Mechanical Performance of Hybrid Rice Straw/Sea Weed Polypropylene Composites

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ABSTRACT: Rice straw (Rs)/polypropylene (PP) composites were prepared in the different ratio of 5:95, 10:90, 15:85, 20:80, 25:75, and 30:70 (Rs wt % : PP wt %) by an injection molding process. This work investigated the tensile strength (TS), bending strength (BS), and impact strength (IS) of the composites. From the results, it is observed that Rs20 : PP80 mixture composite showed better performance with mechanical properties (TS = 26.2 MPa, BS = $58 N/\text{mm}^2$, and IS = 1.7 KJ/mm^2) among the composites prepared. Two hybrid composites were also fabricated using 20% Rs, 10% seaweed with 70% PP and

INTRODUCTION

Natural fibers have recently attracted the attention of scientists and technologists because of the advantages that these fibers provide over conventional reinforcement materials. The natural fibers reinforced thermoplastic composites have gained importance in various applications such as building materials and automotive components. The natural fibers offer advantages of large quantity, annual renewability, low cost, lightweight, competitive specific mechanical properties, reduced energy consumption, and environmental friendliness. The fibers used to reinforce thermoplastics mainly include wood, cotton, flax, hemp, jute sisal, and sugarcane fibers.^{1–3} Rice straw (Rs) fiber can also be considered as important potential reinforcing filler for thermoplastic composite because of its ligno-celluloses characteristics.⁴⁻⁷ Food and Agricultural Organization reported that global paddy production reached 628 millions in 2005 with an additional one percent increase in 2006. The United States rice production in 2006/2007 was at 10 millions tons.⁸ With an approximate rice-to-straw ratio of 1.0, an equivalent amount of Rs (i.e., 10 million tons) was produced and about 230 million tons of Rs is generated annually in central and southern China.9 Chemically, lignocel20% Rs, 30% seaweed with 70% PP. In between the two hybrid composites, superior mechanical behavior showed by the hybrid composite in ratio of Rs20 : Sw10 : PP70 with enhanced results such as TS = 28 MPa, BS = 68 N/mm², and IS = 2.5 KJ/mm². Water uptake, simulating weathering, and soil degradation test of different composites were also performed. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 120: 1843–1849, 2011

Key words: biofibers; composites; injection molding; mechanical properties

luloses Rs fiber has similar compositions as other natural fibers used in thermoplastics. One of the physical properties of these natural fibers is their hydrophilic characteristic. The hydrophilic lignocelluloses fibers do not adhere well to the hydrophobic thermoplastics used as matrix materials. However, the inherent incompatibility of hydrophilic cellulose fibers with hydrophobic matrix like polypropylene (PP) thermoplastic usually yields poor interfacial adhesion, which results in impaired properties to the final products. Much work has been done to improve the interfacial adhesion for efficient transfer of stress from the matrix to the fibers, including physical methods¹⁰⁻¹² (such as corona or plasma discharges) as well as chemical methods^{13–18} (pretreatment of fiber surfaces by coupling agents, such as silanes and isocyanates, and/or modification of the matrix by grafting with reactive moieties, such as acrylic acid, acrylic esters, maleic anhydride, etc.)^{19–25} but much work has yet to be done to guarantee reliability to natural fiber composites. Polyolefin polymers, such as PP, are widely used in automotive industry or for domestic applications when ductility and low cost have to be combined.²⁶⁻²⁷ In addition, natural fibers as fillers are now extensively used in the plastics industry to achieve desired properties or to reduce the price of the finished article. Therefore, sustainable development is an important concept underlying many of today's renewable resource policies.

