

Dynamic Mechanical Analysis of Recycled Polystyrene Composites Reinforced with Wood Flour

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Received 22 March 2011; accepted 5 August 2011

DOI 10.1002/app.36291

Published online 29 December 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: The dynamic mechanical properties of wood flour (WF) reinforced recycled expanded polystyrene (r-EPS) composites were investigated as a function of the WF content and coupling agent used. With the incorporation of WF into the polystyrene the storage modulus (E') and loss modulus (E'') were found to increase and the mechanical damping ($\tan \delta$) decreased. The use of coupling agents improved the storage modulus and reduced the damping peak values of the composites because of the improved WF/matrix interfacial adhesion. The height of

the damping peaks was found to be dependent on the content of maleic anhydride groups and the weight average molecular weight (M_w) of the coupling agent used. Finally, the experimental results were compared with the theoretical predictions for $\tan \delta$ and the storage modulus. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 125: 935–942, 2012

Key words: expanded polystyrene; wood flour; coupling agent; composites; dynamic mechanical analysis

INTRODUCTION

Wood thermoplastic composites have received considerable attention from the materials industry in recent years. The growing commercial importance of these materials has expanded efforts to understand their structure-property relations and to explore new methodologies for their production.¹ Wood flour (WF) is an attractive reinforcement for thermoplastic polymers, mainly because of its low density, low cost, and highly specific properties.^{1,2} It is renewable, biodegradable, and nonabrasive during processing, and improves the stiffness and strength of thermoplastics.^{3–5}

The utilization of recycled plastic for the manufacture of thermoplastic composites has been studied by several authors.^{5–10} These composites are more cost effective than other similar materials. They can also be fabricated using recycled wood fiber, such as that from sawdust (WF) generated in sawmills, providing an additional use for recycled wood fiber and thus further reducing waste disposed to landfills.^{5–7}

Since fiber-reinforced thermoplastic materials are subjected to various types of dynamic stress during service, studies on the viscoelastic properties of these materials are of great importance.^{11,12} Dynamic mechanical analysis (DMA) is an effective method to

study the relaxations in polymers and thereby the behavior of the materials under various conditions of stress, temperature, and phase composition of fiber composites and its role in determining the mechanical properties.¹¹ The temperature-dependent dynamic parameters, such as storage modulus, loss modulus, and mechanical damping, provide an insight into the level of interaction between the polymer matrix and fiber reinforcement.¹³ Several studies have been carried out on the DMA properties of natural fiber-reinforced polymer composites to investigate the effect of the addition of fillers, impact modifiers, coupling agents, and so on, on the fiber matrix interface.^{14–18}

Joseph et al.¹⁹ studied the DMA of short sisal fiber-reinforced polypropylene (PP) composites. They reported that the storage and loss moduli of the composites increase with fiber loading, while the $\tan \delta$ value decreases. Amash and Zugenmaier²⁰ reported on the effectiveness of cellulose fiber in improving the stiffness and reducing the damping of PP/cellulose composites. Hristov and Vasileva²¹ studied the dynamic mechanical and thermal properties of PP/wood fiber composites. They observed that the storage modulus values for compatibilized composites were higher than those for uncompatibilized composites, indicating improved adhesion and interaction between the PP matrix and wood fibers. Manikandan Nair et al.²² evaluated the effects of fiber loading, fiber length, fiber orientation, and fiber modification on the dynamic mechanical properties of polystyrene (PS)/sisal fiber composites. The

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authors reported that the addition of sisal fiber considerably increased the storage modulus; the T_g values for the composites were lower than those for the unreinforced PS and the treated fiber composites showed better properties than the untreated-fiber composites.

In fact as shown above, a considerable amount of studies related with the improved of composite dynamic mechanical properties with the usage of coupling agents has been reported on the literature. However, the literature lacks studies on the characterization of the effect of coupling agents with different molecular weight and different content of maleic anhydride groups on the composite dynamic mechanical properties. In this investigation, the dynamic mechanical properties of recycled PS composites reinforced with WF were examined particularly in relation to the WF loading and use of three different coupling agents with different molecular weights and different contents of maleic anhydride groups, to contribute with the understanding the effect caused by the addition of these coupling agents. The fiber/matrix interactions were evaluated based on the damping peak height and the adhesion factor values calculated. Scanning electron microscopy (SEM) was used to analyze the macroscopic properties measured by DMA.

