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Tribology of composites produced with recycled GFRP waste

Juliana R Souza[1,2], Ruthilene CL Silva[3], Laís V Silva[4,5], João TN Medeiros[1,2], Sandro C Amico[4,5] and Witold Brostow[6]

Abstract

Nowadays, is expected that for most materials to be environmentally friendly. Besides, waste from end-of-life products may be considered a secondary source of materials with an energetic advantage due to its high energy content. This paper deals with the study of friction and wear characteristics of Glass fibre-reinforced polymer (GFRP) composites with polyester/glass fiber (P/GF) waste as filler, replacing the widely used calcium carbonate ($CaCO_3$). Polyester composites based on two or three components, using a combination of polyester, $CaCO_3$, GF, and GF waste, were produced. Pin-on-disc sliding wear test was performed using a polished stainless steel counterface. Roughness, surface energy, and hardness of the composites were characterized before the tests. The GF content (15, 25, 35, and 50 wt.%), the sliding velocity (0.021 and 0.042 m/s), and the normal load (1, 5, and 10 N) were varied. Based on the experimental results, it was observed that the friction coefficient and wear rate were influenced by material composition, surface roughness and energy, adhesive, and abrasive contact mechanisms. P/GF composites having P/GF waste presented enhanced performance considering friction and wear in relation to those with $CaCO_3$ in their composition.

Keywords
Recyclability, wear, glass fiber, polymer composites, waste

Introduction

Nowadays, polymers and their composites are widely used in many situations where machine components are under tribological loading conditions. The use of a polymer and one (or more) solid filler yields a combination of properties of the various phases. Many fillers can be used including calcium carbonate, glass fibers (GFs), talc, kaolin, mica, wollastonite, silica, graphite, high-performance fibers (e.g. carbon, aramid).[1–5] In Europe, approximately 1 mton of composites are manufactured each year and, in Brazil, the estimate for 2011 reaches 211 kton.[6]

There are many successful uses for thermoset composite materials, but recycling at the end of their life cycle is a difficult task. This occurs due to two main reasons: (i) their complex nature, since they are a mixture of materials of mixed nature: polymer, fibrous reinforcement (usually glass or carbon fiber), and often fillers (e.g. cheap mineral powders for a variety of functions such as fire retardants or to lower cost); and (ii) the common use of thermoset resins, which have covalent bonds that do not allow remolding.[1,7,8] Besides, there are few standard formulations and, for most applications, the type and proportion of resin, reinforcement, and filler are tailored for a particular end use.[7]

Much of the currently produced composite waste must be ultimately disposed of via landfill or

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[1] Department of Mechanical Engineering, PPGEM, Federal University of Rio Grande do Norte, Natal, Brazil
[2] Group of Tribology Studies and Structural Integrity, UFRN, Natal, Brazil
[3] Department of Mechanical and Materials, Federal Institute of Rio Grande do Norte, Natal, Brazil
[4] Department of Materials Engineering, PPGE3M, Federal University of Rio Grande do Sul, Porto Alegre, Brazil
[5] Composites and Nanocomposites Group, UFRGS, Brazil
[6] Laboratory of Advanced Polymers and Optimized Materials, Department of Materials Science and Engineering, University of North Texas, Denton, USA

Corresponding author:
Juliana R Souza, Department of Mechanical Engineering, PPGEM, Federal University of Rio Grande do Norte, Brazil, Group of Tribology Studies and Structural Integrity, Center of Technology, Brazil.
Email: julianars.materiais@gmail.com

incineration. Both of them are often considered unsatisfactory (especially the former), because of high cost, technical difficulties, and environmental impact.

Recycling of composites is rapidly becoming an environmental necessity, being sometimes a key barrier to their development or even continued use in some markets.[6] A few techniques are mentioned as potential alternatives to the recycling and/or reuse of end-of-life composites, including low temperature incineration with fiber recovery, dissolution of the polymer component using solvents, and mechanical recycling (grinding or crushing mill).[8]

In the mechanical recycling process, all constituents of the original composite are reduced in size and appear in the resulting recyclate which is a mixture of polymer, fiber, and filler. Recyclates (crushing mill residue) comprised of fine powders could be used as a substitute for calcium carbonate filler in new compounds.[9] An extra advantage, apart from the positive environmental aspect, is that the recyclate has lower density than the calcium carbonate, since it contains a significant content of low density polymer.[7]

Mechanical properties of polymeric matrix composites have been widely investigated in the past decades. In earlier studies, it has been found that the incorporation of fiberglass waste into polyester resin composites considerably enhanced their mechanical properties such as tensile modulus and impact strength, enabling the use of the waste as an efficient alternative for its recycling.[10] Addition of calcium carbonate and fiberglass waste of the automotive industry in a polyester matrix enhanced tensile strength of the composite and although $CaCO_3$ gives good surface finish it may weaken the material if used in excess.[11]

From the application point of view, the actual use of fiber reinforced composites requires good understanding of the correlation between processing and structure and their influence on wear and friction. Sometimes, high coefficient of friction, coupled with low wear, is required (e.g. for brake pads or clutches). In most cases, however, the primary concern is to develop polymeric composites that possess low friction and low wear characteristics under dry sliding conditions against smooth metallic counterparts (e.g. as in gears or bearings).

