Properties of Wood Sawdust and Wheat Flour Reinforced Polypropylene Composites

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In recent years, extensive studies have been carried out on natural fiber reinforced thermoplastics due to their low cost, high specific properties, simple fabrication methods, biodegradable nature and diverse applications touching all commercial sectors. In this research, the effect of incorporating small amounts (5%) of wood sawdust and wheat flour on the mechanical, morphological, chemical, thermal, and water absorption properties of polypropylene (PP) were studied. In terms of mechanical properties the Young’s modulus and flexural modulus increased, whereas the yield strength and elongation at break decreased in both types of composites. However, the wheat flour composites exhibited higher yield strength, Young’s modulus, flexural modulus and elongation at break than wood sawdust composites. Morphological examination of the fracture surfaces by scanning electron microscopy (SEM), carried out in order to investigate the variation of the measured mechanical properties, revealed presence of microvoids resulting from weak filler-matrix interfacial adhesion. Thermogravimetric analysis (TGA) and water absorption tests confirmed the produced composites exhibited higher thermal stability and water absorption in comparison to virgin PP. Changes in the chemical bonding nature of the composites was also investigated by FTIR spectroscopy.

Keywords: Polymer Composites, Natural Fibers, Mechanical and Morphological Properties

Field of Research: Polymer and Composites, Polymer Chemistry, Material Science

1. Introduction

Many of our modern technologies require materials with unusual combinations of properties that cannot be met by the conventional metal alloys, ceramics, and polymeric materials. This is why the interest of scientists has turned to composite materials that provide better property combinations and result in significantly different physical and chemical properties. In recent years, exhaustive research has been carried out on utilizing natural fibers as efficiently as possible to produce superior quality fiber-reinforced polymer composites for a wide range of applications (Premalal et al, 2002; Sharma et al, 2001; Rahman et al., 2010; Yang et al, 2006; Yang et al, 2007; Choi et al, 2006 and Kolybaba et al, 2003). Natural fibers are now exploited as a replacement for conventional fibers, such as glass, aramid and carbon due to their advantages, which include low cost, abundance in nature, high strength-to-weight ratio, fairly good mechanical properties and non-abrasive, eco-friendly and

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The light weight, easy availability, and biodegradability of natural fiber composites afford a variety of uses of these materials for the building and automobile sectors. So, a prime significance of our research is to testify the suitability of wood sawdust and wheat flour reinforced polypropylene composites for these practical applications.

The tensile properties of natural fiber reinforce polymers are mainly influenced by the interfacial adhesion between the matrix and the fibers. Two main factors presently limit the large scale production of natural fibers composites (Westman et al, 2010). Firstly, the strength of natural fiber reinforced composites is very low compared to glass which is often a result of the incompatibility between the hydrophilic fiber and the hydrophobic polymer matrix (Premalal et al, 2002; Westman et al, 2010; Ghani and Ahmad, 2011; Tserki et al, 2006; Mun’aim et al, 2011; Yang 2004; Rahman et al, 2010 and Karmarkar et al, 2007).

Secondly, natural fiber composites have high water absorption tendency. This is because of the presence of hydroxyl groups in natural fibers which absorb water from the air and direct contact from the environment, the ultimate result being lower strength (Westman et al, 2010; Ghani and Ahmad, 2011; Tserki et al, 2006 and Mun’aim et al, 2011). However, a positive outcome of the higher water absorption is the enhancement in biodegradability of the polymer based materials which is an important characteristic required for many modern appliances (Kolybaba et al, 2003, Tserki et al, 2006 and Yang et al, 2004).

The main objective of the research was to delve into the problems associated in manufacturing natural fiber reinforced composites based on synthetic the PP. Moreover, the implications of small additions of natural fibers to the olefin on its mechanical, physical and chemical properties have been explored. For this purpose, the PP and natural fiber composites were fabricated by hot pressing into sheets. The mechanical properties of these composite sheets were then examined using standard tensile and flexural tests. The surface morphology was studied using Scanning Electron Microscope (SEM) and Thermo gravimetric Analysis (TGA) was carried out to evaluate the thermal stability of the composites. Moreover, Fourier Transform Infrared Spectroscopy (FTIR) was performed to study the characteristic peaks of the newly formed bonds in the composites and water absorption tests were also performed to determine the water absorption ability of the composites. All the results obtained have been discussed in detail in the findings and discussion section.

