Usability of Pine Sawdust and Cotton Together as Filler in Recycled Polypropylene Composites

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Keywords
Pine Sawdust
Cotton
Recycled Polypropylene
Physical Properties
Polymer Composites

Abstract:
The use of natural fillers in polymers is a common method used to improve the properties of polymers. Natural fillers can be of plant or animal origin and provide several advantages over polymers. Although natural fillers are mostly used to reduce costs, they also affect other thermal, mechanical and physical properties. The use of natural fillers in polymers is a way to develop a sustainable and environmentally friendly material. Natural fillers are derived from renewable resources and their production requires less energy and resources than the production of synthetic fillers. Wood sawdust is a waste material that occurs extensively in the wood processing industry. Its easy-to-apply nature makes it an ideal choice for a variety of applications. In this study, used pine wood sawdust was used as filler in the composite. Along with sawdust, waste cotton was also added to the composite. Recycled polypropylene was preferred as the matrix material. Composite samples were prepared with extrusion and injection molding methods. Melt flow rate (MFI), density, and Izod impact strength tests of the samples were performed. Scanning electron microscopy (SEM) images taken from the broken surfaces were analyzed. The density values increase with increasing filler ratio. The lowest density value between composites observed in 10% cotton and 10% sawdust filled sample with 0.883 g/cm3. MFR results and impact properties of the composite samples were decreased. 10% cotton and 10% sawdust-filled composite shows optimal results between composites. As a result of the study, it was evaluated that pine sawdust and waste cotton would be used in polypropylene-based composite applications.

1. Introduction

The use of organic fillers instead of inorganic fillers in polymer-based composites has become widespread with environmental awareness, ecological balance, and new research. Features such as being renewable, low-cost, low-density, and nontoxic are among the main reasons for the use of natural fillers in polymer composites. Natural filler-filled polymeric composites are also called green composites [1]. For green composite production, the matrix can be thermoplastic-based or thermoset-based. There are many applications where plant-based fibers and/or fillers are used as reinforcement materials [2–5].

In the industry, plant-based fibers are used as cheap fillers, especially in polypropylene and polyvinylchloride-based composites. It is common to use sawdust, mostly from furniture and construction wood residues. These composites containing wood fiber and/or sawdust are generally used in outdoor floor coverings, window sills, parks, and garden furniture. Since high stress is not required in such applications, low mechanical properties can be accepted. Therefore, the use of recycled plastics finds application for such products. This offers a partial solution to the disposal problems of petroleum-derived plastic waste [6].

Natural fiber/filler-based polymer composites attract more attention than synthetically added polymer composites due to the global energy crisis. The search for sustainability, and their unique features, such as being non-toxic and not irritating the skin, eyes, or respiratory tract. The use of these
Composites in different sectors is becoming increasingly widespread and offers an environmentally friendly approach to recycling [7]. Natural fibers and fillers are seen as alternatives to synthetic-based ones due to reasons such as low production costs, lightweight, abundance of renewable resources, being biodegradable and environmentally friendly, and also showing good physical and mechanical properties. In recent years, natural fibers and fillers have found use in non-structural applications [8].

Natural fibers are also used as reinforcements to improve the strength of polymer matrices. Therefore, they have replaced traditional materials in automotive parts and different technical applications. However, compared to synthetic structures, the morphology and chemical composition of natural structures cannot be completely controlled [9]. Some of the natural fibers used are cotton, banana, coconut, rice, hazelnut shells, sawdust, walnut shells, bamboo, and wheat straw. Polyolefins are mostly preferred in composites containing natural fibers. The use of polyolefins requires the processing temperature to be below 200 °C. Above this temperature, degradation of fibers and fillers has been observed [10, 11].

When natural fibers are classified according to their sources, bast fibers (jute, kenaf, flax, etc.), leaf fibers (palm, sisal, manila, etc.), seed fibers (cotton, kapok, etc.), fruit fibers (coconut, coco fiber, etc.), and stem fibers (bamboo, rice, grass, corn, wheat, etc.) [12–19] Composites made from these natural fibers are also called green composites. Many studies have been done on green composites in the literature [20–27].

