MECHANICAL PROPERTIES OF HYBRID TALC-CELLULOSE
NANOFIBRIL-FILLED POLYPROPYLENE COMPOSITES

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Abstract

There is considerable interest in the automotive industry towards light-weighting vehicles through the application of new material technologies, and polymer matrix composites are of primary importance in meeting the goals of light-weighting. In addition, the application of renewable materials like wood and plant fibers is of interest in meeting sustainability goals and replacement of petroleum-derived feedstocks. This paper presents results of a study examining novel hybrid polypropylene composites using a combination of cellulose nanofibrils and talc for potential use in automobile applications. The composites were compounded using a laboratory scale co-rotating extrusion system, and test samples were prepared by injection molding tensile, flexure and impact test specimens. The tensile and flexural properties of the hybrid composites were determined using an Instron testing machine and the Izod impact strength was determined on Ceast Impact tester. Cellulose nanofibrils can replace a portion of the talc which produces PP composites with improved mechanical properties and lower density.

Background

The concept of utilizing hybrid or mixed fillers in polymer matrix composites has been explored in the research arena for several decades (Zhang et al. 2006; Yang et al. 2010; Gaciua et al. 2005; Mittal et al. 2015). Combinations of fillers can provide enhanced material property performance in polymer matrix composites. For example, filler combinations might be chosen to provide improved mechanical properties and reduced water permeability or enhanced electrical conductivity and lighter weight, etc. Addition of fillers can also typically reduce the cost of the resulting composite especially with the application of nature-derived fillers such as cellulose or clay.

The use of filled polymer composites is common in the automotive industry. Many manufacturers utilize glass-, cellulose-, or talc-filled polypropylene, glass-filled nylon to mention a few. However, manufacturers are looking at automotive composites for lighter weight materials to provide better fuel economy and lower carbon dioxide emissions (Stewart 2010). There is also a significant interest in green composites for automotive applications (Koronis et al. 2013). In many instances, the traditional filled polymer matrix composites are limited in material behavior were the combination of strength and reduced weight are needed. Thus the impetus for the work presented here.
Scope and Objectives

This paper reports on a feasibility study of novel hybrid composites using cellulose nanofibrils and talc in polypropylene for automobile applications. The goal of this project is to present a feasibility of the hybrid composites by comparisons of mechanical properties to polymer matrix composite materials currently used in industry. Several formulations of hybrid composites were prepared, compounded, and molded into ASTM standard specimens for material property characterization.

Materials and Methods

Cellulose nanofibers (CNF) used in this experiment were prepared by the Advanced Structures and Composites Center at the University of Maine. Nanocellulose suspensions were supplied from the University of Maine Process Development Center. The suspension was diluted from the original state 3.0 wt.% to 1.5 wt.% and processed using a pilot-scale spray dryer (VSD 6.3 from GEA Inc.) to produce dry particles. The average circular-equivalent diameter (CED) of the CNF was 18.2 µm which was measured by a laser diffraction method. The particle aspect ratio was around 0.772. The CNF particles were dried at 205°C for 4 hours prior to compounding.

The dry CNF was compounded with polypropylene blends using a lab-scale twin-screw extrusion system (CW Brabender Instruments Inc., Hackensack, NJ, USA). The control PP and its blends were supplied from RTP Co., Winona, Minnesota, USA. Table 1 shows the formulations of the control and hybrid composite samples. All compounds were dried again at 205°C for 4 hours and injected molded into flexural, tensile, or impact testing specimens using a lab mini-injector.

The testing was performed according to ASTM D790, D638, and D256 for flexural, tensile, and impact properties, respectively.

Table 1. The polypropylene (PP) talc (Tc)-cellulose nanofibrils (CNF) sample formulations.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>PP</th>
<th>CNF</th>
<th>Talc</th>
<th>Total filler</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-Tc 20%</td>
<td>80%</td>
<td>-</td>
<td>20%</td>
<td>20%</td>
</tr>
<tr>
<td>PP-Tc5%-CNF5%</td>
<td>90%</td>
<td>5%</td>
<td>5%</td>
<td>10%</td>
</tr>
<tr>
<td>PP-Tc10%-CNF5%</td>
<td>85%</td>
<td>5%</td>
<td>10%</td>
<td>15%</td>
</tr>
<tr>
<td>PP-Tc10%-CNF10%</td>
<td>80%</td>
<td>10%</td>
<td>10%</td>
<td>20%</td>
</tr>
<tr>
<td>PP-Tc5%-CNF10%</td>
<td>85%</td>
<td>10%</td>
<td>5%</td>
<td>15%</td>
</tr>
</tbody>
</table>
Results and Discussion

The mechanical properties of the samples are listed in Table 2. The replacement of the talc with CNF affected the material properties. The tensile and flexural properties increased with the addition of CNF while the impact strength was decreased. The specific gravity of the samples made with CNF also decreased. The increase in flexural and tensile is attributable to the better mechanical properties of CNF.

