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Processing-Properties Correlation for a Rubber Toughened Wood Plastic Composite

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Issues of environmental sustainability attract more and more attention to polymeric systems that can be considered environmentally friendly. A class of materials that meets the demand for a lower environmental impact are Wood Polymer Composites, or WPCs. These are thermoplastic polymers compounded with wood fibers or flour, usually coming from industrial or agricultural waste. With these materials, the greatest benefit is obtained by increasing the wood content, thus reducing the percentage of polymeric matrix. On the other hand, two of the main drawbacks coming from an increase in the wood content are brittleness and a significant reduction in processability. In this work, concentration will be on reducing brittleness by adding different amounts of a toughening agent (i.e., a thermoplastic elastomer) and on finding the optimal combination between processing conditions and elastomer content. This target will be obtained by comparing the tensile properties of all composites that are produced with different extrusion conditions with their apparent density.

1. Introduction

Wood Polymer Composites (WPC) are materials constituted by polymers mixed with wood particles or fibers and some additives. In these composites, the filler acts only partially as a reinforcement, its main function being to limit the amount of polymer that is used.^[1] There is interest to increase the quantity of wood filler inside WPCs, using the minimum amount of polymer to bind the wood fibers together, ^[2,3] because on one side the material becomes more environmentally friendly and on the other it allows to reduce the cost of the finished product.^[4,5]

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On the other hand, a high quantity of wood inside the composite introduces two important disadvantages: a significant increase of brittleness and a remarkable reduction in processability.^[6,7] The embrittlement effect is mainly due to presence of defects, voids and stress concentrations at the wood - polymer interface, since it is well known that there is poor interfacial adhesion between these two materials.[8,9] The reduction in processability, on the other hand, is mainly due to viscosity increase, higher porosity in the final product and higher risks of thermo-oxidative degradation.^[7,10] The viscosity increases is partly counterbalanced by an increase of wall slip due to the processing aids often present in the formulation.

A possible solution to reduce the embrittlement effect is blending WPCs with elastomers, ^[11] while from the

processing point of view, some improvement can be obtained by reducing the extruder screw speed or using a particular type of extrusion feeding, i.e., the so-called starve fed condition, in which the initial portion of the extruder is kept empty and the flow rate is determined only by the mass flow rate entering the hopper.^[12,13]

In this work, we will evaluate the effects of different percentages of a thermoplastic elastomer blended into a polypropylene based WPC filled with 50 wt% of wood. For each material, different extrusion conditions such as a starve versus flood fed feeding condition and screw speeds have been studied. The best combination of extrusion conditions and toughening agent content will be obtained by correlating the tensile properties with the apparent density of all materials.

2. Materials and Samples Preparation

Commercial 70 and 30 wt% polypropylene-based WPC (PP CO 68/BZ, PP 30 S) have been purchased from Plasticwood s.r.l. Mazzantica di Oppeano (VR, Italy) and a polypropylene-based thermoplastic vulcanizate (Santoprene 201-55) has been obtained from ExxonMobil, USA. A complete characterization of both WPCs can be found in Mazzanti and Mollica.^[14] The WPCs have been blended varying the percentage of the different materials, using a single screw extruder (P.R.T. Service & Innovation, S. Agostino (FE) Italy), in such a way that all blends are composed of 50 wt% wood fibers from white fir. All the compositions and the theoretical densities are reported in **Table 1**. The theoretical

Table 1. Compositions and theoretical density of the blend.

Name [wt%]	Wood [wt%]	PP [wt%]	Santoprene [wt%]	Theoretical density [g cm ⁻³]
0	50	50	0	0.984
10	50	40	10	0.990
20	50	30	20	0.997

densities have been calculated using the mixture rule on the basis of the materials density declared by the producers.

$$\frac{1}{\rho} = \frac{\phi_{PP}}{\rho_{PP}} + \frac{\phi_W}{\rho_W} + \frac{\phi_{TPV}}{\rho_{TPV}}$$
(1)

where ρ is the composite density, ρ_{PP} , ρ_W , and ρ_{TPV} are the densities of polypropylene, wood, and Santoprene respectively and φ_{PP} , φ_W , and φ_{TPV} are the mass fractions accordingly.

For each material, slabs of 2 mm thickness and 50 mm width have been extruded. The extruder, with a screw diameter of 50 mm and length over diameter ratio of 40, is equipped with a venting zone. Before extrusion, WPC pellets have been dried

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at 80°C for 12 h. A flat temperature profile of 190°C has been set along the barrel, while the die temperature has been fixed at 180°C. For each blend, two different extrusion conditions have been studied, i.e., starve versus flood fed feeding condition. In the case of flood fed feeding condition, the screw speed has been fixed at 25 RPM or 100 RPM, while in starved condition only the screw speed of 100 RPM has been used. In this situation the mass flow rate at the feeder has been chosen in such a way that it is similar to the mass flow rate relative to 25 RPM flood fed conditions.

