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Composites of polyester + glass fiber residues vs. composites with mineral fillers

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Polymer composites, such as those composed of a polyester, glass fibers (GFs), and mineral fillers (e.g. $CaCO_3$), pose a threat to the environment because of the growing amount of residues and due to difficulties in their recycling. Therefore, we have studied effects of incorporation of (polyester + GFs) waste material as a filler into virgin composites. Two types of polyester + glass fiber composites were developed using hot compression molding, one of them with recycled (polyester + glass fiber) material obtained via knife or ball milling; the other, a control group, contained $CaCO_3$, a traditional filler in this field. Dynamic friction and wear rate were determined using a pin-on-disk tribometer and a stylus profilometer, respectively. As expected, the presence of the residues significantly decreases dynamic friction and wear rate when compared to $CaCO_3$, since the main constituent of the residues is a polymeric material. Thus, polyester + glass fiber composite residues are a candidate for a partial substitution of $CaCO_3$. This should lower the environmental contamination caused by discarding the residues as well as provide composites with lower wear rates.

Keywords: polyester resin; glass fibers; calcium carbonate; dynamic friction; wear; recycling

1. Introduction

With increasing world production and consumption of manufactured goods, recycling of materials has become one of the most important environmental control activities – resulting also in lower material costs.[1]

The ever-growing use of fiber + polymer composites [2] has become an environmental concern since their residues (mainly glass fibers [GFs]) cannot be readily recycled. Some methods have been developed aiming at reduction of the amount of residues: incineration, chemical degradation, and/or mechanical grinding.[3]

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Among the materials in landfills, plastics have emerged as 'the villain' – especially so because they have long shelf lives and constitute a large variety of disposable materials. Many polymers are mutually immiscible [4] – hindering the process of separation and, consequently, hindering recycling. Determination of the dependence of glass transition temperature, T_g, on composition allows to determine miscibility, compatibility (partial miscibility), or full immiscibility.[5,6] Another option in miscibility determination is evaluation of structures by scanning electron microscopy and/or transmission electron microscopy.[7–11]

Creation of composites, containing fibers or otherwise, provides often improved properties.[2,12–16] Composites in engineering and production of consumer goods offer unique combinations of properties in addition to numerous economic advantages compared to other competing materials.[2–4] In general, properties of multiphase composites depend strongly on the strength of interfacial interactions.[17] We also need to note that some processes of manufacturing composites generate high amounts of waste or leftovers harming the environment. Recycling polymer blends that do not contain fibers or other fillers is significantly easier.[18]

The European Union is estimated to generate annually one million tons of thermosetting materials.[19] There are several alternatives to target this waste: energy production, pyrolysis to obtain fuel,[19] milling and embedding in asphalt,[20] and usage in mixtures of thermoplastic polymers for various applications.[21–23] Some countries like France, Germany, Italy, and the Netherlands have pilot plants for recycling fiber-reinforced plastics required by government regulations.[20]

There are several reports on reduction of the volume of waste that would otherwise end up in landfills. Thus, Risson and coworkers [12] reported that the incorporation of waste-laminated polyester resin with GFs used as reinforcement in the polymer matrix improved the tensile strength by 23%. Figueiredo [24] used waste products produced with unsaturated polyester resin incorporated with mineral fillers and GFs in new formulations of a bulk molding compound and a sheet molding compound; the quality of the parts was less than that of the parts produced with virgin products.

Composites exhibit a range of applications in industry and, in general, can reduce costs and provide improved properties.[25] In this work, tribological performance of polymer matrix plus polyester/GFs residues and calcium carbonate ($CaCO_3$) as a filler was studied in order to identify the possibility of partial substitution of $CaCO_3$ by residues of polyester/GFs. $CaCO_3$ has been used as a filler before, for instance for popypropylene + high density polyethylene blends.[26] Dynamic friction and wear rate were determined using a pin-on-disk tribometer [27–31] and a stylus profilometer, respectively, in order to evaluate the effects of fillers.