Koyuncu (2022) examined in his study how the amount of sawdust changes the physical properties of the epoxy matrix composite material. As a result of the study, it was observed that the composite material filled with 15% sawdust by weight showed the highest tensile strength and flexural strength compared to other prepared samples, and a good interfacial interaction occurred between epoxy and sawdust. It was understood in termal analysis that the tensile and flexural strengths of composites can be improved by matrix modification [28].

Başboğa et al. (2022) produced a polypropylene matrix waste wood flour-filled composite material using 3% MAPP (maleic anhydride polypropylene) as a compatibilizer and 3% paraffin wax as a lubricant and examined the mechanical and morphological properties of these materials. With the increase in the amount of wood flour, the tensile elastic modulus and flexural strength improved, and a decrease in the elongation at break value was observed [29].

Espinach et al. (2022) examined the flexural strength of polypropylene composite materials with textile waste cotton fiber added. When the flexural strengths of the samples are examined, it is seen that both the samples with and without compatibilizers have good enough properties to replace glass fiber-reinforced composite materials [30].

Hussain et al. (2022) measured the hardness and friction coefficient values of waste cotton, which they added to polypropylene in different weight ratios. It has been observed that as the cotton ratio increases, the hardness and friction coefficient values of the composite material increase [31].

Narhoğlu et al. (2018) added larch sawdust to the polypropylene matrix and examined the mechanical properties of the composite material. An increase was observed in the tensile strength, flexural strength, and impact resistance of the samples [32].

Neher et al. (2020) prepared a sawdust-filled acrylonitrile butadiene styrene (ABS) matrix composite material. It was observed that the water absorption of each sample increased with the addition of fiber, while the tensile strength and hardness decreased. However, it was observed that the elastic modulus increased as the fiber ratio increased [33].

Usta et al. (2003) produced composite materials by adding cotton waste in different fiber sizes to the recycled polycarbonate matrix. With the addition of fiber, a decrease was observed in the elastic modulus, tensile strength, and hardness values of the composite samples, while an increase was observed in the elongation and Izod impact strength [34].

Taşdemir et al. (2003) obtained composite material by adding different ratios of cotton waste to the polypropylene matrix. According to the results of mechanical tests, it was observed that with increasing fiber ratio, there was a decrease in the elastic modulus, tensile strength, and hardness values of the composite samples and an increase in the elongation and Izod impact strength values [35].

In this study, some physical and thermal properties were examined for the co-usability of waste pine wood sawdust and waste cotton in a recycled polypropylene matrix.

2. Material and Methods

Since the use of recycled polypropylene is quite common in the industry, recycled polypropylene was used as the matrix material. Waste cotton was obtained from the textile industry, and waste pine sawdust was obtained from furniture manufacturers. Since waste cotton comes in different sizes, it is preferred to cut it into 1 cm size. Waste sawdust was also preferred in the range of 0-250 microns for use in composites.
2.1 Sample preparation

The contents of the samples are given in Table 1. It is preferred to add a maximum of 20% filler by weight to recycled polypropylene.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Pine</th>
<th>Sawdust</th>
<th>Waste</th>
<th>Cotton</th>
<th>Recycled Polypropylene</th>
</tr>
</thead>
<tbody>
<tr>
<td>rPP</td>
<td>0</td>
<td>0</td>
<td>100</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>5PS15C</td>
<td>5</td>
<td>15</td>
<td>80</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10PS10C</td>
<td>10</td>
<td>10</td>
<td>80</td>
<td></td>
<td></td>
</tr>
<tr>
<td>15PS5C</td>
<td>15</td>
<td>5</td>
<td>80</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20PS</td>
<td>20</td>
<td>0</td>
<td>80</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20C</td>
<td>0</td>
<td>20</td>
<td>80</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Abbreviations: rPP, recycled polypropylene; PS, pine sawdust; C, cotton

The fillers and rPP were mixed using the extrusion method, the final samples were obtained injection method (table 2).

