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Impact of Chemical Treatments and Coupling Agents on the Properties of Jute Fiber Reinforced by Polybutylene Composite

Md. Mohsin Uddin Azad ^a & Md. Islamul Haque ^a

Abstract- Jute and polybutylene have different characters in their properties. When they are blended, their blended composition shows various purposes. The goal of the research is to enhance the properties of jute fiber (JF) reinforced polybutylene (PB) composite. It was made by sample hot press molding method according to the weight percentage of 25 wt% filler (JF), 2wt% coupling agent (compatibilizer) degraded PB (DgPB) and maleated PB (MAPB) and 70 wt% matrix (PB). To improve the interfacial interactions between the fiber and PB, the fiber was chemically modified with 10w/v% NaOH, 6w/v% dicumyl peroxide (DCPO), 0.05 w/v% K2Cr2O7 and 5 w/v% H2O2 solution. The water absorption, tensile and thermal properties of raw and surface treated fibers composites were compared in particular coupling agent. It has found that the fiber treatments and the addition of coupling agents, composites showed better properties. Among the composites, 10 wt/v% NaOH treated jute reinforced PB composite by using MAPB compatibilizer was presented better water absorption and mechanical properties.

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I. INTRODUCTION

The development of composites furnished by lignocellulosic fibers as reinforcing materials of thermoplastic resin become popular since the lignocellulosic fibers are inexpensive; possess sufficient mechanical properties and environmentally friendly due to their biodegradation properties. In tropical countries, plenty of abundance lignocellulosic fibers like jute, hemp, ramie, sisal, okra fibers, plumb Oil fiber, etc. are grown and most of them have been employed for reinforcements in polymer matrices (Albano et al. 1999; Suardana et al. 2011; Khan et al. 2015b; Khan et al. 2016, Ramez A. Al-Mansob et al. 2013, Islam & Ani 2000). Among the lignocellulosic fibers, jute fiber is promising on account of availability, low densities, nonabrasive nature, high specific properties, high modulus and renewable nature. Jute (*Corchorus capsularis*) is one of the most common agricultural fibers is cultivated almost exclusively in Bangladesh.

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Therefore, due to their availability and suitable properties, jute fiber has the right potential for usage in composites.

Polybutylene (PB) is extensively used in engineering plastic materials for tremendous mechanical properties. It also has advantages like the economy (price), ecological (recycling behaviors) and higher thermal stability and the effectiveness of filler reinforced composite. However, during impregnation of PB by lignocelluloses fiber, the interface shows incompatibility i.e. hydrophilic lignocellulose fiber cannot mix properly with hydrophobic PB. Therefore, afford need to reduce incompatibility on the interfacial adhesion between filler and matrix polymers for manufacturing high-value composite materials.

A lot of conventional methods have practiced improving the interfacial adhesion of composites such as modifying the fiber surface before composite fabrication, using compatibilizer during moulding or matrix modification. The use of silane coupling agents, grafting by bifunctional monomers and the plasma treatment of the fiber surfaces are the most common techniques of interface modification of composites (Pothan et al. 2002; Khan et al. 2009; Khan et al. 2013a; El-Sabbagh 2014; Khan et al. 2015a; Khan et al. 2015b). In the present investigation, to improve the fiber matrix interaction, jute fiber surface has been modified by NaOH, dicumyl peroxide, potassium dichromate, and hydrogen peroxide. MAPB and DgPB have also used to improve interfacial properties which are well-known compatibilizers for filler and matrix. It has been reported methane protons of Isotactic PB is often caused oxidative degradation. The degradation reaction proceeds by a free radical chain reactive mechanism and formed carbonyl group (C=O) as well as hydroperoxide group (ROOH) (Alam et al. 2002). Another study used oxidative degraded PB (DgPB) as a compatibilizer of cellulose/PB composite (Miyazaki et al. 2008).

