

Use of Polyester/Glass-Fiber Residues as Fillers for Composites

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ABSTRACT: In this work, the performance of polyester (P)/glass fiber mats (G) and P/G/calcium carbonate (CaCO₃) composites was compared with that of P/G/fiberglass waste composites. The residues used were conventional P/G postconsumer light resin-transfer-molding parts, obtained via knife or ball milling. Composites with up to 50 wt % reinforcement were prepared by hot compression molding and characterized via physical (density and water sorption), thermal (thermogravimetry and burn-out), and mechanical (impact, Barcol hardness, and tensile) testing. The results show that the simple grinding and reincorporation of the composite residues yielded new composites with generally worse characteristics than the ones with calcium carbonate. Then, the waste was sorted

by removing most of the pure resin particles from it. This yielded a resin-rich fraction, which could be better used for energy recovery and resin-covered fibers. The use of the latter as a filler yielded composites with better overall properties than those with calcium carbonate for a controlled amount of W; thus showing potential use as a replacement for the commonly used inorganic filler, maintaining the mechanical properties, decreasing the raw material cost, and reducing the amount of composite waste discarded in the environment. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 302–310, 2012

Key words: composites; recycling; waste

INTRODUCTION

Polymer composites are used worldwide in a variety of sectors,¹ and as a consequence, there is a large amount of composite waste currently discarded into the environment. Indeed, according to the Brazilian composite association (Abmaco), the residue generation alone reached 13,000 tons in 2007 and grew to 18,000 tons in 2009.

Composites with thermoset matrices are usually considered to be materials of difficult recyclability,² and among those, polyester composites have attracted the greatest attention because they are, by far, the composites most discarded in landfills nowadays.³ Various polymer recycling methods are also being applied to thermoset composites, such as recycling by depolymerization (monomerization) and thermal decomposition with energy recovery. In the former, only the organic fraction of W is recovered, whereas, in the latter, only the inorganic constituents, such as glass fibers and fillers [e.g., CaCO₃], are recovered.^{4,5} Limitations also apply to chemical dissolution with solvents,⁶ for example,

acid solutions for glass fiber composites⁷ and nitric acid solution for carbon fiber composites,⁸ and pyrolysis,³ which involves high temperatures and the release of products as toxic gases, with associated large energy use and environmental impact.

The mechanical recycling of thermoset composites is perhaps the most exploited method.^{9–11} In this type of recycling, usually via grinding, all of the constituents of the original composite appear in the resulting recycle, which is a mixture of polymer, fiber, and fillers. The recovered material can be broadly classified in two fractions: one that contains most of the reinforcing agents (fibers) and the other that contains most of the matrix. Typically, the finely graded fractions are powders and contain a higher content of filler and polymer than the original composite. The coarser fractions tend to be of a fibrous nature, with a higher aspect ratio, which may be incorporated as reinforcement in new thermoset¹⁰ or thermoplastic⁹ composites.

Mechanical recycling does not require treatment of the recycled fibers by solvents or any heat-aggressive route; this prevents the loss of mechanical properties. Also, it is a relatively simple route for large-scale use, being generally considered to be more compatible with than daily industrial practices. Nevertheless, it requires careful attention regarding the separation and reincorporation procedures. The mechanical performance of the recycled composites is not necessarily much lower than that of the original part, and a reasonably high content of recycled

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fractions can be successfully incorporated. This technology reduces the use of virgin materials and the cost of raw materials for processing and may even lead to an economically viable closed-loop thermoset composite recycling operation.¹⁰

Sheet-molding and bulk-molding compounds, largely used in the automotive sector,¹² comprise Polyester resin, styrene monomer, and chopped glass, along with inorganic fillers, such as zinc stearate and calcium carbonate (CC), which are usually added at a significant ratio.¹ In Brazil, light resin-transfer-molding (RTM) parts, commonly employed in a variety of sectors, also use this filler for cost purposes. In this context, the aim of this work was to study the recycling of thermoset composite waste as a substitute for CC through a comparison of the characteristics of Polyester/G/W and Polyester/G/CC composites in an attempt to promote the use of this waste.

EXPERIMENTAL

Materials

The materials used during this study included:

- Polyester/Glass composite waste: This waste, called W, was obtained from polyester composites with around 12 wt % glass fiber, which were produced by light RTM.
- Orthophthalic polyester resin: UCEFLEX UC 5518 from Elekeiroz, Sao Paulo/Brazil (density = 1.09 g/cm³) was used as the composite matrix, being called P.
- Glass fibers: Various layers of glass mats were used as reinforcements (area density of the mat = 300 g/m², fiber density = 2.54 g/cm³), being called G.
- Calcium Carbonate (CC) was used as an inorganic filler. Laser granulometry (Cilas brand, model 1180, Madison, WI) showed a mean diameter of 18.22 μm, and gas pycnometry (multipycnometer model MVP-1-Quantachrome) yielded a density of 2.82 ± 0.01 g/cm³.
- Curing agent: BUTANOX M-50 (methyl ethyl ketone peroxide, 33% in dimethyl phthalate) was used (1.5% v/v of resin) as an initiator.

