PHYSICAL PARAMETERS AND MECHANICAL PROPERTIES IMPROVEMENT FOR JUTE FIBER/POLYPROPYLENE COMPOSITES BY MALEIC ANHYDRIDE COUPLER

Byung S. Hwang*, Byung S. Kim*, Jung H. Lee*, Joon H. Byun*, Jong M. Park**

***Composite Materials Group, KIMM, Changwon, Korea 641-010**
****Gyeongsang National University, 900 Gajwadong, Jinju, Korea 660-701**

Keywords: *Jute Fiber, Maleic anhydride grafted polypropylene, Melt flow index, contact angle*

Abstract

In order to improve the mechanical properties of jute fiber/polypropylene (PP) composites, the property change with the addition of a coupling agent, maleic anhydride, polypropylene (MAPP) was examined experimentally. The maleated coupler acts as an intermediate to chemically connect the polar nature of the fiber and non-polar nature of the polyolefin polymer resin. Furthermore, the decrease in viscosity of the resin which results from the melting point reduction by the MAPP, leads to an increase of contact area with the fiber interface. We discussed the improvement of the PP composite blend of the maleated coupler with the 80mm jute long fiber mat in conjunction with the change of physical parameters in the thermoplastic resin. We confirmed the extent of contribution to the mechanical physical enhancement by using the following parameters: melting flow index(MI) and viscosity, contact angle, thickness of the composite, interfacial shear strength and morphology observation etc. Especially it was observed that the MI and viscosity, MAPP mixture had a very strong relationship with the tensile and flexural strength and modulus, and interfacial shear strength (IFSS).

1. Introduction

Natural fibers, well known as environmental friendly materials, have many applications around. Scientific researches have been performed for more than a decade. When compared with glass fibers, Cellulose fibers posses compatible strength/density and stiffness/density ratios, respectively. Strength/density for glass fiber is 1.35 while that of cellulose is 1.6~2.95 [1~3]. The well known weaknesses of natural fiber reinforced composites are interfacial strength between the fiber and the resin, uneven thicknesses of the fibers, easy moisture absorption and limited temperature applications.

To improve interfacial issues, applying silane [4, 5], NaOH [4, 6, 7] and plasma treatments [8] were studied. However, in the mid eighties to early ninties, with coupling agent maleated polyolephine provided great improvement in composites' properties [9~12].

Numerous researches were performed with addition of maleic anhydride(MA) coupling agent. However, not many researches were performed with 80 mm long natural fibers. Thus, in present work, the property variation with MA addition was carried out. Especially, the effect of MAPP-PP blends to the jute fiber mats were investigated experimentally.

2 Experiments

2.1 Materials

Thermoplastic matrix used in this research is Polypropylene (Polymirae) and E-43 (Eastman) was used as a MAPP coupling agent. Fibers were Bangladesh jute fibers. Their properties are summarized in Table 1. For single fiber test, fibers with similar diameter were selected. PP-MAPP blend contained 1, 3, 5wt% of MAPP, respectively. PP-MAPP blends were prepared by twin screw extruder.

Table 1 Specification of ingredient materials

Materials	Jute	Polypropylene (PP)	Maleic anhydride grafted polypropylene (MAPP)
Vendors	KSTECH	PolyMirae	EASTMAN
Type	Fiber	Fiber	Fiber
Model	Short	Moplen HP652P	E-43
Diameter	40~120 ㎛	6~15 Denier (1 D =0.05g/450m)	6~15 Denier (1 D =0.05g/450m)
Length	80±10 mm/ Continuous	80±10 mm	80±10 mm
Density	1.35 g/cm^3	0.9 g/cm^3	0.934 g/cm^3
Mn	-		3,900 g/mol

2.2 Test Specimen Preparation

In preparing IFSS (interfacial shear strength) test specimen, melted PP at 190℃ was poured into a mold containing the fibers lined up. Extruder was utilized to obtain the test specimens of resin blends. Composite panels were made by adding PP to fiber mat. 200 x 190 mm composite panel was pressure cured at 200℃ and 34kg$_f$/cm^2 for 2 minutes. 5 test specimens were prepared according to ASTM D-638 and D-790.