EXPERIMENTAL

Materials

The expanded PS waste was obtained from a sorting unit called Associação de Recicladores Serrano, Caxias do Sul, Brazil and it had a melt flow index of 20 g/10 min (200°C/5 kg, ASTM D1238). The recycled expanded polystyrene (r-EPS) was a density of 1.07 ± 0.01 g/cm³ (ASTM D792). WF of *Pinus elliottii* was obtained from Madarco, Caxias do Sul, Brazil, with a particle size distribution of 53–105 μ m (ASTM D1921) and density of 1.41 ± 0.01 g/cm³ (based ASTM D297). The poly(styrene-co-maleic anhydride) oligomers, SMA, used as coupling agents were supplied by Sartomer Company, Exton. Three different types of SMA were used, as shown in Table I. The amount of coupling agent incorporated was fixed at 2 wt %.

Sample preparation

The methodology for recycling expanded PS waste has been described in a previous article.¹⁰ The WF was dried in an oven at 105°C for 24 h, before being used in the composite formulations. Samples with 10, 20, 30, and 40 wt % of WF with and without 2 wt % of SMA were processed in a corotating twin-screw extruder COR-20-32-LAB MH Equipment-Bra-

TABLE I
Physical Properties of the Coupling Agents

| Oligomer | Chemical structure | Acid number (mg KOH/g) | M_w (g/mol) |
|----------|--------------------|------------------------|---------------|
| SMA 2000 | | 355 | 7500 |
| SMA 3000 | | 285 | 9500 |
| SMA EF40 | | 215 | 10,500 |

zil at 200 rpm, with L/D ratio of 32. The nine-barrel temperature zones were controlled at between 160 and 190°C. Specimens for dynamic mechanical tests were injection molded at a barrel temperature of 180°C, mold temperature of $40 \pm 2^\circ\text{C}$, with screw speed of 100 rpm and pressure of 650 bar. The composites were denoted by the symbols U100, S202, M202, E202, and so on. In these notations, the first letter denotes the nature of the oligomer used as the coupling agent, U – untreated, S – SMA2000, M – SMA3000, and E – SMAEF40. The first and second digits together denote the weight percentage of the WF and the third digit denotes the amount of coupling agent used.

Dynamic mechanical analysis

Specimens of recycled PS and composites with and without coupling agents, with dimensions of $50 \times 13 \times 3.5$ mm, were subjected to dynamic mechanical testing using an Anton Paar Physica MCR 101 oscillatory rheometer operating in DMA mode. The measurements were carried out in the torsion mode of the equipment and the corresponding viscoelastic properties were determined as a function of temperature. The temperature range used in the experiment was 30–140°C, with a heating rate of 3°C/min, under nitrogen flow. The samples were scanned at a fixed frequency of 1 Hz, with a dynamic strain of 0.1%.

Scanning electron microscopy (SEM)

Studies on the morphology of the composites were carried out using a SHIMADZU Superscan SS-550, scanning electron microscope, with acceleration voltage of 15 kV. The cryo-fractured surface of the specimens was sputter-coated with gold before analysis to eliminate electron charging.

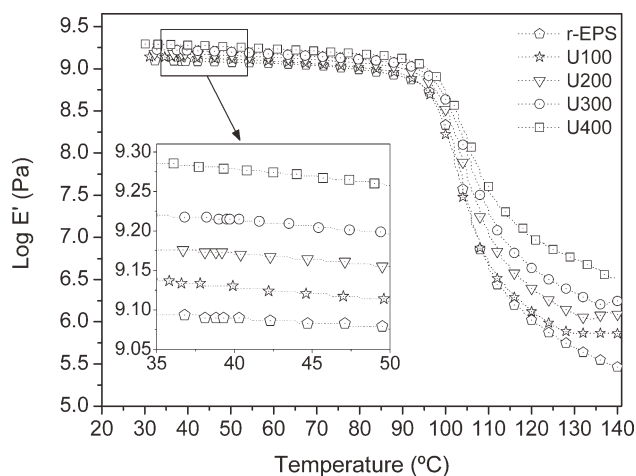


Figure 1 Effect of WF loading with temperature on the storage modulus of r-EPS/WF composites.

