

Characterization of Composites Based on Recycled Expanded Polystyrene Reinforced with Curaua Fibers

Cleide Borsoi,¹ Lisete C. Scienza,² Ademir J. Zattera¹

¹Postgraduate Program in Engineering Processes and Technologies (PGEPROTEC), Centre of Exact Science and Technology (CCET), University of Caxias do Sul (UCS), CEP 95070-560 Caxias do Sul, Rio Grande do Sul, Brazil

²Laboratory of Corrosion and Surface Protection (LCOR), University of Caxias do Sul (UCS), Caxias do Sul, Rio Grande do Sul, Brazil
Correspondence to: A. J. Zattera (E-mail: ajzatter@ucs.br)

ABSTRACT: This study evaluated the mechanical, thermal, rheological, and morphological properties of virgin and recycled matrices and their composites with 20 wt % of curaua fiber. The recycling process of postconsumer polystyrene was carried out by grinding and extrusion. It was found that the recycling of expanded polystyrene did not have a major influence on the mechanical properties; however, the thermal stability was increased. The addition of curaua fibers led to increases in the tensile strength, modulus of elasticity, rigidity, thermal stability and melt viscosity of the composites. The composites made with the recycled matrix revealed higher thermal stability and melt viscosity than those made with the virgin matrix. Scanning electron microscopy characterization showed empty spaces where the curaua fibers had pulled out of the matrices in the fractured regions, indicating poor interfacial adhesion without the use of a coupling agent. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

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INTRODUCTION

Expanded polystyrene (EPS) is widely used in insulation materials for protecting goods in transport, for food and electronics packaging, and for insulation in air conditioning systems, among many other applications.¹ EPS products have relatively short life cycles compared to products made of polystyrene (PS); as a consequence, EPS is used only in special applications. Although a large volume of EPS waste is generated, its relatively low density (ranging from 10 to 25 kg m⁻³) makes it inconvenient for reuse.²

The polyolefins used for packaging materials are often discarded after a single use, which results in a large amount of polymer waste.³ As postconsumer polymeric waste represents one of the largest categories of waste, their alternative use for the production of composite materials is of great economic and environmental importance⁴ and involves reintroducing the waste into new production cycles, which further avoids their environmental disposal. In this way, various recycling methods have been proposed in the literature, such as chemical, thermal, and mechanical recycling,⁵ all of which aim to minimize the environmental impact generated by the final disposal of EPS.

There is recent growing interest in vegetable fibers for use as polymer reinforcements owing to their economical production with few requirements for equipment as well as their low spe-

cific weights, which result in higher specific strength and stiffness values vs. glass-reinforced composites.⁶ Vegetable fibers are nonabrasive to mixing and molding equipment, present safer handling and working conditions compared to synthetic reinforcement materials and represent a positive environmental impact.⁶ Usually, these composites employ natural fibers such as sisal, ramie, cotton, and curaua. Production of thermoplastic matrix composites fundamentally depends on the quality of the natural fiber and should combine good mechanical properties with low density and good dimensional stability.^{7,8}

EPS recycling approaches depend on the final application of the material. Poletto et al.⁹ evaluated the effect of wood flour as a reinforcing filler in composites of recycled EPS matrix. To reduce density without degradation, the authors found that the PS should be hot-pressed at 130°C for 5 min, giving a 25-fold increase in the density of the material compared with its initial density. After the injection process, a 48-fold increase in apparent density was also observed. The authors concluded that the recycled material can be employed to fabricate high mechanical property, low-density composites, which can be used after pressing and milling without any additional processing. Nair and Thomas¹⁰ evaluated short sisal fiber-reinforced PS composites with different coupling agents for their behavior and mechanical property dependence on water absorption and aging