Moreover, the relationship between formulation and performance is not clear, and complex problems and instabilities in the coefficient of friction, excessive wear, vibration, and noise may all be present during friction processes of polymer matrix composite materials.[12] For this reason, the tribological behavior of these materials has to be investigated in the laboratory, following standard tests.[13]

Thus, the aim of this work is to study the recycling of polyester/glass fiber (P/GF) composites incorporating their ground waste in new P/GF composites. The ability of this waste was mainly evaluated to substitute calcium carbonate used as filler in new compounds focusing on tribological tests carried out under different conditions.

Experimental details

P/GF waste

The waste was obtained from waste (W) or end-of-life unsaturated P/GF composites, consisting of plates with 12 wt.% of fiber, which had been produced by light resin transfer molding. The plates were cut and ground in a knife mill with 8 mm × 8 mm screen, then finely ground in a ball mill for a minute, yielding waste retained in the 16 mesh sieve.

Preparation of composite samples

The tests were performed on various samples consisting of two or three components, varying the type and amount of filler and/or fiber. The formulation of each composite is displayed in Table 1.

Composites were produced in the laboratory with unused polyester resin (UCEFLEX UC 5518 from Elekeiroz), fiber glass mats (300 g/m^2), and calcium carbonate. The composites were hot compression molded, with 6 ton distributed in a metal mold (inner cavity: 270 mm × 170 mm) at 90°C for 60 min.[9] The $CaCO_3$, when used, was added to the polyester resin and mixed using mechanical steerer (Fisatom model 713D) at 340–360 r/min for 10 min. Then, the initiator BUTANOX M-50 (methyl ethyl ketone peroxide, 33% in dimethyl phthalate) was incorporated and mixed again for 10 min prior to molding.

The waste, when used, was incorporated by randomly distributing it between the fiber layers. The detailed methodology can be found in Silva et al.[13]

Table 1. The formulation of each composite.

Composite	Designation/Volume fraction (%)
P/$CaCO_3$	P/$CaCO_3$ (50:50)
P/W	P/W (50:50)
P/GF	P/GF (50:50)
P/GF/$CaCO_3$	P/GF/$CaCO_3$ (50:15:35)
	P/GF/$CaCO_3$ (50:25:25)
	P/GF/$CaCO_3$ (50:35:15)
P/GF/W	P/GF/W (50:15:35)
	P/GF/W (50:25:25)
	P/GF/W (50:35:15)

Surface properties of the composites

Composite surfaces were characterized as molded, without any surface treatment. Average surface roughness (*R*a) measurements were carried out using a Taylor Robson Precision, Surtronic 25 model with a cut-off length of 0.08 mm. A goniometer Ramé-Hart Instrument Co. was used to determine surface energy of the samples based on the contact angle obtained using the harmonic method, which is suitable to small energy materials, such as polymers.[14]

In addition, Rockwell M hardness surface measurements were performed using a steel sphere (diameter: 1/4″) in a Pantec equipment. Seven indentations were made at several locations for each specimen and an average value is reported. The indentation load was 100 kgf with a preload of 10 kgf.

Test apparatus and experimental procedure

Tribological properties were evaluated using a pin-on-disc tribometer Nanovea (Micro Photonics Inc.). The configuration consisted of a stainless steel (SS302) sphere with 3.2 mm in diameter sliding against composite discs in dry condition. Tests were performed at room temperature with a disc rotation velocity of 100 r/min, radius of the wear track of 2.0 mm, three normal loads (1.0, 5.0, and 10.0 N), and for 5000 revolutions. Friction coefficient was provided by the tribometer and wear rate of the disc was determined with profilometry measurements in the wear track region. Seven profiles were taken for each wear track and averaged values are reported.

The wear volume loss, V_m (in mm^3), of the discs was calculated according to ASTM G99-05, as shown in equation (1)

$$V_m = 2\pi R A^2 \tag{1}$$

where R is the radius of the wear track (mm), in this case 2 mm, and A is the wear area width (mm^2). The specific wear rate, K (in mm^3/N m), was calculated as per equation (2)

$$K = \frac{V_m}{wx} \tag{2}$$

where X is the sliding distance (m) and W is the load (N).

Three repetitions were conducted for each set of frictional pairs and the average of the results is reported.

Results and discussion

Roughness

Figure 1 shows the roughness values R_a of the composites. Comparing the two-component materials, the P/CaCO$_3$ (50:50) composite has the highest roughness ($R_a = 2.4\,\mu$m), followed by P/W (50:50) and P/GF (50:50). This can be justified considering that the P/CaCO$_3$ composites were developed from the mechanical mixing of CaCO$_3$ powder with the resin, thus CaCO$_3$ particles are present on the surface, significantly increasing roughness, whereas the others are arranged in layers.