RESULTS AND DISCUSSION

Effect of WF loading

Storage modulus (E')

The variation in the storage modulus of the recycled PS/WF composites as a function of temperature can be observed in Figure 1. There is an increase in the modulus of the PS matrix with the incorporation of WF over the entire region. This may be due to the increase in the stiffness of the matrix due to the reinforcement effect imparted by the fibers, which allowed a greater degree of stress transfer at the interface.^{23,24} However, the small wood particles increase the contact surface area with the polymer matrix, which might provide more interaction between r-EPS matrix and WF leading to better performance in modulus with the WF loading.¹⁹ As the temperature increases, E' decreases and thus there is a sharp decline in the E' value in the glass transition region. This behaviour can be attributed to the increase in the molecular mobility of the polymer chains above T_g .^{23,24} The drop in the modulus on passing through the T_g is lower for the reinforced composites compared with the PS matrix. That is, the difference between the moduli in the glassy state and rubbery state is smaller in the composites than in the neat PS, which clearly indicates the reinforcing effect of the WF.

The influence of the addition of WF to the PS matrix can be better understood by studying the relative (normalized) storage modulus (E'_c/E'_m),¹² where E'_c and E'_m are the storage moduli of the composite and the matrix, respectively, with temperature at different WF loadings. From Figure 2 it is clear that the storage modulus of the composites increases with WF loading and the relative change with loading is more pronounced at high temperatures (rubbery region) than low temperatures (glassy region). As the temperature increases from 30°C in the glassy

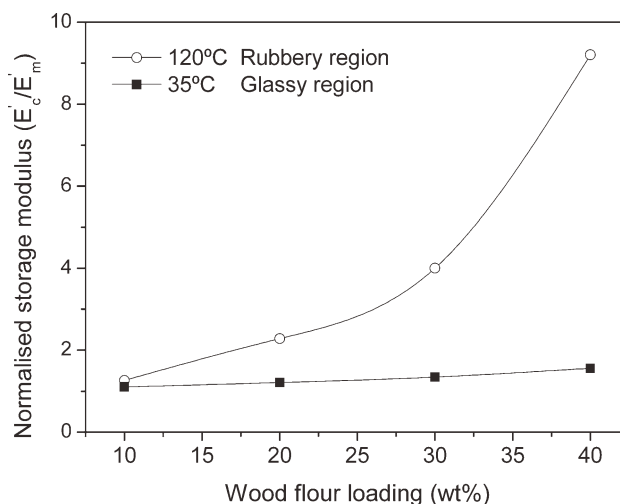


Figure 2 Variation in normalized storage modulus values with temperature and different WF loadings.

state to 120°C in the rubbery state the presence of small particles of WF restrict the flow of the polymer matrix and the restriction increases with increasing the fiber loading,¹² as showed in Figure 2.

The effectiveness of fillers on the storage modulus of the composites can be represented by a coefficient C given in the following equation^{11,12}:

$$C = \frac{(E'_g/E'_r)_{\text{comp}}}{(E'_g/E'_r)_{\text{resin}}} \quad (1)$$

where E'_g and E'_r are the storage modulus values in the glassy and rubbery regions, respectively. The E' values measured at 50 and 120°C were employed as the E'_g and E'_r , respectively. A high C value indicates that the filler is less effective. The values obtained for different composites are given in Table II. It can be observed that for composites with the WF loading the C value decreases and the lowest values were obtained for the composites with 40 wt % of WF. Hence, the WF content with maximum effectiveness is 40 wt %, where the maximum stress transfer between the filler and matrix takes place.

Loss modulus (E'')

The loss modulus is a measure of the energy dissipated or lost as heat per cycle under deformation

TABLE II
Variation of C with Wood Flour Loading

| Sample | C |
|--------|------|
| U100 | 0.99 |
| U200 | 0.95 |
| U300 | 0.92 |
| U400 | 0.88 |

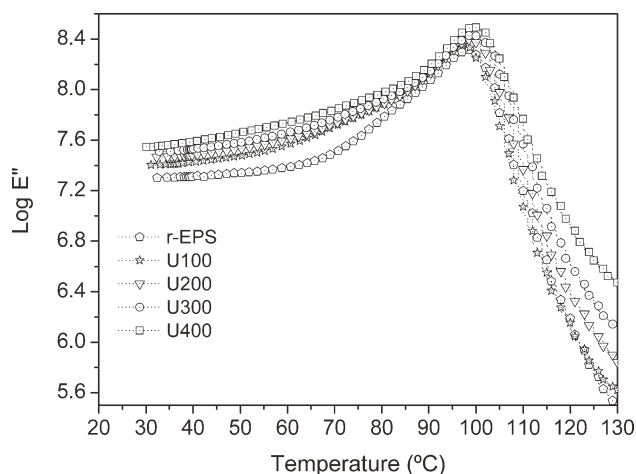


Figure 3 Variation in loss modulus values with temperature for composites with different WF loadings.