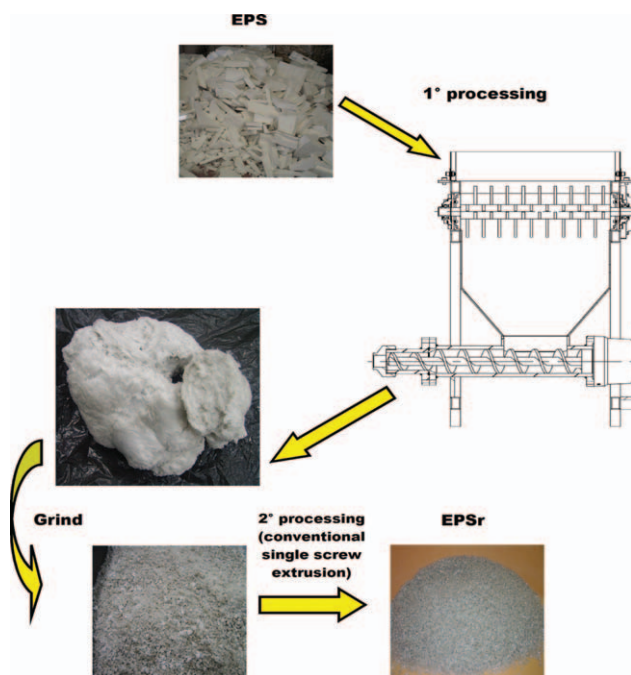


Figure 1. Process used for recycling EPS. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

effects. The authors found that the addition of a coupling agent promoted better dimensional stability and tensile strength of the composites compared to composites free of coupling agents.

Most studies directed toward developing thermoplastic matrix–fibers composites make use of a virgin matrix material.^{11–14} Several current studies are underway toward the development of composites that incorporate curaua (*Ananas erectifolius* L.B. Smith) fiber in different matrices such as high-density polyethylene,^{15,16} polypropylene,¹⁶ recycled polypropylene,¹⁷ polyurethane,¹⁸ and polyamide-6.¹⁹ However, no study has yet been reported concerning curaua fiber (CF)-reinforced PS. This study aims to obtain and characterize CF-reinforced PS composites based on both virgin PS and postconsumer EPS matrices.

EXPERIMENTAL

Materials

CFs were obtained from CEAPAC (Support Center for Community Action Projects) in Santarem, Pará, Brazil. Virgin PS with a melt flow index of 20 g/10 min (200°C, 5 kg) and a deflection temperature under load of 81°C (1.8 MPa, 120°C h⁻¹) was supplied by Innova S/A (Triunfo, Rio Grande do Sul, Brazil). EPS was supplied by ARCS (Association of Recyclers of Caxias do Sul) in Rio Grande do Sul, Brazil.

Fiber Preparation

CFs were first subjected to a cleaning step for husk removal and then were cut into 2.5 cm long pieces, washed in distilled water at room temperature for 1 h, and dried at 70°C for 12 h. The medium diameter of fibers was 73 μm.

Recycling of EPS

EPS was recycled in a single-unit piece of equipment consisting of a mill coupled to a heated endless screw. EPS was first

worked in the grinder and then conveyed to the thread where it was heated to a maximum temperature of 100°C. The recycled expanded polystyrene (EPSr) was ground in a Primotecnica knife mill model 1001 and processed in a Seibt single-screw extruder model ES35FR. The temperatures at the different heating zones varied according to the following profile: 140, 160, and 180°C at a screw speed of 60 rpm. The process used for recycling EPS is shown in Figure 1.

Preparation of Composites

To obtain the composite, the matrix and CF were premixed in a single-screw extruder (Seibt model ES35FR) at a screw speed of 40 rpm. The temperatures at the different heating zones varied according to the following profile: 140, 160, and 180°C. The composites were ground in a knife mill (Primotecnica, model 1001) and dried in an oven for 12 h at 70°C. The resulting material was processed in a twin-screw extruder (MH Equipamentos, model COR 20-32-LAB) at a screw rotation speed of 200 rpm. The extruder had eight heating zones with the following profile: 115, 150, 185, 185, 180, 175, 175, and 170°C. The previously dried samples were made by injection (Himaco Hidráulicos e Máquinas, Novo Hamburgo, Rio Grande do Sul, Brazil, model LHS 150-80) with the following temperature profile: 150, 165, and 180°C. The screw speed was 100 rpm and the mold temperature was approximately 20°C. The identities of the composites are listed in Table I.