2. Literature Review

The rapidly expanding use of composite components in automotive, construction, sports and other mass production industries, has influenced the focus of present day researches on sustainable and renewable natural fiber reinforced composites. Recently various researches have been carried out on different natural fiber reinforced composites with the intention to produce material of desired properties.

In 2002 Premalal et al studied the comparison between the mechanical properties of rice husk powder filled polypropylene composites (RHP) with talc filled polypropylene
The RHP composites exhibited poorer mechanical properties than the talc composites. SEM studies of tensile fracture surfaces of the composites indicated the poorer interfacial adhesion of RHP composites compared to talc composites. A major factor contributing to this difference in adhesion was the smaller mean diameter and larger specific area of talc, in comparison to RHP, that resulted in improved filler-matrix adhesion.

In 2008, Fu et al investigated in detail the effects of particle size, particle/matrix interface adhesion and particle loading on the mechanical properties of particulate polymer composites. In this paper it was reiterated that smaller particles having a higher total surface area for a given particle loading resulted in increased mechanical properties due to a more efficient stress transfer mechanism.

Yang et al reported the tensile strengths and Izod impact strength of rice-husk flour filled polypropylene composites decreased while the tensile modulus improved with increasing filler loading and crosshead speed. Increased filler loading worsened the filler-matrix interfacial bonding therefore acceptable strengths were obtained up to a filler loading of 40 wt. % only.

Karmakar et al, 2007 investigated the effects of incorporation of Eucalyptus wood-fibers both with and without a compatibilizing agent (m-TMI-g-PP) on the processing and mechanical properties of polypropylene composites. Mechanical properties, measured in tensile, flexural and impact tests, demonstrated that the fibers used in this work act as effective reinforcing agents for PP. Addition of wood–fibers, at all levels, resulted in more rigid and tenacious composite, but had lower impact energy and percentage of elongation as compared to the matrix polymer. It was concluded that the increase in mechanical properties demonstrated that m-TMI-g-PP is an effective compatibilizer for wood-fiber polypropylene composites.

Rahman et al, 2010 examined the physico-mechanical properties of jute fiber reinforced polypropylene composite. In this research raw jute fiber was oxidized and manufactured composites were post-treated with urea to increase the compatibility of the jute fiber with the polypropylene matrix. Results revealed that the tensile strength of the composites decreased while the Young's modulus, flexural strength, flexural modulus, Charpy impact strength, and hardness of the composites increased with an increase in the fiber loading. Post-treated jute fiber reinforced specimens yielded better mechanical properties compared to the oxidized and raw ones.

Later in 2010 Rahman et al investigated the influence of benzene diazonium salt fiber treatment on the mechanical and morphological properties of sawdust reinforced polypropylene composites and in 2011 Islam, M.N and Islam, M.S studied the mechanical properties of cetyltrimethyl ammonium bromide treated sawdust reinforced polyethylene. In both cases the chemically treated composites exhibited better mechanical properties than the untreated opponent. The authors believe that chemical treatment resulted in better filler-matrix adhesion.

Ghani and Ahmad, 2011 investigated the difference in water absorption of composites of a hybrid fiber, containing rice husk and sawdust, with recycled high-density polyethylene prepared by two different extruder machines, namely, counter rotating and co-rotating twin screw. The counter rotating twin-screw extruder gives a smaller value of water absorption in comparison to the co-rotating twin-screw
Thus the authors conclude that different forming techniques play an important role in water uptake by improving the filler-matrix adhesion.

In this research we have investigated and compared the effect of easily available natural fillers, wheat flour and sawdust, on the different properties of polypropylene. Since both fillers have a different particle size, the influence of particle size on the composites were also examined to determine optimum properties for the natural fiber reinforced composites.

3. Methodology

3.1 Materials

Commercial PP, bought from the local market in Dhaka, was used as the polymer matrix. The PP in the form of homopolymer pellets had a specific gravity of 0.91 and melting temperature in the range 165-171°C. The wood sawdust (mesh 80-100) used was medium wood obtained from a local sawmill in Dhaka. The wheat flour (mesh 230-270) used was also of local grade. Moreover, polyester and methanol were used during the composite preparation to improve interfacial adhesion in the composite.

3.2 Natural Fiber Preparation

The sawdust and flour were then dried in the furnace at 80°C for 24 hours to remove moisture and other volatile impurities present.