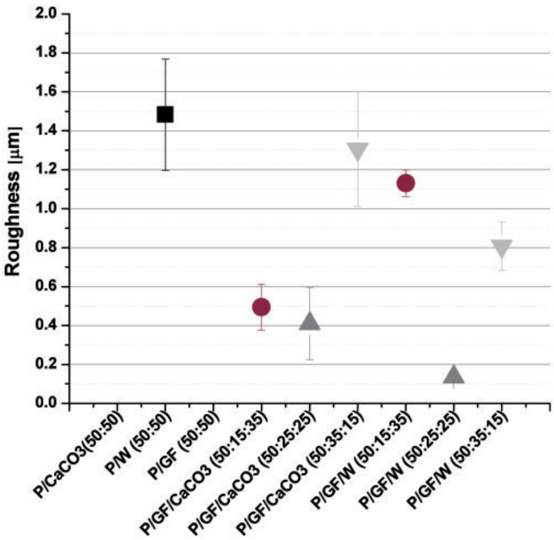

Figure 1. Average roughness (Ra) of the studied composites.

In the three-component group, P/GF/W (50:25:25) has the best surface finish that is the lowest roughness ($R_a = 0.11\,\mu$m). A considerable improvement of surface finish of the composites was observed with an increase in the amount of CaCO$_3$, from 1.3 μm (15% CaCO$_3$) to 0.25 μm (50% CaCO$_3$). These results agree with those of Tonela et al.[10]

Surface energy

Figure 2 presents the surface energy values of the composites before testing. The P/W (50:50) composite has the highest surface energy of the two-phase systems, along with the lowest roughness (previously shown). The presence of waste in the three-component composites yields higher surface energies when compared with those containing CaCO$_3$. Thus, the P/GF/W (50:25:25) composite, which has the lowest R_a as discussed earlier, has the second highest surface energy (48.5 mN/m) which is the highest of the three-phase systems. Small amount of CaCO$_3$ results in low surface energy, as seen when comparing P/GF (50:50), with P/GF/CaCO$_3$ (50:35:15). Surfaces with low roughness and high energy usually exhibit high adhesion and therefore high friction values.[15,16]

Previous studies reported that low roughness leads to high surface energy. However, when the surface energy of polymeric composites is measured by contact angle, the composite surface is not comprised of a polymeric resin only, and the nature of the filler plays a nonnegligible role on the surface characteristics.[17]

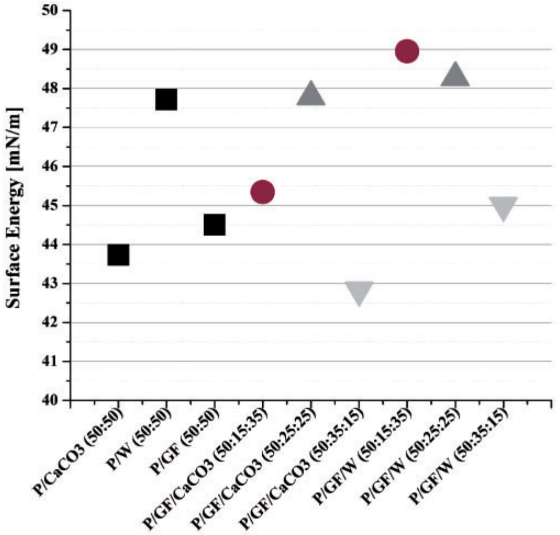

Figure 2. Surface energy of the studied composites.

Hardness analysis

Figure 3 shows the hardness values of the composites. It can be seen that addition of GF causes significant increase in hardness for the composites with two components. The same was found when comparing the $CaCO_3$ family (P/$CaCO_3$ (50:50), P/GF/$CaCO_3$ (50:15:35), P/GF/$CaCO_3$ (50:25:25), and P/GF/$CaCO_3$ (50:35:15)), which showed higher hardness for higher glass content in the composite. As for the composites with waste, not much difference is found in the three-component systems, since the waste is mostly encapsulated by the glass layers.

Coefficient of friction

Two component systems. Figure 4 presents the mean coefficient of friction values of two-component composites for variable normal load, which varied within the 0.25–0.35 range. The composite with waste presented the highest values, within 0.30–0.35 μm, which is consistent with the highest roughness value (Ra = 1.5 μm) previously shown (Figure 1). It is noticeable in Figure 4 a slight increase in coefficient of friction with the increase in normal load.

In Figure 5, the friction values are shown as a function of roughness for two speeds. Considering the results on the left-hand side for 100 r/min speed, at low roughness values (up to 1.0 μm), friction decreases