Table 2. Mechanical properties of the PP-Tc-CNF composite samples.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Tensile</th>
<th>Flexural</th>
<th>Izod impact</th>
<th>Specific gravity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Modulus</td>
<td>Strength</td>
<td>Modulus</td>
<td>Strength</td>
</tr>
<tr>
<td></td>
<td>GPa</td>
<td>MPa</td>
<td>GPa</td>
<td>MPa</td>
</tr>
<tr>
<td>PP</td>
<td>2.060</td>
<td>23.77</td>
<td>1.220</td>
<td>45.59</td>
</tr>
<tr>
<td>PP-Tc 20%</td>
<td>2.647</td>
<td>27.49</td>
<td>1.445</td>
<td>40.45</td>
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<tr>
<td>PP-Tc5%-CNF5%</td>
<td>2.754</td>
<td>33.96</td>
<td>1.321</td>
<td>52.25</td>
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<tr>
<td>PP-Tc10%-CNF5%</td>
<td>3.412</td>
<td>35.40</td>
<td>1.630</td>
<td>60.78</td>
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<tr>
<td>PP-Tc10%-CNF10%</td>
<td>3.39</td>
<td>35.27</td>
<td>1.656</td>
<td>60.36</td>
</tr>
<tr>
<td>PP-Tc5%-CNF10%</td>
<td>3.173</td>
<td>35.72</td>
<td>1.679</td>
<td>60.09</td>
</tr>
</tbody>
</table>

Figure 1 shows the differences of the various composite formulation tensile properties. The sample with a filler content of 15% (Sample PP-Tc10%-CNF5%) showed the highest strength and modulus and is comparable to the sample with 20% talc content. The partial replacement of talc with CNF either maintained the mechanical properties or provided improved tensile properties in some cases. The positive effect of CNF as a talc substitute can be found in flexural properties too (Figure 2). The comparison between two samples with the same filler content, 20%, showed that the CNF clearly increased the flexural modulus and strength as well up to 15% and 50% greater, respectively. The use of CNF can reduce the use of talc without a reduction in tensile and flexural properties.
Figure 1. Tensile properties of the (PP)-(Tc)-(CNF) samples.

Figure 2. Flexural properties of the (PP)-(Tc)-(CNF) samples.
From the Izod impact testing, a strength reduction was observed from the hybrid composite samples. It is challenging to explain the degradation since the mechanical properties of CNF are not supposed to be inferior to talc. The decrease could be related to the compounding methods. The hybrid composite samples were re-compounded to dilute the concentration of CNF which was first master-batched with pure PP, while the composite sample with 20% talc (PP-Tc20%) was processed only once. The master-batching of CNF and pure PP was conducted to assure the maximum effect of coupling between PP and CNF using maleic anhydride modified PP (MAPP) coupling agent. During the multiple processing steps at the temperature around 200°C, CNF can be thermally degraded and may result in impact strength reduction. The reduction can be more noticeable in impact testing than tensile/flexural testing since impact properties are more sensitive to the formulations and materials. Especially in highly-filled thermoplastics, the decreases of impact strength have been continuously reported (Yang et al. 2011).

Figure 3. Izod notched-impact properties of the polypropylene (PP) talc (Tc)- cellulose nanofibril (CNF) samples.
The specific gravity of talc is reported in the range of 2.5 to 2.8 and that is higher than CNF of which specific gravity is about 1.5. It is reasonable to form a hypothesis that talc is heavier than CNF. By replacing talc with CNF, however, the specific gravity was not significantly decreased as Figure 4 shows. It may be that the sample specific gravity is closely related to the fillers’ bulk density and the degree of packing during the compounding and subsequent injection molding.

![Figure 4. Specific gravity of the polypropylene (PP) talc (Tc)- cellulose nanofibrils (CNF) samples.](image)

Overall, the replacement of talc with CNF significantly improved mechanical properties except the impact strength. The increases in mechanical are up to 50% by only replacing 50% of talc portion.

**Conclusions and Future Work**

This study reported on the feasibility of producing novel hybrid composites using cellulose nanofibrils and talc in polypropylene for potential use in automobile applications. The goal of the project was to explore how the manufacture of hybrid composites influenced the mechanical properties of polymer matrix composite materials currently used in industry. It was shown that cellulose nanofibrils can replace a portion of the talc which produces PP composites with improved tensile and flexural properties and lower density. Future work will explore additional compounding studies to optimize composite formulations with enhanced mechanical properties, and lighter weight.
Acknowledgments

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Bibliography