3.3 Fabrication of PP-Wood Sawdust Composites and PP-Wheat Flour Composites

The PP and the natural fibers (5% by weight) were vigorously mixed with polyester and methanol to obtain a uniform mixture. The mixture was then heated in the furnace at 110°C for 10-15 minutes to remove moisture adhering to the surface and finally hot pressed by compression molding into sheets measuring 220 mm x 220 mm x 3mm according to the heating cycle shown in Fig 1.

Figure1: Heating Cycle Used in Hot Press
3.4 Fourier Transform Infrared Spectroscopy (FTIR)

The infrared spectra of raw sawdust, raw wheat flour, virgin PP, PP-sawdust and PP-wheat flour composites were taken and the characteristic peaks were studied.

3.5 Mechanical Tests

Tensile and flexural tests were performed using a Universal Testing Machine of model INSTRON 3369 and at a cross-head speed of 4mm/min. The tensile tests were conducted according to ASTM D 638-01 and the flexural tests were conducted according to ASTM D 790-00, and for each test and type of composite, ten replicate samples were tested and the average values were reported.

3.6 Scanning Electron Microscopy (SEM)

The surface morphology of the fracture surfaces of the tensile specimens were examined by a scanning electron microscope. The samples were sputter coated with gold and observed at various magnifications ranging from 200 to 3000.

3.7 Water Absorption Test

Samples made for water absorption testing were according to ASTM 570-98 [35]. The samples were first dried at 80°C for 24 hours. The percentage of water absorption of pure PP and the composites were then calculated according to the formula;

$$\text{Water Absorption (\%)} = \left( \frac{M_t - M_o}{M_o} \right) \times 100$$

Where $M_o$ is the weight of the samples before soaking in water and $M_t$ is the weight after soaking in water for a time period $t$.

3.8 Thermo Gravimetric Analysis (TGA)

TGA analysis was carried out, using a Thermo-Gravimetric Analyzer of model TGA Q50 W/FMC, to evaluate the effect of addition of sawdust and wheat flour on the thermal stability of PP.

4. Findings and Discussion

4.1 Tensile and Flexural Properties

The tensile test results of the samples, as shown in Fig 2., revealed that the average tensile strength of pure PP was decreased by the addition of both sawdust and wheat flour as has been confirmed by the other researchers[Premalal et al, 2002; Tserki et al, 2006; Mun’aim et al, 2011; Yang and Kim, 2004 and Rahman et al, 2010]. This may be mainly attributed to the poor interfacial adhesion between the hydrophobic polymeric matrix of PP and the hydrophilic lignocellulosic fillers, which does not allow efficient stress transfer between the two phases of the material [Premalal et al, 2002; Westman et al, 2010; Ghani and Ahmad, 2011; Tserki et al, 2006; Mun’aim et al, 2011; Yang and Kim, 2004; Rahman et al, 2010 and Karmarkar et al, 2007]. Moreover, there is a minimum critical fiber length below which, the fibers
do not act as reinforcing agents (Tserki et al, 2006 and Baiardo et al, 2004). The higher percent decrease in tensile strength upon sawdust incorporation (28.9%) in comparison to wheat flour addition (15.9%) can be explained most explicitly by the particle size of the fillers. The particle size of the wheat flour (mesh 230-270) being considerably smaller than sawdust (mesh 80-100), resulted in better mechanical properties of the PP-wheat flour composites due to higher total surface area of the filler particles that lead to a more efficient stress transfer mechanism than the larger sawdust particles (Fu et al, 2008).

The elongation at break follows the same trend as the tensile strength as shown in Fig 3. Incorporation of as little as 5% filler resulted in a decrease in the elongation at break from 38% in pure PP to 5-2% in the composites which is in accordance to other researchers (Premalal et al, 2002; Tserki et al, 2006; Karmarkar et al, 2007; Kaczmarek et al, 2005 and Chawla and Shen, 2001). This steep decline in elongation on filler addition is because the fibers in the matrix restrict the plastic flow of the polymer (Karmarkar et al, 2007).