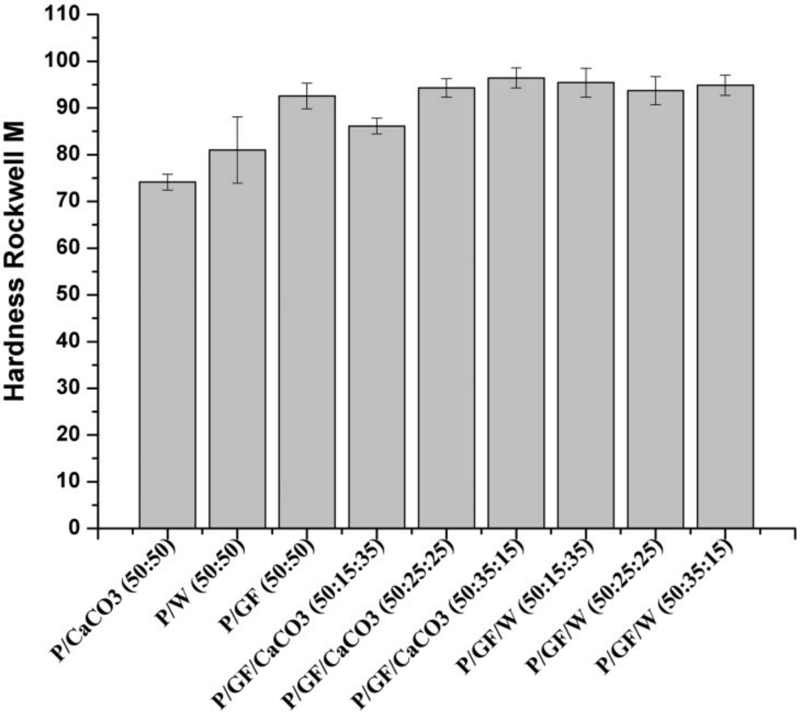

Figure 3. Rockwell M hardness of the studied composites.

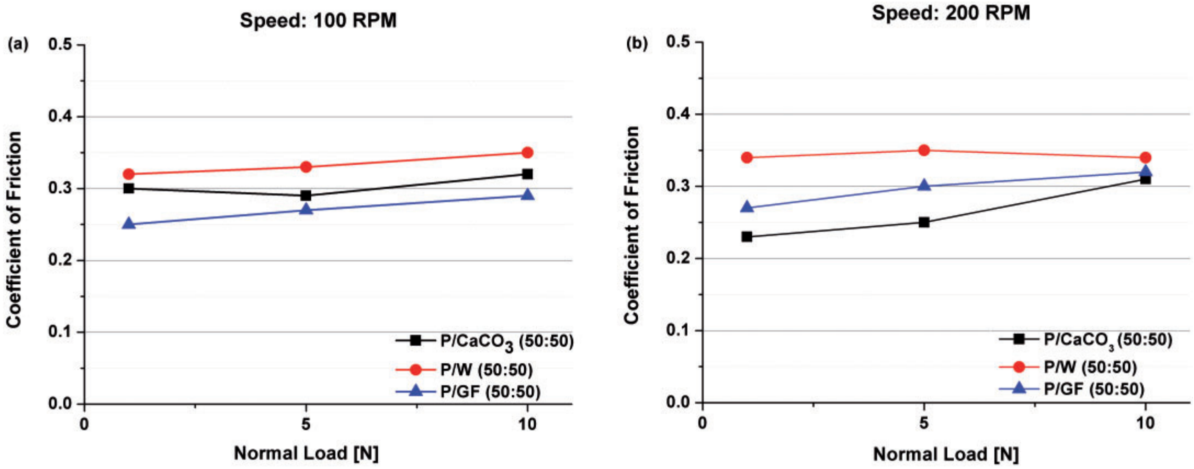

Figure 4. Variation of coefficient of friction with normal load for the two-component systems: (a) speed 100 r/min, and (b) speed 200 r/min.

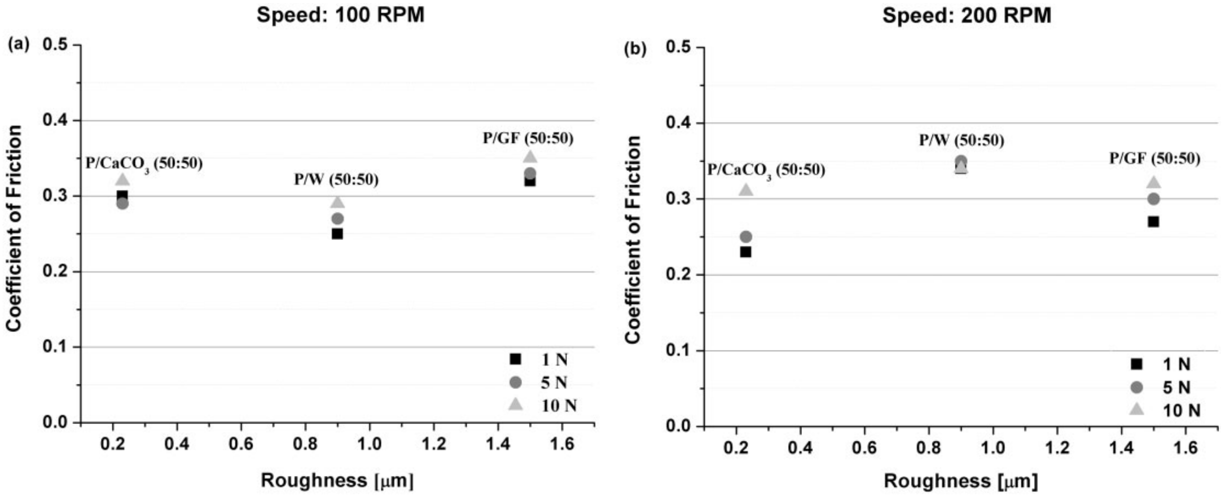

Figure 5. Variation of coefficient of friction with roughness for: (a) speed 100 r/min, and (b) speed 200 r/min.

with increasing surface roughness of the steel counterface. This can be explained based on the *bump model*.[18,19]

Asperities which impact on roughness cause a decrease in the contact surface area and thus yielding less adhesion between the interacting surfaces. Consequently, friction decreases and so does the amount of heat generated during testing. However, a further increase in roughness results in the appearance and later in the prevalence of abrasion as the main mechanism of friction. At this stage, asperities undergo degradation. When abrasion begins to dominate, there is an increase in friction. This explanation is consistent with the literature.[16,20] The overall outcome is a minimum in the dynamic friction versus roughness curves for all loads.

Coefficient of friction of polymer–steel contacts decreases with increasing surface roughness of the steel counterface until a critical value is attained, and then friction begins to increase. For low surface roughness, adhesion forces dominate, whereas abrasion dominates at higher roughness. Between them, there is a roughness range that these components (adhesive and abrasive) overlap, resulting in low coefficient of friction.[16,20]

It was found in this work an intermediate roughness range (0.8–1.0 µm) where the friction coefficient was minimum (Figure 5). Also, a correlation appears between coefficient of friction and surface roughness of the composite (not the steel counterface, as usually mentioned in the literature). The P/W (50:50) composite, with higher roughness, presented higher coefficient of friction due to the deformation component of friction, which caused the rupture of asperities, fragmentation of the matrix, and of the GF.

P/GF/CaCO$_3$ composites. Figures 6 and 7 present the coefficient of friction for *P/GF/CaCO$_3$* composites. The presence of GF yielded higher coefficient of friction, due to its high stiffness, ratifying the hardness results. The P/GF/CaCO$_3$ (50:15:35) composite presented higher coefficient of friction (ca. 0.40) with 1 and 5 N normal load, whereas for a normal load of 10 N, the three composites yielded similar results, probably because, in this case, just the deformation component exerted influence on the coefficient of friction. Nirmal et al.[21] found a coefficient of friction of about 0.7 for polyester/betel nut fiber when sliding it against stainless steel in a dry contact (normal load: 5 and 10 N).

Surface energy was also found to influence the coefficient of friction. Surfaces with lower roughness tend to have higher adhesion, hence, higher coefficient of friction, as reported in the literature.[15,16]

P/GF/waste composites. The coefficient of friction of the composites with waste as filler is shown in Figure 8. The composite P/GF/W (50:25:25) displayed higher coefficient of friction (between 0.30 and 0.37) in all test conditions, which can be explained by its higher surface energy (48.5 mN/m) and lower roughness (0.11 μm), which intensifies the adhesive component. This also indicates a correlation between surface parameters and coefficient of friction.

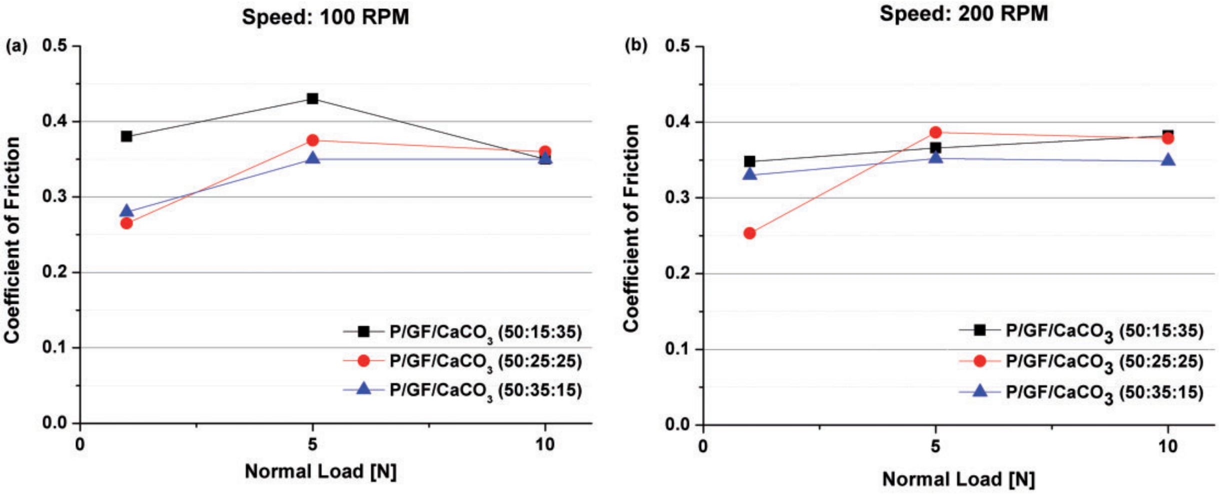

Figure 6. Coefficient of friction versus normal load for composites with CaCO$_3$ for: (a) speed 100 r/min, and (b) speed 200 r/min.