2. Experimental

2.1. Materials

Polyester/GFs residues: The material used as a residue was obtained from polyester composites (density 1.09 g/cm^3) with 12 wt.% GFs which had been molded by a variant of the resin transfer molding (RTM) process called light RTM.

In this process, a liquid resin, precatalyzed for curing later, is injected into a closed mold with the help of a vacuum pump, impregnating dry fibers. Once the fibers are well impregnated and the mold filled, curing is performed.

To mold new composites, the following materials were used: (i) A medium viscosity (90–120 cPoise) polyester resin (UCEFLEX UC 5518 from Elekeiroz); (ii) GF mats with an aerial density of 300 g/m^2; (iii) $CaCO_3$ with an average density of 2.82 ± 0.01 g/cm^3; and (iv) Butanox M-50 (methyl-ethyl-ketone peroxide, MEKP, 33% dimethyl phthalate), 1.5% v/v, as the initiator.

2.2. Methodologies used

The polyester with GF composite wastes were ground in a knife mill with a 8 × 8 mm screen, then in a ball mill (for 1 min), reaching a particle size 9–16 mesh. This material was incorporated into virgin polyester + glass fiber composites by distributing them in the center region (in-between glass mat layers).

The composites were molded by hot compression (using six tons distributed on a 270 × 170 mm metallic mold at the temperature of 90 °C) and the following formulations were used: two control groups (polyester + GFs and polyester + $CaCO_3$, 50 wt.% polyester in both cases), binary (polyester + residues), and two ternary families of composites: polyester + GFs + $CaCO_3$ (50/35/15, 50/25/25 and 50/15/35, weight basis) and polyester + GFs + residues (50/35/15, 50/25/25 and 50/15/35, weight basis).

2.4. Friction determination

Nanovea pin-on-disk tribometer from Micro Photonics, Inc., was used for determining dynamic friction. An SS 302 grade stainless steel ball with the diameter 3.20 mm was used as the pin. The pin was loaded onto the test sample with a known weight of 1.0 and 10.0 N. The highly stiff elastic arm insures a nearly fixed contact point and, thus, a stable position in the friction track. Dynamic friction is determined during the test by measuring the deflection of the elastic arm by direct measurement of the change in torque.[27,31] The rotation speed of the disk was 200.0 rpm and the radius of wear track was 2.0 mm. The tests were performed for 5000 revolutions under room temperature conditions. The results reported are averages each from three runs.

2.5. Wear determination

As said, 5000 revolutions were run in a pin-on-disk machine. The areas of the cross section of wear track after each tribological test were determined with a Veeco Dektak 150 profilometer. The profilometer amplifies and records the vertical motions of a stylus displaced at a constant speed by the surface to be measured. As the stylus moves, the stylus rides over the sample surface detecting surface deviations.[13] A stylus with tip radius of 12.5 μm was used. The load applied to the sample was 1.0 mg and the scan rate was 26.7 μm/s. The scan length was 800 μm and the measurement range was 65.5 μm.

Seven values of wear track width were measured at different locations on each sample and averaged to achieve accuracy. All samples were cleaned by high pressure air to remove all debris before each test.

The volume loss due to wear, V_m, was then calculated using the following formula according to the ASTM G99-05 standard:

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

where V_m is the volume loss in mm^3, R is the wear track radius in mm (2.0 mm in this case), and A is the wear area width in mm^2.

Wear rate k_{wear} was then calculated using:

$$k_{\text{wear}} = V_m / WX \tag{2}$$

where k_{wear} is the wear rate in mm^3/Nm, V_m is the volume loss due to wear in mm^3, W is the load in N, and X is the sliding distance in m.

3. Dynamic friction results

3.1. Binary systems

For brevity, we present dynamic friction results as block diagrams (an example of a pin-on-disk output is provided below).

We see in Figures 1 and 2 that at both loads the polyester + residues (50/50) have the highest dynamic friction, followed by GFs and then by $CaCO_3$ containing materials.