Composite molding

This study was divided into three parts: (1) evaluation of the grinding method and the particle size range of interest, (2) molding of P/G composites filled with either CC or composite W for comparison, and (3) process optimization to obtain a higher content of glass fibers by sorting of the W. These are detailed next.

P/G (12 wt % glass fiber) composites molded by light RTM were initially coarsely ground with a knife mill (Rone-SRB2305, Osasco/Brazil, with an

opening screen of 8 × 8 mm²). Two other equipments were selected for the fine grinding of the W: a second knife mill (Retsch-SM2000, with an opening screen of 2 × 2 mm²) and a ball mill (Servitech-CT-242) with 20 balls (21.1 mm in diameter) running for 1 min.

The W was classified with a mechanical shaker (Bertel, Pauliceia/Brazil) with the following sieves: 9, 16, 32, and 60 mesh. Two granulometric ranges were used: the W retained in the 16-mesh sieve (called 16 mesh) and that retained in the 60-mesh sieve (called 60 mesh). The glass and polyester content for each range of particle size were evaluated with burnout tests (4 h at 600°C) in a Sanchis furnace on the basis of ASTM D 2584. Fiber length was evaluated with the aid of a magnifying glass and a Carl Zeiss (Axio Scope model) optical microscope coupled with digital cameras.

The polyester resin was mixed with the initiator just before molding and degassed for 5 min in a Thornton T14 ultrasound, Sao Paulo/Brazil. The layers of glass mats were assembled in the mold in a way that, to each layer of resin, a layer of W was added. Composites with 50 wt % W were produced by hot compression molding (6 ton) for 60 min at 90°C in a Marconi MA 098/A 3030 hydraulic press. The resulting fiber orientation was in-plane random because the fibers from the W were very short, the mats were also random, and the chosen manufacturing process did not orient the fibers.

To define the most adequate grinding process and waste particle size, characterization of these preliminary composites was carried out with density, Barcol hardness, and Izod impact tests. Next, composites with ground W or CC were molded and compared. The molding procedure was the same as before, except that for the composites with CC, the resin was first mixed with CC with a Fisatom 713D, Sao Paulo/Brazil mechanical stirrer (340–360 rpm for 10 min) for homogeneity and then with the initiator. The composite was molded, in a way that the fiber mats were always placed at the top and bottom layers of the assembly.

In the third part of this study, the W was manually sorted [hereafter called sorted waste (SW)]. The obtained polyester-rich W fraction could be more adequately used for energy recovery, whereas the fiber-rich fraction is more suitable for incorporation as a filler and, therefore, was used to produce new composites following the same methodology detailed previously. The compositions of all of the produced composites are shown in Table I.

Composite characterization

The density and water sorption measurements were carried out to assess the void content of the composites and, thus, indirectly evaluate the quality of the new composites and their reliability if they were to

TABLE I
Formulations of the Bicomponent and Tricomponent Composites Studied

[P] (wt %)	[G] (wt %)	[CC] (wt %)	[W] (wt %)	[SW] (wt %)
50	50			
50	35	+	15	or 15
50	25	+	25	or 25
50	15	+	35	or 35
50		+	50	or 50

be used in parts exposed to the environment during their life cycle. The densities of all composites were measured with an ordinary glass pycnometer. Water sorption at room temperature was evaluated on the basis of ASTM D 5229; that is, the samples ($100 \times 25 \text{ mm}^2$) were cut from the composites, dried, weighed, and immersed in distilled water at room temperature. Periodically, the samples (three for each composition) were taken out of the immersion, and the excess water was quickly removed before weighing. The amount of absorbed water was calculated by comparing the original and the new weights.

Thermogravimetric analysis (TGA) and burnout tests were performed to estimate the weight fraction of the constituents of the samples. Thermal analysis was carried out in a TGA instrument (TA Instruments-TGA 2050 model, New Castle/DE) at a $20^\circ\text{C}/\text{min}$ heating rate from 20 to 875°C with synthetic air. Burnout tests were conducted as mentioned before.

Mechanical testing was carried out to compare the performance of the various composites. These included Barcol hardness (Bareiss BS 61 II), according to ASTM D 2583, Izod impact testing on unnotched specimens (Ceast Impactor II instrument, Pianizzia, Italy), according to ASTM D 256, and tensile testing with an EMIC (LD-2000) instrument (20-kN load cell), Sao Jose dos Pinhais/Brazil, with a crosshead speed of 5 mm/min on the basis of ASTM D 3039 (all tests were conducted at room temperature). Dynamic mechanical analysis (DMA) was performed in a TA thermoanalyzer 2980 with single-cantilever mode at a $2^\circ\text{C}/\text{min}$ heating rate from room temperature to 150°C and at a frequency of 1 Hz.

Optical microscopy of the polished cross section of the composites was carried out in a Carl Zeiss optical microscope. The cross section of the Izod fractured composites was analyzed by scanning electron microscopy (SEM) in a JEOL (6060 model) instrument, Tokyo/Japan.