3. Methods

Tensile tests have been performed at room temperature using a universal testing machine (INSTRON 4467, MA, USA). All tensile samples (ISO 37-2011, type 1-A) have been obtained by punch cutting from the extruded profiles. All tests have been performed using a 500 N load cell with a crosshead speed of 2 mm min⁻¹. Digital imaging correlation (DANTEC DYNAMICS, Denmark) has been used to measure strain. Six dog bone specimens have been tested for each composite obtained at any processing conditions.



Figure 1. Stress versus strain for all materials in a) starve fed, b) flood fed at 25 RPM, and c) 100 RPM feeding mode.

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Figure 2. SEM micrographs of samples with 0 and 20 wt% of Santoprene. The red arrows indicate the presence of pull-out, while the red square reveals the presence of polymers ligaments.



Figure 3. a) Strength and b) Stiffness as a function of the content of Santoprene for each feeding condition.

Directly from each tensile test sample, two 10 mm diameter disks have been punch cut to evaluate the apparent density. The mass and the dimensions of each sample have been measured using a precision scale (Mettler AE240 with a \pm 0.01 mg resolution) and a digital micrometer (Mitutoyo 293 with a \pm 2 μ m resolution), respectively.

The fracture morphology of samples has been investigated by Scanning Electron Microscopy (SEM) by using a FEG-SEM Mira3 by Tescan. The samples have been sputter coated with gold prior to analysis.

4. Results and Discussion

In **Figure 1**, typical stress versus strain curves for all materials in a) starved, b) flood fed at 25 RPM, and c) flood fed at 100 RPM processing conditions are shown. As expected, for any processing condition if the content of Santoprene increases, the deformation at break increases, while strength and stiffness decrease. In fact, the consequence of the presence of Santoprene is particularly evident at 20 wt%, in which the deformation at break reaches 3%. This macroscopic behavior has been confirmed also by fractographic analysis by SEM. **Figure 2** shows the fracture surfaces of samples at two different contents of Santoprene, namely 0 and 20 wt%. The specimen with the highest content of toughening

agent has exhibited a morphology characterized, also on a microscopic scale, by a much more ductile behavior as evidenced by the presence of polymer ligaments. In both cases wood fibers appear to be well dispersed and embedded in the polymer matrix, even though their interfacial adhesion has been found to be poor as confirmed by many instances of fiber pull-out and debonding phenomena.

The effect of the processing conditions is not negligible and is particularly evident when the WPCs are processed at the higher screw speed (Figure 1c). All the mechanical properties decrease, and the effect of toughening is strongly reduced. Comparing Figure 1a) with Figure 1b), fewer differences are appreciable. Strength and stiffness are comparable although slightly lower values have been observed in the case of starved feeding.

In **Figure 3** average and standard deviation of the a) strength and b) stiffness as a function of the Santoprene percentage are presented. For both strength and stiffness, the presence of the toughening agent at the maximum percentage makes the processing condition effects less significant. In the other cases the flood fed condition at the highest screw speed produces the worst results, the other two conditions being comparable.

A possible explanation of this behavior can be due to the higher likelihood of introducing defects such as voids and porosity in

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Figure 4. Apparent density versus the content of Santoprene for each feeding conditions.

the material that is processed at high screw speed and mass flow rate. To verify this hypothesis, in **Figure 4** the apparent density as a function of the content of Santoprene at all processing conditions is shown. All the apparent densities calculated from the tensile samples are lower than the theoretical ones (Table 1), but the lower values can be found in the samples that have been obtained in flood fed condition at the highest screw speed, while the values measured from the samples extruded in starve fed and the flood fed at 25 RPM conditions are comparable. These results are similar for any content of the toughening agent.

5. Conclusions

The presence of Santoprene has the effect of decreasing stiffness and strength at all percentages, while elongation at break increases only at 20 wt%. The effectiveness of the TPV at 10 wt% is not sufficient to increase the ductility of the composite at such high wood fiber content (50 wt%). Unfortunately, all the samples had a relatively low apparent density, and this hints toward the presence of defects in the samples that inevitably conditioned the results in terms of mechanical properties. Since the lowest apparent density appears for specimens that were processed in flood fed conditions and highest screw speed, one can speculate about the importance of processing these materials in starve fed conditions and at moderate screw speed.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

No data are available.

Keywords

mechanical properties, processing conditions, toughening agent, Wood Polymer Composite

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