

Mechanical, Thermal, and Morphological Properties of Maleic Anhydride-*g*-Polypropylene Compatibilized and Chemically Modified Banana-Fiber-Reinforced Polypropylene Composites

Sanjay K. Chattopadhyay,¹ R. K. Khandal,¹ Ramagopal Uppaluri,² Aloke K. Ghoshal²

¹*Shriram Institute for Industrial Research, Delhi 110 007, India*

²*Indian Institute of Technology, Guwahati 781 039, Assam, India*

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ABSTRACT: Composites were prepared with chemically modified banana fibers in polypropylene (PP). The effects of 40-mm fiber loading and resin modification on the physical, mechanical, thermal, and morphological properties of the composites were evaluated with scanning electron microscopy (SEM), thermogravimetric analysis (TGA), Infrared (IR) spectroscopy, and so on. Maleic anhydride grafted polypropylene (MA-*g*-PP) compatibilizer was used to improve the fiber-matrix adhesion. SEM studies carried out on fractured specimens indicated poor dispersion in the unmodified fiber composites and improved adhesion and uniform dispersion in the treated composites. A fiber load-

ing of 15 vol % in the treated composites was optimum, with maximum mechanical properties and thermal stability evident. The composite with 5% MA-*g*-PP concentration at a 15% fiber volume showed an 80% increase in impact strength, a 48% increase in flexural strength, a 125% increase in flexural modulus, a 33% increase in tensile strength, and an 82% increase in tensile modulus, whereas the heat deflection temperature increased by 18°C. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 117: 1731–1740, 2010

Key words: compatibility; composites; fibers; morphology; poly(propylene) (PP)

INTRODUCTION

Composite materials based on the natural-fiber reinforcement of thermoplastics have been the subject of extensive research and development worldwide. Because of an increase in environmental concerns, natural fibers are fast replacing synthetic fibers as reinforcing agents in thermosets and thermoplastic matrices; for example, banana-fiber-reinforced polypropylene (PP) composites are now being commercialized in the automotive industry for interior components, panels, spare wheel recesses, and so on. Because composites cannot be easily recycled or reused and usually end up in landfills, the large-scale production of fully biodegradable composites will reduce the harmful effects of plastics waste on the environment. Natural fibers are lignocellulosic in nature and are the most abundant renewable biomaterial of photosynthesis on earth. These fibers have a low density and a high specific strength and modulus, are relatively nonabrasive, have surfaces that are easily modified, and are widely available.¹

India, which has a typical tropical and humid climate, is the largest banana producing country in the world. Bananas are used primarily as an agricultural crop, whereas banana fibers are a waste product of banana cultivation. Table I shows some of the typical properties of banana fibers.^{2,3} Banana fibers were selected as reinforcement materials in PP matrices for the following reasons. First, banana fiber, the cellulosic fiber obtained from the pseudostem of the banana plant (*Musa sapientum*), is a bast fiber with relatively good mechanical properties. Second, banana fibers have a relatively high cellulose content (66%), are crystalline in nature, and are available in abundance. Third, little work has been reported on the properties of banana-fiber-reinforced thermoplastics.

Sapuan et al.⁴ observed that woven banana-fiber-reinforced epoxy composites have stable mechanical behavior under tensile and flexural tests. Generally, a poor composite strength is observed because of a lack of stress transfer from the polymer matrix to the load-bearing natural fibers. This has been attributed to weak adhesion as a result of poor dispersion and incompatibility between the hydrophilic natural fibers and the hydrophobic polymer. Pothan and Thomas⁵ used various silanes and alkalis to modify the fiber surface in banana-fiber-reinforced polyester resin composites and found improved fiber/matrix

Correspondence to: A. K. Ghoshal (aloke@iitg.ernet.in).

TABLE I
Physical, Chemical, and Mechanical Properties of Banana Fibers

Natural fiber	Diameter (μm)	Density (kg/m^3)	Cell length/ diameter ratio	Moisture:		Cellulose (%)	Hemicellulose (%)	Lignin (%)	Microfibrillar angle ($^\circ$)	Initial modulus (GPa)	Ultimate tensile strength (MPa)	Elongation (%)
				Raw/ treated (%)								
Banana	20–60	1350	150	11/7	63–64	19	5	10 ± 1	7.7–20	54–754	10–35	

interactions resulting in better mechanical properties and low damping properties of the composites. Kumar and Varma⁶ reported banana surface modification by alkalization with NaOH and treatment with sebacoyl chloride and toluene diisocyanate during studies on banana-fiber-reinforced high-density polyethylene/polycaprolactone (80 : 20 w/w) blend composites. The grafting of maleic anhydride (MA) on polyethylene or PP is one popular method for improving the interfacial adhesion because of the formation of covalent bonds. According to Keener et al.,⁷ among coupling agents, newly developed maleated couplers demonstrate superior performance compared to other potential polyolefin coupling agents. In our earlier studies⁸ on pineapple leaf fiber (PALF)–PP composites, surface chemical modifications such as dewaxing followed by alkali treatment of the fibers and then treatment with maleated thermoplastic considerably improved the fiber–matrix adhesion and physicomechanical properties of the resulting biocomposites.

According to our previous studies^{8,9} and other literature reports,⁷ the optimum dosage of MA-g-PP as a compatibilizer into PP is 5%. Therefore, in this study, banana fiber–PP composites were fabricated after surface chemical treatment of the fiber by dewaxing followed by alkali treatment to improve the binding capability of the fiber with the PP matrix in the presence of 5% MA-g-PP as a compatibilizer. The compatibility between the banana fiber surface and the PP matrix was caused by esterification of the anhydride groups of MA-g-PP with the hydroxyl groups of the banana fiber. The physicomechanical and thermal properties of the composites at different volume fractions of fiber loading both from treated and untreated fibers were compared. Morphological observations were carried out with scanning electron microscopy (SEM). Thermogravimetric analysis (TGA) and heat deflection temperature (HDT) measurements were carried out to assess the thermal stability and thermal transitions of the developed composites.

EXPERIMENTAL

Materials

Banana fibers with fiber diameters in the range 25–60 μm were obtained from the Mushroom Growers

Welfare Society (Agartala, Tripura, India). PP (injection grade, Repol, H110MA), was obtained from Reliance Industries, Ltd. (Jamnagar, Gujarat, India). Other chemicals, namely, ethanol, hydrochloric acid, benzene, sodium hydroxide, xylene, isopropyl alcohol, benzoyl peroxide, and MA, were obtained from S. D. Fine Chemicals (Mumbai, India) through authorized agents in New Delhi, India.

Resin modification

PP was grafted with MA by a grafting process reported by Kasandariya et al.¹⁰ and Hujuri et al.⁸ to result in MA-g-PP. PP (50 g) was dissolved in 500 mL of xylene at 100°C. Maleic acid (4 g) dissolved in a minimum quantity of isopropyl alcohol and 0.4 g of benzoyl peroxide were then added to the solution of PP in xylene and continuously stirred. The resulting solution was then heated to 100°C for 7 h. Methanol was then added to obtain a white precipitate of MA-g-PP. This whitish powder-like material was dried in a vacuum oven at 60°C for 24 h. The Fourier transform infrared (FTIR) spectra of the material confirmed the presence of peaks characteristic of the carbonyl group and PP in MA-g-PP. The MA-g-PP was used as a compatibilizer during the blending of the fiber and resin.

Fiber surface modification

The banana fibers were dewaxed and then treated with alkali.¹¹ Dewaxing was carried out with a 1 : 1 mixture of ethanol and benzene at 60°C for 36 h in a hot-air oven. The fibers were then washed with cold distilled water and dried in a vacuum oven at 60°C for 24 h. The defatted fibers were then treated with 2N NaOH at 23°C in a climatic chamber for 60 h and washed with acidified (dilute HCl) water until the fibers showed no alkalinity reaction. The fibers were then washed again with cold distilled water and dried in a vacuum oven at 60°C for 24 h.¹²

Chopping of the banana fibers

Joseph et al.³ found that, in banana fiber–phenol formaldehyde composites, the maximum flexural strength and modulus values were obtained for 40-mm fibers. In this case, the untreated fibers and chemically modified fibers, as described previously,

were separated into strands and chopped into lengths of 40 mm with a fiber cutting machine. The banana fiber diameters after dewaxing and alkali treatment in this study were in the range 9–20 μm , as determined microscopically.

Preparation of the biocomposite

The required amounts of banana fibers and PP to be mixed for each volume fraction of the fibers were calculated before composite fabrication. For this, at first, the density of the banana fiber was determined according to ASTM D 729. Then, the required amounts of fiber and polymer for different fiber volume fractions, namely, 5, 10, 15, and 20%, were weighed, with an allowance given for flash in the sheet mold of the compression-molding press.

The melt mixing of the chopped banana fiber and PP were carried out in a two-roll open mill (Pyrotech Engineers, Delhi, India) heated with rows of cartridge heaters. The temperature of the front and back rollers were maintained at 140 and 160°C, respectively. Composites were prepared with untreated fibers and PP without compatibilizer, treated fibers with PP without compatibilizer, untreated fibers with PP and compatibilizer, and treated fibers with PP and compatibilizer. MA-g-PP compatibilizer was incorporated at a 5% level. After the uniform mixing of the fibers and the polymer matrix, we scraped out the resulting fiber–polymer matrix mix from the roller without allowing it to harden and immediately pressed into a $205 \times 205 \times 3.5 \text{ mm}^3$ sheet mold in a compression-molding press (SANTEC, Delhi, India) by applying a pressure of 25 T. The material was kept at a contact temperature of 160°C for 5 min. After a sufficient time was given for the composite sheet to cool and attain ambient room temperature, it was taken out, and the edges were trimmed. Two polyester sheets were put above and beneath the mold platens to facilitate a better surface finish of the molded composite sheets.

Characterization techniques

SEM analysis

The surface topographies of the banana fiber–PP interface and the fractured surfaces of the flexural specimens of the composites were scanned with the aid of a variable-pressure scanning electron microscope (model LEO 435 VP, Carl Zeiss SMT, Inc., Peabody, USA.) at the Scanning Electron Microscope Center of the All India Institute of Medical Sciences, New Delhi. The samples were mounted on special stubs and given a coating of gold, which rendered them conductive to be studied under the scanning electron microscope. Coatings were applied at a

thickness of about 20 nm, which was too thin to interfere with the dimensions of the surface features.

Spectral and thermal analysis

Raw and alkali-treated banana fibers were pulverized and compressed in KBr to form pellets. Pure PP and MA-g-PP granules were made into thin films in slides under heated conditions. The FTIR spectra of the raw and alkali-treated banana fiber samples and pure PP and MA-g-PP were recorded in an FTIR (attenuated total reflection) Bomem spectrometer (model FTLA 2000-100, ABB Inc., Quebec, Canada). TGA was carried out with a simultaneous DSC-TGA instrument (model SDT 2960, TA Instruments, New Castle, DE, USA) for the raw and alkali-treated fibers, virgin PP, and the composites with 15% loading of both untreated and treated fibers with compatibilizers to compare their thermal stabilities.

Physicomechanical properties

Water absorption

Water absorption was determined with ASTM D 570-98 (reaffirmed 2005). For each test, three test specimens in the form of bars with dimensions of $76.2 \times 25.4 \times 3 \text{ mm}$ were cut from the composite sheets. The specimens were then dried in an oven at 50°C for 24 h, cooled in a desiccator, and immediately weighed (w_1) to the nearest 0.001 g. The specimens were then entirely immersed in a container of distilled water at room temperature ($23 \pm 1^\circ\text{C}$). At the end of 24 h, the specimens were removed from water one at a time, all of the surface water was wiped off, and they were immediately weighed (w_2) to the nearest 0.001 g. The percentage increase in weight during water immersion was calculated to the nearest 0.01% as follows:

$$\text{Increase in weight (\%)} = \frac{w_2 - w_1}{w_1} \times 100\%$$

Tensile strength and modulus

For each tensile test, five replicate dumbbell-shaped test specimens (overall length = 165 mm, width of the narrow section = 13 mm, thickness = 3 mm, and gauge length = 50 mm) as per ASTM D 638 (Type 1) were prepared from the composites of untreated fiber plus PP, chemically treated fiber plus PP, untreated fiber plus PP plus MA-g-PP, and chemically treated fiber plus PP plus MA-g-PP. The tensile tests were carried out with a Universal Testing Machine (H 50 K-S, Tinius Olsen, Surrey, England.) A load range of 10 kN and a crosshead speed of 5 mm/min were used for the test. From the experimental data obtained, the average data and the

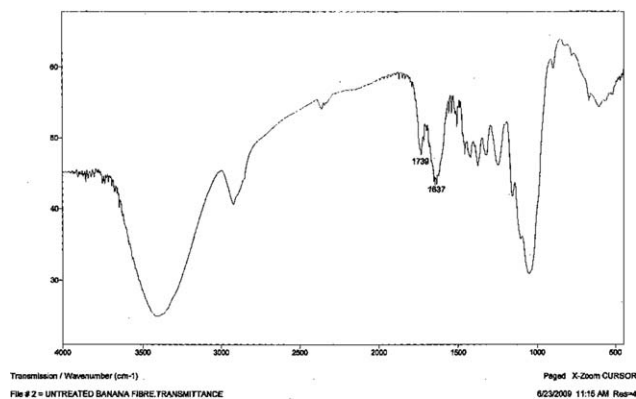


Figure 1 FTIR spectra of the raw banana fiber (%T vs wave number).

corresponding maximum standard deviation were calculated.

Flexural strength and modulus

Flexural tests under three-point bending were carried out on similar composite samples of untreated fiber plus PP, chemically treated fiber plus PP, untreated fiber plus PP plus MA-g-PP, and chemically treated fiber plus PP plus MA-g-PP with dimensions of $80 \times 12.7 \times 3 \text{ mm}^3$ with the same universal testing machine as per IS 13360 (part 5/section 7, 1996) at a load range of 500 N and a cross-head speed of 1.0 mm/min. For each test, five replicate test specimens were taken, and from the experimental data obtained, the average data and the corresponding maximum standard deviation were calculated.

Impact strength

Notched Izod impact testing on specimens with dimensions of $63.5 \times 12.7 \times 3 \text{ mm}^3$ was carried out with a Ceast impactometer (code 6545/000, Akron, Ohio, USA) as per ASTM D 256 with a notch angle of 45° and a depth of 2.54 mm. For each test, five replicate test specimens were taken, and from the experimental data obtained, the average data and the corresponding maximum standard deviation were calculated.

Thermal properties

HDT was determined on composite samples of untreated fiber with and without MA-g-PP and chemically treated fiber with and without MA-g-PP with an HDT/Vicat softening temperature apparatus (Toyo Seiki Seisaku Sho, Ltd., Tokyo, Japan) as per ASTM D 648-07 with a span value of 100 mm (method B) and a flexural load of 1.82 MPa. The test sample, in the form of a bar of rectangular cross sec-

tion having dimensions of $127 \times 13 \times 3 \text{ mm}^3$, was tested in the edgewise position. The temperature of the heat-transfer medium when the test specimen deflected 0.25 mm was recorded as the deflection temperature. For each test, two replicate test specimens were taken, and from the experimental data obtained, the average data and the corresponding maximum standard deviation were calculated.

RESULTS AND DISCUSSION

Characterization of the fiber and the resin

SEM micrographs of PP and MA-g-PP carried out in our earlier studies⁸ showed a considerable amount of grafted polymer in the polymer backbone of MA-g-PP. The SEM micrographs of raw, dewaxed, and alkali-treated fibers showed a fibrillated smooth surface in untreated fibers because of the presence of a waxy coating of cuticle, the swelling of fibers on dewaxing, and much thinner fibers with surface roughness after alkali treatment. In this study, the weight losses and visibly clear color observed in the defatted and alkali-treated banana fibers suggested the removal of natural and artificial impurities along with the nonpolar, waxy, and aliphatic cuticle layer. The dewaxing followed by alkali treatment and the incorporation of MA-g-PP compatibilizer led to the enhancement of fiber wetting and impregnation to give an increase in the effective surface area of contact.

Various authors, such as Herrera-Franco et al.¹³ and Hujuri et al.,⁸ clearly identified that the band around 1740 cm^{-1} corresponding to hemicellulose disappeared when the fiber was treated by a dilute NaOH aqueous solution. In this study, similar observations were noticed in the case of the FTIR spectra of untreated and alkali-treated banana fibers, as shown in Figures 1 and 2, where absorption was noticed in the untreated fibers at about 1739 cm^{-1} ; this vanished in the alkali-treated fibers because of

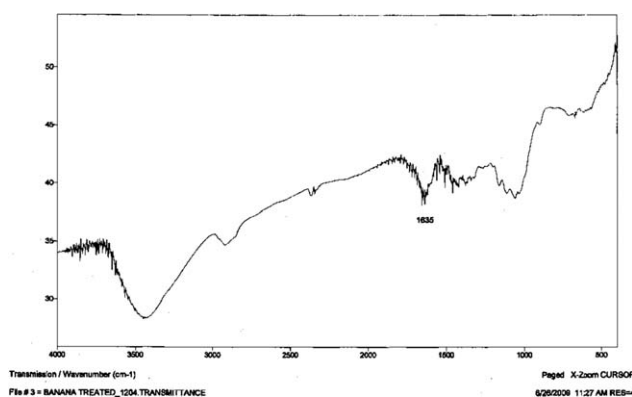


Figure 2 FTIR spectra of the treated banana fiber (%T vs wave number).

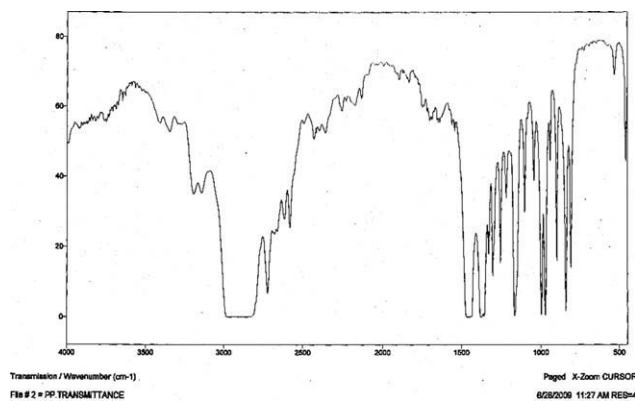


Figure 3 FTIR spectra of PP (%T vs wave number).

the removal of hemicellulose constituent groups, which absorbed at an IR wave number corresponding to those for carbonyl groups.

SEM micrographs of MA-g-PP showed a considerable amount of grafting in the polymer backbone in which we carried out grafting in a similar manner earlier.⁸ The FTIR spectra of PP and MA-g-PP are shown in Figures 3 and 4, respectively. The absorption peak at about 1729 cm⁻¹ in MA-g-PP (Fig. 4), which was absent in virgin PP (Fig. 3), was characteristic of the carbonyl group of the anhydride and indicative of the grafting in the polymer backbone.

The results of TGA along with those of differential thermogravimetry (DTG) of the fibers before and after treatment are presented in Figure 5. The higher DTG peak for the treated fiber ($T_{max} = 357.5^{\circ}\text{C}$) than that of the untreated fiber ($T_{max} = 324^{\circ}\text{C}$) demonstrated the higher thermal stability of the treated fiber compared to the untreated one. The slow but steady and continuous loss in weight for the untreated fiber at temperatures beyond 370°C was ascribed to the loss of the hydroxyl and polar components. In addition, the treated fiber did not contain the constituents of hemicellulose and lignin, which were responsible for the almost 14% of residues in the untreated fiber. The results of TGA

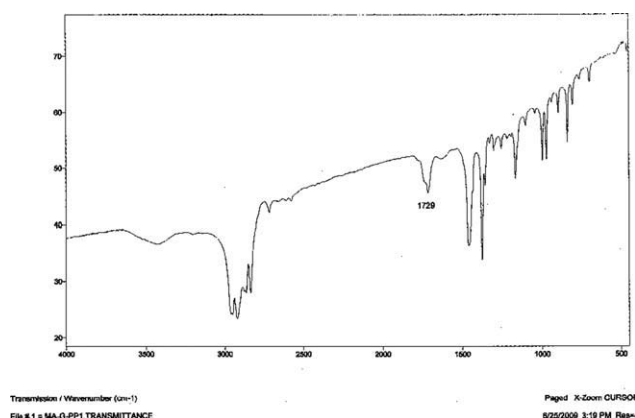


Figure 4 FTIR spectra of MA-g-PP (%T vs wave number).

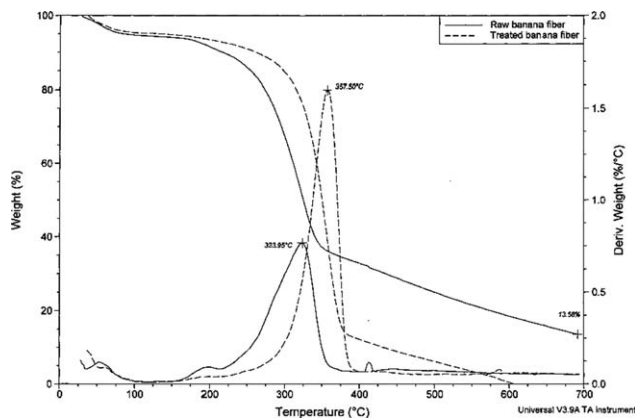


Figure 5 TGA-DTG of the raw and treated banana fibers.

along with the DTG results of the composites, both of the treated and untreated fibers with compatibilizers and virgin PP, are presented in Figure 6. It was clear that the treated fibers with compatibilizers had a higher thermal stability ($T_{max} = 445^{\circ}\text{C}$) compared to the untreated fiber composites with compatibilizers ($T_{max} = 433^{\circ}\text{C}$) and virgin PP ($T_{max} = 441^{\circ}\text{C}$).

Effect of the surface modification and compatibilizer on the physicomechanical and thermal properties of the composites

The variation in the physical and mechanical strength as a function of fiber volume percentage is represented in Tables II–V. Water absorption in the untreated fiber composites increased with increasing fiber volume loading (Table II). This was quite obvious because of the presence of hydroxyl and other polar groups in the various constituents of natural fibers. Water absorption was comparatively less in the composites with treated fibers (Table II) without compatibilizer than in the untreated fiber without compatibilizer because of the fact that, by the action of alkali,¹¹ a substantial portion of uronic

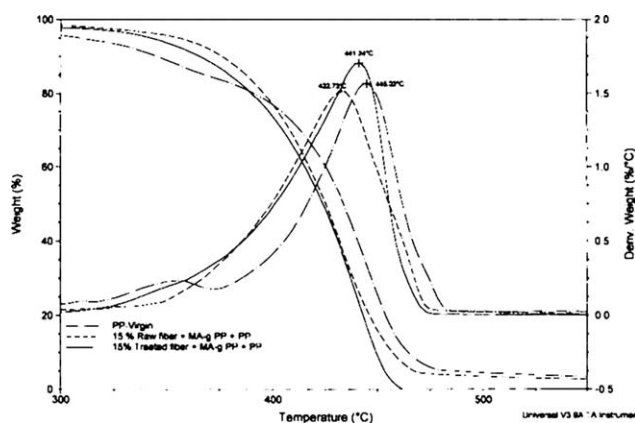


Figure 6 TGA-DTG of the virgin PP and composites with 15% volume fraction of raw and treated banana fibers + MA-g-PP + PP.

TABLE II
Water Absorption of the Banana-PP Composites at Various Fiber Volumes

Fiber volume (%)	Water absorption (%)			
	Raw fiber + PP	Raw fiber + PP + MA-g-PP	Treated fiber + PP	Treated fiber + PP + MA-g-PP
0	0.09	0.09	0.09	0.09
5	0.64	0.61	0.45	0.24
10	1.27	1.20	0.85	0.62
15	1.78	1.65	1.25	0.95
20	1.96	1.80	1.34	1.12

acid, a constituent of hemicellulose and other polar groups, was removed, which caused a reduction in moisture absorption. Furthermore, the incorporation of MA-g-PP imparted hydrophobicity to the fibers because of the esterification of the anhydride groups of MA-g-PP with the hydroxyl groups of the natural fibers. Therefore, water absorption for the composite with compatibilizer was less in comparison to that without compatibilizer.

We also observed that the mechanical properties, such as impact strength (Table III), flexural strength and flexural modulus (Table IV), and tensile strength and tensile modulus (Table V) of the untreated banana fiber-PP composites with and without compatibilizer increased with increasing fiber loading from 5 to 15% and decreased for the fiber loading of 20%, except in the case of impact strength. The impact strength continuously increased with increasing fiber loading. This increase was due to an initial reinforcement effect¹⁴ by the natural fibers, which allowed stress distribution from the continuous polymer matrix to the dispersed fiber phase. Beyond 15%, a downward trend in the mechanical properties, except in the impact strength, was noticed, possibly because of an agglomeration effect that resulted in poor dispersion of the fibers in the polymer matrix at higher volume loadings, which caused the non-uniform transmission of the applied stress and poor fiber-matrix adhesion. The continuous increase in impact strength was also observed in our recent study on PALF-PP composites.⁸ This was explained on the basis of the fact that fibers reduced the crack propagation rate by forcing a crack around the fiber and bridging the crack through fiber pullout, which led to an increase in impact strength. However,

impact strength was found to decrease at a relatively higher percentage of fiber in the PP matrix. Because we expected a similar behavior in this case, impact strength with further increasing fiber content was not studied because such high fiber content would deteriorate the other mechanical properties, as suggested by the trends in the tables.

The mechanical properties of the untreated and chemically treated fiber composites without compatibilizer at different fiber loadings were worse than those with compatibilizers. The dewaxing followed by alkali treatment resulted in the removal of hemicellulose and lignin (the amorphous components) in the treated fibers. Cellulose was not compatible with PP, and this incompatibility or poor fiber-matrix chemical adhesion caused poor mechanical properties. Untreated fiber had a lignin component that made it a little bit compatible with PP. Possibly because of that reason, a marginal improvement in the mechanical properties was observed compared to the treated fiber composites (without lignin), although the treated fibers were rough, which could have caused more mechanical interlocking within the PP matrix. The treated fibers, being crystalline, had a higher strength and modulus. The improved fiber matrix chemical adhesion in the composites of the treated fibers with MA-g-PP compatibilizer in the PP matrix resulted in higher mechanical properties, as shown in Tables III-V.

In the untreated fiber composites with MA-g-PP, the incorporation of compatibilizer reduced the surface hydrophilicity of the fibers and, thereby, enhanced the fiber wetting and dispersion within the matrix. This caused a decrease in water absorption and an increase in the mechanical properties, as

TABLE III
Impact Strength of the Banana-PP Composites at Various Fiber Volumes

Fiber volume (%)	Impact strength (J/m)			
	Raw fiber + PP	Raw fiber + PP + MA-g-PP	Treated fiber + PP	Treated fiber + PP + MA-g-PP
0	26.54 ± 0.15	26.54 ± 0.15	26.54 ± 0.15	26.54 ± 0.15
5	31.32 ± 0.21	36.42 ± 0.24	25.68 ± 0.17	41.02 ± 0.24
10	33.42 ± 0.11	36.42 ± 0.21	28.44 ± 0.15	45.18 ± 0.24
15	36.57 ± 0.33	41.50 ± 0.34	27.97 ± 0.23	47.75 ± 0.35
20	40.02 ± 0.23	43.40 ± 0.22	30.64 ± 0.23	49.98 ± 0.41

TABLE IV
Flexural Properties of the Banana-PP Composites at Various Fiber Volumes

Fiber volume (%)	Raw fiber + PP		Raw fiber + PP + MA-g-PP		Treated fiber + PP		Treated fiber + PP + MA-g-PP	
	Flexural strength (MPa)	Flexural modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)
0	38.00 ± 0.30	1650 ± 3	38.00 ± 0.30	1650 ± 3	38.00 ± 0.30	1650 ± 3	38.00 ± 0.30	1650 ± 3
5	39.50 ± 0.22	1890 ± 5	39.60 ± 0.20	2320 ± 7	37.30 ± 0.35	1786 ± 6	40.95 ± 0.45	2700 ± 5
10	39.70 ± 0.45	2030 ± 7	42.30 ± 0.41	2263 ± 5	38.60 ± 0.48	1940 ± 4	45.75 ± 0.65	2887 ± 4
15	42.00 ± 0.35	2445 ± 5	49.10 ± 0.52	3125 ± 6	36.20 ± 0.50	2308 ± 9	56.20 ± 0.72	3720 ± 4
20	38.50 ± 0.52	2250 ± 8	44.00 ± 0.36	2815 ± 8	37.90 ± 0.42	2211 ± 6	45.20 ± 0.60	3135 ± 6

TABLE V
Tensile Properties of the Banana-PP Composites at Various Fiber Volumes

Fiber volume (%)	Raw fiber + PP		Raw fiber + PP + MA-g-PP		Treated fiber + PP		Treated fiber + PP + MA-g-PP	
	Tensile strength (MPa)	Tensile modulus (MPa)	Tensile strength (MPa)	Tensile modulus (MPa)	Tensile strength (MPa)	Tensile modulus (MPa)	Tensile strength (MPa)	Tensile modulus (MPa)
0	24.50 ± 0.21	560 ± 4	24.50 ± 0.21	560 ± 4	24.50 ± 0.21	560 ± 4	24.50 ± 0.21	560 ± 4
5	24.05 ± 0.47	745 ± 6	25.30 ± 0.64	790 ± 7	23.98 ± 0.53	725 ± 7	26.85 ± 0.26	825 ± 6
10	24.35 ± 0.29	820 ± 4	25.45 ± 0.43	860 ± 6	24.20 ± 0.49	780 ± 4	28.60 ± 0.41	910 ± 4
15	24.70 ± 0.25	890 ± 8	26.00 ± 0.39	970 ± 5	24.48 ± 0.58	750 ± 3	32.65 ± 0.22	1020 ± 5
20	24.40 ± 0.38	875 ± 5	25.40 ± 0.52	865 ± 6	24.45 ± 0.27	790 ± 5	31.36 ± 0.54	1050 ± 4

TABLE VI
HDT Values of the Banana-PP Composites at 1.8 MPa and with a Span Value of 100 mm

Fiber volume (%)	HDT (°C)			
	Raw fiber + PP	Raw fiber + PP + MA-g-PP	Treated fiber + PP	Treated fiber + PP + MA-g-PP
0	72	72	72	72
5	78	80	76	86
10	80	82	78	89
15	80	85	77	89
20	79	87	78	90

shown in Tables II–V. In the chemically treated fiber composites with MA-g-PP, the incorporation of compatibilizer also reduced the surface hydrophilicity of the fibers. The MA groups of MA-g-PP covalently linked with the hydroxyl groups of the fibers to form an ester linkage, whereas the nonpolar part (PP) of MA-g-PP became compatible with the virgin matrix, lowered the surface energies of the fibers, and thereby, increased their wettability and dispersion within the matrix.¹⁴ This was reflected in the considerably increased values of the mechanical properties with increasing fiber loading in comparison to the untreated or chemically treated composites at the same volume percentage of fiber content.

The composites prepared with 5% MA-g-PP concentration at an optimum fiber content of 15 vol % had an 80% increase in impact strength, a 48% increase in flexural strength, a 125% increase in flexural modulus, a 33% increase in tensile strength, and an 82% increase in tensile modulus, as was evident from the experimental data shown in Tables III–V, respectively.

Rana et al.¹⁵ also found sharp increases in all of the mechanical properties with increasing jute fiber loadings from 30 to 40% for jute-PP composites with compatibilizer, but when the jute content was increased from 40 to 50%, there was no considerable increase in the mechanical properties. This was due to the agglomeration and poor dispersion of the fibers in the polymer matrix at higher fiber loading concentrations, which resulted in inadequate fiber-matrix adhesion and nonuniform transmission of the applied stresses.

The variation in HDTs as a function of fiber volume percentage in the raw fiber composite with and without MA-g-PP compatibilizer and in the treated fiber composites with and without MA-g-PP compatibilizer are represented in Table VI. We observed that the HDTs of both the untreated and chemically treated banana fiber-PP composites without MA-g-PP showed a marginal increase with the increase in fiber volume percentage from 5 to 20%. On the other hand, the HDTs of the raw and chemically treated fiber composites with MA-g-PP compatibilizer showed much higher values. Compared to the HDT of virgin PP, the HDT of the chemically treated fiber

composites with MA-g-PP compatibilizer showed an increase from 14 to 17°C with fiber volume percentages from 5 to 10%, with the values tending to saturate thereafter.

Nielsen et al.¹⁶ suggested that the incorporation of fibers or fillers into polymer matrices restricts the mobility of polymer chains and leads to an increase in their thermal-transition properties, such as the glass-transition temperature. Hristov and Vasileva¹⁷ observed an increase in the glass-transition temperature in PP-wood flour (10%) composites when compatibilizer was added because of more restricted macromolecular mobility in the amorphous phase. In our earlier study,⁹ an increase in the Vicat softening temperature (a property related to thermal transition) of chemically treated PALF-PP composites with compatibilizers was observed because of similar reasons of restricted macromolecular chain mobility. In this study, the increase in the HDT in the raw and treated banana fiber-PP composites without compatibilizer was primarily due to the restricted mobility of the polymer chains in the amorphous phase. The HDT was further augmented in the presence of the MA-g-PP compatibilizer because of the enhanced interfacial strength as a result of improved fiber matrix chemical adhesion. The saturation was due to the fact that no further restriction in mobility of the polymer chains was feasible.

Morphology of the interface

The fiber-polymer interface was studied with SEM [Fig. 7(a,b)] for 15 vol % banana fiber composites with compatibilizers. As is evident from the micrographs, poor dispersion was seen in the untreated fiber composites with MA-g-PP [Fig. 7(a)], where the fibers were bunched together. The dispersion was slightly better and comparatively more uniform in the treated fiber composites with MA-g-PP compatibilizer [Fig. 7(b)].

Morphology of the fractured surface

The morphology of the fractured surfaces of flexural specimens was observed under a scanning electron microscope. The fracture characteristics are

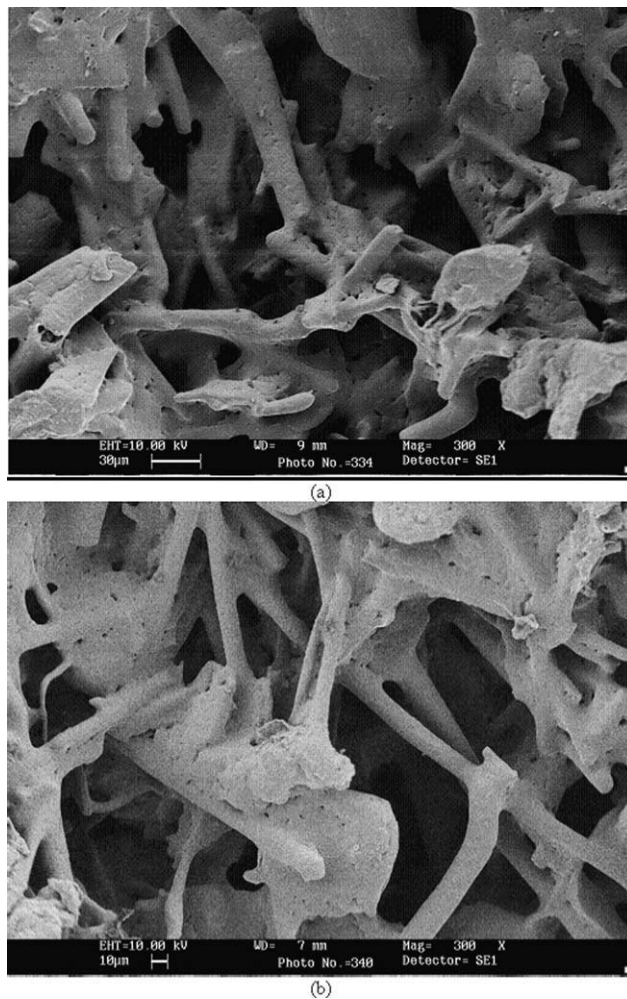


Figure 7 SEM micrograph of the fiber (40 mm long and with 15% fiber volume fraction)–polymer interfaces: (a) untreated fiber composite with MA-g-PP and (b) treated fiber composite with MA-g-PP.

very much evident in the micrographs shown in Figure 8(a,b). As shown in Figure 8(a), poor dispersion was noticed in the untreated fiber composites with compatibilizer, where the fibers were seen to agglomerate into bunches, and many holes were left after fiber pullout from the matrix on account of the application of flexural stress. On the other hand, in the treated fiber composites with compatibilizer [Fig. 8(b)], improved adhesion and fiber dispersion were seen; this resulted in a lower number of fiber pullouts and voids due to fibers not coming out totally out of the matrix. The fractured surfaces were also more uniformly distributed than in the case of the untreated fiber composites with compatibilizer.

CONCLUSIONS

The physical, mechanical, and thermal properties of banana-fiber-reinforced thermoplastic PP composites at various fiber volume percentages were investi-

gated in this study. The alkali treatment of the defatted fibers improved the fiber–polymer interaction by enhancing fiber wetting and impregnation, and these properties were further improved by the action of MA-g-PP compatibilizer, which enhanced the properties of the composite by chemically binding the fiber and the polymer by a grafting mechanism. The composites prepared with 5% MA-g-PP at an optimum 15 vol % fiber had an 80% increase in impact strength, a 48% increase in flexural strength, a 125% increase in flexural modulus, a 33% increase in tensile strength, and an 82% increase in tensile modulus. The HDT of the chemically treated fiber composites with MA-g-PP compatibilizer showed an increase from 14 to 18°C (when compared to virgin PP) with increasing fiber volume from 5 to 20%. Thus, 15 vol % fiber with the MA-g-PP coupling agent is the recommended composition when the fiber length is 40 mm, where the mechanical

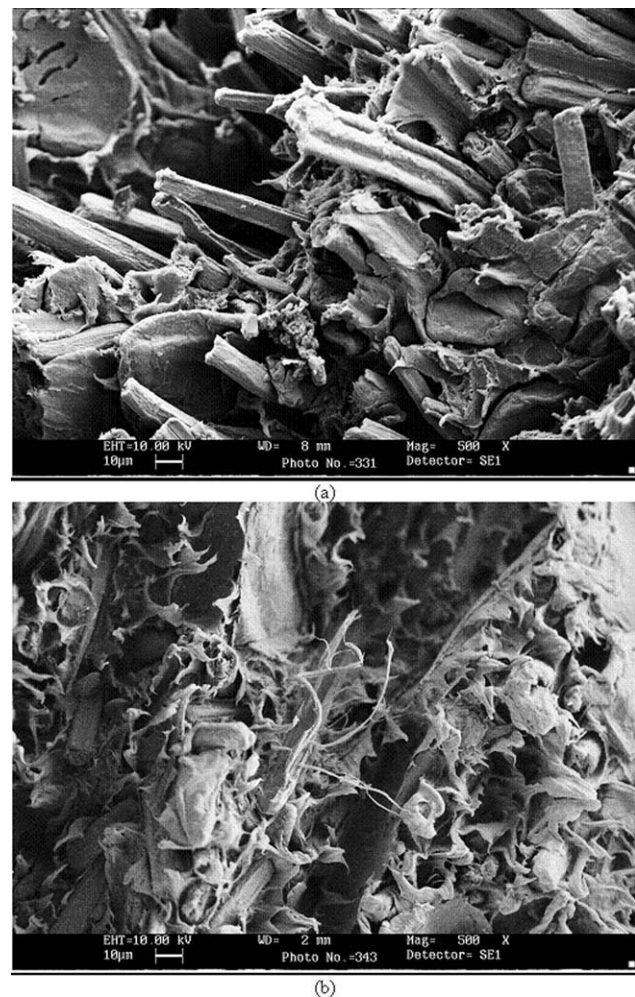


Figure 8 SEM micrograph of the flexural fractured surfaces (40 mm fiber long and with 15% fiber volume fraction): (a) untreated banana fiber composite with MA-g-PP and (b) treated fiber composite with MA-g-PP.

properties were at a maximum and the thermal stability was also quite high.

In forthcoming communications, biodegradability studies of natural-fiber-reinforced composites with a large scope of research will be the main focus area, along with other scopes for improvements in the properties of the composites by optimization of various other parameters, such as fiber length, orientation, appropriate coupling agents, and chemical modification of the matrix and fibers.

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