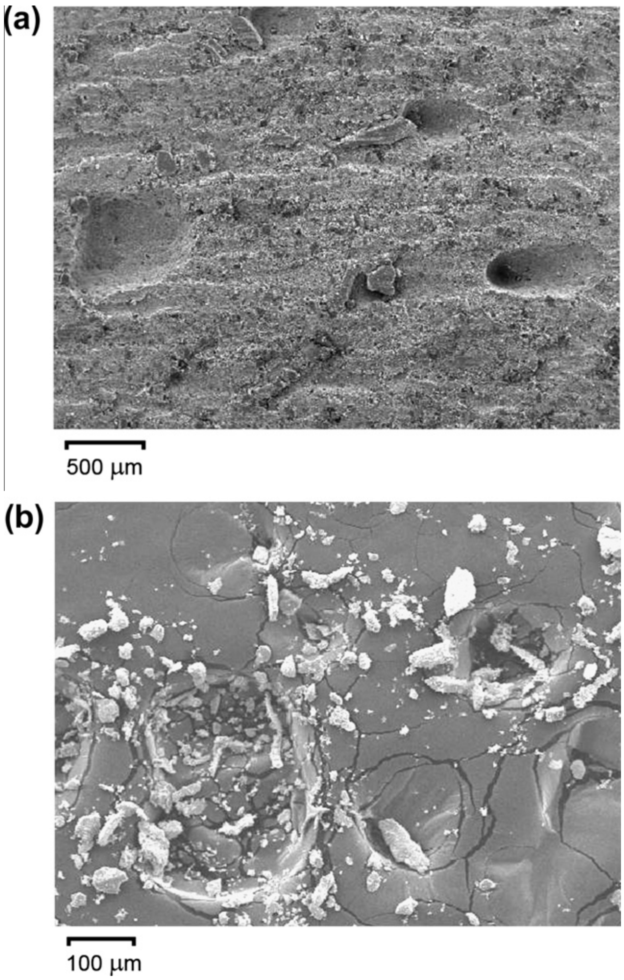

Fig. 10. Abrasive wear and particle detachment with epoxy–matrix deformation at high temperature at 200 N contact load and 2 m/s sliding speed; (a) worn surface (sliding direction from left to right), and (b) debris.

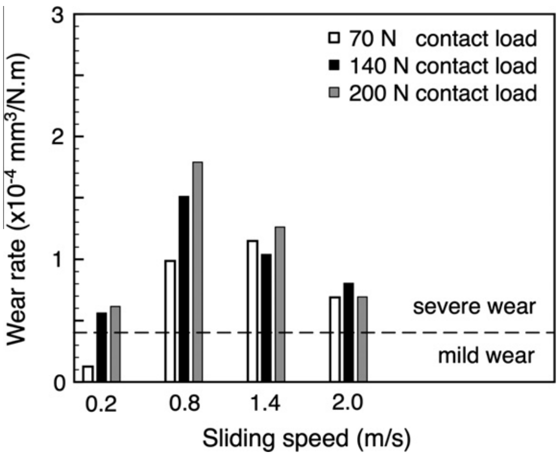

Fig. 11. Relationships between wear rate and sliding speed.

3.3. Wear mechanism map

As a rate of volume loss per contact load and sliding distance, wear rate at each tribological condition are shown in Fig. 11. At 70 N contact load (approximately 1 MPa contact pressure) and

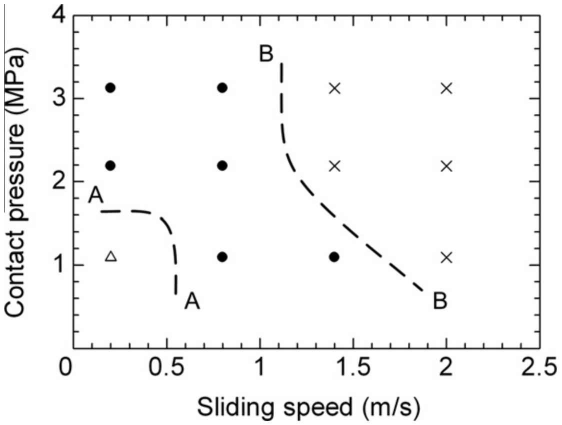

Fig. 12. Wear mechanism map; (△) abrasive wear without particle detachment, (●) abrasive wear and particle detachment with epoxy–matrix deformation at low temperature, and (X) abrasive wear and particle detachment with epoxy–matrix deformation at high temperature.

0.2 m/s sliding speed, the wear rate was 0.12×10^{-4} mm^3/N m, which was significantly lower than that of neat epoxy resin (3.75×10^{-4} mm^3/N m) [13]. While, this wear rate was slightly lower than that of epoxy–matrix composite filled with 2 wt.% of 120 nm silica particles, i.e. 0.2×10^{-4} mm^3/N m [4]. It is clear that the improvement of wear resistance was possible with the filling of crushed-silica particles.

The transitions of wear mechanisms are usually accompanied by changes in wear rate of one or often multiple orders-of-magnitude. Thus, the wear mechanism of this composite could be classified into two main wear regimes, i.e. mild wear regime and severe wear regime. Mild wear regime was defined as wear situations, in which the wear rates were low, and worn surfaces were fine. On the other hand, severe wear regime was defined as wear situations, in which the wear rates were high, and worn surfaces were coarse. In the present work, the abrasive wear was the main mechanism in mild wear regime. While, there were two sub-regimes in the severe wear regime, i.e. abrasive wear and particle detachment with epoxy–matrix deformation at low temperature, and abrasive wear and particle detachment with epoxy–matrix deformation at high temperature.

For comparison, these wear mechanisms were graphically shown using wear mechanism map (Fig. 12). The transition from abrasive wear (mild wear) to abrasive wear and particle detachment with epoxy–matrix deformation (severe wear) was indicated by line AA. On the other hand, the transition from abrasive wear and particle detachment with epoxy–matrix deformation at low temperature ($<T_g$) to abrasive wear and particle detachment with epoxy–matrix deformation at high temperature ($>T_g$) was indicated by line BB. Under abrasive wear (mild wear), the present material could be an alternative selection for sliding applications without additional surface coatings and/or lubricants. On the other hand, the abrasive wear with crushed-silica detachment was severe wear. Using additional surface coatings and/or lubrications may reduce surface traction, reduce abrasion, reduce temperature, reduce crushed-silica detachment, and consequently improve wear resistance.

4. Conclusion

The effects of contact loads (70–200 N) and sliding speeds (0.2–2 m/s) on dry-sliding wear characteristic of epoxy resin filled with 68.5 wt.% of crushed-silica particles (20–40 μm) were evaluated using pin-on-disk tests. The findings are summarized as follows

(1) The volume losses at all sliding speeds increased with increasing contact load. At each contact load, there was an increase in volume losses over a range of low sliding speeds, and then the volume losses decreased after some transition sliding speeds. The transition sliding speeds were approximately 1.4 m/s for wear tests at 70 N contact load, and 0.8 m/s for wear tests at 140 and 200 N contact loads.
(2) At low sliding speeds, the temperatures were below glass-transition temperature (T_g). The wear process was time dependence, and the volume loss increased with increasing sliding speed. The wear mechanisms were either abrasive wear or abrasive wear and particle detachment with epoxy–matrix deformation at low temperature ($<T_g$ of composite).
(3) At high sliding speeds, the temperatures were above glass-transition temperature (T_g). The wear process became temperature dependence; i.e. the volume loss decreased with increasing sliding speed. The wear mechanisms were abrasive wear and particle detachment with epoxy–matrix deformation at high temperature ($>T_g$ of composite).

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