II. EXPERIMENTAL DESIGN

a) Materials

The water retted jute fiber (*Corchorus capsularis*) was collected from Kushtia district, Bangladesh. The polybutylene (PB) was obtained from

Polyolefin Company (Singapore) Pvt. Ltd. in the form of pellets. Maleated polybutylene (MAPB) was purchased from Sigma-Aldrich in the granule form and degraded polybutylene (DgPB) was prepared in a crucible allowed by heating at 1300C for 20 h in the presence of air. All analytical reagent (AR) grade chemicals were used in the investigation.

b) Fiber Processing

Fibers were cut into 25-30 cm and then scoured by (6.5 g/L soap and 3.5 g/L of Na₂CO₃) solution at 70-75°C during 30 min. The fiber to solution weight ratio was maintained at 1:50. After scouring the fiber was several times with distilled water and dried in the open air (Mondal and Khan 2008). The dried fibers were chemically treated with four different chemicals, namely NaOH, dicumyl peroxide (DCP), K₂Cr₂O₇ and H₂O₂.

The dried jute fibers were immersed in 10w/v% NaOH solution at 30°C maintained the fiber to liquor ratio at 1:20 for 2 h. Then the fibers were washed repeatedly by distilled water and finally washed by dilute acetic acid to remove NaOH sticking to the fiber surface.

The neutralized fibers were dried in air and finally stored in desiccator (Khan et al. 2015b).

6w/v% dicumyl peroxide (DCP) solution was prepared with acetone. The NaOH treated fiber was soaked in the DCP solution for 30 min at 30°C maintained the fiber to liquor ratio at 1:11. The treated fiber was washed by distilled water and dried at room temperature. The NaOH treated fiber was treated by 0.055w/v% K₂Cr₂O₇ solution maintained the fiber to liquor ratio 1: 15 for 30 min at 60°C with occasionally added few drops of H₂SO₄. Then the fiber was washed by distilled water and dried in air (Khan et al. 2015b).

The jute fibers were treated by 5w/v% H₂O₂ solution maintained the fiber to liquor ratio 1: 80 for 2 h at 90°C. During the treatment the pH of adjusted by adding 7-8 drops of 0.2 M CH₃COOH solution. The fiber was filtered and washed by distilled water. Then the fibers were immersed in 0.2 % Na₂S₂O₆ solution maintained the fiber to liquor ratio 1:20 for 15 min at 30°C. Finally the fiber was washed and dried in air and finally stored in the desiccator (Li and Wang 2013).

Table 1: Various types of composite sample prepared in different parameters

Sample	Fiber chemical treatments	Fiber wt%	Matrix	Coupling agent	Composite
A	No chemical treatment	30%	PB (70 wt%)	-	Raw JF+PB
B	No chemical treatment	30%	PB	DgPB (2wt%)	Raw JF+PB+DgPB
C	No chemical treatment	30%	PB	MAPB (2wt%)	Raw JF+PB+MAPB
D	10 w/v % NaOH	30%	PB (70 wt%)	-	10% NaOH JF+PB
E	10 w/v % NaOH	30%	PB (68 wt%)	DgPB (2wt%)	10% NaOH JF+PB+DgPB
F	10 w/v % NaOH	30%	PB (68 wt%)	MAPB (2wt%)	10% NaOH JF+PB+MAPB
G	6 w/v % DCP	30%	PB (70 wt%)	-	6% DCP JF+PB
H	6 w/v % DCP	30%	PB (68 wt%)	DgPB (2wt%)	6% DCP JF+PB+DgPB
I	6 w/v % DCP	30%	PB (68 wt%)	MAPB (2wt%)	6% DCP JF+PB+MAPB
J	0.055 w/v % K ₂ Cr ₂ O ₇	30%	PB (70 wt%)	-	0.055% K ₂ Cr ₂ O ₇ JF+PB
K	0.055 w/v % K ₂ Cr ₂ O ₇	30%	PB (68 wt%)	DgPB (2wt%)	0.055% K ₂ Cr ₂ O ₇ JF+PB+DgPB
L	0.055 w/v % K ₂ Cr ₂ O ₇	30%	PB (68 wt%)	MAPB (2wt%)	0.055 K ₂ Cr ₂ O ₇ JF+PB+MAPB
M	5 w/v % H ₂ O ₂	30%	PB (70 wt%)	-	5%H ₂ O ₂ JF+PB
N	5 w/v % H ₂ O ₂	30%	PB (68 wt%)	DgPB (2wt%)	5%H ₂ O ₂ JF+PB+DgPB
O	5 w/v % H ₂ O ₂	30%	PB (68 wt%)	MAPB (2wt%)	5%H ₂ O ₂ JF+PB+MAPB