Characterization

Polymer Matrices Characterization. The molecular weight of PS and EPSr was determined by size exclusion chromatography using a system Waters chromatographer. Tetrahydrofuran was employed as solvent with a flow rate of 1.0 mL min⁻¹, injection volume of 150 μL, and temperature of 45°C. Tensile tests were carried out in a universal testing machine, the EMIC DL 2000, in accordance with ASTM D638-10 at a speed of 5 mm min⁻¹. Thermal analysis was performed in a Shimadzu TGA-50 from 23 to 600°C at a heating rate of 10°C min⁻¹ and a nitrogen flow rate of 50 mL min⁻¹. Fourier transform infrared spectroscopy (FTIR) was performed according to the technique of attenuated total reflectance (ATR) using an IS10 Thermo Scientific Nicolet spectrometer with a measuring range of 4000–400 cm⁻¹. Morphological characterization was carried out with a JEOL JSM 6060 scanning electron microscope (SEM) at an accelerating voltage of 15 kV.

Composites Characterization. Characterization of the composites included tensile tests, SEM, and thermogravimetric analysis (TGA and DTGA), as described previously. Izod impact strength

Table I. Identification of composites with CF and virgin (PS) and recycled (EPSr) matrix

Sample	PS matrix content (wt %)	EPSr matrix content (wt %)	CF content (wt %)
EPSr	–	100	–
PS	100	–	–
EPFC20	–	80	20
PFC20	80	–	20

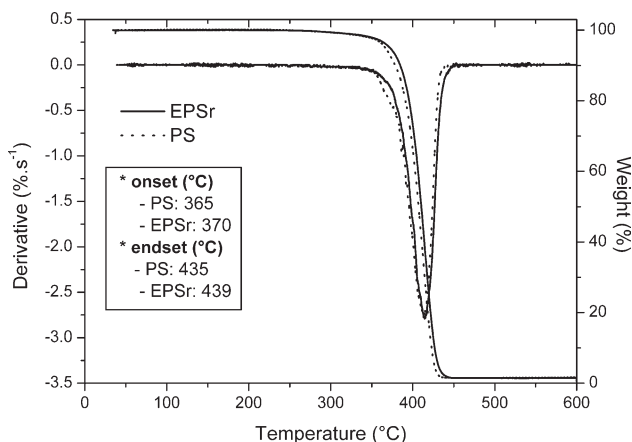


Figure 2. TGA and DTGA for PS and EPSr matrices.

was measured with a CEAST Resil 25 pendulum 1J using unnotched specimens in accordance with ASTM D256-10. Heat deflection temperature (HDT) analysis was performed in a HDT/VICAT 6CEAST at a heating rate of $120^{\circ}\text{C h}^{-1}$ and applied load of 1.82 MPa. The HDT measurement was taken when the specimen underwent a deflection of 0.25 mm. The rheological behaviors of the composites were assessed with a capillary rheometer Galaxi III, model 9052. The tests were performed in triplicate at 190°C with measurements at 14 shear rates between 100 and 2500 s^{-1} . The values of shear rate ($\dot{\gamma}_w$), shear stress (τ_w), and melt viscosity (η) were obtained directly, and the Rabinowitsch correction factor $(3n + 1)/4n$ was applied. The value of n was obtained from the slope of the graph of $\ln \tau_w$ vs. $\ln \dot{\gamma}_w$ (eq. (1)).²⁰

$$n = \frac{d(\ln \tau_w)}{d(\ln \dot{\gamma}_w)} \quad (1)$$

The correction of shear rate on the wall for pseudo-plastic materials was calculated according to eq. (2).²⁰

$$\gamma_{\omega a} = \dot{\gamma}_w \times \frac{3n + 1}{4n} \quad (2)$$

Thus, the apparent viscosity (η_a) was calculated according to eq. (3).²⁰

$$\eta_a = \frac{\tau_w}{\gamma_{\omega a}} \quad (3)$$

RESULTS AND DISCUSSION

Polymer Matrices Characterization

The average molecular weights of 190,935 and $195,041\text{ g mol}^{-1}$ were found for PS and EPSr, respectively. The polydispersion was found around 2.9 for both matrices. These results mean that possible differences found in the thermal and mechanical properties of the composites may not be attributed to the differences in their molecular weight.

