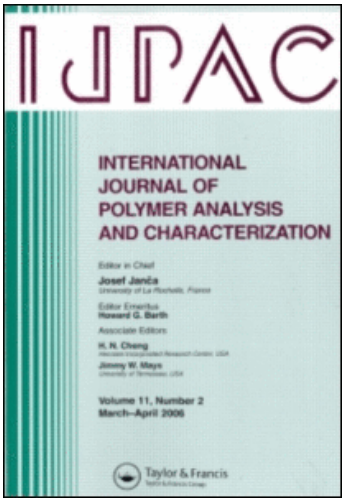

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Characterization of Green Coconut Fiber Composites

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Abstract: Composites of polypropylene and green coconut fiber were prepared in an extruder with temperatures of 140°C in the first zone and 170°C in the other five zones. The screw speed varied from 60 to 140 rpm. Two fiber granulometries were used, X (0,500–0,250 mm) and Y (0,841–0,500 mm). The homogeneity and the effect of the strain history of these composites could be studied by rheological data of molten composites. Flexural modulus was improved by using an adequate fiber granulometry and extruder screw speed. Results showed that the composite with Y fiber, obtained at 140 rpm, was the better one.

Keywords: Extrusion; Flexural modulus; Green coconut fiber; Polypropylene; Rheological properties

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INTRODUCTION

The growing environmental awareness experienced in recent decades has promoted industrial interest in the use of natural fibers as reinforcing material in polymer composites.[1–4] Because of their low density, good mechanical performance, unlimited availability, and problem-free disposal, natural fibers offer a real alternative to synthetic reinforcing fibers. Moreover, they are advantageous in processing because of their low abrasivity and physiological harmlessness.[5]

The green dwarf coconut occupies an estimated area of 65,000 hectares in Brazil and has been responsible for the increase of husk generation. Among the alternatives researched for the productive use of this residue, its use as composites reinforcement stands out. When thermoplastic polymers are mixed with natural fibers, interesting composites can be prepared with obvious economical and environmental advantages. Coir is an important lignocellulosic fiber obtained from coconut trees, which grow extensively in tropical countries. It is used for making a wide variety of floor furnishing materials, yarn, rope, etc.[6] However, these traditional coir products consume only a small percentage of the potential total world production of coconut husk. The high increase in the consumption of green coconuts and the industrialization of the processing of coconut water has increased the generation of green coconut trash, which corresponds to around 85% of the weight of the fruit. Hence, research and development efforts have been underway to find new use areas for coir.

The polymer matrix in a fiber-reinforced composite material binds the fibers together, transfers applied load to these fibers, and protects them from harmful environmental effects. The properties of available polymers, either thermosets or thermoplastics, make them ideal matrices.[7]

Polypropylene was chosen as the matrix material in this study because of its good balance of property range, low processing temperature, low price, and good thermal stability. Taking these considerations as granted, this study validates the proposal of using green coconut residues for the generation of new products. Thus, the aim of this work is to investigate rheological and mechanical properties of green coconut fiber–reinforced polypropylene composites with special reference to the effect of fiber granulometry and shear rate on mechanical and rheological properties.

EXPERIMENTAL SECTION

Materials

Polypropylene (PP1074), with specific mass of $0.9\,g/cm^3$ and melt flow index (MFI) (230°C) of $20\,g/10\,min$, was supplied by ExxonMobil

Chemical (USA). Green coconut fibers (GCF) of fraction X (0.500–0.250 mm) and fraction Y (0.840–0.500 mm) was supplied by Embrapa Agroindustria Tropical (Fortaleza, Brazil).

Methods

Fiber Treatment

Green coconut fibers (GCF) were air-dried for 48 h at 80°C and for at least 1 h at 100°C until a moisture content of around 4% was reached and stabilized. This treatment was employed immediately before preparing PP-GCF composites.

