Mechanical Properties of Biorenewable Fiber/Plastic Composites

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ABSTRACT: Plastic fiber composites, consisting of polypropylene (PP) or polyethylene (PE), and pinewood, big blue stem (BBS), soybean hulls, or distillers dried grain and solubles (DDGS), were prepared by extrusion. Young's modulus, tensile and flexural strengths, melt flow, shrinkage, and impact energy, with respect to the type, amount, and size of fiber on composites, were evaluated. Young's moduli under tensile load of wood, BBS, and soybean-hull fiber composites, compared with those of pure plastic controls, were either comparable or higher. Tensile strength significantly decreased for all the PP/fiber composites when compared with that of the control. Strength of BBS fiber composites was higher than or comparable to that of wood. When natural fibers were added there was a significant

INTRODUCTION

Combining agrofibers (lignocellulosics) with other resources provides a strategy for producing advanced composite materials that take advantage of the properties of both types of resources. The use of plantbased fiber as an additive to plastics has accelerated rapidly over the past decade, primarily as a result of improvements in process technology and economic factors. Most plant fibers used in these applications are derived from wood. However, a growing body of research on use of other plant fibers has shown that many biorenewable fibers may also be suitable for fiber/plastic composites. Further development of these applications of biorenewable fibers for use by the plastic industry could provide attractive new value-added markets for agricultural products while simultaneously displacing petrochemical-based plastic resins.

Thermoplastic resin production in the United States increased by approximately 60% from 1988 to 1998. In 2001 the total sales and captive use of selected therdecrease in the melt flow index for both plastic/fiber composites. There was no significant difference in the shrinkage of all fiber/plastic composites compared to that of controls. BBS/PE plastic composites resulted in higher notched impact strength than that of wood or soybean-hull fiber composites. There was significant reduction in the unnotched impact strength compared to that of controls. BBS has the potential to be used as reinforcing materials for low-cost composites. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 93: 2484–2493, 2004

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moplastic resins by major markets in the United States was: 78,645 million pounds (dry weight), a 3% increase from 1997.9 The tremendous growth in thermoplastic production is largely attributable to the fact that these materials are versatile and economical. Nonetheless, manufacturers who use thermoplastics are continually seeking new ways to reduce costs and improve product performance. It is this impetus that has led to the rapid growth of the fiber/plastic composites industry within the past ten years. Typically, blending thermoplastic resins with fibers, fillers, or other additives in an extruder produces fiber/plastic composites. The extrusion process uses controlled heat and shear to effectively blend dissimilar materials. Blended materials may be postprocessed by injection molding or other manufacturing techniques to form final products or parts.

The fiber/plastic composites industry is based on the premise that the addition of lower-cost materials (fillers and reinforcements) to plastic resins will decrease overall materials manufacturing costs and increase stiffness of the material.⁸ Furthermore, many low-cost fillers actually improve certain materials' properties, such as bending strength. Processing advantages such as lower energy consumption and faster cycle times (i.e., greater production rate) are also typical advantages. Most of the fillers currently in use

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are inorganic or synthetic, but biorenewable or natural fibers are also used. The use of fillers by the U.S. plastics industry in 2000 was estimated as 5.5 billion pounds, of which 0.4 billion pounds (7%) were biobased fibers.⁶ Most biofiber plastic additives are derived from wood. However other natural fibers, such as flax or wheat straw are finding their way into the fiber/plastic composites industry. In fact, industry experts believe that the demand for bio-based or natural fibers for these applications will grow at least sixfold in the next 5 to 7 years.¹² The authors believe that further research aimed at the use of other biorenewable fibers, such as soybean hulls and big blue stem grass, would likely follow a development track similar to that for wood-filled plastics. The result may lead to a new market opportunity for a variety of agricultural fiber sources. Agricultural byproducts, already accumulated in significant quantities at central processing locations, have potential as well.

The main objective of this research was to investigate the mechanical properties of the following in fiber/plastic composites: soybean hulls from soybeanprocessing plants, distillers dried grain and solubles (DDGS) from ethanol-processing plants, and big blue stem (BBS) grass from native prairies. Specific objectives were as follows:

- 1. To evaluate the mechanical properties of mixed pinewood–, BBS grass–, DDGS–, or soybeanhull fiber–polypropylene (PP) plastic composites.
- 2. To evaluate the mechanical properties of mixed pinewood–, BBS grass–, DDGS–, or soybeanhull fiber–polyethylene (PE) plastic composites.
- 3. To evaluate the effect of the amount of fiber and the size of fiber has on the mechanical properties of mixed pinewood–, BBS grass–, DDGS–, or soybean-hull fiber–PP plastic composites.
- 4. To evaluate the effect of the amount of fiber and the size of fiber has on the mechanical properties of mixed pinewood–, BBS grass–, DDGS–, or soybean-hull fiber–PE plastic composites.

EXPERIMENTAL

The four different types of fibers used in this study were big blue stem (BBS) grass, mixed pinewood, distillers dried grain and solubles (DDGS), and soybean hulls. BBS was collected from the South Dakota State University Farm Department (Brookings, SD). Mixed pinewood was obtained from American Wood Fibers Inc. (Pella, IA). Soybean hulls were obtained from South Dakota Soybean Processors Inc. (Volga, SD) and DDGS were obtained from Dakota Ethanol (Wentworth, SD).