2.3 Testing and Instruments

HAAKE Torque rheometer was used for blending MAPP-PP and SPF-100 (Hyundai Heavy Ind.) extruder was used for test specimen extrusion. Due to the diameter irregularity of the fibers, IFSS measurements were carried out by the method suggested by Son T. Q. [13] with 10 specimens each for three MAPP contents, respectively. For MI(melting flow index) measurements, DKS-0125G extruder was used. Melting viscosity was measured by Rheometer system model Gemini 150/200. Composite test specimens were cured by 50 and 150 ton press. Mechanical tests were carried out by Instron 5567 with 3-ton load cell and strain gages (FCA-5-11-1L). The morphology of fracture surface was obtained by Jeol JSM 5900. Moisture absorption condition was at 70℃, 90%RH for 400 hrs.

3. Results and Discussions

3.1 Melt Flow Index and melting viscosity

In Fig. 1, the changes of Melt Flow Index (MI) with MAPP contents are shown. As MI increases, the contact area with the fiber increase since the resin flow between the fibers becomes easier. The increased contact area improves the chemical bonding opportunities and physical bonding between the fiber and resin. This behavior will provide the improvement of the material properties.

MI as well as interfacial bonding and physical interlocking will decide the level of thermo plastic material property improvements depending on the state of resin infiltration.

Fig. 2 represents the changes of viscosity measurements with varying MAPP contents. Since, with shear rate of 0.5 ~ 20, the viscosity decreased with increase in MAPP contents, MI analysis may be applied. Also noticed is the viscosity decrease is distinct with MAPP contents increase at low shear rate and very small viscosity changes at high shear rate. In extrusion or injection process, where high shear rate is applied, the decrease in viscosity is not obtained even with increasing MAPP contents. However, with press process where low shear is applied, the viscosity decrease can obtained efficiently. With MAPP content of 1, 3 and 5%, respectively, similar values of viscosities were obtained indicating that similar wetting effect would be obtained at these MAPP contents.

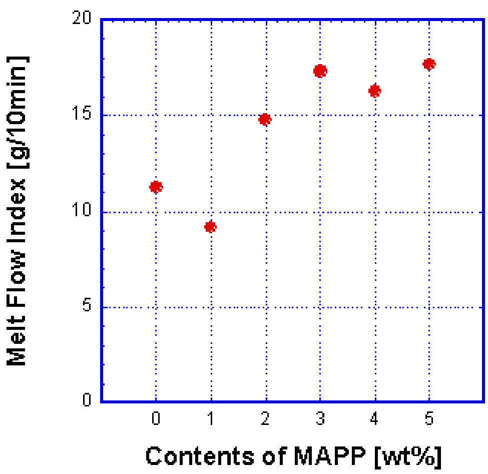


Fig. 1 Change of MI against contents of MAPP

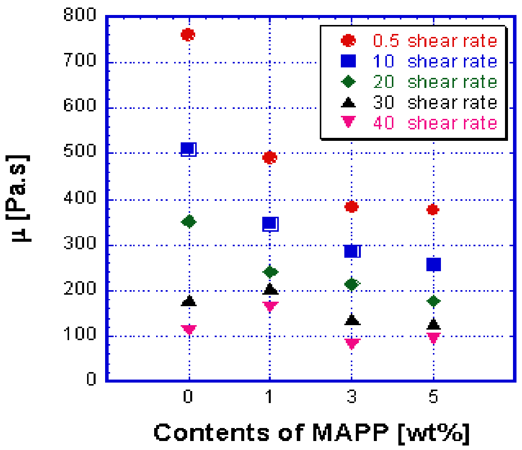


Fig. 2 Viscosity vs contents of MAPP at various shear rate

3.2 Changes in contact angle

Contact angles have been measured to evaluate the compatibility between the natural fiber and polymer. Usually, non-polar polymer posses larger contact angle than polar natural fiber.
Coupling agent to natural fiber reduces the contact angle and improves the interfacial bonding at the interface. Felix J. M.[10] measured the contact angles of 130~140° from MAPP treated cellulose. This says that the contact angle is increased by the coupling agent treatment to natural fiber and then improves the interfacial compatibility. In present study, instead of measuring the contact angles, the compatibility is estimated by observing the contact angle changes with the varying coupling agent contents.
Table 2 summarizes the dynamic contact angles of MAPP-PP matrix measured by Wilhemly plate method. Presence of MAPP showed lower contact angles that of than no MAPP added PP. This indicates that the maleic anhydride link reaction improves with –OH group to reduce the contact angles and to improve wetting with the natural fibers.
From the test results, it may be noticed that the rapid reduction of the contact angles on the fibers with the addition of 3% MAPP.