and measures the viscous response of the material. Figure 3 shows the variation in E'' as a function of the temperature for the different composite systems. The incorporation of WF caused broadening of the loss modulus peak when compared to the polymer matrix. The peak broadening is an indication of a change in relaxation mechanisms. This can be attributed to the increase in relaxation time, caused by the larger concentration of WF and may be due some WF agglomerates originated from the formation of hydrogen bonds between the wood particles due the presence of free hydroxyl groups in wood surface,^{7,21} increasing material heterogeneity.^{11,18}

The values for the loss modulus of the composites increased with increasing WF content. The higher value is due to the increase in internal friction, which enhances the dissipation of energy.^{24–26} Additionally, the presence of WF particles with a high modulus value reduces the flexibility of the composite material by introducing constraints on the segmental mobility of the polymeric molecules^{13,24} at the temperatures studied.

Mechanical damping ($\tan \delta$)

Mechanical damping is an important parameter in relation to the dynamic behavior of fiber-reinforced composites. The damping properties of the material give the balance between the elastic phase and viscous phase in a polymeric structure. In composites, damping is influenced by the incorporation of fibers. The mechanical damping values for the composites are lower than those for the neat PS, as seen in Figure 4. It was found that as the amount of WF in the composite increases, the $\tan \delta$ value decreases. The incorporation of WF reduces the height of the $\tan \delta$ peak. As the WF content increases, the system becomes more rigid. As a result, the restriction of

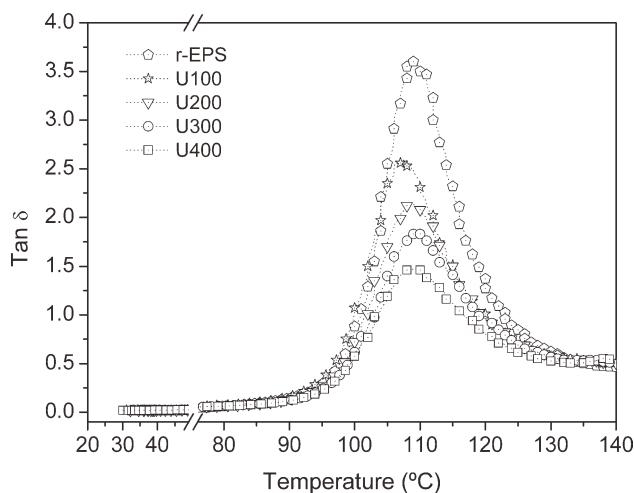


Figure 4 Effect of temperature on the $\tan \delta$ value for composites with different WF loading.

the polymer chains increases. The lowering of the damping curves on the addition of WF, compared with the neat PS, is also due to the decrease in the volume fraction of the matrix.^{11,12} This affects the mechanical properties and, in turn, the damping.

The T_g values for the neat matrix and for the composites with WF loading are presented in Table III. The T_g values obtained from the loss modulus peaks were lower than those obtained from the damping peaks. The T_g values for the composites were found to be slightly shifted to higher temperatures with the incorporation of WF, which may be attributed to the restriction imposed by the fibers on the molecular motion in the matrix,²² which increases with WF loading.

Effect of the utilization of coupling agents

Storage modulus

The dynamic mechanical properties of composites are influenced by an improvement in the fiber/matrix adhesion. The effect of the use of coupling agents on the storage modulus of recycled PS/WF composites at 20 wt % of WF loading is shown in Figure 5.

TABLE III
Effect of Wood Flour Loading on T_g Values of r-EPS/WF Composites

| Sample | T_g (°C) | |
|--------|------------------|--------------------------|
| | From E''_{Max} | From $\tan \delta_{Max}$ |
| r-EPS | 98.2 | 109.3 |
| U100 | 97.2 | 107.6 |
| U200 | 98.5 | 108.8 |
| U300 | 98.9 | 109.2 |
| U400 | 99.3 | 109.7 |

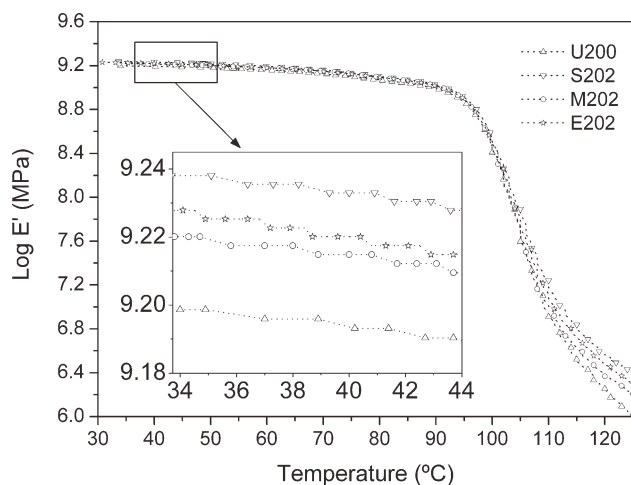


Figure 5 Effect of coupling agents on storage modulus of r-EP/S/WF composites.