<table>
<thead>
<tr>
<th>Process</th>
<th>Injection Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>190-220</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>100-110</td>
</tr>
<tr>
<td>Mold waiting time (s)</td>
<td>12</td>
</tr>
</tbody>
</table>

The melt flow rate determination (MFR) test of the samples was carried out in the Devotrans brand device of Efeler Polimer company, at 230 °C and 2.16 kg weight. A density determination test was carried out by Efeler Polimer company with Precisa XB 220A brand balance with 0.0001 g sensitivity. The impact test was carried out on the Zwick B5113.30 brand test device with a 5.4 J hammer.

3. Results and Discussions

In this study, natural filler-filled thermoplastic-based composites were produced, and pine tree sawdust, which is abundant in our country, and cotton scraps, which come from recycling and emerge as waste in the textile industry, were used as natural fillers. Thus, an environmentally friendly green composite was produced. Recycled polypropylene, which is widely preferred in the composite industry, was used as the matrix material.

MFR and MVR (melt volume ratio) are two common rheological testing methods used to measure the melt flow properties of thermoplastic polymers. These tests are used to evaluate the processability and flow behavior of polymers. Both MFR and MVR are tools that help us determine important properties of polymers, such as their processability and suitability for injection molding, extrusion, and similar processes. The effects of natural fillers on the polymer flow index may vary depending on the filler type, amount, distribution, and processing process. Understanding these effects is important to optimize the machinability of composite materials [36].

Figure 1 shows the MFR and MVR results of the samples. When both MFR and MVR results are examined, it is seen that the highest values are in the unfilled recycled polypropylene sample with 18.588 g/10min and 24.8 cm³/10min, respectively. It was observed that the values decreased significantly in all filled composites. It is seen in other studies in the literature that the melt flow rate decreases with increasing cellulose content [34, 37–39].

It can have significant effects on the polymer density of natural fillers. These effects may vary depending on the type, amount, and distribution of the natural filler material and its interaction with the polymer matrix.

It can be seen in Figure 2 that the density values of the samples vary between 0.874 g/cm³ and 0.946 g/cm³. Looking at the density results, it was observed that the unfilled recycled polypropylene sample was 0.874 g/cm³. The highest density value was seen in the 20% cotton-filled composite sample, with a value of 0.946 g/cm³. It has been determined from other articles that natural fillers added to the polymer matrix do not change the density value much, and the density values of the samples are compatible with the literature [40, 41]. In figure 3, the values of Izod impact strength according to filler ratios are given. As expected, the impact strength of recycled Polypropylene was the highest. It was observed that the impact strength of pure recycled polypropylene (23.15 kJ/m²). The highest value in filler-added composite samples was measured with the sample coded 10PS10C. The lowest impact strength was observed in the 20PS sample. Many factors such as the properties, structure and ratio of fillers added to the polymer matrix affect the impact resistance of
composites. This decrease is also seen in other studies in the literature [42].

SEM images of the fracture surfaces of the samples are given in figure 4. Pure recycled PP showed high elongation (4a). In figure 4b, pine sawdust and cotton fillers are seen in the same image. Cotton fibers in the polypropylene matrix are seen at figure 4c (the fibers diameter of around 10 microns). In figure 4d, the sawdust in the sample filled with 20% pine sawdust is shown. It is seen that the length of the sawdust is approximately 200 microns. It can be understood from the SEM images that there is not a good adhesion between rPP and pine sawdust.

4. Conclusions

Polymer composites containing natural fillers attract great attention due to their many advantages. Therefore, it replaces traditional structural materials in many applications. In this study, the effect of waste cotton and pine sawdust fillers on recycling polypropylene was examined.

- When MFR values are examined, it is observed that there is a decrease in filled composite materials compared to rPP.
- No obvious change was observed in the density data. The lowest density was obtained in the rPP sample.
- From the SEM images, it can be said that the samples were prepared homogeneously, but in some samples, a good bond was not formed between the matrix and the filler.
- Impact strength decreased slightly with the fillers added to the polymer matrix.
As a result of the study, it was evaluated that cotton and pine sawdust can be used together as fillers in polypropylene matrix composite materials.

Author Statements:

- **Ethical approval**: The conducted research is not related to either human or animal use.
- **Conflict of interest**: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
- **Acknowledgement**: The authors declare that they have equal right on this paper.
- **Funding information**: The authors declare that there is no funding to be acknowledged.
- **Data availability statement**: The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

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