Preparation of PP-GCF Composites

Polypropylene composites containing 20% of green coconut fiber (GCF), of either fraction X or Y, were prepared using a twin-screw extruder at 170°C on all zones, except the feeding zone (140°C). The screw speeds were set at 60, 80, 100, or 140 rpm. The extrudate was pelletized and then air-dried for 1 h at 100°C. Afterwards, 3 mm thick plaques were compression molded under 7 MPa at 180°C for 2 min, then cooled for 5 min under the same pressure before demolding. The resultant plaque was cut into test samples and polished for flexural tests, according to ISO 178. For rheological testing, disks of around 5 cm diameter were first cut out of the plaques, their volumes being controlled so as to exceed the test cavity volume by 5% (i.e., 3.15 cm^3). Table I shows all the samples prepared.

Rheological Analysis

PP-GCF composites are expected to exhibit a strong nonlinear viscoelastic character that needs special testing techniques to be studied. Close cavity torsional rheometers allow for large amplitude harmonic testing to be performed on a variety of stiff and heterogeneous materials that cannot be conveniently tested with standard open-gap rheometers. Because direct strain-stress proportionality is lost in the nonlinear domain, appropriate modifications were brought to a commercial torsional rheometer, initially developed for rubber materials, i.e., the Rubber Process Analyser RPA® (Alpha Technologies), in order to capture strain and torque signals and to treat them with a suitable calculation technique.[8,9]

All tests were perfomed according to a protocol that consists in testing two samples per compound, tests A and B, using a strain sweep

Table I. PP-GCF (80/20) composites

Sample code	Shear rate (RPM)
Neat PP	
P60	60
P80	80
P100	100
P120	120
P140	140
Fraction X	
60X	60
80X	80
100X	100
120X	120
140X	140
Fraction Y	
60Y	60
80Y	80
100Y	100
120Y	120
140Y	140

procedure, as shown in Table II. The nonlinear behavior is consequently documented, as well as any strain history effects, and the quality of test samples is checked.

Such a strain sweep procedure consists of two subsequent tests with a resting time of 2 min between run 1 and run 2. Run 1 lasts approximately

Table II. Strain sweep procedure

Test A			Test B		
Run 1 Strain (°)		Run 2 Strain (°)	Run 1 Strain (°)		Run 2 Strain (°)
0.5		0.6	0.6		0.5
0.9		1.4	1.4		0.9
2.3		4.3	4.3		2.3
7.0		12.0	12.0		7.0
18.0	2 min at rest	24.0	24.0	2 min at rest	18.0
30.0		34.5	34.5		30.0
39.0		43.0	43.0		39.0
47.0		51.0	51.0		47.0
55.0		60.0	60.0		55.0
64.0		68.0	68.0		64.0

9 min, after a preheating period of 5.5 min. Consequently, when run 2 starts, the test material has already been in the test cavity for around 16.5 min. Therefore, one might question the relative importance of the thermal stability of test materials and of the mechanism of strain history effects when they are observed. In fact, during the last step of run 1, the material is submitted to around 1000% of shear strain, then left at rest for 2 min. If the strained material fully recovers during this 2 min period, then run 2 starts with a strain modulus that is nearly equal to the initial low strain modulus as measured at the beginning of run 1. In such a case, no strain history effect is detected. Consequently, one would consider that there are strain history effects when run 1 and run 2 curves do not superimpose. The "homogeneity" of test samples can be checked when there are no differences between tests A and B. All RPA-FT tests were made at 180°C and a frequency of 0.5 Hz.

Flexural Modulus

Three point bending tests were carried out using an Instron universal testing machine according to ISO 178, with crosshead speed of 2 mm/min, span of 32 mm, and cell load of 1 kN. At least five samples for each composite of dimensions ($12.7 \times 65 \times 3$) mm were tested.

RESULTS AND DISCUSSION

Rheological Analysis

RPA strain sweep tests yield essentially complex modulus versus strain data that were fitted with the model showed by Equation (1):

$$G^*(\gamma) = G_f^* + \left(\frac{G_0^* - G_f^*}{1 + (A\gamma)^B} \right)$$

where G_0^* and G_f^* are the extrapolated complex modulus values at zero and infinite strain, respectively, A and B are fitting parameters, where A is the reverse of a critical strain that corresponds to $(G_0^* + G_f^*)/2$ and B is a parameter describing the strain sensitivity of the material, γ is the shear strain, and $G^*(\gamma)$ is the complex modulus calculated at each shear strain. G_f^* has relatively little physical meaning in the experimental context, but G_0^* can be considered as the "linear" modulus when a linear plateau is clearly observed.