3.2. Ternary systems

We present the diagram of friction averages in Figures 3 and 4 for 1.0 and 10.0 N, respectively, for composites without residues.

We find that under both loads, the polyester + $CaCO_3$ material has the lowest friction. Replacing one half of the carbonate by GFs (the second block from the left) has a very small effect on friction at 1.0 N but causes a significant increase of friction at 10.0 N. Other compositions have higher friction. Thus, if low loads are seen in service, partial replacement of $CaCO_3$ by GFs is worthwhile.

We now report results of introduction of polyester + glass fiber residues on friction. We begin by showing in Figure 5 the block diagram for 1.0 N load.

The GFs have been used for polymer reinforcement in various combinations.[32–36] We have seen in earlier figures that largely, the inclusion of GFs does not lower friction. However, Figure 5 tells us that the presence of glass fiber residues *in combination with our polyester and* GFs changes the situation. Fifteen percent of residues together with 35% GFs results in the lowest value of friction. Replacing even more GFs (15% GFs + 35% residue, second block from the right) causes a relatively small increase of friction, while 35% residue makes this composition quite interesting from the point of view of low material costs and less waste going into the environment. A residue is a combination of polyester + GFs; since the recycled material contains a polyester, it is evidently well miscible with virgin polyester.

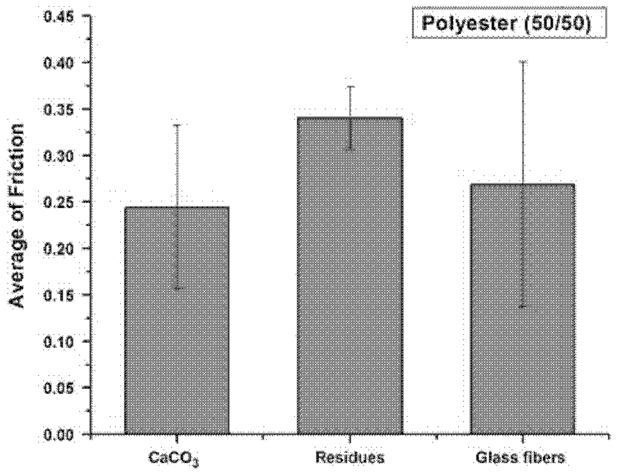

Figure 1. Averages of dynamic friction for binary composites for 1.0 N and 200 rpm.

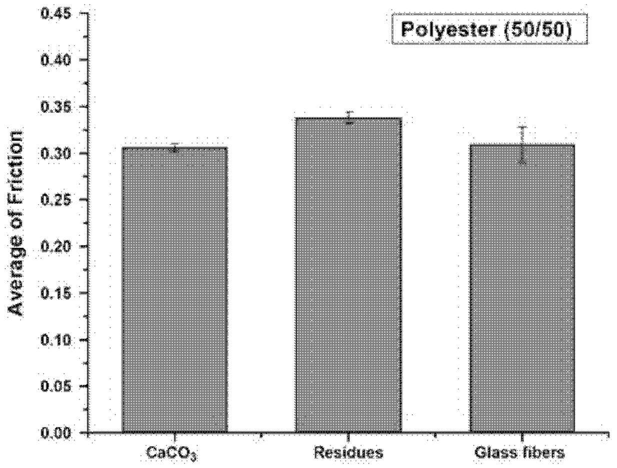

Figure 2. Averages of dynamic friction for binary composites for 10.0 N and 200 rpm.