Tensile analysis revealed tensile strength values of 37.57 ± 0.21 and $32.03 \pm 0.88\text{ MPa}$ for the virgin (PS) and recycled (EPSr) matrices, respectively. Thus, it appears that the recycling process entails an approximate 15% reduction in tensile strength. This behavior could result from either the possible presence of impurities in the recycled material or the EPS storage conditions. Natural environmental conditions such as variations in sunlight, moisture, and temperature have a direct influence on the mechanical, physical, and chemical properties of polymers; as such, they are critical factors for special applications.²¹

In Figure 2, EPSr shows slightly better thermal stability than PS, which can be observed as a weight loss temperature of the EPSr at approximately 5°C above that of the virgin PS degradation temperature. This result agrees with the studies by Samper et al.,² which describe similar values in the initial degradation temperatures of virgin and recycled PS. The derivative curve shows that the maximum degradation rates for PS and EPSr both occur at 417°C , which confirms that the PS was not thermally degraded during the recycling process.

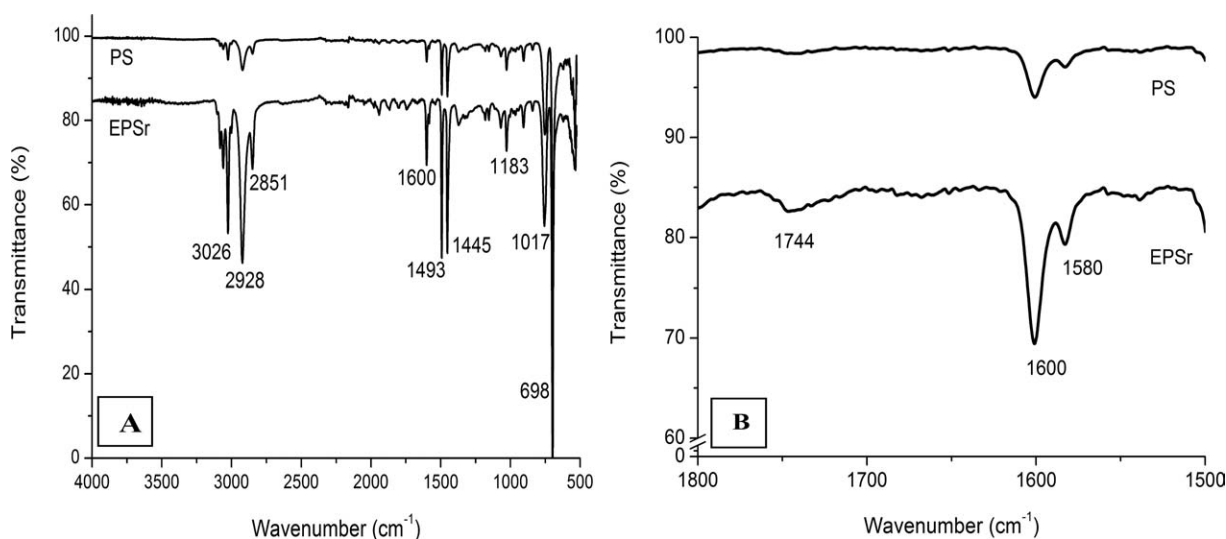


Figure 3. (A) FTIR-ATR spectra of PS and EPSr matrices from 500 to 4000 cm^{-1} and (B) ATR-FTIR spectra of PS and EPSr matrices from 1500 to 1800 cm^{-1} .