There is a recent idea of hybrid fiber reinforced composites which has been pursued for a series of material combinations before. By using hybrid composites made

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 TABLE I

 Average Chemical Composition of the Rice Straw as

 Determined by Fiber Analysis Method²³

	5
Composition	Average amount (%)
Cell content	21
Hemi cellulose	26
Cellulose	33
Lignin	7
Silica	13
Elements	
Calcium	0.19
Potassium	1.2
Magnesium	0.11
Sulphur	0.10

of natural fibers and carbon fibers or natural fibers and glass fibers, the properties of natural fibers reinforced composites can be improved significantly.^{17–23} The advantages of natural fiber incorporated hybrid composites are reported by Clark and Ansell.²² Sisal and glass are good examples of hybrid composites^{24,25,28-30} possessing very good combined properties. Jute/cotton woven fabric reinforced polyester composite,³¹ sisal/ sawdust hybrid composites,³² hybrid composite and wood flour/fiber reinforced thermoplastic composites, and bamboo/glass fiber reinforced polymer composites³³ were reported. Our earlier work reported on exploratory study on seaweed as novel filler in PP composite and observed the effect of thermoplastic elastomer as additives and maleic anhydride treatment on seaweed in which results corroborate the preparation of hybrid composites belongs to enhanced mechanical performance.^{34,35} This is the extension work on the preparation of hybrid composites using seaweed and Rs due to its high silica content and very much abundance in nature, which hopefully possess good mechanical properties and sufficient water resistance and biodegradability that could be extensively used in so many structural applications.

EXPERIMENTAL

Material

PP (Sabic, 579S, Melt Flow Index (MFI) (230, 2.16 kg) = 47 g/10 min) was used as matrix polymer. Rs and seaweed (Sw) were dried at 80°C in a vacuum oven for 24 h before the preparation of the composites. The chemical composition of the Rs³⁶ is presented in Table I.

Composite fabrication

Composites were prepared by compounding with extrusion and subsequent processing with injection molding. The PP matrix and the fillers were taken in different weight fractions (Table II). Composites were prepared by passing the mixtures through a twinscrew extruder (L/D = 20, LEISTRITZ, GmbH,

Germany) at 175°C, screw rotation speed 80 rpm (passing time in the barrel \sim 30 s).

The extruded composites were cut into small pieces of 15–20 cm length. The small pieces were then crashed into small granules using a grinding mill (Herbold SML 180/100). The granulated products were dried in an oven at 105°C for 24 h and used in an injection molding machine (BOY 30A, Germany) under 180/175/175/175°C, mold temperature 40°C, cooling time 45 s, injection and molding pressure 60 bar for making specimen. The dried granulated products were molded as per ISO 527-2 Type 1A standard specimen.

Determination of mechanical properties of the composites

To investigate the mechanical properties of the prepared composites, mechanical test properties like tensile, bending, and impact tests were performed. Tensile testing was done using dumbbell shaped injection molded specimen on a Zwick 1446 universal testing machine according to ISO 527-1. Bending testing was also done on the machine Zwick 2201 following ISO 178 methods. Impact strength (IS) of samples was measured on the machine Zwick 5102 according to ISO 179. All testing were done at room temperature 20°C. At least five samples were tested for each composition and results were averaged.

Water absorption test

Samples of a dumbbell specimen were used for the measurements of water absorption. After being oven dried at 105°C for 24 h, the specimens were kept in the desiccators using silica gel at room temperature. Then the specimens were weighed before being immersed in distilled water. The mass was recorded as the mass of samples before immersion. The specimens were periodically taken out of the water, surface

 TABLE II

 Relative Amount (% wt) of Reinforcing Materials and Polymer Matrix in Composites

Fillers as reinforcing materials (%)	Polymer matrix (%)	Composites		
None	PP: 100	PP		
Rice straw: 5	PP: 95	Rs5		
Rice straw: 10	PP: 90	Rs10		
Rice straw: 15	PP: 85	Rs15		
Rice straw: 20	PP: 80	Rs20		
Rice straw: 25	PP: 75	Rs25		
Rice straw: 30	PP: 70	Rs30		
Rice straw: 20, Seaweed: 10	PP: 70	Rs20Sw10		
Rice straw: 20, Seaweed: 20	PP: 60	Rs20Sw20		
Seaweed: 30	PP: 70	Sw30		



Figure 1 TS of the composites.

dried with absorbent paper, reweighed, and immediately put back into the water. Water absorption was calculated according to ASTM D 5229/D 5229/M-92.

Simulated weathering test

The composites were treated by using a simulated weathering tester from Q-Lab, Cleveland, USA (model Q.U.V.). The weathering test was performed in alternating cycles of sunshine >4 h ($65 \pm 2^{\circ}$ C) and dews and condensation 2 h ($65 \pm 2^{\circ}$ C). This treatment was carried out for a period of ~ 720 h.

Soil degradation test

Cellulose possesses the tendency to be degraded when buried in soil (having at least 25% moisture). For this purpose, the composite samples are weighed individually and buried in soil for 1–16 weeks. After the period, samples are withdrawn carefully, washed with distilled water, dried at



Figure 2 Eb (%) of the composites.



105°C temperature for 20 min, kept at room for 24 h, and weight is recorded. Finally, weight loss of various degraded samples is calculated.

RESULTS AND DISCUSSION

Mechanical properties of Rs PP composite

Tensile strength and elongation at break

The tensile properties like tensile strength (TS) and elongation at break (Eb) of the Rs (as filler) PP composites containing 5, 10, 15, 20, 25, and 30% filler were studied and the results are presented in Figures 1 and 2, respectively. It is observed that with an increase of filler content from 5 to 20% the TS gradually increased but the TS of the composites are found to insignificant decrease with increasing filler loading by weight fraction from 25-30% (W/W). This may be due to the lack of stress transfer from the PP matrix to filler. The Eb of the composites





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Modulus of the composites [N/mm²]

2300

2100

1900

1700

1500

1300

0

-BM

Figure 5 Tensile modulus (TM) and bending modulus (BM) of the composites.

20

Rice straw content [%] in the composites

30

40

10

shows the similar trend as shown as TS performance and maximum Eb obtained for 20% Rs sample. An increase of the Eb of the composites increases the toughness and ductility of the composite.³⁷ The composites might be characterized by a moderate physical-mechanics adhesion (better known as interdiffusion) that allows a kind of bonding between two polymeric surfaces via diffusion of the macromolecules of both polymers.³⁸ But further research is needed to clarify the probable type of bonding between the two polymeric surfaces.

Bending strength

The bending strength (BS) of the different composites are depicted in Figure 3 in which like the enhancement of TS the similar trends for BS enhancement of the composites have observed. The most obvious reason for the identical results of the tensile and bending properties is due to Rs content in different proportions. BS increases up to 20% Rs content and thereafter remains constant.



Figure 6 Mechanical Properties of the hybrid composites.



Figure 7 BS and IS of the hybrid composites.

Impact strength

Natural fibers play an important role in the IS of the composites as they interact with the crack formation in the matrix and act as stress transferring medium. Many studies have been reported on the impact behavior and factors affecting the IS of laminated composite materials.³⁹⁻⁴¹ On considering the IS of the composites as shown in Figure 4, an increasing trend with increasing Rs content from 10 to 20% is observed, followed by a decreasing trend with insignificant change. It might be noted that the optimum filler content (highest IS) varies with the nature of filler and matrix, filler aspect ratio, filler/matrix interfacial adhesion, etc. The lower value at high filler content may be due to the presence of so many filler ends in the composites, which could cause crack initiation and hence potential composite failure.^{42,43}

Tensile and bending modulus

The tensile and bending modulus of the Rs PP composites are increasing with increasing filler loading



Figure 8 Eb (%) of the hybrid composites.





Figure 9 Water uptake of the composites.

as compared to 100% PP (Fig. 5). Although filler loading >20% Rs has adverse effect on TS, at the same time, it has direct proportional effect on tensile modulus and bending modulus. Both effects may be due to the high stiffness and the presence of silica in the Rs fibrils. Moreover, being nonpolar thermoplastics matrix, the fibers are not compatible as such; however, they impart their properties along with that of the matrix.

Hybrid composites

On linkage of our earlier works, seaweed (Sw) has been chosen as another filler to prepare hybrid composite with Rs in PP and thereafter two hybrid composites were prepared namely Rs20Sw10 and Rs20Sw20 by the formulation of 20% Rs, 10% Sw with 70% PP and 20% Rs, 20% Sw with 60% PP, respectively. There is an enhancement of the tensile behavior exhibited by the hybrid composites, which is depicted in Figure 6. The highest TS (28 MPa), maximum Eb value (50%) and Young's modulus (1.7 GPa) could be produced by the hybrid composite Rs20Sw10 and similar trend also showed by the other composite Rs20Sw20. It is also apparent from the result presented in Figure 7, which proved that the BS (68 N/mm²) and IS (2.5 kJ/mm²) values of the Rs20Sw10 hybrid composites are higher than that of the other composites. The property enhancement of the hybrid composites may be caused due to better compatibility of the two fibers and PP matrix. The remarkable behavior on Eb of the hybrid composite shown in Figure 8 might be due to increased fiber/matrix adhesion and better fiber dispersion which demonstrated hybrid reinforcing effect as well as the positive hybrid effect.⁴⁴ Additional research demands the interpretation of the activities of hybridization in the composite during extrusion and injection molding process.

Water absorption test

Water absorption capacity is an important characteristic of composite material, which determines the end use application of composite. The results of water uptake are shown in Figure 9 as water uptake versus soaking time. As expected the samples uptake the highest amount of water compared to the other samples depending on the filler loading in the composites. The lower water uptake of the Rs PP composite may be attributed to the fact of water retention capacity due to presence of silica in Rs. However the lowest uptake of water by the composites indicate that more OH group of cellulose content in the fibers of the composites being blocked by their interaction with the PP matrix, hence hindering them from being accessed by water.^{35,44}

Simulating weathering effect

The three types of composites such as Rs PP composite (Rs20), seaweed PP composite (Sw30), and Rs/seaweed PP hybrid composite (Rs20Sw10) were exposed to accelerating weathering tester over a period of about 720 h to study the degradation properties. The loss of weight and tensile properties (TS and Eb) of the samples due to weathering is shown in Table III. The loss of TS of the Sw30 composite is about 8%, whereas that of Rs20 is ~ 7% and for the hybrid composite Rs20Sw10 is ~ 5%. Similarly, Eb

 TABLE III

 Loss of Weight and Mechanical Properties of the Composites due to Simulating Weathering

	0		-		-		0	0		
	l.	Weight loss (%)			Loss of TS (%)			Loss of Eb (%)		
Weathering time (h)	Sw30	Rs20	Rs20Sw10	Sw30	Rs20	Rs20Sw10	Sw30	Rs20	Rs20Sw10	
24	3 ± 1	2.5 ± 1	2 ± 0.7	1 ± 0.3	0.9 ± 0.2	0.9 ± 0.1	2 ± 0.2	2 ± 0.2	1.5 ± 0.1	
48	5 ± 1	4 ± 1	1.5 ± 0.7	2 ± 0.3	1.2 ± 0.2	1 ± 0.1	4 ± 0.2	3 ± 0.2	2 ± 0.1	
72	7 ± 1	6 ± 1	5 ± 0.7	3.5 ± 0.3	1.8 ± 0.2	1.2 ± 0.1	7 ± 0.2	6 ± 0.2	5 ± 0.1	
96	9 ± 1	7 ± 1	6 ± 0.7	4 ± 0.3	2 ± 0.2	1.9 ± 0.1	10 ± 0.2	10.2 ± 0.2	8 ± 0.1	
120	12 ± 1	9 ± 1	6.5 ± 0.7	4.2 ± 0.3	2.6 ± 0.2	2 ± 0.1	15 ± 0.2	16 ± 0.2	13 ± 0.1	
240	13 ± 1	11 ± 1	8 ± 0.7	4.5 ± 0.3	3.5 ± 0.2	3 ± 0.1	20 ± 0.2	19 ± 0.2	16 ± 0.1	
480	15 ± 1	14 ± 1	9 ± 0.7	6.5 ± 0.3	4.5 ± 0.2	4.2 ± 0.1	$24~\pm~0.2$	22 ± 0.2	18 ± 0.1	
720	$18~\pm~1$	16 ± 1	$10~\pm~0.7$	8 ± 0.3	7 ± 0.2	5 ± 0.1	26 ± 0.2	24 ± 0.2	20 ± 0.1	

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TABLE IV								
Loss of Weight and Mechanical	Properties of th	e Composites	due to Soi	I Degradation				

Degradation time (weeks)	Weight loss (%)		Loss of TS (%)			Loss of Eb (%)			
	Sw30	Rs20	Rs20Sw10	Sw30	Rs20	Rs20Sw10	Sw30	Rs20	Rs20Sw10
1	4 ± 2	2 ± 1	1.5 ± 0.8	3 ± 0.4	3 ± 0.3	2.3 ± 0.3	4 ± 0.4	4 ± 0.2	3.5 ± 0.2
2	6 ± 2	5 ± 1	3 ± 0.8	7 ± 0.4	6.2 ± 0.3	4 ± 0.3	6.2 ± 0.4	6 ± 0.2	4 ± 0.2
4	8 ± 2	9 ± 1	5 ± 0.8	11 ± 0.4	9 ± 0.3	7.2 ± 0.3	10 ± 0.4	12 ± 0.2	9 ± 0.2
8	11 ± 2	10 ± 1	8 ± 0.8	12 ± 0.4	11 ± 0.3	9 ± 0.3	14 ± 0.4	13 ± 0.2	12 ± 0.2
16	12 ± 2	11 ± 1	9 ± 0.8	14 ± 0.4	12 ± 0.3	10 ± 0.3	$21~\pm~0.4$	20 ± 0.2	17 ± 0.2

Sw30: composite formulated by 30% seaweed and 70% PP.

Rs20: composite formulated by 20% rice straw and 80% PP.

Rs20Sw10: composite formulated by 20% rice straw, 10% seaweed and 70% PP.

loss is 26% for the Sw30 sample and that of the Rs20 and Rs20Sw10 sample are 24 and 20%, respectively. The simulating weathering experimental result shows that extend of tensile property loss of hybrid composite is less than that of the other composites like Rs20 and Sw30 composites. The observed changes can be attributed to the combination of the breakdown of lignocellulosic material by ultraviolet light and the removal of these material by the water spray, exposing previously in the simulated weathering system.⁴⁵

Soil degradation

The composites such as Rs PP composite (Rs20), sea weed PP composite (Sw30), and Rs/sea weed PP hybrid composite (Rs20Sw10) were buried in soil (25% water) for a period of 16 weeks to study the effect of environmental condition on the degradability of the samples. Weight loss and tensile properties (TS and Eb) of the composite samples were periodically measured, and the results are tabulated in Table IV. The weight loss is minimum for the hybrid composite Rs20Sw10 (9%) as compared with the others composite samples. TS and Eb loss due to degradation is also minimum for the hybrid composite at the maximum period of observation. These results are not surprising since lignocelluloses materials are usually biodegraded quite slowly, due to the structural and chemical complexity of the materials.46

CONCLUSIONS

Composite properties such as TS, Eb, BS, IS, and modulus of Rs PP composites have been studied. It is observed that Rs effectively reinforces in PP matrix in the Rs PP composites. In fact, the composite properties increased with the increase in the fiber loading in the composites due to the reinforcement imparted by the fibers that allowed greater stress transfer at the interface. The results presented in this work indicate that it is possible to enhance mechanical properties of hybrid fiber-reinforced composites through hybridization of Rs and seaweed with PP matrix by extrusion and injection molding process. The results of the water uptake, simulating weathering and soil degradation test of different composites demonstrate that the prepared composites are quite durable in water, soil, and simulated environmental condition, but they could be decomposed by keeping the composites in garden soil after its use.

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