The storage modulus values for the treated composites are higher than the untreated composites. The improvement in the storage modulus observed when SMA was used is due to the better interfacial adhesion between the WF and the PS matrix. The SMA increases the fiber matrix adhesion, causing reduced molecular mobility in the interfacial region. The maximum improvement was observed with the utilization of SMA2000. The sufficiently lower M_w of SMA2000 allows better diffusion in the polymer matrix, which indicates easier entanglement with the PS matrix.^{17,27} Furthermore, the sufficient number of maleic anhydride groups (higher acid number) attached to the PS chains causes strong interfacial interaction, probably due to the formation of chemical bonds between the maleic anhydride groups and the hydroxyl groups of the WF, which could result in an increase in E' because improve compatibility between WF and polymer matrix, as can be seen in Figure 5. A hypothetical model of the interface of the r-EP/S/SMA/WF composites is shown in Figure 6.

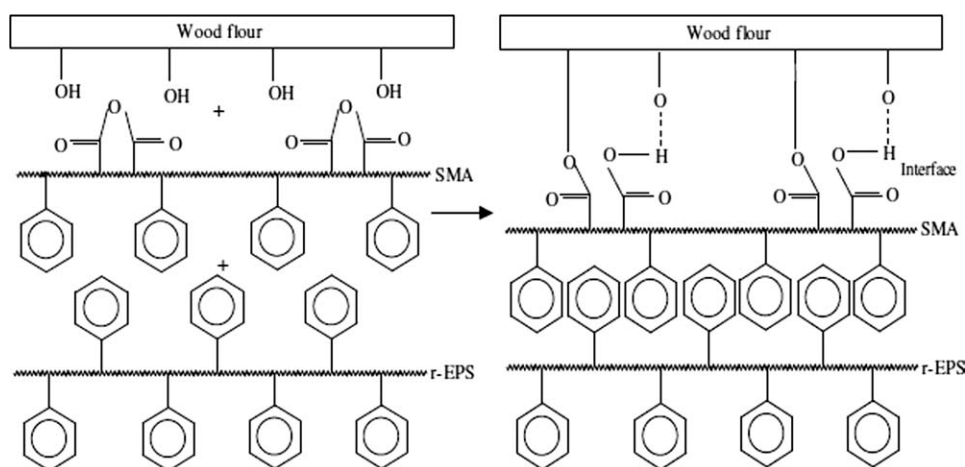


Figure 6 A hypothetical model of the interface of r-EP/SMA/WF composites.

Loss modulus

Figure 7 shows the effect of three different types of SMA on the loss modulus of the composites with 20 wt % of WF. The composite S202 presented the highest value of loss modulus throughout the temperature range studied. This composite was treated with SMA2000 that have more maleic anhydride groups than two others oligomers studied. The loss modulus is a measure the viscous response of the material, thus the sufficient number of maleic anhydride groups in SMA 2000 can lead to a formation of strong chemical bonds between de coupling agent and the WF and increasing the viscous response of the composite.

However, the composite E202 shows similar behavior that composite S202, however this composite was treated with SMA EF40 oligomer that have the lowest content of maleic anhydride groups but has the longest PS chain and in turn the highest molecular weight. In this composite, the longest PS chain of the coupling agent may be resulting in more entanglement with the PS matrix increasing the viscosity of the composite and resulting in a increasing of the loss modulus when compared to untreated composite. The loss modulus of the composites treated with different types of SMA was probably dependent on the amount of maleic anhydride groups in the oligomer and dependent of the size of the PS chain, and in consequence, dependent of the molecular weight of the coupling agent.

Mechanical damping

The variation in $\tan \delta$ for neat PS and composites with 20 wt % of WF with and without coupling agents as a function of temperature is represented in Figure 8.

As can be seen in Figure 8, the $\tan \delta$ peaks for the treated composites were lower than the neat PS and untreated composite. The lowest $\tan \delta$ peak height was observed for the composite treated with SMA2000, according to Table IV.

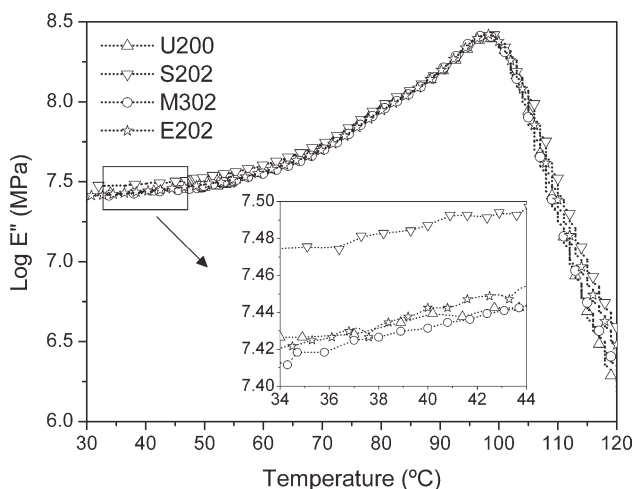


Figure 7 Variation in loss modulus values with temperature for composites with different coupling agents.

The energy dissipation will occur in the polymer matrix at the interface and a strong interface is characterized by less energy dissipation.²⁴ Thus, the $\tan \delta$ peak of the untreated composite was lower in comparison to the neat PS and higher when compared with the treated composites. This behavior demonstrates that a composite material with poor interfacial bonding between the fibers and matrix will tend to dissipate more energy, showing a higher damping peak in comparison to material with a strongly bonded interface.¹³ This fact is substantiated by the SEM micrographs in Figure 9. Figure 9(a) showed the composite with 20 wt % of WF without coupling agent (U200) that indicates the presence of pulled-out areas and bigger gaps between the WF and matrix, which is evidence of weak interfacial adhesion at the interface.²⁸ The SEM micrograph of the SMA2000-treated composite with 20 wt % of WF (S202) shown in Figure 9(b) clearly shows the strong bonding and good wetting between the WF and PS matrix.

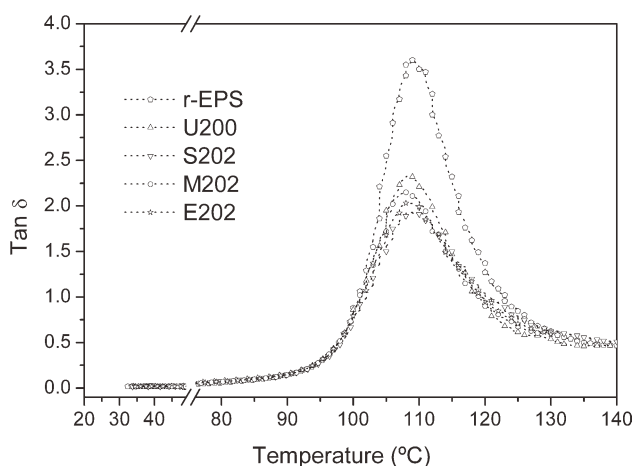


Figure 8 Effect of coupling agents on $\tan \delta$ values for r-EPS/WF composites.

TABLE IV
Values for Peak Height and Adhesion Factor for r-EPS/WF Composites

| Sample | Peak height | Adhesion factor |
|--------|-------------|-----------------|
| r-EPS | 3.07 | – |
| U200 | 1.92 | –0.246 |
| S202 | 1.45 | –0.383 |
| M202 | 1.74 | –0.287 |
| E202 | 1.59 | –0.330 |

Through DMA, the adhesion factor, A , proposed by Kubát et al.,²⁹ can be calculated to investigate the effects of different surface treatments on the adhesion between filler and matrix. The adhesion factor can be expressed in terms of the relative damping of the composite and the polymer matrix and the volume fraction of the filler as follows:

$$A = [(\tan \delta_c - \tan \delta_m)/(1 - V_f)] - 1 \quad (2)$$

where the subscripts c and m denote composite and matrix, respectively, and V_f is the corresponding volume fraction of the filler. At high levels of