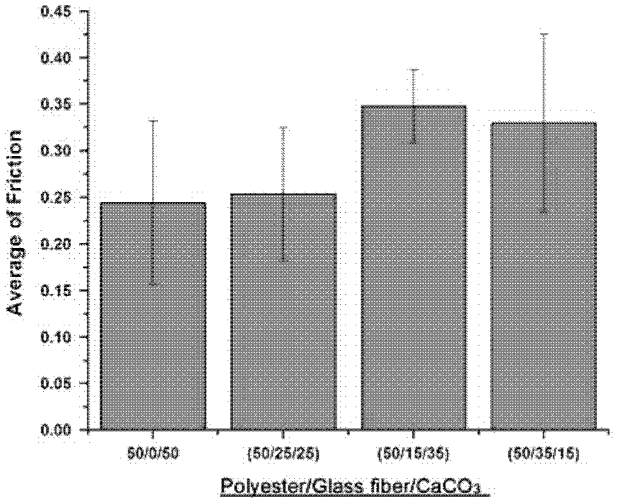

Figure 3. Averages of dynamic friction for ternary compositesfor 1.0 N and 200 rpm.

We now consider results for the same ternary composites but under the load of 10.0 N. We show dynamic friction values as a function of the number of revolutions in Figure 6 and the block diagrams of friction averages in Figure 7.

The difference between Figures 5 and 7 is that under the low load of 1.0 N, the composite that does not contain 'virgin' GFs has the highest friction while under 10 N, the same material exhibits the second highest friction. However, under both loads, the composite containing 35% GFs + 15% residue has the lowest friction. The composite with 15% GFs and 35% residue has the second lowest friction. Thus, even at high loads, there is the opportunity to include the residue that would otherwise be discarded.

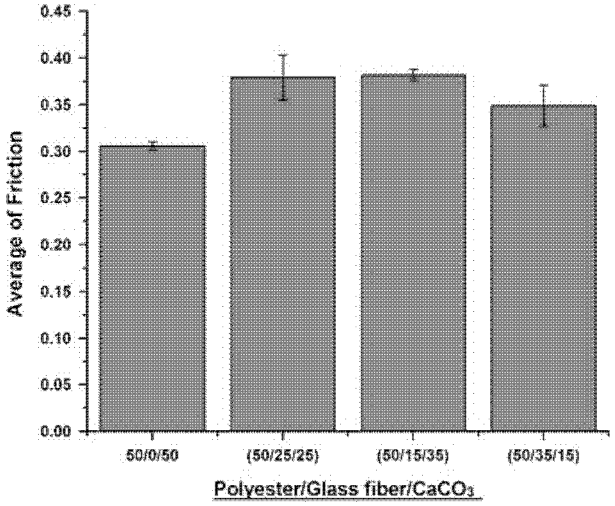

Figure 4. Averages of dynamic friction for ternary composites for 10.0 N and 200 rpm.

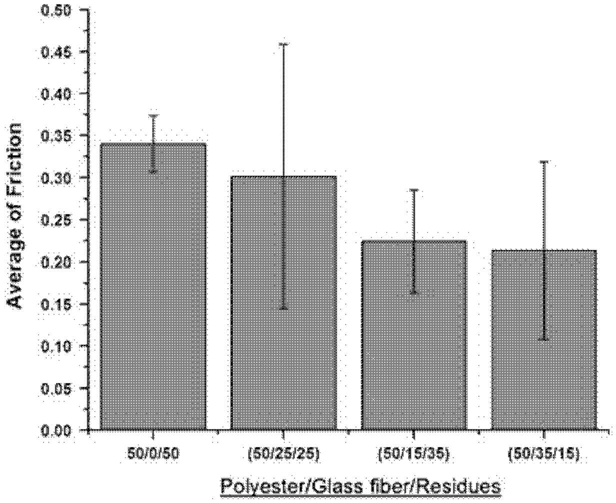

Figure 5. Averages of dynamic friction for ternary composites with residues for 1.0 N and 200 rpm.

4. Wear rates

Here, also we begin with binary systems.

We see in Figure 8 that $CaCO_3$ provides the highest protection against wear; residues are somewhat worse, while GFs offer the least resistance to wear. Apparently, at the low load, $CaCO_3$ acts as a lubricant of sorts – mitigating wear. Our results confirm once more that wear cannot be estimated on the basis of friction values. Since the results in Figure 8 pertain to the load of 1.0 N, we present respective results for 10.0 N in Figure 9.