Figure 1 shows complex modulus versus strain amplitude curves, as obtained for pure PP samples extruded at various screw speeds. As can be seen, the lack of homogeneity (incomplete melting) of certain samples

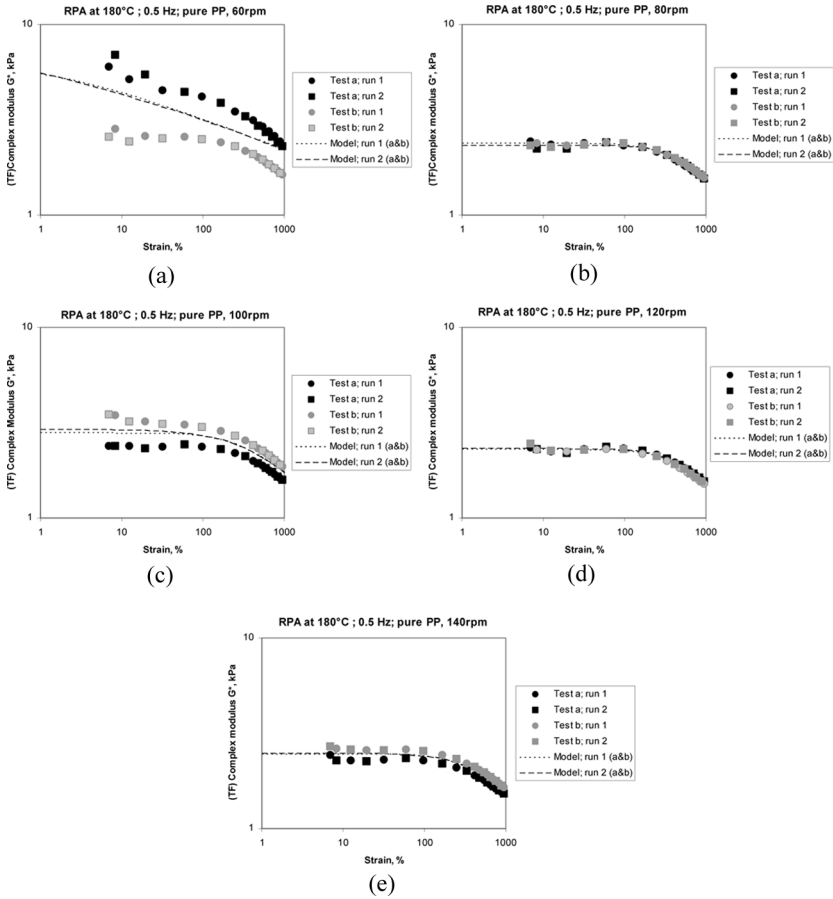

Figure 1. Pure polypropylene extruded at different screw speeds: a, 60 rpm; b, 80 rpm; c, 100 rpm; d, 120 rpm; e, 140 rpm.

(i.e., a and c, e to a lesser extent) is the main information provided by RPA tests on this series of samples. Indeed, a low strain plateau is generally observed at around 2.4 kPa without any effect of the screw speed. The linear viscoelastic behavior is observed up to around 100% strain.

Figure 2 compares G^* versus γ curves obtained on the two series of GCF-filled composites. No significant effect of screw speed is detected, but irrespective of the preparation conditions, clear strain history effects are observed, since run 2 data are systematicaly shifted downwards, when compared with run 1 data.

GCF consists of fibers of various lengths mixed with a powdery material, and it is believed that these two components come from different parts of the coconut. Fraction X has, in fact, a higher powder content

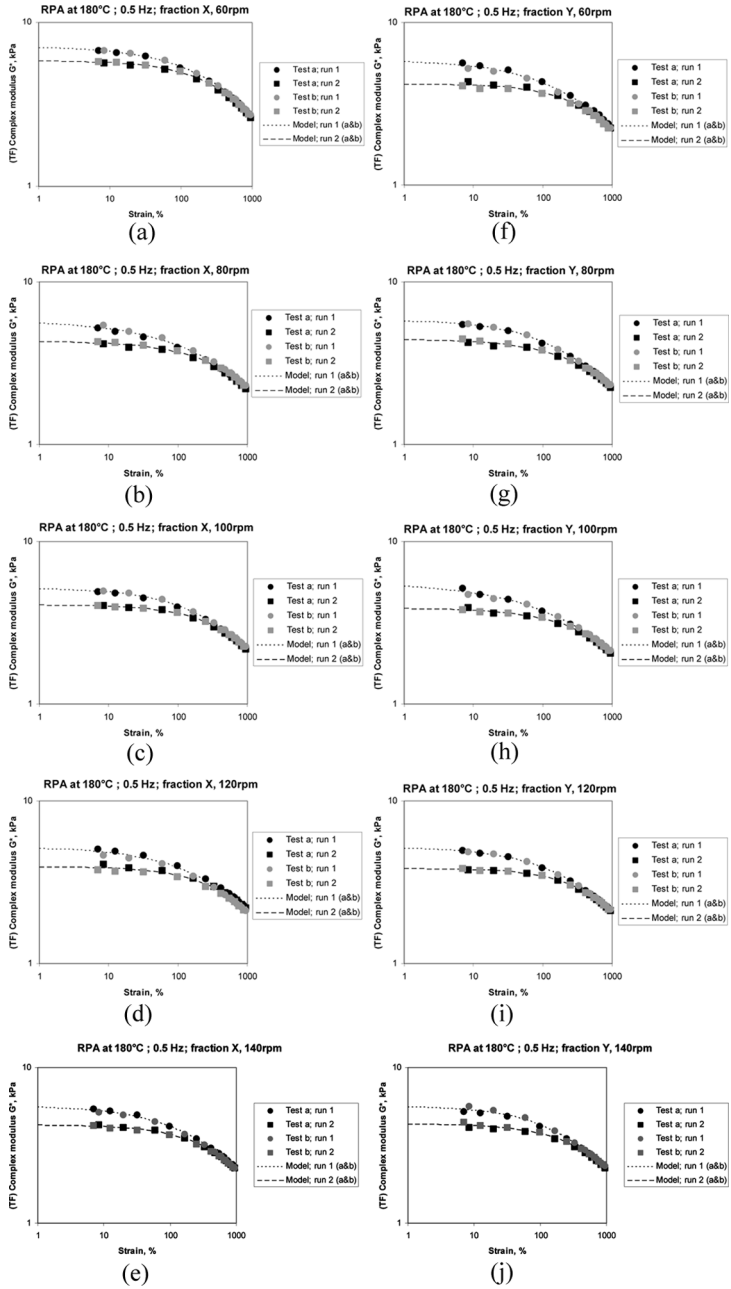

Figure 2. GCF composites of fraction X (a–e) and fraction Y (f–j) at different screw speeds: a, f: 60 rpm; b, g: 80 rpm; c, h: 100 rpm; d, i: 120 rpm; e, j: 140 rpm.

than fraction Y, and Figure 3, tests A and B, clearly demonstrates that the powdery fraction is responsible for the poor homogeneity of PP/GCF composites. This obviously suggests that a careful preparation procedure for GCF material is a fundamental condition for obtaining good quality composites.

As can be seen on the left-hand side graph of Figure 3, strong strain history effects are observed when fraction X is used in preparing the composites (compare run 1 and run 2 data), in addition to poor homogeneity (compare test A and test B). On the right side of Figure 3, almost no differences are seen between tests A and B (indicating good homogeneity), but the strain history effect is well observed (compare run 1 and run 2).

With complex polymer systems, strain history effects are usually associated with strain-induced modification of a "structure" assumed to exist in the unstrained material. In the case of GCF composites, such a structure can readily be assigned to a network of percolated fibers. By noting that thermal degradation could also give rise to a similar effect, the following test procedure was designed: first a two-run procedure is

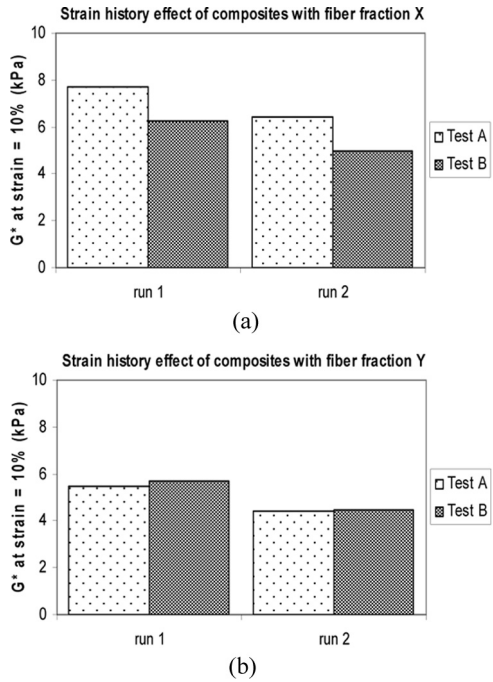

Figure 3. Strain history effect of composites containing fiber fraction X (a) and Y (b).

performed, but the test sample is maintained in the cavity for at least 5 min, then another two-run procedure is applied. Results obtained with this procedure are illustrated in Figure 4. As can be seen, no significant thermal degradation effects can be considered as the source of differences between run 1 and run 2. Indeed, results a and c confirm that sample inhomogeneity is the main reason for the non-superimposition of G^* versus γ curves. Small differences observed on results b, d, and e are not significant and only confirm that the nonlinear behavior of pure PP remain unchanged whatever the exposition time at the test temperature (180°C).

Figure 5 shows G^* of composites with fiber fraction X (a) and fiber fraction Y (b) at 10% of strain. The values obtained at 37 min are the results after a two-run procedure is performed. In the case of composites,

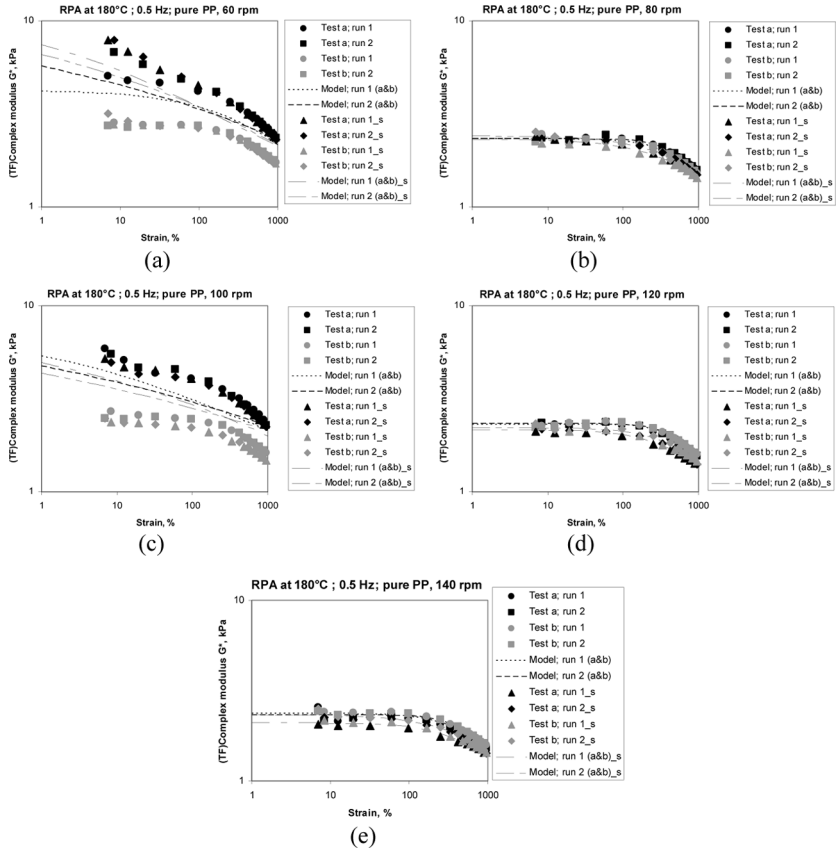

Figure 4. Pure PP at different screw speeds: a, 60 rpm; b, 80 rpm; c, 100 rpm; d, 120 rpm; e, 140 rpm.

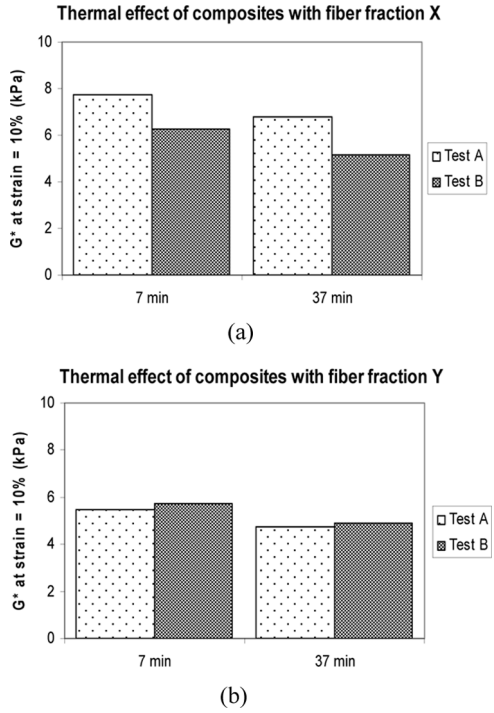

Figure 5. Thermal vs. strain effect in composites with fiber fraction X (a) and fraction Y (b).

some thermal effects are observed that are clearly related to the composition of GCF batches.

Indeed, in the left graph, results obtained on the composite prepared with fraction X (high powder content) show significant discrepancies when testing two samples (tests A and B), and comparing results obtained at 7 min with those obtained at 37 min demonstrates the temperature exposure effect. The right graph shows that testing two samples gives essentially the same results (tests A and B), but that extending the exposure time to test temperature induces a small decrease of G^* (10%).

Flexural Test

Figure 6 shows flexural modulus of pure PP (a) and composites made with fraction X (b) and fraction Y (c) as a function of the extruder screw speed. In general, flexural modulus of pure PP is around 1800 MPa. No significant effect of the shear rate was observed in the range studied.

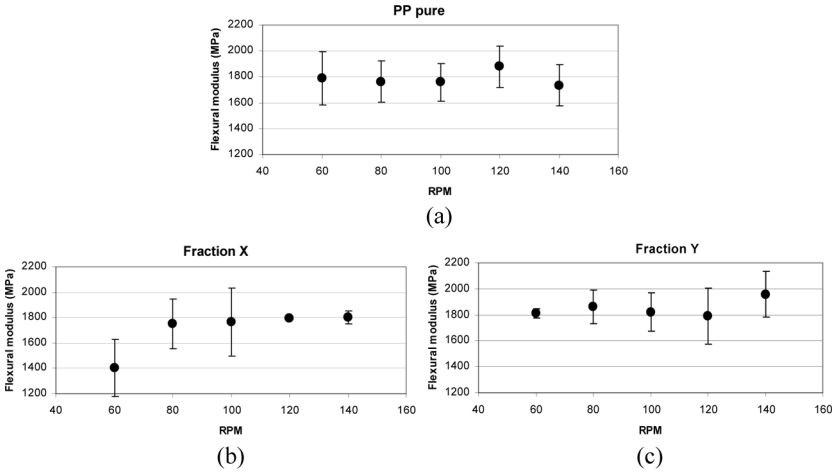

Figure 6. Flexural modulus of pure PP (a) and composites with fraction X (b) and fraction Y (c).

In the case of composites prepared with fraction Y, higher average values of flexural modulus than those of fraction X are observed (see Figure 6(b) and (c)). These results are in agreement with RPA tests, from which it was seen that composites made with fraction Y are more homogeneous than composites made with fraction X. The homogeneity of the composites is a key factor in obtaining materials with good mechanical properties.

The highest average values of flexural modulus were given by the composites extruded at 140 rpm, containing either fraction X or Y. Such a behavior indicates that when high rotation rates are used, better dispersion and wetting of the fibers by the molten polymer are achieved, and consequently higher modulus is obtained.

CONCLUSIONS

Rheological data of molten composites can be conveniently applied to study the homogeneity and effect of strain history of the composites.

The flexural modulus of green coconut fiber–reinforced polypropylene can be improved by using adequate fiber granulometry and preparation conditions, such as the extruder screw speed.

Considering the conditions used in this study, it is concluded that the best performance was achieved when using granulometry Y (0,840–0,500 mm) and 140 rpm for PP-green coconut fiber composites.

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