c) Composite Preparation

The fiber bundles were cut into approx. 10cm length, and then placed in an air oven for a week to remove the fiber surface moisture. Virgin PB sheet was prepared in an open S-S mold. The mold was electrically heated by thermostat at 180°C for 115 min. Then it was placed in hydraulic press and pressure was set to 50 kN. It was then cooled by cold tap water flow and transparent PB sheet was obtained. The sheet was cut into 10×10 cm² size and placed in a closed mold with the same dimension. The predetermined amount fibers

were unidirectionally sprayed on the PB sheet (Table 1). The fixed amount of MAPB or DgPB was added as a compatibilizer. The fibers were then covered with PB sheets. The mold was put in between two hot plates of compression molding machine. Pressure and temperature were fixed at 50 kN and 180°C respectively. The heating was gradually increased up to 180°C. After 30 min, heating was stopped and, mold was cooled by tap water. Finally, the composite sample was taken out from the mold and cut for the mechanical testing.

d) Measurements

Tensile properties were measured using Universal Testing Machine (Hounsfield UTM 10KN) by standard ASTM D638 methods. The crosshead speed was 50 mm/min and gage length was 50 mm. The results are taken from the average of 10 measurements. The water absorption property was measured by ASTM D570 method. The specimens were dried in an oven at 80°C for 24h before testing. Then those samples were soaked into the water at room temperature.

The composites were taken out from the water after every 24h and all surface moisture was removed with tissue paper. The weight gain was calculated using Equation 1:

$$\text{Water absorption \%} = \frac{(w_t - w_0) \times 100}{w_0} \dots \dots \dots (1)$$

Where, W_0 and W_t are the initial weight and weight after water absorption, respectively. The data reported are an average value obtained from ten separate samples of each composite. The thermal properties of composites were assessed by thermogravimetric analyzer supplied by TA Instrument (EXTAR 6000 STATION, Seiko Instrument, Inc. JaPBn).

About 20 mg of composite sample was taken for each analysis. The heating was increased by the rate of 20°C/min from 25°C to 600°C in nitrogen environment (gas flow rate 50 ml/min).

III. RESULT AND DISCUSSION

The surface of jute fiber has been chemically modified through different chemical process. The performance of DgPB and MAPB coupling agents at interface modification of composites were investigated regarding their water absorption, mechanical and thermal properties.

a) Water Absorption Properties

Figure 1 shows the effect of immersion time on water absorption of raw and treated jute fiber PB composites. Raw JF+PB composite (Sample A) is taken as control for comparison. It shows that the lowest water absorption is found in case of 10% NaOH JF+PB+MAPB composite and water absorption is increased with the increase of immersion time. It is also observed that the addition of coupling agents MAPB and DgPB in composites give lower water absorption than JF+PB composite. With the addition of MAPB and DgPB coupling agents in composites, the percentage of water absorption is decreased. It may be due to the enhancement of surface adhesion of fibers and PB matrix that reduced the water consumption in the interfacial voids. Probably the coupling agents took part in the esterification reaction with the -OH group in jute fiber (Khan et al. 2013a).