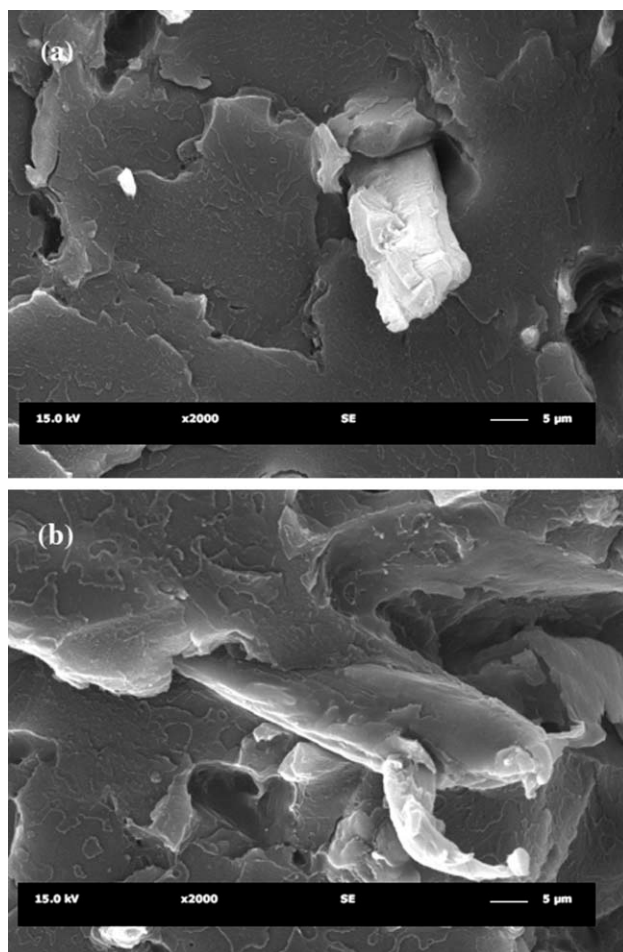


Figure 9 (a) SEM micrograph of untreated sample at 20 wt % of WF. (b) SEM micrograph of SMA-treated sample at 20 wt % of WF.

interface adhesion, the molecular mobility surrounding the filler is reduced, and this reduces the $\tan \delta_c$ values and consequently the A values. Thus, a low value for the adhesion factor A suggests improved interactions at the matrix-filler interface.^{29,30}

The adhesion factor values for the treated and untreated composites calculated at a temperature of 110°C are shown in Table IV. These values show that independent of the type of SMA used in the composites and its maleic anhydride content, in terms of the adhesion factor, all formulations seem to have improved matrix-WF adhesion when compared with the untreated composite. The lowest adhesion factor value was obtained for the composite treated with SMA2000, in agreement with the higher storage modulus obtained in the previous section. It was found that the improvement in the dynamic mechanical properties of the composites using SMA as a coupling agent was dependent on the amount of maleic anhydride groups and the M_w of the SMA.

Theoretical modeling of viscoelastic properties

Theoretical modeling of storage modulus

The storage modulus of recycled PS/WF composites with varying WF content was calculated theoretically using three different equations. The simplest equation for the reinforcement of a material due to an inclusion is given by Einstein^{19,31}:

$$E_c = E_m(1 + 1.25V_f) \quad (3)$$

where E is the storage modulus, subscripts c and m represent the composite and matrix, respectively, and V_f is the volume fraction of the filler. Another relation proposed by Einstein^{11,19,32} is:

$$E_c = E_m(1 + V_f) \quad (4)$$

The modification of Einstein's equation has been carried out by several authors such as Guth^{31,32}:

$$E_c = E_m(1 + 1.25V_f + 14.1V_f^2) \quad (5)$$

The experimental and theoretical storage modulus values for composites with different loadings at 35°C are shown in Figure 10. It can be observed that as the WF content increases all of the curves show an increasing trend. However, the experimental values show a greater agreement with the Einstein equations rather than the Guth equation. Therefore, the deviation from Einstein theoretical curves is more pronounced with the addition of WF. However, at 10 wt % of WF loading, the deviation is smaller for both of the Einstein models. Theoretical modeling thus indicates that the models are valid at lower WF loading, and deviations are highest with a WF content of 30

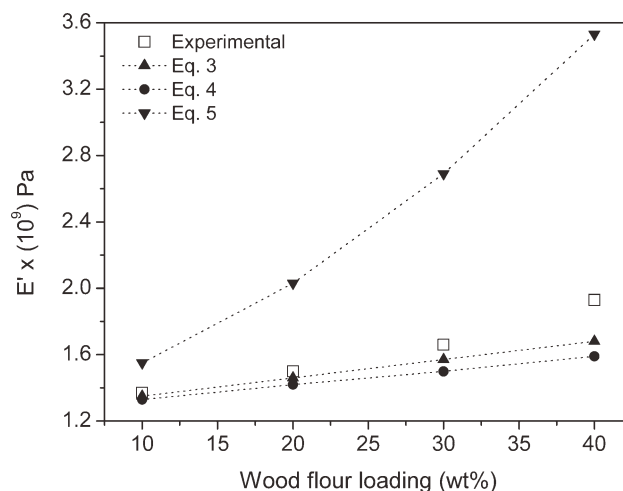


Figure 10 Experimental and theoretical storage modulus values for r-EPS/WF composites.

wt %. The deviations to theoretical models may be caused by more restraint at the interfaces imposed by the WF loading whereas the theoretical models did not consider the interactions at the interface.