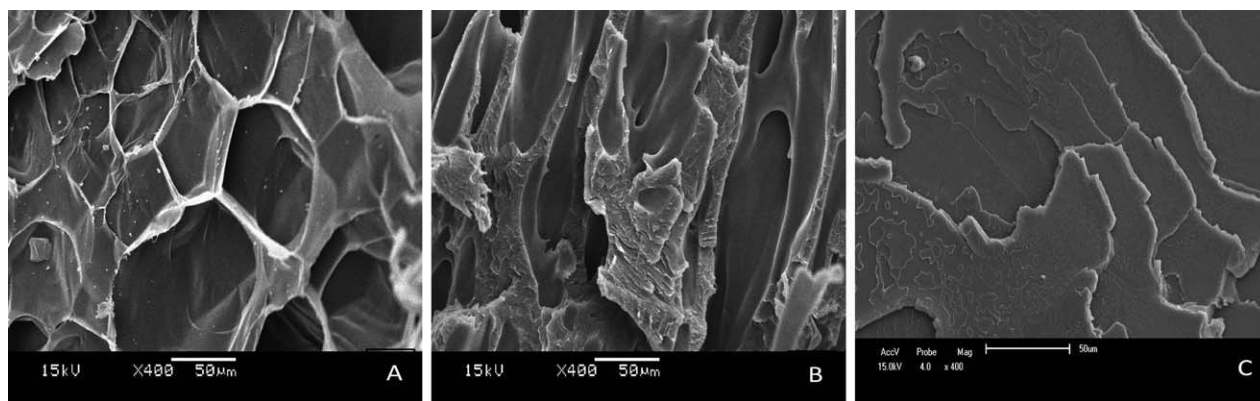


Figure 4. Morphology of PS: (A) before processing (EPS), (B) after the first processing step, and (C) after the second processing (EPSr) step.

Absorption bands for PS and EPSr are shown in Figure 3(A) in which the 3026 cm^{-1} band is related to the axial deformation of the aromatic CH and the bands at 2928 and 2851 cm^{-1} are related to stretching vibrations of asymmetric and symmetric CH and CH_2 groups, respectively. The 1600 and 1493 cm^{-1} bands are related to the $\text{C}=\text{C}$ stretching of the aromatic ring, the 1445 cm^{-1} band is related to the symmetric and asymmetric CH_2 angular deformation, the 1183 and 1017 cm^{-1} bands correspond to the $\text{C}-\text{C}$ stretch, and the 698 cm^{-1} band corresponds to the CH aromatic out-of-plane deformation.²² In Figure 3(B), a new band corresponding to the carbonyl group ($\text{C}=\text{O}$) can be observed at 1744 cm^{-1} in the EPSr, which can be related to the oxidation products originating from the degradation process.²³ The bands at 1600 and 1580 cm^{-1} , corresponding to $\text{C}=\text{C}$ stretching of the aromatic ring, show increased intensities. Although the appearance in the carbonyl group owing to recycling can be observed, this has no effect on the thermal stability of EPSr.

The loss of properties exhibited by the recycled materials results from the degradation processes that take place during processing, service life, and mechanical recycling.²⁴ Thermal, mechanical, and oxidative degradation may occur during processing, whereas photo- and thermo-oxidative degradation and hydrolysis may occur during exposure to the natural environment.²⁵ Thus, when analyzing recycled materials, it is important to determine whether the degradation of the polymer has occurred during the waste recycling process or during the material's life cycle.²

Figure 4 shows SEM images of the EPS structure (A) before processing, (B) after the first processing step, and (C) after the

second processing step. Before processing, the expanded PS contains cells resulting from the expansion process, and consequently, the EPS density decreases. After initial processing, the cells have undergone deformation, leading to a more compact structure. An even denser structure is obtained after the second processing step [Figure 4(C)], where the extruded material exhibits a fragile and uniform structure without voids and can be further used as a matrix for developing polymer composites.

Composites Characterization

Table II lists the HDT, tensile strength, tensile modulus, and impact strength analysis for PS and EPSr matrices as well as PFC20 and EPFC20 composites. It can be observed that the presence of the fiber leads to increases in the deflection temperature values for both composites. The HDT of the recycled matrix is higher than that of the virgin matrix. According to Huda et al.²⁶ and Garcia et al.,²⁷ the HDT of a polymer can be increased, among other ways, by adding natural fibers to polymers increases the stiffness of the material as verified in this study.

The tensile strength appears to increase with the addition of CFs for both composites. For the matrices, there is a 26% increase in the PS matrix strength relative to that of the EPSr matrix, but this difference drops to 10% following the addition of 20 wt % of CF. The mechanical properties of composite materials are influenced by many factors, including the aspect ratio of the fiber, the fiber/matrix interaction, and the processing temperatures.³

Table II summarizes that the modulus of elasticity for EPSr increases compared to that of PS. This is achieved at the

Table II. HDT, Tensile Strength, Tensile Modulus, and Impact Strength of Composites as a Function of CF Content