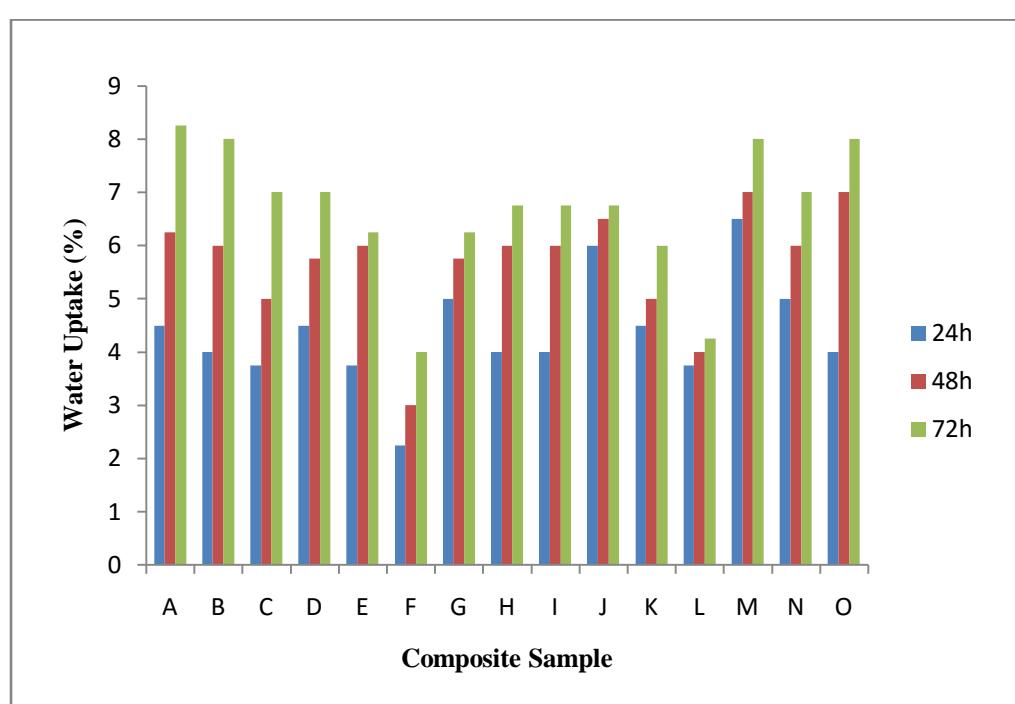


Fig. 1: Effect of immersion time on water absorption of raw and treated jute fibers PB composite

b) Mechanical Properties

The tensile properties of unidirectional jute fiber reinforced PB composites were investigated by means of the effect of coupling agents (DgPB, MAPB) and

chemical treatments of fiber. The tensile strength, Young's modulus and elongations were obtained from tensile stress-strain data.

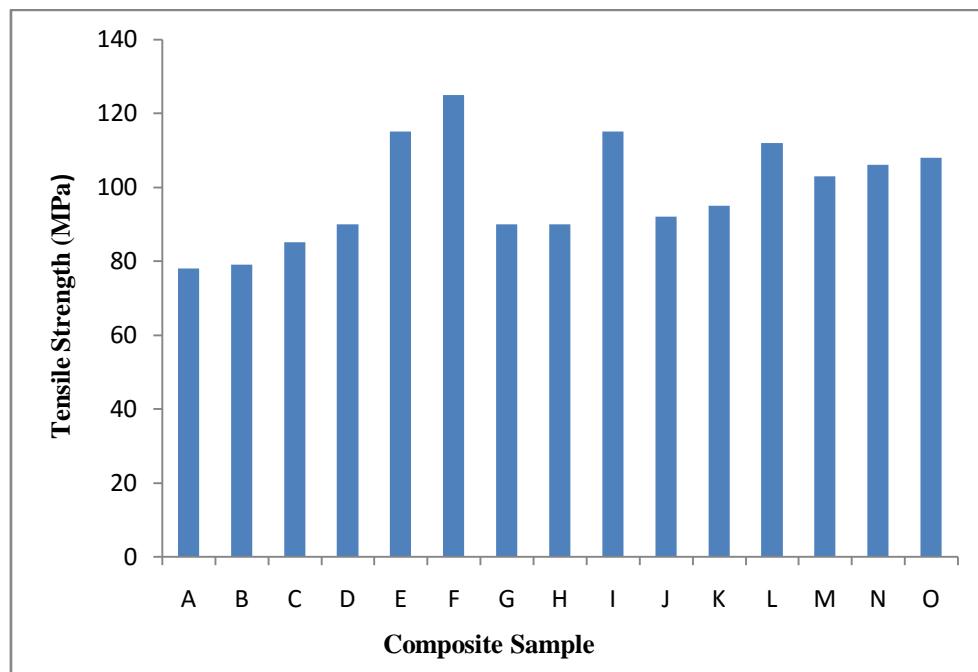


Fig. 2: The effect of chemical treatments and coupling agents on the tensile strength of raw and treated jute fiber PB composites

Fig. 2 shows the effect of chemical treatments and coupling agents (DgPB and MAPB) on tensile strength (TS) of raw and treated jute fibers PB composites. It is observed that the coupling agents added in both raw and treated jute fibers - PB composites yield higher TS than without coupling agents. Among the composites, the highest value of TS is found for 10% NaOH JF+PB+MAPB composite and the lowest value is obtained for raw JF+PB composite. The chemical treatments are employed in this investigation such as NaOH, DCPO, K₂Cr₂O₇, H₂O₂ are all oxidative treatments process. These treatments can make the fiber surface 'clean' due to the removal of impurities, pectin, waxes, hemicellulose and part of lignin. The removal of these substances enhances the surface roughness. Therefore, the jute fiber easily wetted by PB matrix. Also, interlocking between fiber and matrix is increased. The result shows, the presence of coupling agents caused better adhesion of jute fibers and polybutylene matrix and formed a strong interfacial bond between them. The coupling agents MAPB and DgPB have a similar molecular arrangement with extra functional groups which forms a chemical bond with hydrophilic jute fiber. The main PB chain of the coupling agents entangle the mutual chains of PB and therefore builds stronger interfacial adhesion between the PB matrix and jute fiber (Khan et al. 2013a; Miyazaki et al. 2008, Khan et al. 2015a). Both coupling agents have a similar mechanism of bond formation via esterification between the -OH group in jute fiber and the reactive γ -lactone groups in the coupling agents (Khan et al. 2013a).

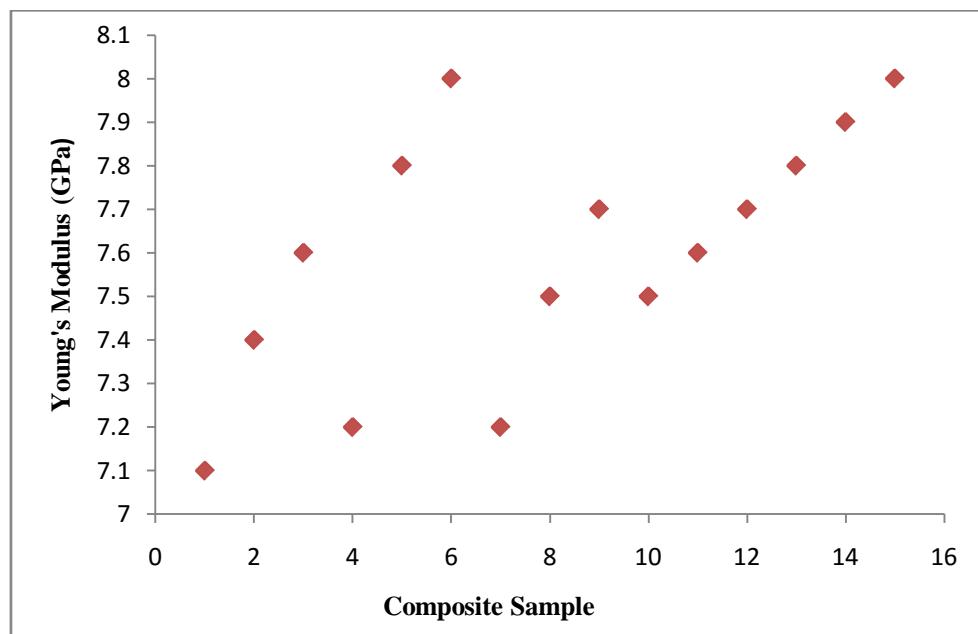


Fig. 3: The effect of chemical treatments and coupling agents on Young's modulus of raw and treated jute fiber PB composites

Fig. 3 shows the comparison of the effect of chemical treatments and coupling agents on YM of raw and treated jute fibers PB composites. It shows that the addition of coupling agents increases YM of raw and

treated jute fibers PB composites. The highest and lowest values of YM are obtained for 10% NaOH JF+PB+MAPB and raw JF+PB composites, respectively.