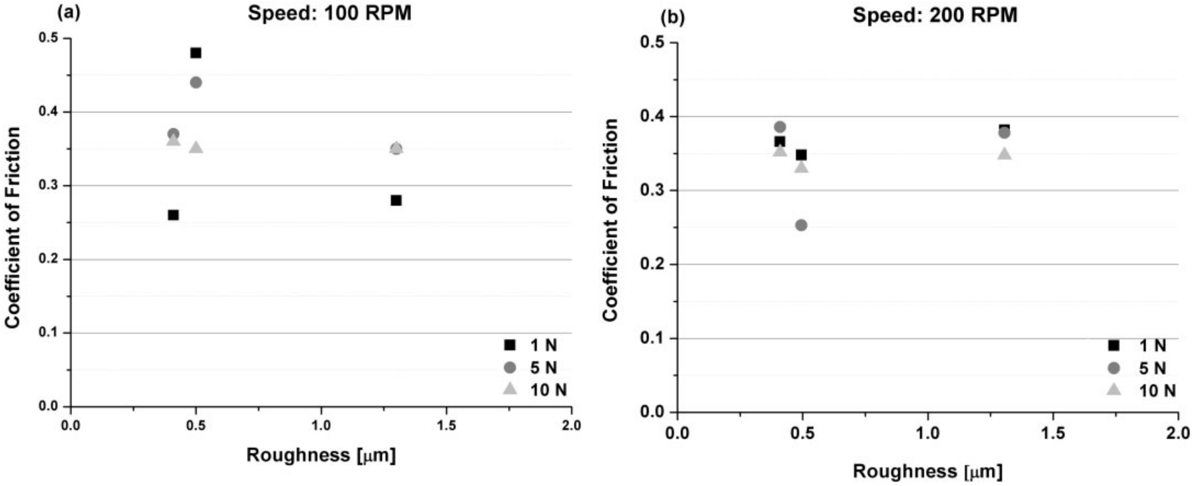

Figure 7. Coefficient of friction versus roughness for composites with CaCO$_3$ for: (a) speed 100 r/min, and (b) speed 200 r/min.

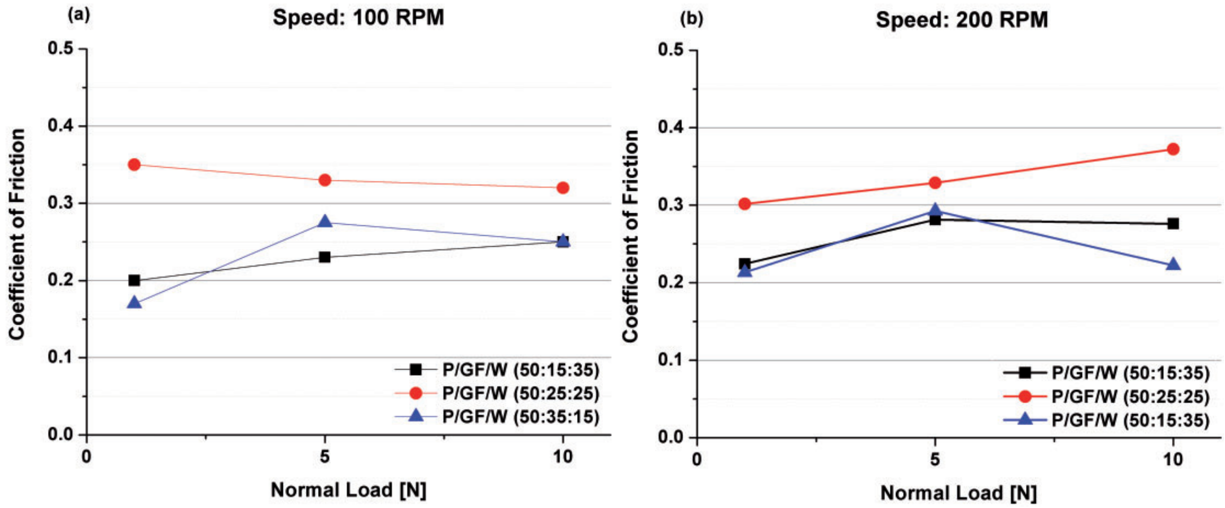

Figure 8. Coefficient of friction versus normal load for composites with waste as filler for: (a) speed 100 r/min, and (b) speed 200 r/min.

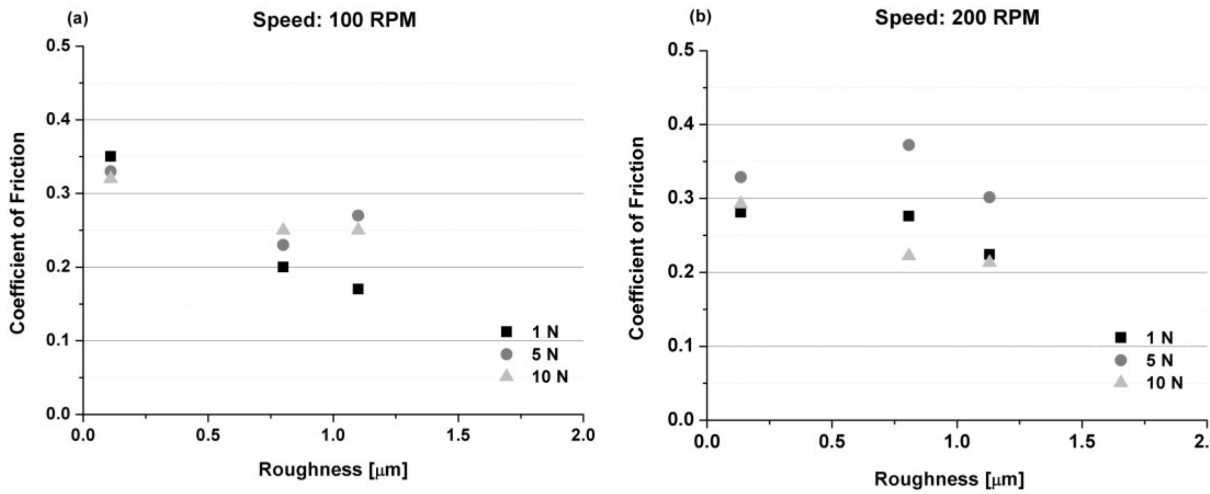

Figure 9. Coefficient of friction versus roughness for composites with waste as filler for: (a) speed 100 r/min, and (b) speed 200 r/min.