Table 2 Contact angles of jute fibers and matrices

Materials	Conditions	Contact angle (θ)			
		Formamide	Ethylene glycol	Diiodomethane	Water
Jute fiber	Untreated	68.18	62.80	63.90	78.46
	Silane	76.11	63.70	64.10	79.74
	NaOH	63.34	49.50	52.70	71.10
MAPP(%)	0	81.10	71.46	74.10	85.60
	1	76.70	71.03	73.74	84.78
	3	76.70	68.72	73.50	84.31
	5	74.39	65.79	66.67	83.68

*Probe liquids for the contact angle measurements
-FM: formamide
-EG: ethylene–glycol
-DO: diiodomethane

3.3 Changes in the composites' thickness

Following the preheating at 200℃, Jute/PP panels (220 x 190 mm) is press formed with $34kg_f/cm^2$ pressure. Press formed plate thickness have varied with different MAPP contents as shown in Fig. 3. This behavior indicates that there is a close relation with melt flow index and viscosity.

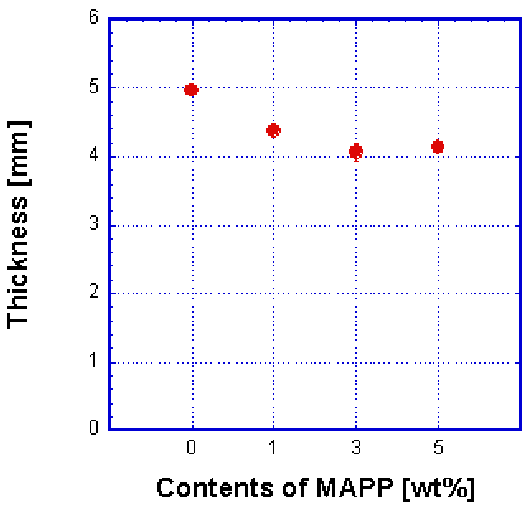
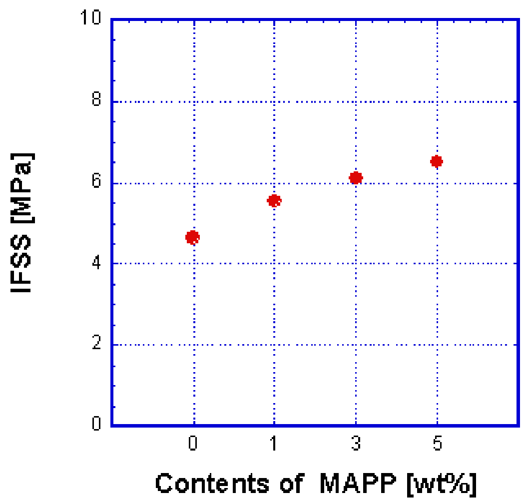


Fig. 3 Thickness change of specimens vs. contents of MAPP

3.4 Improvement of interfacial shear strength, IFSS

IFSS test is carried out to evaluate the interfacial strength between the fiber and the PP. Interfacial shear strength of an impregnated single fiber was measured. Fig. 4 shows that IFSS increased with increasing MAPP contents. With 1, 3 and 5% of MAPP, IFSS have increased 20, 32 and 40%, respectively. Higher IFSS was obtained from higher MFI and lower viscosity. With 3 and 5% of MAPP, MFI have increased 54 and 57%, respectively. Melting point have changed, under 10 and 20 shear rate test conditions, to 44 and 50%, respectively, with 3 and 5% MAPP contents.



Fig, 4 Increase of IFSS vs. contents of MAPP

3.5 Increase in tensile and bending strength

The tensile strength of composite, with mat composed of long fibers, is increased by 31, 94 and 63%, respectively with 1, 3 and 5% MAPP contents. Among the different MAPP contents, 3% content showed highest increase. Fig. 5 and 6 shows the comparison between present work and that of Keener [14]. Although Keenr have used different fibers and different MAPP contents, the changes in the tensile strength changes are in similar trend. After initial MAPP addition, the additional MAPP did not contribute to the increase in strength as much. Even in our result, exceeding 3% of MAPP, the strength declined. The excessive MAPP, instead of improving the adhesiveness, act as an inhibitor to the chemical reaction.[11, 15]

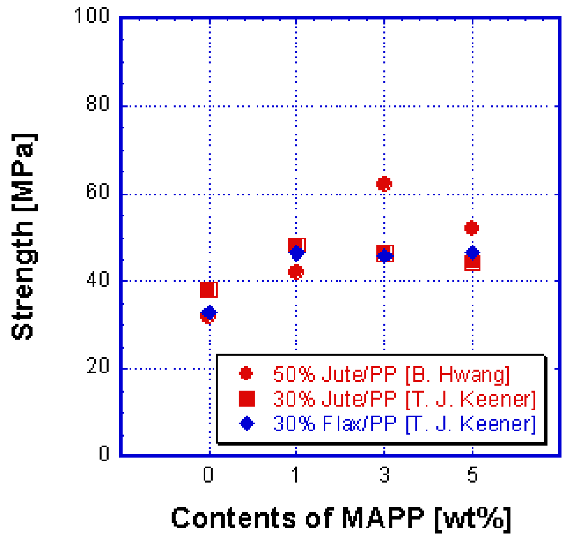


Fig. 5 Comparison of tensile strength of natural fibers composites

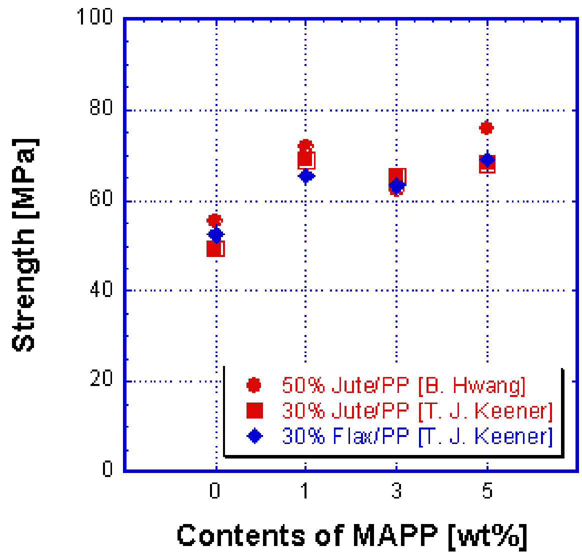


Fig. 6 Comparison of flexural strength of natural fibers composites

3.6 Effect of humidity conditioning

The changes in Flexural strengths after the humidity conditioning are shown in Fig 7(a) and (b). Moisture absorbed fibers leads the composite to have lower strength. However, the maleated coupler seems to somewhat protect the fibers from the moisture absorption. The strength and modulus have decreased after the moisture absorption but still in same pattern.

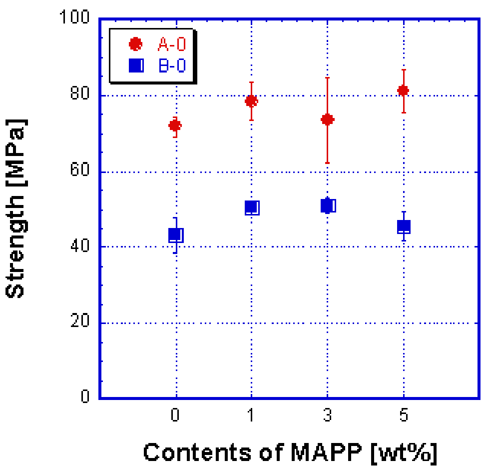


Fig 7(a) Flexural strength changes after humidity conditioning.
A: before conditioning, B: after conditioning

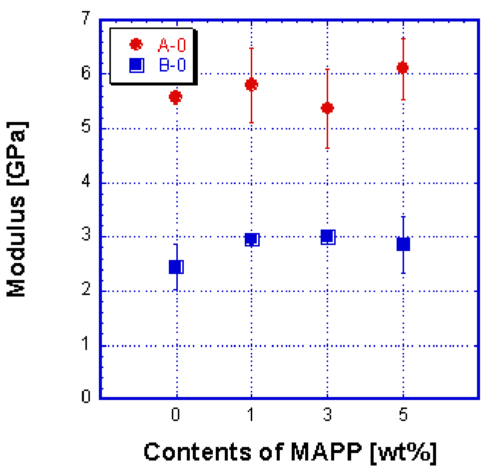
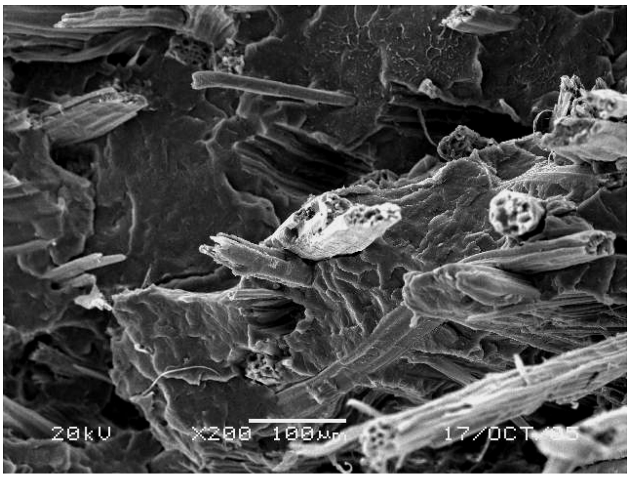


Fig 7(b) Flexural moduli changes after humidity conditioning.
A: before conditioning, B: after conditioning

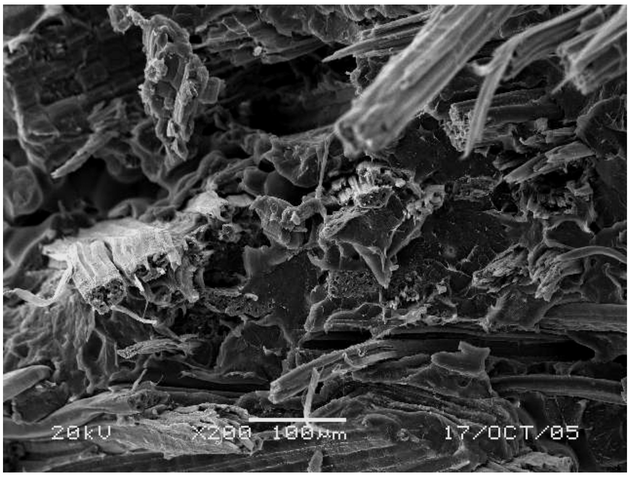
3.7 Morphology

In Fig. 8 the resin may be seen between the fibers and interface. In 0% MAPP specimen, the fibers are not as compacted as compared with the specimen with 1, 3, and 5% MAPP contents. MAPP contents lower the melting point and allow easier resin flow. With 5% MAPP, the gap between the fibers is much smaller. In 0% MAPP specimen, the resin shrinkage is observed which resulted from cooling in fabrication. In case of 5% MAPP specimen, the failure occurred along the fiber length since the interfacial strength between the fiber and the resin is much better. Fig. 9 shows the differences in the interfacial state, resin residue and the smoothness of the resin on the fiber.

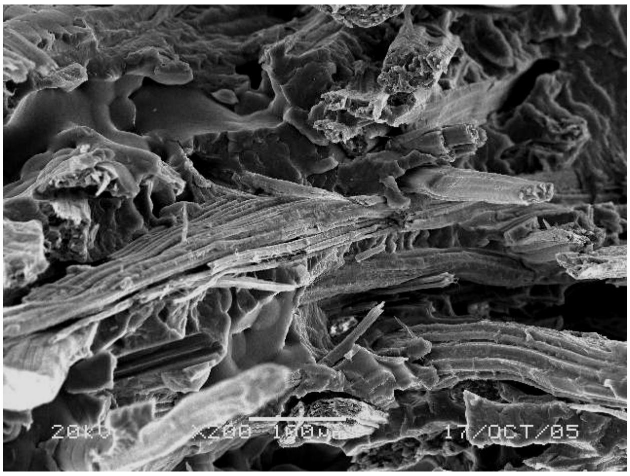
(a)



(b)

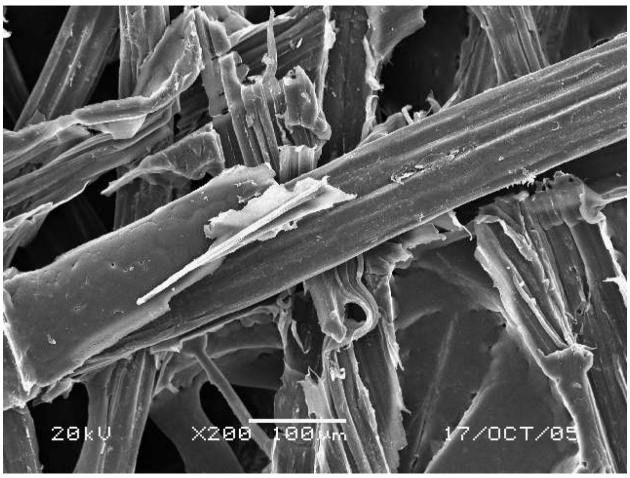


(c)



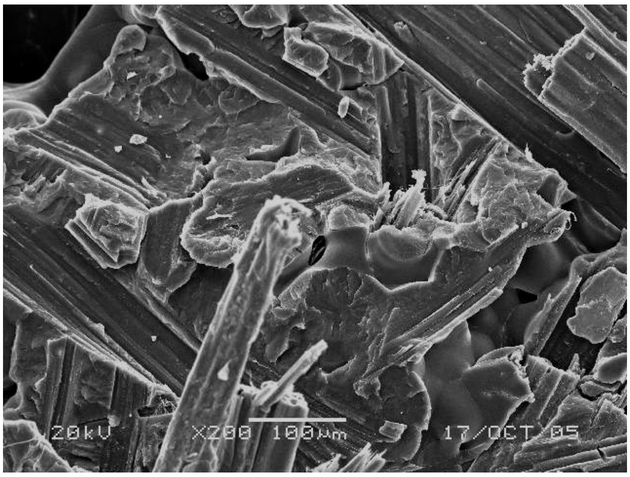
(d)

Fig. 8 Scanning electron micrograph of composite cross section, x200, (a) 0%, (b) 1%, (c) 3%, (d) 5% of MAPP contents. Numbers indicate contents of MAPP.

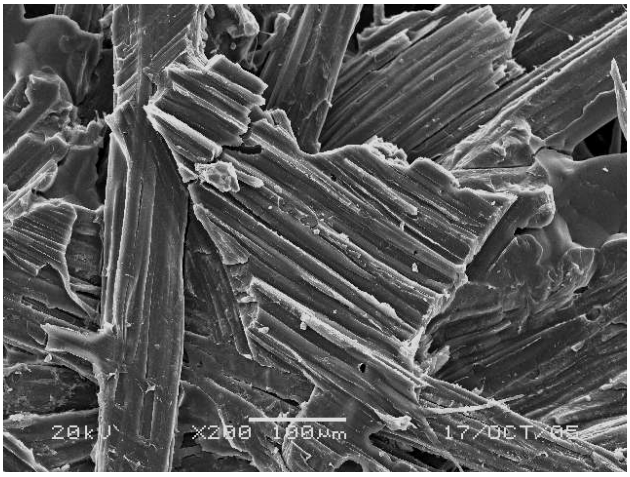


(a)

(b)



(c)



(d)

Fig. 9 SEM photos of interlayers: (a) 0%, (b) 1%, (c) 3%, (d) 5% MAPP

4. Conclusions

1) Highest Melt flow index and viscosity were obtained with 5% MAPP content. From this effect, IFSS increased in 1, 3 and 5% MAPP contents order. Similarly, the composite thickness also changed with MAPP contents as well. Since more than 3% MAPP showed some decrease in strength and considering economics, 3% MAPP contents may be most appropriate one.

2) With fiber length of about 80mm, MAPP contents have improved the tensile and interfacial shear strength.

3) Contact angle reduction by maleic anhydride addition improved the chemical bonding with the fibers.

5. References

[1] O'Dell, J. L., *"Natural fibers in resin transfer molded composites,"* Proceedings of the Fourth International Conference on Woodfiber-Plastic Composites, May 12-14, 1997, pp. 280-285 Madison, WI. U.S.A.

[2] Bolton, A. J., "Natural fibers for plastic reinforcement," