Mechanical Behavior of Agro-Residue-Reinforced Polypropylene Composites

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ABSTRACT: In this research, fully environment-friendly, sustainable and biodegradable composites were fabricated, using wheat straw and rice husk as reinforcements for thermoplastics, as an alternative to wood fibers. Mechanical properties including tensile, flexural, and impact strength properties were examined as a function of the amount of fiber and coupling agent used. In the sample preparation, three levels of fiber loading (30, 40, and 50 wt %) and two levels of coupling agent content (0 and 2 wt %) were used. As the percentage of fiber loading increased, flexural and tensile properties increased significantly. Notched Izod results showed a decrease in strength as the percentage of fiber increases. With addition of 50% fiber, the impact strengths decreased to 16.3, 14.4, and 16.4 J/m respectively, for wheat straw-, rice husk-, and poplar-filled composites. In general, presence of coupling agent had a great effect on the mechanical strength properties. Wheat straw- and rice husk-filled composites showed an increase in the tensile and flexural properties with the incorporation of the coupling agent. From these results, we can conclude that wheat straw and rice husk fibers can be potentially suitable raw materials for manufacturing biocomposite products. J Appl Polym Sci 111: 2616–2620, 2009

Key words: fibers; mechanical properties; polypropylene; reinforced composites

INTRODUCTION

During last two decades, people have been concerned about deforestation and forest degradation and its impact on biodiversity, global warming, and productivity for wood.[1,2] Additionally, in many developing countries, such as Iran, forest wood is not sufficient to meet their demands for wood-based industries. However, many of these countries do have relatively large quantities of other lignocellulosic materials in the form of agro-residues from annual crops.[3–5] Agro-residues or byproducts such as cereal straw, corn stalks, rice husk, and bagasse represent a potentially valuable source of fiber that could be used as a supplement or could be used as a direct substitute for wood fiber in the manufacture of composites. These lignocellulosic biomasses are produced in billions of tons around the world and represent as abundant, inexpensive, and readily available raw materials.[5] Among these enormous amounts of agro-residues, only a minor quantity of residues is reserved as animal feed or household fuel and a major portion of the straw is burned in the field creating environmental pollution. The exploration of these inexpensive agro-residues as bioresource for making industrial products will open a new avenue for the utilization of such residues by reducing the need for disposal and environmental deterioration through pollution, fire, and pests and, at the same time, add value to the creation of rural agricultural-based economy.[6]

Wood fiber reinforced composites have been extensively studied over the last two decades.[7–9] Use of the cereal straw and other agro-residues as a filler or reinforcement in the production of plastic composites alleviate the shortage of wood resources and can have the potential to start a natural fiber industry in countries where there are little or no wood resources left. Apart from particleboards, straw has been used for making composites with polyester, polypropylene (PP), polyurethane, poly(3-hydroxybutyrate-*co*-3-hydroxyvalearate), and Novolac resin.[5] The main factors that restrict the use of the cereal straw and other agro-residues in composites are problems associated with collection, storage, transportation of these materials, and economics for the overall production of composites. However, it is believed that the research focused at the use of these materials could follow a developmental market, and, at the same time, can lead to a new market opportunity for these surplus-inexpensive field crop leftovers.[10,11]

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TABLE I
Morphological and Chemical Properties of Used Fibers

Properties	Wheat straw	Rice husk	Poplar
Fiber morphology			
Fiber length (mm)	1.2	0.8	1.0
Fiber width (μm)	15.3	9.7	23.9
Aspect ratio[a]	76.4	88.9	39.2
Chemical			
Cellulose (%)	51.1	48.9	51
Lignin (%)	19.5	19.1	22.3
Extractives[b] (%)	3.0	2.5	2.1
Ash (%)	4.8	12.3	0.7

[a] The ratio of fiber length to fiber width.
[b] Cold water.

Wheat straw is one of the most important agricultural crop residues. It is an annually renewable fiber resource that is available in abundant quantity in many regions of the world. In Iran, 1,000,000 tones of unused wheat straw residues are generated every year. Straw is similar to wood and could also be considered as a natural composite material. It consists mainly of cellulose, hemicellulose, and lignin.[2,12]

The main objectives of this research work were (i) to prepare wood plastic composites from wheat and rice stalks as reinforcements, (ii) to evaluate the mechanical properties of composite materials, (iii) to evaluate the effectiveness of the agro-residues/PP composites; they have been compared with poplar as an important fast growing wood species in Iran.

EXPERIMENTAL

Materials

Fibers

The three different types of fibers used in this study, to obtain the comparative data, were wheat straw, rice husk, and poplar (*Populus deltoides*) (Table I). The agro-residues were obtained from local farmers. Poplar fiber was used as a filler without any modification to compare with the properties of agro-residue-based composites. The fibers were provided by refiner mechanical pulping process. The chips were steamed for 15 min at 7 MPa and 175°C, disc–refined, and then dried in a laboratory-made hot air tube dryer. Before composite preparation, fibers were oven-dried at 95°C for 24 h. The moisture content of oven-dried fiber was lower than 3%.

TABLE II
Properties of Homopolymer Polypropylene (PP)

Appearance	White
Density (g/cm^3)	1.36
Elongation at yield (%)	4.75
Tensile strength (MPa)	23.5
Flexural strength (MPa)	38.5
Flexural modulus (MPa)	1150

Polymer matrix

Polymer used for making composites was homopolymer pellet PP, with a melt flow index of 7–10 g/10 min. The polymer was obtained from Bandar Imam Petrochemical Company, Iran. The important characteristics of the polymer matrix used are given in Table II.

Coupling agent

Maleated polypropylene (MAPP) was supplied by Eastman Chemical Products, Inc.; as Epolene G-3003™ polymer with 8% acid anhydride and a molecular weight of 103,500. MAPP acts as a coupling agent in polymer blends and is particularly effective when one polymer is hydrophilic and the other is hydrophobic.

Panel manufacturing

The weight ratios of the fiber to polymer were 30 : 70, 40 : 60, and 50 : 50 (w/w), and the weight of coupling agent in the formulation was 0 and 2 wt %. Fiber and PP was kneaded with and without coupling agent (depending on compositions) in a corotating twin-screw extruder (Collin) for 10 min to produce homogeneous fiber/PP composite mixture. The melt temperature at the die was 180°C, and the rotation speed was 60 rpm.

After compounding, the extrudate in the form of strands were allowed to cool to room temperature and then granulated using a CW Brabender Granulator. The resulting granules were dried at 105°C for 24 h before being injection molded into ASTM test specimens. Tests specimens including tensile,

TABLE III
Injection Molding Conditions in This Study

Heater (°C)			Injection pressure (MPa)	Holding pressure (MPa)
Front	Middle	Nozzle		
170	180	200	3	5

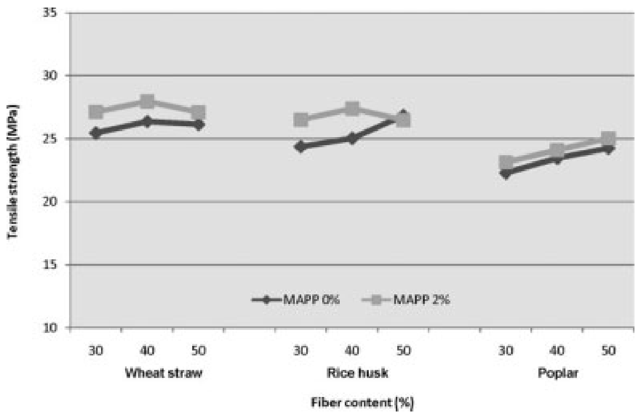

Figure 1 Comparison of tensile strength for various fibers with different fiber loading.

flexural, and impact samples were prepared by an Imen injection-molding machine. The injection-molding conditions are shown in Table III.

Mechanical testing

The following mechanical properties were evaluated: (a) modulus of elasticity (MOE); (b) modulus of rupture (MOR); (c) tensile strength; (d) tensile modulus; and (e) impact strength. Test specimen dimensions were according to the respective ASTM standards. The specimens were stored under carefully controlled conditions (50% relative humidity and 23°C) for 3 days before testing. At least six specimens of every composite were tested to obtain a reliable average and standard deviations.

Tensile testing

Tensile tests were conducted to measure the short-term strengths at same conditions in accordance with ASTM D 638 98. The tests were conducted using an Instron Universal Testing Machine (model 1186) at a speed of 1.5 mm/min.

Flexural testing

Flexural properties were measured at a temperature of 23°C and humidity of 50% for MOE and MOR tests according to ASTM D 790 98. Tests were conducted using an Instron Universal Testing Machine (model 1186) at a speed of 2 mm/min.

Izod impact testing

Notched Izod impact tests were conducted at a temperature of 23°C according to ASTM D 256 97. The tests were performed with a pendulum apparatus (Zwick 1446) using conventional V notched Izod notched specimens (notch depth: 2 mm).

RESULTS AND DISCUSSION

Tensile strength

Figures 1 and 2 show the results of tensile tests done on the composite specimens with and without coupling agent treatment. In general, composites with high fiber content and treated with MAPP exhibited better tensile strength than the untreated ones. The increase in tensile strength is due to the crosslinking network (chemical bonding) formation between the fibers and PP polymer chains. Results suggest that modifying the polymer or fiber surface could enhance the compatibility of hydrophobic polymer and hydrophilic cellulose fiber.

The maximum tensile strength value was observed 27.96 MPa for wheat straw specimens. It was observed that tensile strength showed a slight decrease due to the addition of 50 wt % agro-residues fibers to polymer. Since it was well known that the tensile strength increases for a polymer when any mineral or wood fiber is incorporated into it; in this case, for filler contents above 50 wt %, the results follow a different trend. The decrease in tensile strength is governed by the fact that the agro-residues fibers with 50% concentration need more coupling agent to give good reinforcement with the polymer matrix.

Because of high modulus of lignocellulosic fibers, they can reinforce the PP matrix. Figure 2 shows the variations of modulus of composite versus wood content-coupling agent. It is clear that, to improve the reinforcing effect of fiber, the presence of coupling agent is vital. At the constant concentration of fiber (50 wt %) with increasing of coupling agent from 0 to 2 wt %, the increment of tensile modulus was about 10 and 12% for wheat straw and rice husk, respectively. All the compositions showed a tensile modulus higher than the virgin polymers. Some researchers attribute that the chemical reaction between maleic anhydride in nonpolar and polar

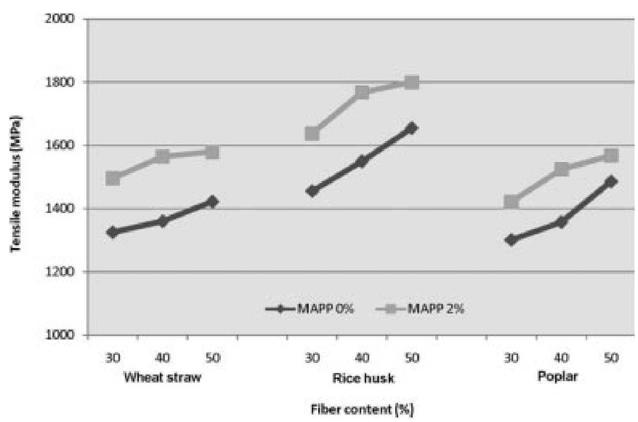

Figure 2 Comparison of tensile modulus for various fibers with different fiber loading.

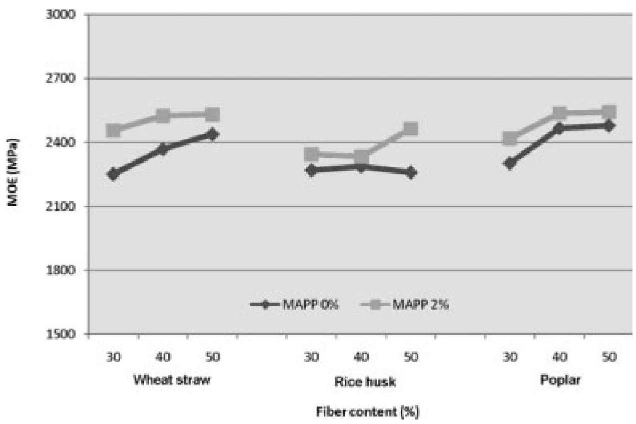

Figure 3 Comparison of modulus of elasticity (MOE) for various fibers with different fiber loading.

group in fiber structure results in a strong coupling between them, and they tracked this reaction with IR, NMR, and other characterizations methods.[9,13,14]

Flexural strength

Figures 3 and 4 show the MOE and MOR of fiber reinforced PP composites with fiber loading of 30, 40, and 50 wt % and the polymers with and without MAPP. Adding fiber to the PP matrix markedly increased the MOR of the composites. As expected, the results of flexural strength testing are similar to that of tensile strength testing; composites with high fiber content and treated with MAPP exhibited better flexural strength than the untreated ones.

The maximum flexural MOE and MOR were shown in wheat straw fiber/PP (2531 and 44.64 MPa, respectively). It was found that composites with 2 wt % MAPP provided significantly higher flexural MOE and MOR, compared with untreated samples. Moreover, wheat straw fiber/PP showed the highest flexural properties. This indicates that, in composites with wheat straw initially due to the presence of a good interaction between the wheat straw fiber and PP as a result of the hydrophobic rich layer attached to wheat straw, the flexural properties is higher than rice husk- and poplar-filled composites. Generally, the strength of fiber reinforced composites depends on the properties of constituents and the interface interaction. However, when considering the flexural properties, homogeneity of the overall composite needs to be taken into account. This is mainly because in bending, the convex side of the specimen is extended and the concave side is compressed.[15]

Impact strength

Unlike the tensile and flexural properties, the Izod impact strength of composite decreased as the fiber loading increased and fairly raised when the concentration of the coupling agent increased (Fig. 5). Composites with high fiber content possess low impact strength. Lignocellulosic fiber is a kind of stiff organic filler, so adding fiber could decrease the impact strength of composite. Composite treated with MAPP exhibited better impact strength than the untreated ones, since the debonding behavior between the interface of fiber and PP matrix absorb larger impact energy in modified composites than the unmodified ones.

As observed in mechanical properties, the reduction in impact strength is higher in rice husk-filled composites compared to wheat straw-filled composites. With addition of 50 wt % fiber, the percentage reduction in the impact of MAPP treated composites is 33.7%, 45.9%, and 41.2% respectively, for wheat straw-, rice husk-, and poplar-filled composites. This result may attribute to the increased stress concentration by the decreased uniformity of polymer and liquefied mixture. It is also considered that the reduced flexibility of polymer matrix is another reason for decreased impact strengths. These results are

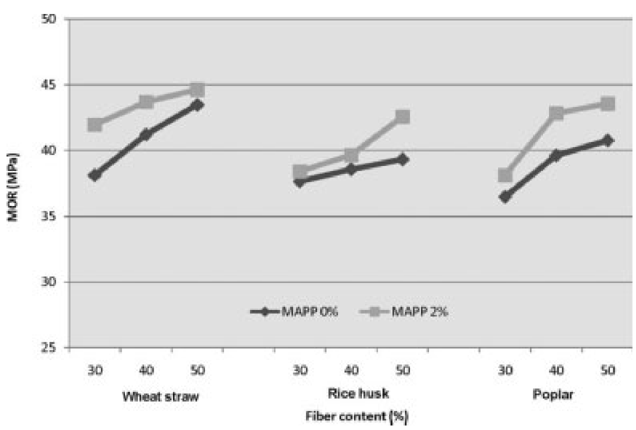

Figure 4 Comparison of modulus of rupture (MOR) for various fibers with different fiber loading.

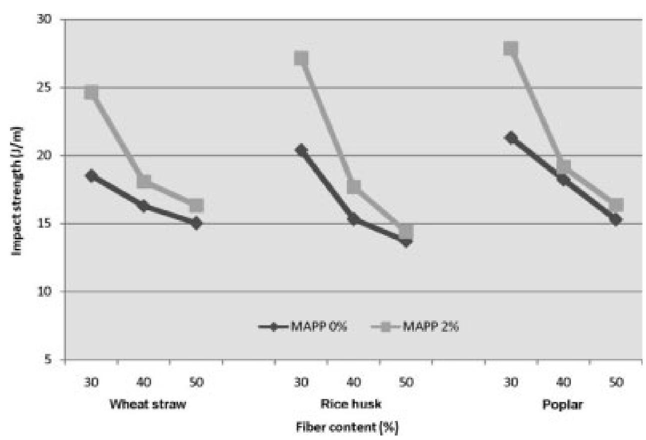

Figure 5 Comparison of tensile strength for various fibers with different fiber loading.

quite accepted for wood plastic composite and have been reported by many researchers.[4,7,8,15,16]

CONCLUSIONS

Ecological concerns and the impending depletion of wood forest are driving the development of new bio-based, green products. Agro-residues are excellent alternative materials to substitute wood, because they are plentiful, widespread, and easily accessible. Aside from their abundance and renewability, utilization of agro-residues has advantages for economy, environment, and technology. The main objective of this research was to study the potential of agro-residues such as wheat straw and rice husk as reinforcements for thermoplastics as an alternative to wood fibers.

Incorporation of wheat straw into PP, despite the addition of coupling agent, gave higher values of tensile and flexural strength than did rice husk, and poplar indicating a good interfacial interaction between wheat straw and PP. This is in accordance with the more hydrocarbon-rich wheat straw surface made the straw more compatible with that of nonpolar PP compared to other fibers, which leads to better stress transfer from the polymer to fiber. Rice husk fiber-reinforced composite exhibited better notched Izod impact strength compared to that of composite with that of wheat straw, while it showed comparable values with that of poplar fiber-filled composites.

The effect of coupling agent on the mechanical behavior of composites with different fibers was investigated. The coupling agent used offered improved tensile and flexural strength properties relative to the composites without coupling agent and is attributed to the improvement in fiber matrix adhesion in the presence of coupling agent. Though flexural strength showed an improvement in the case of composites with wheat straw and wood fiber, no noticeable difference was observed in composites with rice husk and poplar.

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