Sample	HDT (°C)	Tensile strength (MPa)	Tensile modulus (MPa)	Impact strength (J/m)
EPSr	77.6 ± 0.8	26.5 ± 0.3	3712 ± 756	148.9 ± 27.7
PS	70.2 ± 1.1	35.9 ± 1.5	3284 ± 342	257.9 ± 22.3
EPFC20	86.7 ± 1.8	43.5 ± 0.7	5738 ± 339	166.6 ± 28.1
PFC20	76.9 ± 1.3	48.1 ± 1.6	5799 ± 422	182.5 ± 21.5

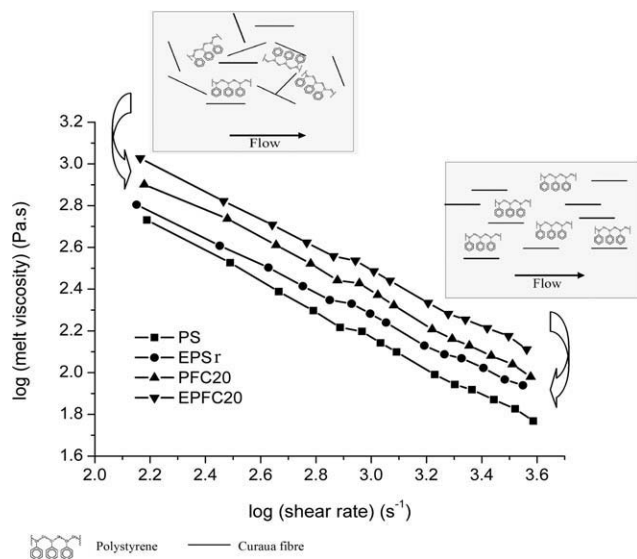


Figure 5. Variation of the melt viscosity in relation to the shear rate of composites as a function of the CF content at 190°C.

expense of a wider standard deviation and increases the stiffness of the material. However, with the addition of 20 wt % of CF, no significant difference in the modulus as a function of matrix type can be observed. The highest modulus of elasticity value can be observed for the PFC20 composite and is 43% higher than that of the matrix (PS). Poletto et al.⁹ observed that by adding 40 wt % of wood flour to a recycled PS matrix, the modulus was increased by 100% compared to the matrix alone.

The absorption of moisture by the fibers can yield a plasticity effect in the composite, reducing the tensile modulus and tensile strength. However, if sufficient fiber–matrix interaction is provided, the plasticity effect is no longer observed.²⁸

The impact energy is important because limitations under impact are very common in rigid polymers during use. In composites, the addition of fibers may result in decreased polymer chain mobility and act as stress concentrators in the polymer

matrix, reducing the energy of initiation of cracks, which reduces both the ability of the composite to absorb energy⁹ and the impact strength properties. Among other ways, the deformation and fracture energy can be absorbed by fiber breakage and fiber pull-out.²⁹ In our study, a significant reduction in impact strength can be observed for the virgin matrix composites with 20 wt % of CF; meanwhile, the recycled matrix exhibits a trend toward increased impact strength.

Figure 5 shows the behavior of melt viscosity in relation to shear rate for composites of the virgin and recycled matrices at 190°C. Viscosity decreases with increasing shear rate for all composites. According to Joseph et al.,³⁰ this behavior is characteristic of pseudo-plastic materials because the polymer molecules and the fibers orient along the flow direction. The decrease in viscosity with increasing shear rate as the fibers are incorporated can be explained by fiber–fiber and/or fiber–matrix contact. For lower shear rates, the fibers are distributed with more disorder, which causes more fiber–fiber contact, further leading to increased viscosity.³¹ In relation to the addition of the CF in PS virgin or recycled matrix, the viscosity increases in relation to the polymer only. According to Nair et al.,³¹ adding the fiber to the polymer disrupts normal flow and hinders the mobility of the polymer segments, which increases the viscosity of the polymer until a critical fiber concentration level is reached. At a shear rate of $\log 2.9 \text{ s}^{-1}$, it is possible to observe a decrease in viscosity for all composites. The non-linearity of the system may be owing to the fiber orientation in the matrix as well as the effect of the sliding wall.²⁰