In addition, Figure 9 shows that, in general, 0.8–1.0 μm roughness produces lower coefficient of friction values. Empirical studies have shown that even a small change in roughness of the counterface may significantly affect the coefficient of friction.[22] Zsidai et al. studying the tribological behavior of different engineering plastics concluded that all of them exhibited lower coefficient of friction using rougher surfaces (Ra = 0.10–0.30 μm) than smoother ones (Ra = 0.05–0.10 μm).[23] They also observed that, for some plastics, the roughness influence was larger for low contact pressures whereas, in the present work, the coefficient of friction did not vary significantly when the normal load was increased. Coefficient of friction of P/GF/waste composites was smaller than those for P/GF/$CaCO_3$, which may be a positive factor considering the application.

Wear rate

Wear rate of P/$CaCO_3$ (50:50), P/W (50:50), and P/GF (50:50) composites is presented in Figure 10 for 1, 5, and 10 N normal loads. The trend observed for 1 N was different from that for 5 and 10 N. For 1 N load, the steel sphere just slides on the surface of the composite and therefore the material on the surface influences the wear rate. The P/$CaCO_3$ (50:50) composite was prepared by mixing polyester and calcium carbonate using a mechanical steerer, which resulted in a surface more resistant to sliding wear than P/W (50:50) and

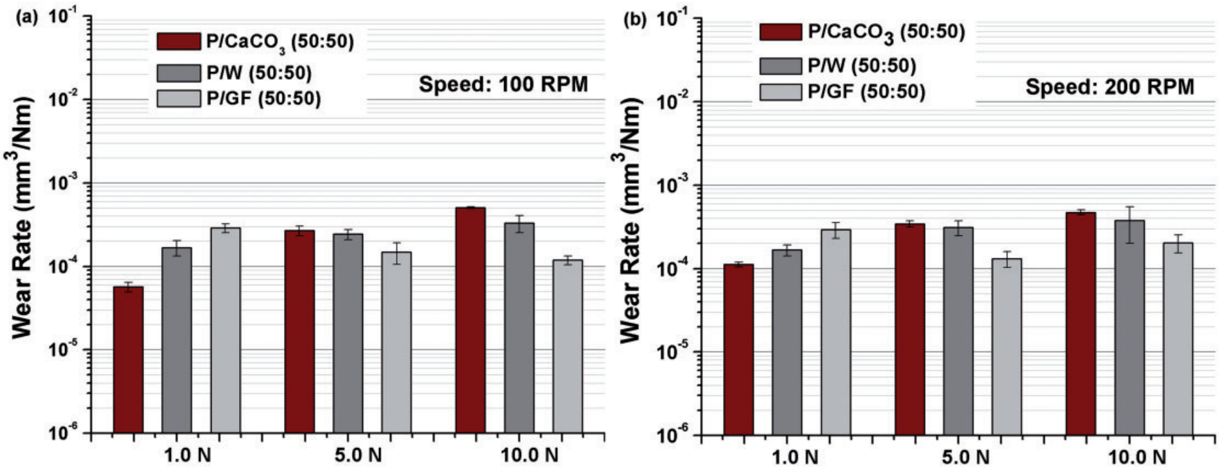

Figure 10. Wear rate as a function of load for the two-component composites P/W (50:50) and P/$CaCO_3$ (50:50) for: (a) speed 100 r/min, and (b) speed 200 r/min.

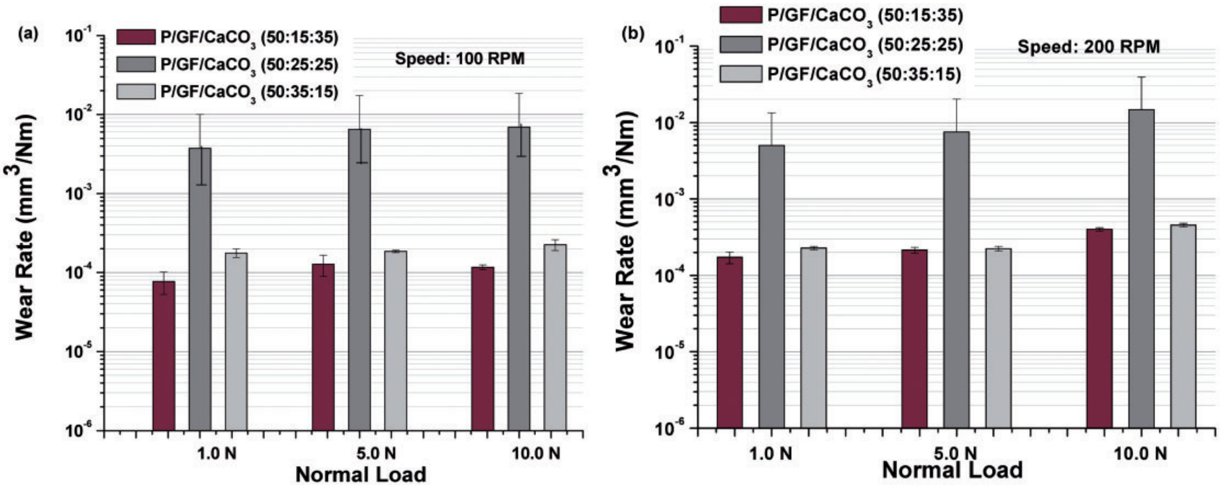

Figure 11. Wear rate as a function of load for the P/GF/$CaCO_3$ composites for: (a) speed 100 r/min, and (b) speed 200 r/min.