Fig 3. shows that Young’s modulus of both composites increased in comparison to pure PP which is in agreement to previous studies (Premalal et al, 2002; Rahman et al, 2010; Tserki et al, 2006; Mun‘aim et al, 2011; Yang and Kim, 2004; Rahman et al, 2010 and Karmarkar et al., 2007). Incorporation of the rigid fibers into the soft PP matrix results in increased stiffness as the poor interfacial bonding creates
partially separated micro-spaces between the filler and matrix polymer thereby obstructing stress propagation during tensile stress loading (Rahman et al, 2010; Yang and Kim, 2004 and Fu et al, 2008). From Fig 3, it is also evident that the modulus of the wheat flour composite is slightly higher than the modulus of the sawdust composite. This can be attributed to the smaller particle size and larger surface area of the wheat flour particles resulting in better filler-matrix adhesion and poorer mobility of PP matrix (Premalal et al, 2002).

From Fig 4, it is evident that the flexural strength remains almost unchanged by the small (5%) additions of both the natural fibers which is in agreement with other researchers [Premalal et al, 2002; Mun’aim et al, 2011 and Rahman et al, 2010]. However, the flexural modulus of the composites is higher than pure PP thereby following the same trend as the Young’s modulus as shown in Fig 6. Wheat flour and sawdust being high modulus material, when incorporated into pure PP demands higher stress for the same deformation.

4.2 Morphological Study

The SEM images of the tensile specimen fracture surfaces are shown in Fig 7. The fracture surfaces of pure PP are characteristic of brittle fracture with cleavage planes (Fig 7a.). The fracture surface of both composites, revealed the presence of voids which indicate the poor interaction between the non-polar matrix and polar fillers. This results in poor stress transfer from the matrix to filler resulting in poor properties. Moreover, both composites revealed the presence of fibrils which suggest plastic deformation of the matrix as shown in Fig 7c. As the adhesion of the matrix
and the fillers is poor so the matrix can deform independently until the filler particles restrict the deformation (Premalal et al, 2002).

**Fig 7:** SEM Micrographs Showing Fracture Surfaces of (a) PP at a Magnification of 1000x, (b) PP-Sawdust Composite at a Magnification of 1000x, (c) PP-Wheat Flour at Magnification of 3000x.

4.3 Water Absorption Test Results

From the water absorption test results it is evident that pure PP is extremely hydrophobic and its water absorbing ability is increased by the addition of sawdust and flour. Water absorption is higher in the PP-sawdust composite, i.e. almost twice as high, than the PP-flour composite. This may be attributed to the fact that wood is extremely hydrophilic in nature due to the presence of the hydrophilic hydroxyl groups of cellulose, hemicelluloses and lignin that responsible for water absorption. The results are expressed in a bar chart in Fig 8.

**Figure 8:** Bar Chart Showing Percent Water Absorption of the Samples
4.5 FTIR Analysis

Fig 9. shows the overlapping FTIR spectra of pure PP, wheat flour and PP-wheat flour composites. From the pure PP spectrum major peaks are found at 2922.6 cm\(^{-1}\), 1458 cm\(^{-1}\) and 1377.3 cm\(^{-1}\) that correspond respectively to the sp\(^3\) C-H stretch, CH\(_2\) (methylene) bend and CH\(_3\) (methyl) bend. The IR spectrum of wheat flour shows strong & broad absorption bands at 3418.6 cm\(^{-1}\) that corresponds to O-H stretching in starch. The peak at 2928.3 cm\(^{-1}\) corresponds to aliphatic saturated (sp\(^3\)) C-H stretching vibration due to lignin polysaccharides (cellulose and hemicelluloses). Absorption band at 1654.6 cm\(^{-1}\) may either correspond to the C=O vibrations of acetyl or uronic ester groups of hemicelluloses or to the scissoring or bending of two O-H bonds of absorbed water. The aromatic C=C stretch from aromatic ring of lignin gives two peaks at 1541.6 and 1458.2 cm\(^{-1}\). The peaks in the range 1022.4 to 1155.9 cm\(^{-1}\) range may be attributed to C-O (ester) vibration. From the IR spectrum of the PP-wheat flour composite we can see that the characteristic absorption bands of pure PP have been retained but some of the characteristic bands of flour have disappeared or become weaker. The peak at 1654.6 cm\(^{-1}\) in wheat flour corresponding to O-H bond vibration of absorbed water has been significantly suppressed in the composite. Peaks in the range 1022.4-1155.9 cm\(^{-1}\) corresponding to C-O vibrations have also been suppressed drastically in the composite along with the peaks in the range of 520.7-575.5 cm\(^{-1}\).