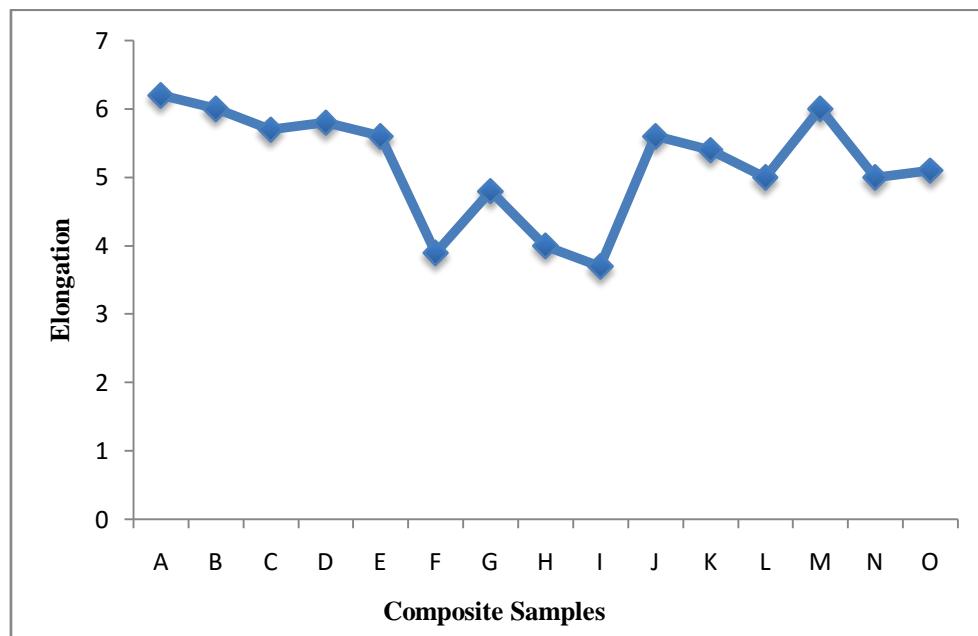


Fig. 4: The effect of chemical treatments and coupling agents on elongation at break of raw and treated jute fiber PB composites

Fig. 4 shows the effect of chemical treatments and coupling agents on elongation properties (PE) of raw and treated jute fibers PB composites. From the Figure 4, it is observed that the highest PE value was found for raw JF+PB composite, then 10%NaOH JF+PB+MAPB and the lowest for 0.05% K₂Cr₂O₇

JF+PB+MAPB composites. The lower values of elongation for composites which were made by using chemical treated fiber and coupling agents may be due to the increase of brittleness of the composite.

Table 2 shows the properties of sawdust reinforced polybutylene composites of some previous

research. Since the properties of composites mostly depend on the properties of matrix use on it. The improvement of mechanical properties of our

composites is much higher than others. On the other hand, a great extent of water uptake was also found in this study.

Table 2: Comparative Result from Some Previous Study

Amount of Sawdust	Chemical treatments	Compatibilizer	Method of Molding	Tensile strength(MPB)	Tensile Modulus (GPB)	Water absorption (%)	Ref.
15 wt%	Diazonium salt		Injection	28.4	1.2	0.2	Rahman et. al 2010
40 wt%		DCP or BPO peroxide/maleic anhydride(MAH)	Compression	24.8	-	-	Santos et al. 2016
50 wt%	3- Aminopropyl tri ethoxy silane	MAH	Injection	49	-	-	Kim et al. 2010
	3-Methacryloxy propyl trimethoxy silane (MPS)	MAH	Injection	49	-	-	
	Vinyl trimethoxy silane	MAH	Injection	49.2	-	-	
30 wt%	NaOH	MAPP	compression	127	8.1	2.1	
	NaOH	DgPP	compression	118	7.9	3.9	

IV. CONCLUSION

The study illustrates the properties of composites fabricated by hot press molding method using polybutylene (PB) as matrix and jute fibers. The 2wt% DgPB and MAPB are used as a compatibilizer to improve the surface adhesion between the hydrophilic jute fibers and hydrophobic PB matrix. As a result, composites showed higher mechanical properties than that of composites prepared by without coupling agent. In case of, the raw JF+PB composite have higher ability to absorb water and 10%NaOH JF+PB+MAPB composite has lower ability to absorb water.

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