Theoretical modeling of mechanical damping

Several equations for modeling mechanical damping have been discussed in the literature, for instance, the modified rule of mixtures equation proposed by Nielsen^{11,22}:

$$\tan \delta_c = \tan \delta_m(1 - V_f) \quad (6)$$

where the subscripts c and m represent the composite and matrix, respectively, and V_f is the volume fraction of the filler. This equation can be modified by introducing the stiffness term. The modification is based on the assumption that in the presence of fibers the matrix offers a stiffness equivalent to the minimum elastic modulus of the composites.^{19,32} Thus, the equation becomes:

$$\tan \delta_c = V_m(E_m/E_c) \tan \delta_m \quad (7)$$

where E_m and E_c are the storage modulus values for the matrix and the composite, respectively.

The plots of the experimental and theoretical values at the $\tan \delta$ maximum for the composites with different WF content are shown in Figure 11.

Rigid fillers and fibers usually decrease the damping, as can be seen from the theoretical and experimental equations. However, the experimental value is not in agreement with the theoretical prediction. The deviation is because both equations neglect the localized constraints imposed by the fibers on the matrix deformation¹⁹ and due to the heterogeneous nature of the composite.³² However, a specific interaction between the filler and the polymer matrix

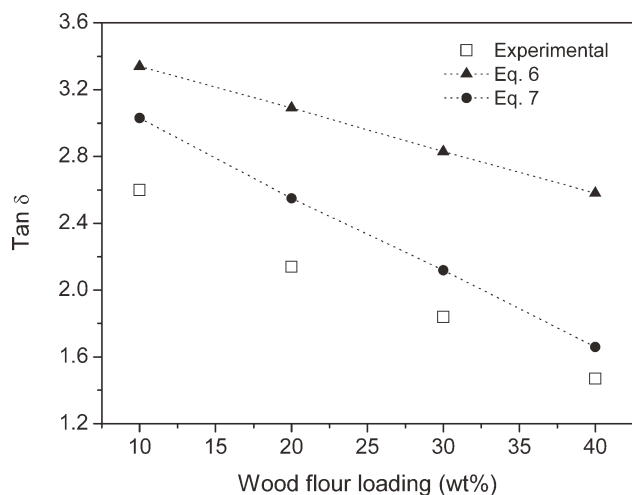


Figure 11 Experimental and theoretical $\tan \delta$ values for r-EPS/WF composites.

might be create an immobilized layer of polymer²⁵ around the fibers and caused a deviation on the theoretical modeling. The presence of weak agglomerations of WF and surface irregularities of the fiber may be the other reasons.¹²

CONCLUSIONS

The study on the dynamic mechanical behavior of WF reinforced PS composites revealed that the behavior of a composite is dependent on the WF loading and the nature of the interface. The incorporation of WF into the PS matrix gave rise to an increase of the storage modulus and loss modulus and a decrease in the damping values. The storage modulus showed a maximum for the composite with a WF content of 40 wt %, which showed lower C values. The loss modulus curve broadened as the WF increased when compared with the neat PS. The $\tan \delta$ value peak height is lower for the composite with a WF content of 40 wt % and the T_g values were slightly shifted to higher temperatures with the incorporation of WF. The use of coupling agents further improved the storage modulus of the composites because of the improved WF/matrix interfacial adhesion. SEM micrographs of the composites indicated strong bonding and good wetting between the WF and PS matrix. The lower adhesion factor values obtained for the treated composites demonstrate that in all formulations the matrix-WF adhesion was improved when compared with the untreated composite. The lowest adhesion factor value was obtained for the composite treated with SMA2000. The improvement in the dynamic mechanical properties of the composites obtained using SMA as a coupling agent was dependent on the amount of maleic anhydride groups and the M_w of the SMA. Finally, in the theoretical modeling of the storage modulus and

damping factor, the experimental value shows deviation due to the reinforcement effect imparted by the WF and the heterogeneous composite nature.

The authors would like to thank to Associação de Recicladores Serrano, Madarco S.A., and Sartomer Company for supplying materials.

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