The TGA and DTGA analysis for PS and EPSr matrices and PFC20 and EPFC20 composites are shown in Figure 6. In Figure 6(A), thermal degradation of the matrices starts at approximately 305°C and ends at 435°C. For CFs, the first mass loss occurs before 100°C and can be attributed to the loss of moisture. The second mass loss begins nearly at 240°C and extends to approximately 375°C. It is related to the degradation of cellulose and hemicellulose as well as to the slow degradation of lignin. Cellulose fibers begin to lose weight at approximately at 200°C, starting at the amorphous region and depending on the

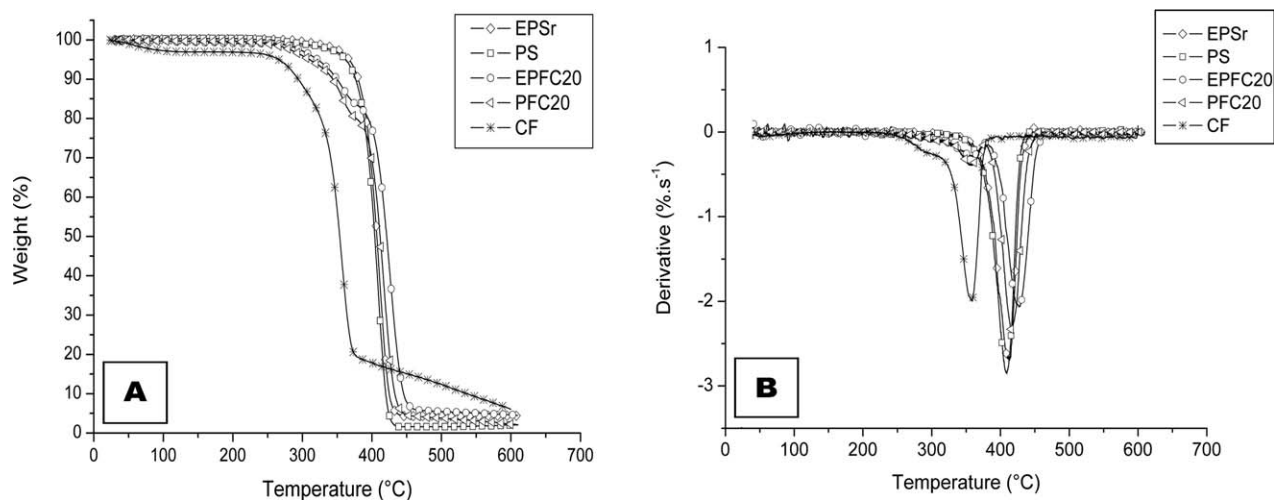


Figure 6. Thermal analysis: (A) TGA and (B) DTGA of CFs and composites as a function of the CF content.