RESULTS AND DISCUSSION

Preliminary study

Table II shows the results of the granulometric study carried out on the W sample, along with the fiber and resin content found in each sample. It can be

TABLE II
Burnout Results for All of the Particle Size Ranges Studied

Sample	Mesh	Fraction (%)	Resin (%)	Glass fiber (%)
W	9	10.7	83.7	16.3
	16	10.7	90.4	9.6
	32	22.0	92.7	7.3
	60	28.4	95.0	5.0
	>60	28.2	83.6	16.4
SW	Fiber-rich fraction	16.2	63.5	36.5
	Resin-rich fraction	83.7	97.2	2.8

seen that the ball mill preferably yielded finer particles. Also, the glass fraction was mostly found either as large fibers or as a fine powder. The manual sorting of W produced a fiber-rich fraction with a 13 times larger fiber content than the resin-rich fraction (fiber contents of 36.5 and 2.8%, respectively).

The results of the preliminary study are shown in Table III. The 16-mesh particle size showed twice the impact strength of the composites produced with the 60-mesh W fraction which was mainly associated with the variation in the fiber aspect ratio between them. Indeed, the measured mean fiber lengths were, respectively, $4.0 (\pm 0.6) \text{ mm}$ and $0.24 (\pm 0.05) \text{ mm}$. The longer fibers were more effective in deviating cracks in an impact event, requiring more energy in the process.¹³ The contrary was found when particles were incorporated in polymers; that is, the finer particles usually yielded the best results because the particles and fibers presented distinct reinforcement mechanisms.

Regarding the grinding process, the differences in hardness and impact strength were small, but a greater uniformity in the composite with ball-milled W was noticed. In addition, the ball mill allowed greater flexibility than the knife mill, with greater control of the grinding process by variation of the diameter, the number of balls, and the grinding time. These results led to the selection of 16-mesh particle size and the ball mill for the remaining experimental work.

Bicomponent and tricomponent composites

The results of theoretical and experimental density for all formulations are shown in Table IV. It can be

TABLE III
Test Results for the Preliminary Composites

	Mesh	Density (g/cm^3)	Barcol hardness	Impact strength (kJ/m^2)
Ball mill	16	$1.185 (\pm 0.002)$	$42 (\pm 2)$	$45.1 (\pm 9.8)$
	60	$1.196 (\pm 0.004)$	$42 (\pm 2)$	$15.8 (\pm 4.8)$
Knife mill	16	$1.185 (\pm 0.002)$	$44 (\pm 2)$	$42.2 (\pm 18.4)$
	60	$1.192 (\pm 0.013)$	$45 (\pm 2)$	$24.0 (\pm 7.7)$

TABLE IV
Density and Hardness Results for the Bicomponent and Tricomponent Composites

Formulation (wt %)	Density (g/cm ³)		Barcol hardness
	Theoretical	Experimental	
P/G (50 : 50)	1.53	1.47	56 ± 3
P/CC (50 : 50)	1.56	1.56	66 ± 1
P/G/CC (50 : 15 : 35)	1.55	1.57	62 ± 2
P/G/CC (50 : 25 : 25)	1.55	1.54	60 ± 3
P/G/CC (50 : 35 : 15)	1.54	1.54	60 ± 3
P/W (50 : 50)	1.18	1.18	46 ± 3
P/G/W (50 : 15 : 35)	1.27	1.25	50 ± 3
P/G/W (50 : 25 : 25)	1.33	1.29	51 ± 2
P/G/W (50 : 35 : 15)	1.40	1.39	57 ± 2
P/SW (50 : 50)	1.23	1.23	47 ± 3
P/G/SW (50 : 15 : 35)	1.31	1.33	52 ± 4
P/G/SW (50 : 25 : 25)	1.37	1.41	58 ± 3
P/G/SW (50 : 35 : 15)	1.43	1.46	61 ± 4

clearly seen that density increased with the CC or G content (when CC was not in the composition). Also, the experimental values were similar to the theoretical ones (within 4% error), and the difference between them could be easily explained when we consider that this analysis evaluated small samples that may not have been representative of the part. In fact, not even the virgin commercial glass mat was homogeneous when one considers small samples (and a stack of a few layers only).

Also, the composites with W showed a lower density than the composites with CC, which was advantageous. The composites with SW showed a higher density than those with ordinary W because of the expected higher glass content of the former. In addition, because the composition of the W was considerably variable, the difference between the experimental and theoretical values tended to increase.

Regarding the Barcol hardness (shown in Table IV), the highest values were found for the composites with higher CC content because this inorganic filler had a high hardness and distributed homogeneously in the resin, which was clearly not the same for the samples with glass mats. Also, hardness was measured at the surface, where the resin content (along with the CC when present) was comparatively higher, to the detriment of the fibers. Among the composites with incorporated W, the ones with SW showed a higher hardness because of their higher glass content.

To assess the composition of the various samples, TGA was carried out. As an example, the thermogravimetric curve of the P/CC (50 : 50) sample is shown in Figure 1. The thermal degradation of the polyester resin usually occurs in two stages,^{14,15} and in this work, the main decomposition occurred at 280–460°C.