P/GF (50:50) composites, both with a polyester-richer layer at the surface which is less resistant to wear.

When the normal load increased to 5 and 10 N, the P/$CaCO_3$ (50:50) composite showed higher wear rate, followed by P/W (50:50) and P/GF (50:50), respectively. In this case, load was supported by material matrix and stiffness of the composite was determinant to wear resistance, being in accordance with the Rockwell M hardness results presented earlier in a way that higher hardness yielded lower wear rate. For the P/$CaCO_3$ (50:50) and P/W (50:50) composites, it can be seen that wear rate increased for higher normal load.

Figure 11 presents the wear rate of the three-component composites with $CaCO_3$ as filler. The P/GF/$CaCO_3$ (50:15:35) composite presented the lowest wear rate and the highest coefficient of friction (Figure 7). Besides, the P/GF/$CaCO_3$ (50:25:25) composite showed a much higher wear rate (between 3 and $7 \times 10^{-3}\,mm^3/N\,m$). In these cases, it is seen that the increase of the applied force and velocity practically did not change the values of wear rate. In general, the wear rate values of composite tri-component with $CaCO_3$ were superior to composites bi-component. According to Kukureka, the composite reinforced with GF increases the wear rate and decreases the coefficient of friction, and this only occurs due to the wear of composite, releasing temperature and debris.[24]

The wear rate results for composites with waste as filler are presented in Figure 12. An increase in the amount of GF resulted in a decrease in wear resistance for all three normal loads tested. It can also be observed that composites with waste presented a lower wear rate, again indicating its potential as a substitute for $CaCO_3$.

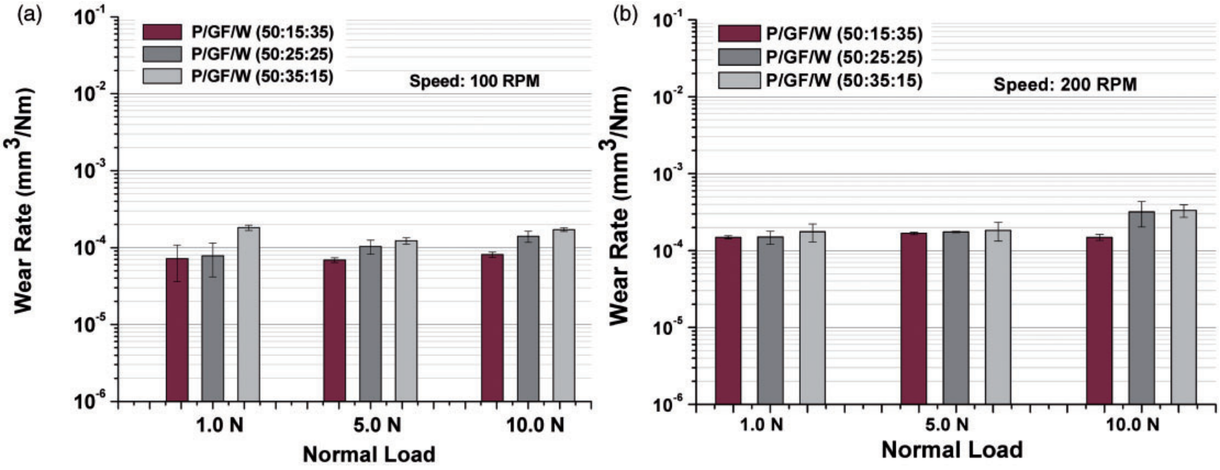

Figure 12. Wear rate as a function of load and their speed test for the P/GF/W composites for (a) speed 100 r/min and (b) speed 200 r/min.

Conclusions

Tribological behavior of P/GF composites using P/GF waste as filler has been successfully studied and their performance was compared to that of P/GF composites with calcium carbonate as filler. The incorporation of P/GF waste proved to be more beneficial than calcium carbonate with respect to both friction and wear performance, justifying its use for tribological applications. Regarding formulation, P/GF/W composites with higher content of waste presented higher wear resistance.

The present investigation has shown quite encouraging results and opened an extra possibility for the recycling of P/GF composites. Among the composites with two components only, the P/GF composites presented better tribological results for 5 and 10 N normal load. A correspondence between coefficient of friction and surface roughness for these composites was proposed, which differ from that usually found in the literature, which reports a correlation between roughness of the metallic counterface and coefficient of friction. A minimum in coefficient of friction was found when roughness of the composites was between 0.8 and 1.0 µm.

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Conflict of interest

None declared.

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