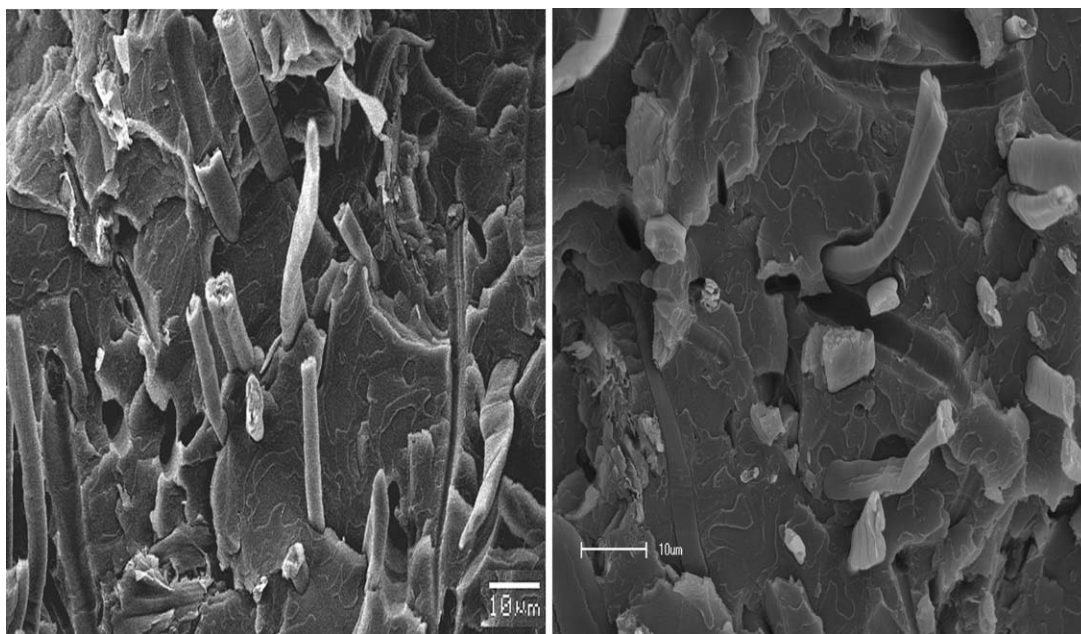


Figure 7. Morphological analysis for 20 wt % of CF in virgin (left) and recycled matrix (right) composites.

residence time at this temperature.³² The composites exhibit an intermediate behavior between that of the matrix and the CF. Adding fiber to the polymer results in a reduction of the thermal stability of the material, which starts approximately at 260°C. This reduction does not compromise the extrusion and injection molding maximum temperatures of 185 and 180°C, respectively. The second process begins at 385°C and ends at 460°C for the EPFC20 composite, whereas for PFC20, it begins at 376°C and ends at 450°C. Therefore, it appears that the recycled matrix composites possess better thermal stability compared to the virgin matrix composites.

Figure 6(B) shows the curve of the first derivative of mass loss for CF and the 20 wt % CF composites in virgin and recycled matrices. The maximum decomposition rate occurs at 411°C for EPSr and at 408°C for PS. For the CF composite, two degradation temperatures were observed, one approximately at 237°C and the other at 300°C. The maximum degradation temperature occurs approximately at 356°C; according to Joseph¹¹, this can be attributed to a larger cellulose crystal size, a higher crystallinity index, and the degree of polymerization. DTGA analysis shows two degradation temperatures, the first approximately at 355°C for both composites and the second at 427 and 416°C for EPFC20 and PFC20, respectively. These results indicate a higher thermal stability for the composite obtained from the recycled matrix compared to the virgin matrix.

The cross-sectional morphologies of the virgin (PS) and recycled (EPSr) matrix composites are shown in Figure 7. The images reveal good dispersion of the CF in the matrices; however, fiber dispersion appears to be less uniform in the recycled matrix than in the virgin matrix. Similar behavior can be observed for both matrices in relation to the voids observed around the pulled-out fibers of the matrix in the fractured regions. The addition of a coupling agent, among others proce-

dures,^{17,33–34} would improve fiber/matrix adhesion by contributing to a decrease in fiber pull-out and making the fiber break closer to the matrix. The weak fiber/matrix interaction is related to surface energy, where the CF is predominantly polar and PS is nonpolar.³⁵

CONCLUSIONS

Recycling of PS produces changes in its physical, thermal, and morphological properties. It was shown here that the recycling process causes losses in the tensile strength (approximately, 15%), increases in the thermal degradation temperature, and induces deformation in the PS cells (or cavities), which then gives rise to a denser structure. The degradation caused by the recycling process was demonstrated by a new band corresponding to the carbonyl group (1744 cm^{-1}), which confirms that the polymer undergoes a thermo-oxidative process.

The incorporation of CF into PS (virgin matrix) led to increases in the tensile strength and tensile modulus and a reduction in the impact strength. The impact resistance of the recycled matrix composites exhibited an upward trend, which was in contrast to observations for the virgin matrix composites. In relation to the rheological behavior, it was observed that the viscosity increased for all composites with the incorporation of CF and that this effect was highest when the recycled matrix was used. The recycled matrix composites presented higher thermal stability with a higher HDT than the plain matrix and a thermal degradation behavior intermediate between the matrix and the CF alone. However, the composite made from the recycled matrix exhibited higher thermal stability than the composite made from the virgin matrix. With respect to morphology, similar behavior was observed for both matrices.

This study evaluated the properties of recycled PS matrix vs. virgin matrix as related to the addition of CFs to obtain fiber-reinforced composites. It was found that composites made from EPSr and CFs are technically feasible alternative to the reuse of polymer waste into more valuable products.

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