At about 780–800°C, CC decomposed into calcium oxide (CaO) and carbon dioxide (CO₂; also shown in

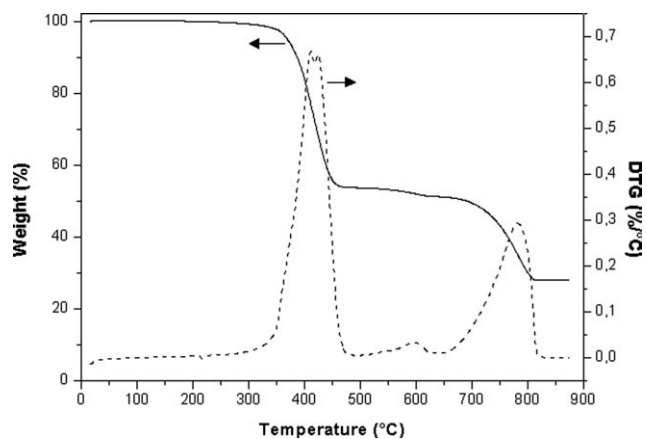


Figure 1 TGA of the composite with 50 wt % CC.

Fig. 1). The amount of released CO₂ was used to quantify the original amount of CC in each sample via stoichiometric calculations.¹⁶ In addition, the amount of CaO present in the residue (>800°C) was analogously estimated, and this value was subtracted from the weight of the residue to reveal the glass fiber weight content because the fiber itself shows only minor weight loss in this temperature range.¹⁷ The estimated amount of CC for all composites with this filler is presented in Table V. The results were in good agreement with the expected values, except for P/G/CC (50 : 35 : 15), probably because of the difficulties in homogenizing a small amount of CC into the resin.

Both the TGA and burnout results, shown in Table V, could be used to infer the glass content of the various samples. The advantage of the burnout method over TGA, apart from being cheaper and more readily available, is that the former evaluates a much larger amount of sample (typically, 5 g) than the latter (typically, 20 mg) and is, therefore, more

TABLE V
TGA and Burnout Test Results for the Bicomponent and Tricomponent Composites

Formulation	TGA			Burnout
	Weight at 600°C (%)	G (%)	Residue at 875°C (%)	Weight at 600°C for 4 h (%)
P/G (50 : 50)	51.7	—	51.7	48.1
P/CC (50 : 50)	52.0	—	52.5	49.1
P/G/CC (50 : 15 : 35)	47.1	12.1	37.9	47.9
P/G/CC (50 : 25 : 25)	51.1	25.1	24.9	39.7
P/G/CC (50 : 35 : 15)	51.9	38.8	11.3	47.1
P/W (50 : 50)	10.1	—	—	5.5
P/G/W (50 : 15 : 35)	6.3	—	—	6.3
P/G/W (50 : 25 : 25)	23.1	—	—	22.9
P/G/W (50 : 35 : 15)	35.8	—	—	35.8
P/SW (50 : 50)	20.5	—	—	19.7
P/G/SW (50 : 15 : 35)	26.4	—	—	25.5
P/G/SW (50 : 25 : 25)	32.5	—	—	31.8
P/G/SW (50 : 35 : 15)	45.8	—	—	41.4

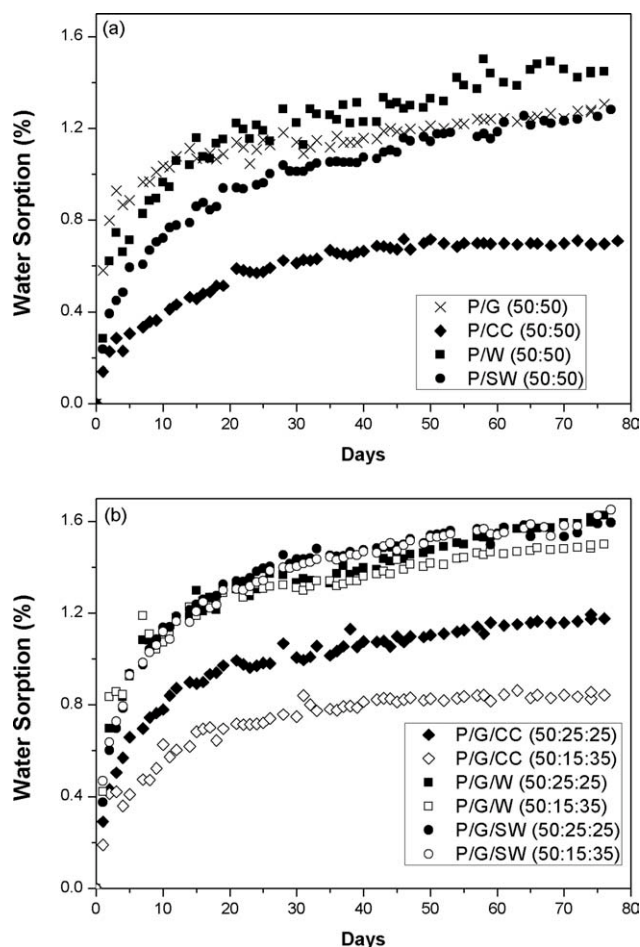


Figure 2 Water sorption results for (a) bicomponent and (b) tricomponent mixtures.

representative of highly heterogeneous samples. Also, when the distinct periods of exposure to high temperature are taken into account, a comparison between the results of the TGA at 600°C and of the burnout test is not straightforward.

According to the TGA, the cured unsaturated polyester resin left just 4.5 and 1% at 600 and 875°C, respectively. In the burnout test of polyester, less than 0.1% residue was found. Considering that and the fact that the residue at 600°C did not only refer to glass fiber (especially for the samples with CC), we observed that the amount of fibers increased with the amount of residue.

It was also possible to contrast the glass content of the general W and the sorted one, which were 3.9 and 17.9 wt % for P/W (50 : 50) and P/SW (50 : 50), respectively. This was a fourfold increase in the glass content, which showed the efficiency of the manual separation of fibers and ratified the density results previously shown because the G displayed more than twice the density of the polyester. Indeed, the fiber-rich SW was basically comprised of Gs with resin residue adhered to their surface. Also, because of the very low amount of G in the general W, poor mechanical properties could be expected for these samples.

Figure 2 shows the median water sorption curve obtained for each composite. Water retention in the polymer composites could have been a consequence of hydrolysis of the chain ends of unsaturated polyester and also of hydrophilization, which increased the sorption through swelling and plasticization of the matrix.¹⁸ The sorption process is usually considered to follow non-Fickian diffusion with two stages: first, the composite absorbs water very quickly, and after that, the sorption becomes slower; this delineates a sigmoid curve that approaches equilibrium state, with the first one known as quasi-equilibrium.¹⁹

In this work, only one type of polymer matrix was used; that is, the amount of polar sites²⁰ available varied only with the polymer content. Thus, the sorption of water basically depended on the amount of polymer, the crosslinking efficiency, and the availability of voids in the polymer due to processing particulars in each case. As shown in Figure 2(a,b), it can be said that

1. The composites with G only or G/CC approached equilibrium faster than the others.
2. The composites with CC reached the lowest sorption values, in the same range reported in the literature.²¹ This could be associated with the size of the CC particles (18.22 μm), which were much finer than the covered glass or the polyester particles of the residues (minimum of 1.19 mm), and the latter caused a much more significant disruption of the polymer network and, therefore, promoted water sorption.
3. The water sorption increased with the introduction of W. The addition of W, even when encapsulated in the midlayers of the sample, promoted sorption when the water managed to reach those layers after a certain period of time. Indeed, the P/G (50 : 50), P/W (50 : 50), and P/SW (50 : 50) curves were comparable in the first few days and differentiated after that.

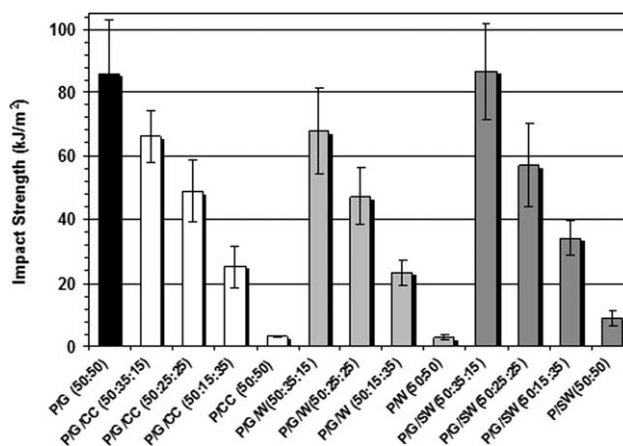


Figure 3 Izod impact strength of the bicomponent and tricomponent composites.

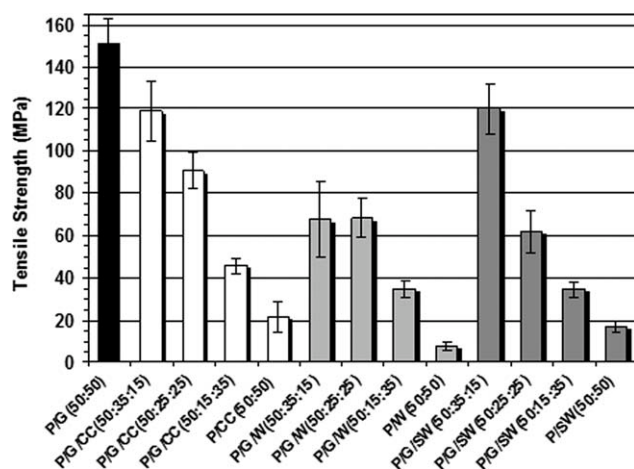


Figure 4 Tensile strength of the bicomponent and tricomponent composites.

- The introduction of W or SW caused a similar increase in water sorption.

Figure 3 shows the Izod impact strength of the composites. It can be seen that the amount of virgin glass was determinant of the impact strength, and its substitution with either CC, W, or SW significantly reduced this value. The rigid inclusions (e.g., CC) hindered polymer movement in the surroundings of the particle, increasing the stiffness of the sample and, when this was not compensated by adequate adhesion at the interface (as found for the virgin fibers), with associated energy use to enable debonding, ultimately leading to a decrease in the impact strength.

The incorporation of W caused a similar reduction in the impact strength to that found for CC. However, the use of SW (with a higher content of recycled fibers in it) was responsible for a consistent recovery of this property, and ultimately, the inclusion of 15% SW did not significantly alter the impact strength of the composite when compared to the one produced with virgin materials only, that is, P/G (50 : 50). This was very interesting for technological purposes because a controlled addition of SW was able to maintain the performance of the composite with 50% virgin G; this decreased the cost of raw material used and reduced the W discarded into the environment.²²

With regard to tensile strength (Fig. 4), the highest value was found for the composite produced with virgin materials only, as expected. Here again, the inclusion of either CC, W, or SW significantly reduced this value, which was associated with a lower fiber content, poorer dispersion and distribution in the matrix, and higher void content. Unfortunately, the void content could not be estimated using the density results because of the considerable heterogeneity of the W used. Another parameter affecting

the strength results was fiber length, which was significantly higher in the virgin glass mats in comparison to the W.

The use of 15% CC caused c. a. 20% reduction in tensile strength, which was similar to that found for the inclusion of 15% SW. For higher CC loading, however, the SW produced inferior results. Thus, with regard to this property, we inferred that SW could be used to replace only a controlled amount of CC in the composites.

With regard to the DMA results for the 50 : 50 mixtures [Fig. 5(a)]. The storage modulus curves for P/CC, P/W, and P/SW were similar and much lower than that of the P/G composite, even though small differences could be noticed, including: (1) the P/CC and P/SW were slightly higher than the P/W up to 70°C; that is, the higher amount of glass in the SW in comparison with the P/W was sufficient to yield a similar response to P/CC, and (2) P/SW and P/W, both with an overall higher amount of resin in the sample than P/CC, decayed similarly from 80°C, whereas the P/CC showed a greater resistance to temperature variation.

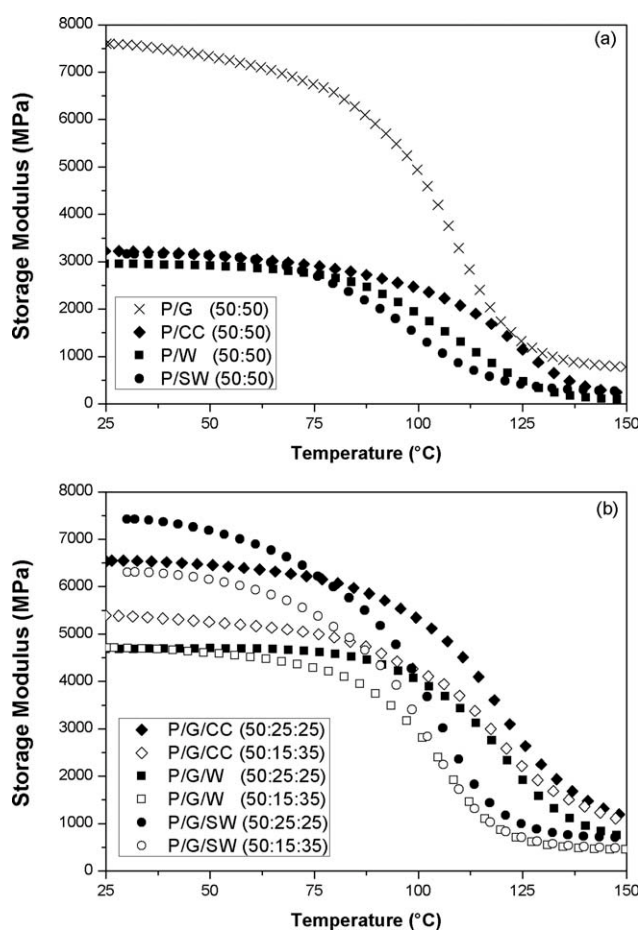


Figure 5 Storage modulus results for the (a) bicomponent and (b) tricomponent mixtures.

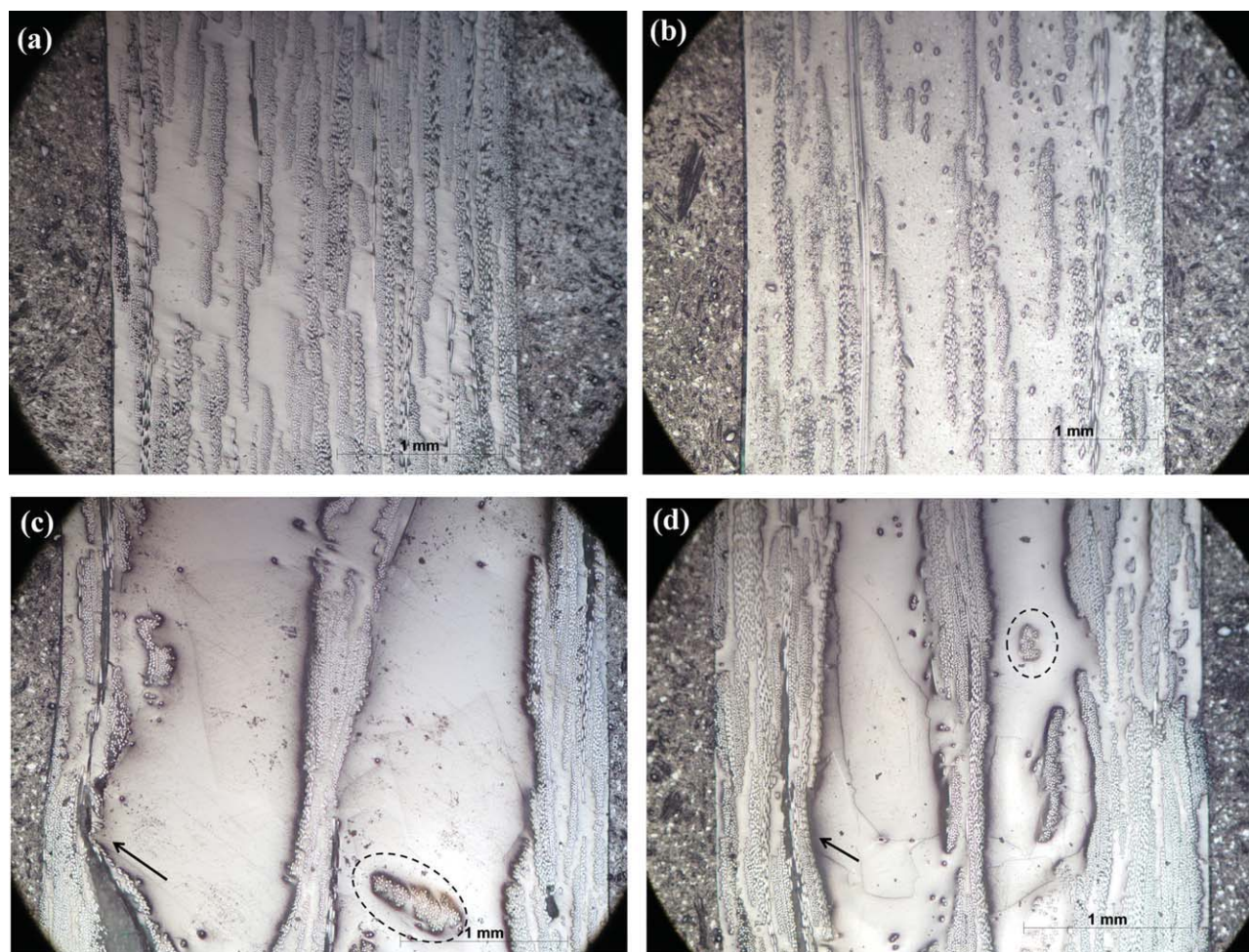


Figure 6 Optical microscopy obtained for (a) P/G (50 : 50), (b) P/G/CC (50 : 35 : 15), (c) P/G/W (50 : 35 : 15), and (d) P/G/SW (50 : 35 : 15). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Focusing on the DMA results of the tricomponent mixtures [Fig. 5(b)], the storage modulus increased in each mixture whenever the G content increased because of a more restricted movement of the polymer chains brought about by the inclusion of the rigid reinforcement. Also, with regard to the 50 : 15 : 35 and the 50 : 25 : 25 mixtures of P/G/W, the small increase in G content and the inclusion of a considerable amount of soft particles were unable to produce an increase in modulus. This was not noticed for the P/G/SW mixture, where the 50 : 25 : 25 mixture was superior than the 50 : 15 : 35 one. The P/G/SW was even higher than the P/G/CC, possibly because of the better glass/resin adhesion in comparison with CC/resin adhesion, although this occurred only up to 75–85°C, where the higher amount of P and the greater disruption of the virgin crosslinked resin network started to play a more significant role on the composite behavior. A comparison of Figure 5(a,b) led to a very interesting result: P/G (50 : 50) was actually very similar to P/G/SW (50 : 25 : 25) up to about 60°C; that is, we were able

to substitute 25% of G with 25% of SW without compromising the modulus response of the composite.

Microstructural analysis was performed in the bicomponent mixture and the tricomponent composites that showed the best mechanical performance, that is, with 35% G. In Figure 6(a–d), the clearest regions are the P resin, and the near circular shapes represent the cross section of glass fiber (from the mats) arranged at distinct angles. The dark regions, especially within fiber bundles, are microvoids.

Figure 6(a), for P/G (50 : 50), shows that the six glass mat layers were homogeneously distributed throughout the thickness of the sample. The same could be said about the P/G/CC (50 : 35 : 15) sample [Fig. 6(b)] with five glass layers because the CC was mixed with the resin before molding. On the other hand, Figure 6(c,d), also with five glass layers, shows a contrasting feature: the solid W particle forced the outer glass layers toward the surface of the composite and led to a heterogeneous fiber distribution throughout the thickness of the sample, which resembled a sandwich structure rather than a laminate. It was also

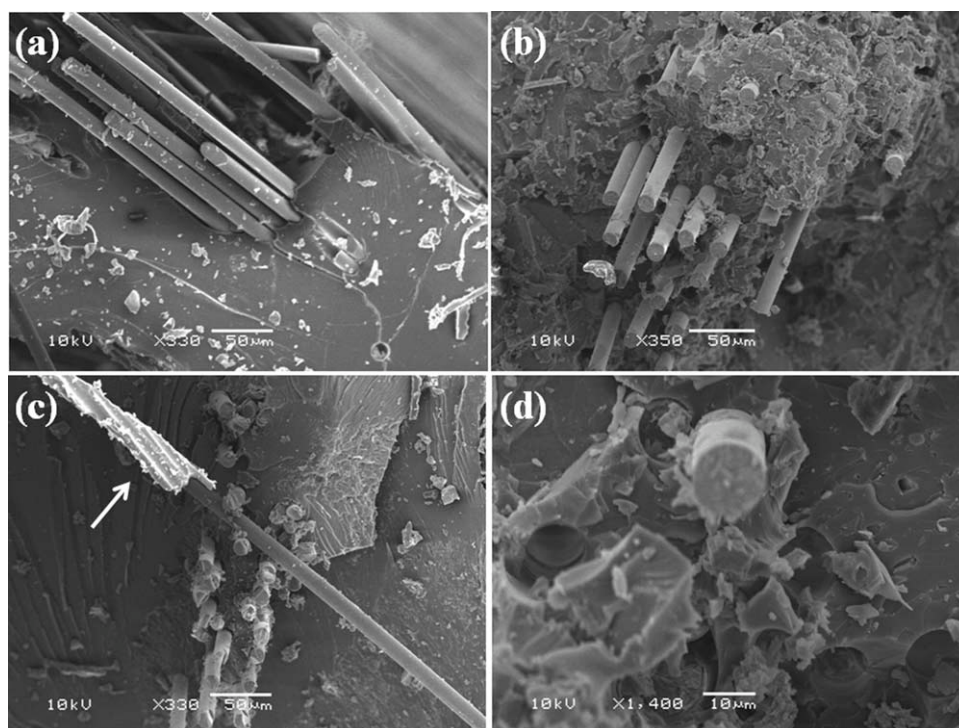


Figure 7 SEM images obtained for (a) P/G (50 : 50), (b) P/G/CC (50 : 35 : 15), and (c) P/G/W (50 : 35 : 15) and for (d) P/G/CC (50 : 35 : 15) at higher magnification.

interesting to notice that the P/G/W sample, which had larger particles than the P/G/SW sample, caused a greater disturbance of the lay-up (see arrows). In Figures 6(c,d), it is also possible to notice some glass (see dashed ellipses) near the resin particles, both from the original W fraction. This sandwichlike arrangement undoubtedly affected the results, such as those from flexural and DMA testing.

Figures 7(a–d) shows SEM images obtained from the Izod fractured surfaces of P/G (50 : 50), P/G/CC (50 : 35 : 15), and P/G/W (50 : 35 : 15) samples. The good adhesion between the glass and the polyester resin was indicated by the presence of resin residues adhered at the fiber surfaces, even though pullout sites could also be noticed. Comparing Figures 7(a) and 7(b), it was clear that when the CC was present, the fractured resin surface became less smooth. A similar effect was reported by Yuan et al.²³ for the inclusion of organic rectorite, a type of layered silicate, in a polyester resin. At higher magnification [Fig. 7(d)], the CC particles at the surface of the P/G/CC (50 : 35 : 15) sample became more evident. In Figure 7(c), for the P/G/W (50 : 35 : 15) sample, it is easy to notice a glass fiber (previously coated with resin), which came from the original W fraction.

CONCLUSIONS

The use of W in the 9–16-mesh range yielded more homogeneous composites, with higher impact

strength, than that of the finer mesh; therefore, some degree of fiber–matrix interaction, rather than only the particle–matrix, is expected to have occurred. The simple grinding of the composite residues (W) yielded composites with generally worse characteristics than the ones with calcium carbonate; this was partly attributed to their heterogeneity and very low fiber content. This was, to some extent, overcome by the removal of a large part of the pure resin particles from the W to reveal resin-covered fibers. The W fraction richer in polymer could be more successfully used in an energy recycling process rather than as filler for composites.

In general, the use of the SW, that is, the W fraction richer in glass fibers, as filler for composites yielded higher water sorption and impact strength but similar hardness and tensile strength in comparison with calcium carbonate filled composites. Also, the reduction in impact and tensile strength of the composite with 15% SW was within just 20% of those of the fully virgin composite.

In all, the sorted waste showed potential to be used as general purpose filler, reaching or sometimes surpassing the overall properties obtained with CaCO₃ for a controlled amount of waste addition. This substitution could decrease the cost of raw materials used and reduce the amount of W currently discarded in the environment. It is important to add that the manual sorting of glass fibers, as employed here, is adequate for laboratory-scale experiments only. For the industrial scale, this step should be optimized with a more efficient method,

for example, the use of a saline solution to create a separation fluid of the desired density²⁴ or via cascade air classification.¹⁰

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