Fig 10. shows the overlapping FTIR spectra of pure PP, sawdust and PP-sawdust composite. The FTIR spectrum of sawdust reveals a strong broadband at 3396.5 cm\(^{-1}\) characteristic of the OH bond stretching (due to the presence of cellulose, hemicelluloses and lignin). Doublet peaks at 2917.1 cm\(^{-1}\) and 2849.8 cm\(^{-1}\) correspond to C-H stretching vibration. The Peak at 1735.8 cm\(^{-1}\) corresponds to C=O (carbonyl group) stretching vibrations and the absorption bands at 1054.8 cm\(^{-1}\) and 1032.3 cm\(^{-1}\) corresponds to C-O (ester) bond vibrations. From the IR spectrum of the PP-sawdust composite it can be seen that all the characteristic bonds of PP and sawdust are present in the composite, so the major peaks at 2958.5-2919.8 cm\(^{-1}\), 1458.3 cm\(^{-1}\), 1376.6 cm\(^{-1}\) correspond to C-H stretch, CH\(_2\) (methylene) bend and CH\(_3\) (methyl) bend respectively. In a similar manner all the characteristic peaks for sawdust are also present but the characteristic peaks of sawdust in the region of 668.6 to 611.5 cm\(^{-1}\) have become suppressed and the characteristic peak of sawdust at 1725.1 cm\(^{-1}\) and 1288 cm\(^{-1}\) has become greatly exaggerated in the composite.

From these results it can be concluded that the amount of wheat flour and sawdust was too insignificant to bring about any significant changes in the actual bonding of nature PP. Therefore, the PP bonds remain dominant in both composites and the natural fibers and PP coexist in the composite maintaining their individual characteristic.
4.6 TGA Analysis

All the three TGA curves obtained are one-stage decomposition curves and therefore can be used to define the stability limit of the samples.

From Fig 11 it can be seen that PP, PP+5%wheat flour and PP+5% sawdust start to
decompose respectively at 295.67°C, 370.49°C, 380.51°C. From this observation it can be concluded that both composites have better thermal stability than PP. In general, PP has a comparatively low thermal stability due to its volatile nature. Additions of sawdust or wheat flour blanket the PP grains and therefore decrease mass loss by volatilization.

Another observation made from the TGA analysis is that the virgin PP contains no decomposition end-products thereby confirming it is pure and contains no other non-decomposable substances. However, in the case of PP-sawdust composite almost 2.167% residual products are obtained after decomposition. This is because sawdust contains a considerable amount of non-decomposable minerals and metallic oxides. The PP-wheat flour composite contains about 0.9% residual substances after decomposition.

**Figure 11: TGA Curve of Pure PP, PP-Wheat Flour, PP-Sawdust Composite**

5. Conclusion

The addition of wheat flour and sawdust to PP clearly altered its mechanical, morphological, physical and thermal properties considerably. The tensile strength and percent elongation of both composites were lower in comparison to the virgin PP mainly due to the poor adhesion between the natural fibers and plastic matrix confirmed by the morphological study, revealing the presence of blow holes in the composite fracture surfaces. The wheat flour composites exhibited better mechanical properties than the wood sawdust composites due to their smaller particle size that provided higher surface area for efficient filler-matrix interfacial adhesion.

The poor filler-matrix adhesion could be overcome considerably by using a compatibilizing or coupling agent to improve adhesion between natural fiber & PP.
matrix and further research using a suitable coupling agent is suggested. Also, performing compression molding under vacuum can considerably reduce the concentration of blow holes and improve mechanical properties. Further research can also be carried out to compare the efficiency of different forming processes on the properties of the composites. On this account, twin-screw extrusion and injection molding is suggested. In addition, since finer filler size has a positive effect on the mechanical properties the effect of nano scale fibers on the properties of natural fiber reinforced composites can also be investigated in future work.

Results have also revealed increased water absorption in both composites. This unique characteristic makes natural fiber composites significantly eco-friendly in comparison to virgin PP and hence they have become viable for use in many modern appliances which thrive on biodegradability.

Furthermore, the thermal properties of both composites were significantly improved. However, the small addition of natural fibers had little effect on the bonding nature of the PP revealed by the FTIR results. Extended research is suggested by increasing filler loading to study the resultant chemical bonding nature of the composites.

References

Fakhru1, Mahbub and Islam


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