BBS was ground in a hammer mill (Speedy Jr. Winona Attrition Mill Co., Winona, MN) using a 20

TABLE I Experimental Design for One Type of Fiber

1 habite	Size of fiber	Percentage of fiber
Polypropylene	40 Mesh	20
51 15		30
	60 Mesh	20
		30
	80 Mesh	20
		30
Polyethylene	40 Mesh	20
		30
	60 Mesh	20
		30
	80 Mesh	20
		30
	Polypropylene	Polypropylene 40 Mesh 60 Mesh 80 Mesh Polyethylene 40 Mesh 60 Mesh 80 Mesh

mesh sieve. Soybean hulls and DDGS were ground first using a Wiley mill (Model No. 2; Arthur H. Thomas Co. Philadelphia, PA) followed by a Cyclone Sample Mill (Model 3010-030; UDY Corp., Fort Collins, CO). Wood was obtained already partitioned as 40, 60, and 80 mesh fiber size from the American Wood Fibers, Inc.

Ground fibers were separated using a nest of American Society for Testing Materials (ASTM) standard 40 mesh, 60 mesh, and 80 mesh sieves (Model CL-392-B; Soil Test Inc., Chicago, IL). The nest of sieves was shaken for 10 min. The sieved fibers were oven-dried at 60°C for 16 h to reduce the moisture content between 1.5 to 1.8%. This is done to aid in homogeneous mixing of fibers with plastic during extrusion.

High-density PE plastic beads (1469 PE, melting point: 130–150°C; Exxon Mobil, Houston, TX) and PP plastic beads (8004-ZR PP, melting point: 210–227°C; Equistar Chemicals, Houston, TX) were the two plastics used for this study of biocomposites. Their low melting points allowed processing below the degradation temperature of the fibers. The experimental design was a factorial arrangement of treatments conducted in a randomized design. Table I shows the example outline of the experimental design for one type of fiber. The same design was used for the other fibers as well.

Extrusion

Extrusion was conducted at the Center for Crop Utilization and Research (CCUR), Iowa State University (Ames, IA), using a Leistritz Micro-18 multimode twin-screw extruder (American Leistritz Extruder Corp., Somerville, NJ). The temperatures at the six different heating zones and the screw speeds of the feeder for all the fibers (Schenck AccuRate Co., Whitewater, WI) and the extruder, for each plastic tested, are shown in Table II.

		Temperatures and	d Screw-Sp	eed Setting	;S			
Type of	Barrel screw	Feeder screw	Temp	erature setti	ngs for each (°	heating zor C)	ne ^a of the ex	truder
plastic	speed (rpm)	speed (rpm)	3	4	5	6	7	8
Polyethylene Polypropylene	100 150	105 95	110 140	140 185	145 195	165 205	150 200	145 195

TABLE II

^a Heating zone 3 is closest to feed entry point and zone 8 is closest to the exit die.

The plastic and fiber were mixed well before being placed in the feeder. A pelletizer (Type 12-72-000; C.W. Brabender Instruments, South Hackensack, NJ) was used to form the fiber/plastic biocomposites pellets that were used in the injection molder.

Injection molding

Dumbbell-shaped test samples were prepared using an injection-molding machine (Model 22S Dipronic; Dr. Boy GmbH and Co., KG, Neustadt-Fernthal, Germany). The temperatures at the entrance and tip of the injector are listed in Table III.

Conditioning of samples

All the samples were conditioned in a conditioning chamber (Model STC-III; Sanplatec Corp., Osaka, Japan) for 48 h at a constant relative humidity of 52% and temperature of 25°C before performing the tensile, flexural, and impact tests.

Tests

Five samples were tested for each blend. The tensile test was conducted in compliance with ASTM Standard D 638,¹ using an Instron Testing Machine (IX series, Model 4500; Instron Corp., Canton, MA). The flexural test was conducted in compliance with ASTM Standard D 790,² using an Instron testing machine. The melt flow index (MFI) test was conducted in compliance with ASTM Standard D 1238,⁴ using a Melt Indexer (Dynisco, Kayeness Polymer Test Systems, Morgantown, PA). The parameters for the MFI test are shown in Table IV. The shrinkage test was conducted in compliance with ASTM Standard D 955,⁵ using the

TABLE III **Temperature Zones for Injection Molding**

1	,	0
	Temperature (°C)	
Plastic	Entrance	Tip
Polypropylene	185	195
Polyethylene	150	165

injection-molder. The impact test was conducted in compliance with ASTM D 256.³

RESULTS AND DISCUSSION

Fifty different types of biocomposite blends were prepared and their Young's modulus, tensile strength, flexural strength, shrinkage, MFI, and impact energy were measured. SAS Institute's general linear model (GLM)¹¹ was used to perform the statistical analysis. Least-square means of the mechanical properties were evaluated for all mesh sizes and fiber levels of different fiber composites. (In Tables V-XVIII, numbers having different letters are significantly different at the 0.05 level.) While DDGS was being milled to a smaller size it formed a hard layer, which blocked the sieves. As a result we were unable to collect all the mesh sizes of DDGS to blend with both PP and PE plastics. We collected enough 40 mesh size fiber to blend with PP and PE. Therefore we evaluated fibers of wood, BBS, soybean hulls, and DDGS of 40 mesh size.

Tensile test

Wood and BBS composites generally exhibited better properties than those of soybean hulls and DDGS composites (Tables V and VI). Additionally, increasing the fiber content increased the tensile modulus but decreased or did not change the tensile strength. The Young's modulus and stress at maximum load were the tensile properties analyzed under tensile load. Young's modulus increased significantly, when wood, BBS, or soybean hull fibers were added to plastic, compared to that of the control. In a comparison of all

TABLE IV Parameters for Melt Flow Test

	Plas	stic
Parameter	Polypropylene	Polyethylene
Temperature, °C	230	190
Melt time, s	240	240
Load, g	2060	2060
Cut time, s	30	30

TABLE V
Least-Square Means of Young's Modulus, Under Tensile
Load, at 40, 60, and 80 Mesh Sizes and 20 and 30% Fiber
Levels of Different Fiber/PP Composites

Tensile test, PP, Young's modulus $ imes$ 10 ² , MPa					
		Size of the fiber, mesh			
Fiber type	Percentage	40	60	80	
Wood	20	10.52 ^{d,e}	10.96 ^d	10.37 ^e	
Wood	30	13.02 ^{a,b}	13.07 ^{a,b}	13.28 ^a	
BBS	20	12.24 ^c	10.40 ^e	9.68^{f}	
BBS	30	13.21 ^{a,b}	12.71 ^{b,c}	12.30 ^c	
Soybean	20	7.89 ^g	7.81 ^{g,h}	6.58 ^j	
Soybean	30	7.76 ^{g,h}	7.69 ^{g,h}	7.65 ^{g,h}	
DĎGS	20	6.97 ^{i,j}	NA	NA	
DDGS	30	7.36 ^{h,i}	NA	NA	
Control	0	6.97 ^{i,j}	6.97 ^{i,j}	6.97 ^{i,j}	

the composites wood 80 mesh size fiber and 30% fiber content resulted in the highest modulus: 1330 MPa for PP (Table V). BBS 40 mesh size fiber and 30% fiber content resulted in the highest modulus: 1120 MPa for PE (Table VI).

Young's modulus increased for both types of the plastic, as the percentage of all fibers increased from 20 to 30% fiber content, with the exception of PP/ soybean-hull composites. This increase can be attributed to the increase in volume fractions of high-modulus fibers in plastic composites.¹⁴ A reduction in BBS fiber size resulted in a greater decrease in Young's modulus than that from a reduction in wood fiber size for PP blends. This may be attributable to the aspect ratio of BBS fiber becoming closer to that of wood at the smaller sizes. The effect of size was not significant at 30% fiber content when wood or soybean hulls were added to PP plastic. This indicates any size of these two fibers can be used, depending on availability, and not affect the Young's modulus of composites. The addition of BBS 40 mesh fiber size to PE resulted in a significantly higher Young's modulus than that of wood 40 mesh fiber size for both 20 and 30% fiber content and for 20% fiber content in PP composites. This indicates BBS can replace wood at 40 mesh fiber size and at both 20 and 30% fiber content in PE and 20% fiber content in PP with no effect on the composites' Young's modulus.

Wood flour and other low-cost agricultural-based flour can be considered as particulate fillers that enhance the tensile moduli. Sanadi et al.¹⁰ found similar results using agricultural-based flour as particulate filler in plastics and the specific Young's modulus with natural fibers such as kenaf was significantly higher than that of wood fibers.

Wood 80 mesh fiber size and 20 or 30% fiber content is not significantly different from BBS 80 mesh fiber 2487

size and 20 or 30% fiber content, respectively, for PE plastic. This may be because the aspect ratio of both fiber types became more nearly equal to each other as the fiber size is reduced. This indicates BBS can be used as a substitute for wood at 80 mesh fiber size with no effect on the composites' Young's modulus. DDGS 40 mesh fiber size and 20 or 30% fiber content resulted in higher or comparable Young's modulus, compared with that of both PP and PE plastic controls (Tables V and VI). This indicates DDGS at 40 mesh and 20 or 30% fiber content can be used as a filler in plastics, resulting in comparable or higher Young's modulus. Wood, soybean hulls, and BBS fiber at 40 and 60 mesh fiber size, when combined with PP, are not significantly different in Young's modulus at 20 and 30% fiber content, except for BBS at 20% fiber content. This indicates we can use either 40 mesh fiber size or 60 mesh fiber size, depending on the availability, either the 20 and 30% fiber content level for wood, soybean hulls, or BBS.

Stress at maximum load was considered tensile strength. In a comparison of all the composites, BBS 40 mesh size fiber and 20% fiber content resulted in the highest tensile strength, 34.56 MPa (Table VII), when combined with PP. BBS 40 mesh size fiber and 30% fiber content resulted in the highest tensile strength, 22.62 MPa, when combined with PE (Table VIII). Tensile strength significantly decreased for all the PP composites, compared with that of the control (Table VII). The trend of lower strength may be attributable to the higher melt temperature required for the PP plastic (Table II), thus resulting in increased pyrolytic degradation. Wood and BBS fiber composites exhibited comparable or higher tensile strength, compared with that of PE plastic control, except BBS 80 mesh fiber size and 30% fiber content. PE/soybean-hull fiber composites resulted in significantly lower tensile strength

TABLE VI Least-Square Means of Young's Modulus, Under Tensile Load, at 40, 60, and 80 Mesh Sizes and 20 and 30% Fiber Levels of Different Fiber/PE Composites

		1	
le test, PE, Youn	ıg's modulus	\times 10 ² , MPa	
	Size o	of the fiber, 1	nesh
Percentage	40	60	80
20	6.99 ^f	7.07 ^f	7.11 ^f
30	8.54 ^d	9.77 ^b	9.03 ^c
20	9.36 ^{b,c}	7.30 ^f	7.14^{f}
30	11. 2 4 ^a	8.08^{e}	9.13 ^c
20	5.48 ^h	5.00 ⁱ	4.33 ^j
30	6.30 ^g	5.36 ^{h,i}	5.56 ^h
20	3.92 ^{j,k}	NA	NA
30	4.25 ^j	NA	NA
0	3.60 ^k	3.60 ^k	3.60 ^k
	Percentage 20 30 20 30 20 30 20 30 20 30 20 30 20 30 20 30 20 30 20 30 20 30 0	$\begin{tabular}{ c c c c c c } \hline le test, PE, Young's modulus \\ \hline \hline le test, PE, Young's modulus \\ \hline \hline Percentage & 40 \\ \hline 20 & 6.99^{f} \\ 30 & 8.54^{d} \\ 20 & 9.36^{b,c} \\ 30 & 11.24^{a} \\ 20 & 5.48^{h} \\ 30 & 6.30^{g} \\ 20 & 3.92^{j,k} \\ 30 & 4.25^{j} \\ 0 & 3.60^{k} \\ \hline \end{tabular}$	$\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ Size of the fiber, 1}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ Size of the fiber, 1}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$ $\frac{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}{ e \text{ test, PE, Young's modulus} \times 10^2, \text{ MPa}}$

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TABLE VII
Least-Square Means of Tensile Strength at 40, 60, and 80
Mesh Sizes and 20 and 30% Fiber Levels of Different
Fiber/PP Composites

Tensile test, PP, tensile strength, MPa							
		Size	Size of the fiber, mesh				
Fiber type	Percentage	40	60	80			
Wood	20	33.05 ^{c,d}	33.15 ^c	29.09 ^f			
Wood	30	31.62 ^e	32.42 ^d	28.67 ^{f,g}			
BBS	20	34.56 ^b	32.56 ^{c,d}	28.98^{f}			
BBS	30	32.61 ^{c,d}	31.63 ^e	27.39 ^h			
Soybean	20	28.09 ^{g,h}	27.53 ^h	25.51 ⁱ			
Soybean	30	23.18 ^k	23.98 ^j	23.40 ^{j,k}			
DĎGS	20	27.80 ^h	NA	NA			
DDGS	30	24.90^{i}	NA	NA			
Control	0	36.68 ^a	36.68 ^a	36.68 ^a			

than that of the control, BBS, and wood fiber plastic composites across all fiber sizes and levels. Tensile strength either decreased or was comparable as the percentage of wood, BBS, or soybean-hull fiber increased from 20 to 30% fiber content for both plastics evaluated. This decrease could be a result of the decrease in plastic matrix material as the fiber content increases.

BBS 40 mesh size fiber and 20% fiber content has significantly higher tensile strength than that of wood at both 20 and 30% of fiber content (Table VII) for both types of plastic composites. This indicates we can replace 30% wood by 20% BBS, which results in higher tensile strength. Size has no significant effect when 20% soybean-hull fiber was added to both types of plastics, except when 80 mesh size soybean hulls were added to PP (Tables VII and VIII). This can be attributed to the aspect ratio of the particles; even as the

TABLE VIII Least-Square Means of Tensile Strength at 40, 60, and 80 Mesh Sizes and 20 and 30% Fiber Levels of Different Fiber/PE Composites

		-				
	Tensile test, PE,	, tensile stre	ngth, MPa			
		Size	Size of the fiber, mesh			
Fiber type	Percentage	40	60	80		
Wood	20	21.36 ^d	22.39 ^{a,b}	21.10 ^{d,e}		
Wood	30	20.59 ^e	22.75 ^a	20.64 ^e		
BBS	20	22.47 ^{a,b}	22.05 ^{b,c}	20.61 ^e		
BBS	30	22.62 ^a	21.56 ^{c,d}	19.21^{f}		
Soybean	20	16.98 ^{g,h}	17.17 ^g	16.95 ^{g,h}		
Soybean	30	16.22 ⁱ	14.87 ^j	14.25^{k}		
DĎGS	20	16.95 ^{h,i}	NA	NA		
DDGS	30	14.25^{k}	NA	NA		
Control	0	21.05 ^{d,e}	21.05 ^{d,e}	21.05 ^{d,e}		

Note: Numbers having different letters are significantly different (p < 0.05); NA, not available.

TABLE IXLeast-Square Means of Young's Modulus Under FlexuralLoad at 40, 60, and 80 Mesh Sizes and 20 and 30% FiberLevels of Different Fiber/PP Composites

Flexural test, PP, Young's modulus $ imes$ 10 ² , MPa					
		Size o	Size of the fiber, mesh		
Fiber type	Percentage	40	60	80	
Wood	20	15.28 ^k	17.04 ^{g,h}	17.30 ^g	
Wood	30	19.59 ^e	21.28 ^d	22.83 ^c	
BBS	20	19.60 ^e	17.02 ^{g,h}	18.78^{f}	
BBS	30	22.61 ^c	23.54 ^b	24.68 ^a	
Soybean	20	16.43 ^{g,h,i}	16.06 ^{i,j}	14.37^{1}	
Soybean	30	16.46 ^{g,h,i}	16.70 ^{g,h,i}	15.83 ^{j,k}	
DĎGS	20	10.20°	NA	NA	
DDGS	30	11.10 ⁿ	NA	NA	
Control	0	12.20 ^m	12.20 ^m	12.20 ^m	

Note: Numbers having different letters are significantly different (p < 0.05); NA, not available.

soybean particle size decreased, the aspect ratio remained identical. This indicates we can select either 40, 60, or 80 mesh size soybean hulls and obtain the same tensile strength in either PP or PE plastic composites.

Flexural test

BBS and wood generally exhibited better flexural properties than those of soybean hulls and DDGS composites. Additionally increasing the fiber content increased the flexural modulus and tensile strength for PE/fiber composites. Moreover, for PP composites, increasing the fiber content increased the flexural modulus and did not change, or only slightly decreased, the flexural strength. Young's modulus increased substantially, compared to that of either the pure PP or PE plastic controls, when wood, soybean hulls, or big blue stem were added to the plastic. In a comparison of all the composites, BBS 80 mesh size fiber and 30% fiber content resulted in the highest Young's modulus: 24.67 MPa for PP plastic (Table IX). BBS 40 mesh size fiber and 30% fiber content resulted in the highest Young's modulus: 14.50 MPa for PE plastic (Table X).

Young's modulus under flexural load significantly increased, or was comparable, for both types of plastic composites as the amount of fiber increased from 0 to 30% fiber content. This can be attributed to the large volume fractions of high-modulus fibers that increase the Young's modulus.¹⁴ This indicates increasing fiber content from 0 to 30% will result in an increase in Young's modulus. Young's modulus is significantly higher for PP composites, using BBS fiber at 30% fiber content, compared to that of all the wood fiber sizes evaluated. This indicates BBS at 30% fiber content at all sizes can be used as a substitute for wood at 30%

TABLE X
Least-Square Means of Young's Modulus Under Flexural
Load at 40, 60, 80 Mesh Sizes and 20% and 30% Fiber
Levels of Different Fiber/PE Composites

Flexural test, PE, Young's modulus $ imes$ 10 ² , MPa					
		Size o	Size of the fiber, mesh		
Fiber type	Percentage	40	60	80	
Wood	20	7.53 ^h	8.32 ^f	7.41 ^{h,i}	
Wood	30	10.37 ^d	12.01 ^b	10.23 ^d	
BBS	20	9.30 ^e	8.52 ^f	8.06 ^{f,g}	
BBS	30	14.45 ^a	11.30 ^c	12.00 ^b	
Soybean	20	7.31 ^{h,i}	5.65 ^k	5.60 ^k	
Soybean	30	7.75 ^{g,h}	6.46 ^j	7.02 ⁱ	
DĎGS	20	4.16^{1}	NA	NA	
DDGS	30	4.52^{1}	NA	NA	
Control	0	4.32^{1}	4.32^{1}	4.32^{1}	

fiber content and result in comparable or higher Young's modulus. There was no significant difference in Young's modulus between PE/wood fiber composites at 40 mesh size and 20% fiber content or PE/ soybean-hull fiber at 40 mesh size and either 20 or 30% fiber content. This indicates wood can be replaced by soybean hulls at 40 mesh fiber size and 20% fiber level. PE plastic/wood composites containing 40 mesh or 80 mesh size and 20 or 30% fiber content levels has no significant effect on Young's modulus. This indicates either 40 mesh or 80 mesh wood fiber can be used at the 20 or 30% fiber content level, depending on the availability, and not affect the composites' Young's modulus. Young's modulus of the PP/DDGS fiber composite was significantly lower than that of all other fiber/PP composites as well as the control. This indicates that the use of DDGS 40 mesh fiber in PP composites reduces the Young's modulus. This could be a consequence of clumping of DDGS particles as they are heated to higher melting point temperature required for PP plastic.¹⁴ PE/DDGS fiber composite at 40 mesh fiber size had a comparable Young's modulus with respect to that of PE controls. This indicates DDGS can be used as a filler in PE/fiber blends at 40 mesh fiber size and still result in comparable Young's modulus.

Stress at yield was recorded as the flexural strength during the flexural test. In a comparison of all the composites, BBS 40 mesh size fiber and 20% fiber content resulted in the highest strength: 59.59 MPa for PP (Table XI). BBS 40 mesh size fiber and 30% fiber content resulted in the highest strength: 36.27 MPa for PE (Table XII). There was significantly higher or comparable flexural strength compared to that of the pure PP plastic control, when either wood or BBS was added, across all fiber sizes and fiber content (Tables XI and XII). There was a significant increase in flexural

TABLE XI Least-Square Means of Flexural Strength at 40, 60, and 80 Mesh Sizes and 20 and 30% Fiber Levels of Different Fiber/PP Composites

Flexural test, PP, flexural strength, MPa				
		mesh		
Fiber type	Percentage	40	60	80
Wood	20	51.95 ^g	54.71 ^{c,d}	53.64 ^{d,e,f}
Wood	30	52.50 ^{f,g}	54.30 ^{c,d,e}	53.52 ^{e,f}
BBS	20	59.59 ^a	55.20 ^c	54.19 ^{c,d,e}
BBS	30	56.91 ^b	57.56 ^b	51.55 ^g
Soybean	20	51.72 ^g	51.42 ^g	48.15 ^h
Soybean	30	46.50^{i}	47.06 ^{h,i}	44.95 ^j
DĎGS	20	42.60 ^k	NA	NA
DDGS	30	41.70 ^k	NA	NA
Control	0	52.20 ^g	52.20 ^g	52.20 ^g

Note: Numbers having different letters are significantly different (p < 0.05); NA, not available.

strength of PE plastic when any type of fiber was added at any fiber size and fiber content (Table XII).

Wood/PP composites, 20 or 30% fiber content, were not significantly different at any fiber size. This indicates we can select any wood fiber size and can obtain comparable flexural strength. BBS/PP composites containing either 40 or 60 mesh fiber size at 30% fiber content resulted in significantly higher strength than that of wood 40 or 60 mesh fiber size at 20 and 30% fiber content (Table XI). Again, this may be attributable to a higher aspect ratio for BBS versus wood at those fiber sizes. This indicates wood can be replaced by BBS at 40 or 60 mesh fiber sizes at 30% fiber content and yet result in significantly higher flexural strength.

Wood flour and other low-cost agricultural-based flour can be considered as particulate fillers that enhance the flexural moduli. Sanadi et al.¹⁰ found similar results using agricultural-based flour as particulate

TABLE XII Least-Square Means of Flexural Strength at 40, 60, and 80 Mesh Sizes and 20 and 30% Fiber Levels of Different Fiber/PE Composites

		-		
Fl	exural test, PE, f	flexural strei	ngth, MPa	
		Size	of the fiber,	mesh
Fiber type	Percentage	40	60	80
Wood	20	29.12 ^f	30.67 ^e	28.44 ^g
Wood	30	31.59 ^d	33.81 ^b	31.45 ^d
BBS	20	32.22 ^c	31.55 ^d	29.54^{f}
BBS	30	36.26 ^a	33.79 ^b	31.25 ^d
Soybean	20	25.83 ^h	23.23 ^j	22.62 ^k
Soybean	30	27.31 ⁱ	23.24 ^j	23.13 ^j
DĎGS	20	20.60 ^m	NA	NA
DDGS	30	20.20 ^m	NA	NA
Control	0	21.90^{1}	21.90^{1}	21.90^{1}

filler in plastics and the specific Young's modulus with natural fibers such as kenaf was significantly higher than that of wood fibers.

BBS/PE composites containing 40 mesh size and 20 or 30% fiber content had significantly higher flexural strength than that of wood 40 mesh fiber size and 20 or 30% fiber content (Table XII). This indicates BBS can be used as a substitute for wood at 40 mesh fiber size, although resulting in higher flexural strength. BBS/PE 60 and 80 mesh fiber size at 30% fiber content is not significantly different from wood 60 and 80 mesh fiber size at 30% fiber content. This indicates wood fiber can be replaced by BBS fiber and result in equal or higher flexural strength. There is a significant reduction in flexural strength of BBS/PE composites as the fiber size is reduced from 40 to 60 to 80 mesh size. The aspect ratio may be approaching unity when the sizes of the fibers are reduced. Longer fibers transfer the stress more efficiently, thus improving the flexural mechanical properties of the plastic composites.¹⁴ This is true for both 20 and 30% fiber content levels. DDGS at 40 mesh fiber size resulted in significantly lower flexural strength, compared with that of other fiber blends and controls. This may be because of clumping of the DDGS particles when heated and also its lower fiber content. This indicates DDGS may not be a good fiber in either PP or PE plastic composites for increasing flexural strength.

Melt flow index

When fiber was added there was a significant decrease in the melt flow index for both PP– and PE–fiber composites. The fiber surface is likely to restrict the mobility of the polymer molecules and the entanglements will vary with the type of fiber and the fiber surface characteristics.¹⁵ In a comparison of all the

TABLE XIII Least-Square Means of Melt Flow Index at 40, 60, and 80 Mesh Sizes and 20 and 30% Levels of Different Fiber/PP Composites

Melt flow index test, PP, g/10 min				
		Size o	of the fiber,	mesh
Fiber type	Percentage	40	60	80
Wood	20	7.68 ^j	7.29 ¹	7.53 ^k
Wood	30	5.81 ^p	5.62 ^q	5.61 ^q
BBS	20	8.27 ⁱ	8.41 ^h	7.52 ^k
BBS	30	5.95°	6.22 ⁿ	6.30 ^m
Soybean	20	11.15 ^d	11.03 ^e	10.99 ^e
Soybean	30	10.08^{f}	10.01^{f}	9.82 ^g
DDGS	20	12.60 ^a	NA	NA
DDGS	30	12.50 ^b	NA	NA
Control	0	11.50 ^c	11.50 ^c	11.50 ^c

Note: Numbers having different letters are significantly different (p < 0.05); NA, not available.

TABLE XIV Least-Square Means of Melt Flow Index at 40, 60, and 80 Mesh Sizes and 20 and 30% Levels of Different Fiber/PE Composites

Melt flow index test, PE, g/10 min				
		Size o	of the fiber,	mesh
Fiber type	Percentage	40	60	80
Wood	20	6.40 ^p	8.22^{1}	9.65 ^k
Wood	30	3.59 ^r	4.44 ^q	6.64°
BBS	20	10.17^{j}	10.38^{i}	10.22 ^j
BBS	30	7.08 ⁿ	8.17^{1}	7.75 ^m
Soybean	20	11.95 ^d	14.58 ^c	14.56 ^c
Soybean	30	10.51 ^h	10.93 ^g	11.47 ^e
DĎGS	20	14.70 ^b	NA	NA
DDGS	30	11.30^{f}	NA	NA
Control	0	19.10 ^a	19.10 ^a	19.10 ^a

Note: Numbers having different letters are significantly different (p < 0.05); NA, not available.

PP/fiber blends, PP/DDGS 40 mesh fiber size and 20% fiber content resulted in higher MFI: 12.60 g/10 min (Table XIII). PE/DDGS 40 mesh fiber size and 20% fiber level resulted in higher MFI compared to that of other fiber/PE blends (Table XIV).

Soybean-hull composites had significantly higher melt flow index, for both PP and PE plastic composites, than that of blends containing wood or BBS fiber (Tables XIII and XIV). This could be because of the effect of the heat on the lipids and carbohydrates present in the soybean hulls. In a comparison of wood fiber composites with BBS/plastic composites across all fiber sizes, BBS/plastic had significantly less reduction of MFI, except for the PP 80 mesh fiber size and 20% fiber content composites. This indicates wood can be replaced by BBS, resulting in a higher MFI. There are several economic advantages of having higher melt flow index: decreases in energy costs; reduced cycle times; and, consequently, time savings. Melt flow index significantly increased when DDGS was added to pure PP plastic and significantly decreased when combined with pure PE plastic. This is attributed to the difference in melting temperatures of the two plastics evaluated. The PP/DDGS matrix was exposed to higher temperatures (Table II) than was the PE/DDGS matrix. There was a more dramatic effect on the lipids and carbohydrates, resulting in an increase in the MFI of PP/DDGS composites compared to that of PE/DDGS composites.

Shrinkage test

This shrinkage test method, ASTM D955, is intended to measure uniformity in initial shrinkage from the mold to molded dimensions of either thermoplastic or thermosetting materials when molded by compression, injection, or transfer under specified conditions.

TABLE XV
Least-Square Means of Notched Impact Strength at 40
60, and 80 Mesh Sizes and 20 and 30% Levels of
Different Fiber/PP Composites

Notched impact strength, PP						
		Size	Size of fiber, mesh			
Fiber type	Percentage	40	60	80		
Wood	20	16.13 ^{d,e,f,g}	16.40 ^{d,e,f}	15.53 ^{d,e,f,g}		
Wood	30	13.80 ^{f,g,h,i}	13.067 ^{g,h,i,j}	16.93 ^{d,e}		
BBS	20	31.40 ^a	21.63 ^c	2.27^{k}		
BBS	30	14.87 ^{d,e,f,g,h}	17.47 ^d	0.87^{k}		
Soybean	20	11.13 ^{i,j}	12.07 ^{h,i,j}	21.83 ^c		
Soybean	30	11.13 ^{i,j}	10.53 ^j	21.67 ^c		
DĎGS	20	13.90 ^{e,f,g,h,i}	NA	NA		
DDGS	30	16.60 ^{d,e,f}	NA	NA		
Control	0	26.80 ^b	26.80 ^b	26.80 ^b		

The shrinkage was less in the composites than that in the pure plastic, although the difference was not significant. The lower amount of shrinkage may be explained by the smaller thermal coefficient of expansion of wood, or any other fiber, compared to that of plastic.⁷

Impact test

The agro-based fiber composites showed Izod impact notched properties comparable to those of wood flour composites. With respect to notched tests, the impact strength increases with the amount of fibers up to a 30% fiber content. In the case of unnotched impact values of composites, the presence of the fibers decreases the energy absorbed by the specimens. Addition of the fibers creates regions of stress concentrations such as fiber ends, defects, and at the interface region that require less energy to initiate a crack. The impact strength can be increased by providing strong and flexible interface regions in the composite or by using impact modifiers such as MAPP (maleic anhydride grafted polypropylene).¹⁰

Notched impact strength

BBS fiber composites exhibited higher notched impact strength than that of wood fiber composites. The formulation of BBS 40 mesh size fiber and 20% fiber content resulted in the highest impact strength for both plastics: 31.40 J/m for PP (Table XV) and 34.00 J/m for PE (Table XVI). Compared with the pure plastic, the addition of fiber (with the exception of BBS 40 mesh and 20% fiber content) resulted in a significant decrease in their notched impact strength.

No significant difference in notched impact strength of PP composites occurred as the percentage of wood or soybean-hull fiber was increased, as the fiber size changed, except wood at 60 mesh. This indicates we can obtain comparable notched impact strength for PP composites when wood or soybean-hull fiber is used, irrespective of the fiber size or level added, except wood 60 mesh. There was a significant reduction of notched impact strength of PP/BBS composites at 20% as the size decreased from 40 to 60 to 80 mesh. This may be explained by the reduction in aspect ratio as the fiber size is reduced, thus reducing the resistance to impact.¹³

The notched impact strength of PE plastic increased significantly above that of the pure plastic control, when BBS fiber was added at all levels and mesh sizes, except at 80 mesh size and 30% fiber content (Table XVI). The impact strength of the soybean hull/PE composites containing 30% fiber content does not change significantly as the fiber changes. This indicates we can use any size and achieve the same notched impact strength. Notched impact strength of wood fiber/PE plastic composites at 20% fiber content is not significantly different from that of soybean-hull fiber/PE plastic composites at 30% fiber content for all fiber sizes of PE plastic composites. This indicates that wood 20% fiber can be replaced by soybean hulls 30% fiber for PE composites and still result in comparable notched impact strength. BBS 40 and 60 mesh fiber size and 20 or 30% fiber content resulted in significantly higher resistance to the notched impact test compared to that of wood or soybean-hull fiber composites for PE plastic composites. This indicates wood or soybean hulls can be substituted by BBS and yet result in high notched impact strength.

The notched impact strength of PP/DDGS 40 mesh fiber size and 20 or 30% fiber content were comparable to that of PP/wood 40 mesh fiber size and 20 or 30% fiber content (Table XV). This indicates DDGS at 40

TABLE XVILeast-Square Means of Notched Impact Strength at 40,60, and 80 Mesh Sizes and 20 and 30% Levels ofDifferent Fiber/PE Composites

Notched impact strength, PE				
		Size of fiber, mesh		
Fiber type	Percentage	40	60	80
Wood	20	5.87 ^{k,1}	6.73 ^{i,j,k,l}	4.07^{1}
Wood	30	13.80 ^{f,g}	8.80 ^{h,i,j}	9.40 ^{h,i}
BBS	20	34.00 ^a	29.50 ^b	20.67 ^e
BBS	30	28.00 ^{b,c,d}	25.70 ^d	14.67^{f}
Soybean	20	8.47 ^{h,i,j,k}	4.93^{1}	11.13 ^{g,h}
Soybean	30	5.13 ¹	6.20 ^{j,k,1}	5.40^{1}
DDGS	20	28.80 ^{b,c}	NA	NA
DDGS	30	26.50 ^{c,d}	NA	NA
Control	0	15.30 ^f	15.30 ^f	15.30^{f}

Unnotched impact strength, PP					
			Size of the fiber, mesh		
Fiber type	Percentage	40	60	80	
Wood	20	135.09 ^{b,c,d}	142.91 ^{b,c}	126.11 ^{b,c,d,e}	
Wood	30	87.95 ^{d,e,f}	101.55 ^{b,c,d,e,f}	76.40 ^{e,f}	
BBS	20	116.30 ^{b,c,d,e,f}	120.23 ^{b,c,d,e}	107.69 ^{b,c,d,e,f}	
BBS	30	93.87 ^{c,d,e,f}	81.56 ^{d,e,f}	66.43 ^f	
Soybean	20	107.27 ^{b,c,d,e,f}	105.89 ^{b,c,d,e,f}	129.32 ^{b,c,d,e}	
Soybean	30	77.07 ^{e,f}	93.20 ^{c,d,e,f}	105.95 ^{b,c,d,e,f}	
DDGS	20	149.02 ^b	NA	NA	
DDGS	30	107.31 ^{b,c,d,e,f}	NA	NA	
Control	0	470.00 ^a	470.00 ^a	470.00 ^a	

TABLE XVIILeast-Square Means of Unnotched Impact Strength at 40, 60, and 80 Mesh Sizes and
20 and 30% Levels of Different Fiber/PP Composites

mesh fiber size can be used in place of wood at 40 mesh fiber size, resulting in the same notched impact strength. The notched impact strength of PE/DDGS 40 mesh fiber size and 20 or 30% fiber content was significantly higher compared to that of PE/wood or PE/soybean hulls 40 mesh fiber size and 20 and 30% fiber content (Table XV). This indicates DDGS at 40 mesh fiber size can be used in place of wood or soybean hulls at 40 mesh fiber size and 20 or 30% fiber content to obtain higher notched impact strength. PE/DDGS composites resulted in significantly higher notched impact strength compared to that of the pure polyethylene control.

Unnotched impact strength

There was a significant reduction in the unnotched impact strength of fiber composites compared to that of the pure plastic control, for both types of plastic composites. Among all the composites tested, wood 60 mesh fiber size at 20% fiber content resulted in the highest unnotched impact strength: 142.91 J/m for PP (Table XVII); and soybean hulls 80 mesh fiber size at 20% fiber resulted in highest unnotched impact strength: 138.82 J/m for PE (Table XVIII). As the fiber percentage increased from 20 to 30% the unnotched impact strength decreased, but not always significantly, in all the composite blends.

There is no significant difference between PP/wood fiber, PP/BBS fiber, or PP/soybean-hull fiber composites in a comparison across all fiber sizes of 40 to 60 to 80 and at 20 or 30% fiber contents. For example, the unnotched impact strengths of PP/wood 40 mesh size and 20% fiber content, PP/BBS 40 mesh size and 20% fiber content, and PP/soybean hulls 40 mesh size and 20% fiber content were 135.09, 116.30, and 107.27 J/m, respectively, which are not significantly different. This indicates we can use BBS or soybean hulls in place of wood at equivalent fiber sizes and content and achieve the same unnotched impact strength. There was no significant difference among all the PE/fiber composites, across all sizes and all fiber percentages (Table XVIII). This indicates we can replace wood with BBS or soybean-hull fiber and obtain the same unnotched impact strength. PP/wood, PP/soybean hulls, or PP/ BBS at 40 mesh fiber size and 20 or 30% fiber content is not significantly different from that of PP/DDGS 40 mesh fiber size and 20 or 30% fiber content (Table XVII). This indicates DDGS can be used as a filler to replace wood, soybean hulls, or BBS and result in comparable unnotched impact strength.

CONCLUSION

This research was carried out to study the effect that soybean hulls, wood, DDGS, and BBS biofibers had on

TABLE XVIII Least-Square Means of Unnotched Impact Strength at 40, 60, and 80 Mesh Sizes and 20 and 30% Levels of Different Fiber/PE Composites

Unnotched impact strength, PE					
		Siz	e of fiber, m	esh	
Fiber type	Percentage	40	60	80	
Wood	20	94.51 ^b	110.13 ^b	116.03 ^b	
Wood	30	46.07 ^b	55.41 ^b	64.57 ^b	
BBS	20	75.81 ^b	93.14 ^b	75.02 ^b	
BBS	30	47.45 ^b	44.03 ^b	32.42 ^b	
Soybean	20	68.89 ^b	99.37 ^b	138.82 ^b	
Soybean	30	49.37 ^b	65.70 ^b	63.39 ^b	
DĎGS	20	102.00 ^b	NA	NA	
DDGS	30	51.90 ^b	NA	NA	
Control	0	738.00 ^a	738.00 ^a	738.00 ^a	

the mechanical properties of polypropylene and polyethylene plastic biofiber composites. The mechanical properties evaluated were Young's modulus under both tensile and flexural loads, tensile strength, flexural strength, melt flow, impact energy absorption, and shrinkage as the fiber type, size, and amount varied. The following conclusions are reported:

- 1. Young's modulus under tensile load increased from 40 to 90% when wood or BBS fiber was added to PP plastic and increased from 94 to 212% when wood or BBS fiber was added to PE plastic, compared to that of the controls. Tensile strength reduced from 6 to 36%, compared to that of the controls, when all types of fiber were added to PP plastic. The addition of fibers to PE did not result in either a consistent increase or decrease of tensile strength. Tensile strength for fiber/PP plastic composites, compared to that of the control, ranged from 8 to -32%.
- 2. Young's modulus under flexural load increased from 18 to 102% when wood, BBS, or soybean-hull fiber was added to PP plastic and from 30 to 234% when wood, BBS, or soybean-hull fiber was added to PE plastic, compared to that of the controls. Flexural strength did not consistently increase when wood, BBS, or soybean-hull fiber was added to PP plastic, compared to that of the controls. Flexural strength varied from −13 to 14% for PP composites and the flexural strength increased from 30 to 65% for PE composites compared to that of the pure plastic controls.
- 3. Melt flow index decreased, when compared to the controls, from 26 to 51% when fiber was added to PP plastic. The exception was DDGS/PP composites in which there was a 10% increase in MFI. Melt flow index decreased from 23 to 80%, compared to that of the controls, when all the fibers were added to PE plastic.
- 4. Notched impact strength reduced from 18 to 96% for all the fiber/PP composites, compared to that of the control, except for BBS at 40 mesh fiber size and 20% fiber content. Notched impact strength decreased from 10 to 73%, compared to that of the controls, when wood or soybean fiber was added to PE plastic. BBS/PE composites

5. Unnotched impact strength decreased from 71 to 85% when all types of fiber were added to PP plastic and from 81 to 95% when all fiber types were added to PE plastic, compared to that of the controls.

In general BBS fiber composites showed comparable or higher mechanical properties compared with those of wood fiber composites. BBS could be a good source of fiber for plastic composites. Soybean hulls may be an acceptable source of fiber, depending on the application, but more investigation is required. DDGS may not be an acceptable filler because DDGS/thermoplastic composites have lower mechanical properties as well as problems with grinding of the raw material.

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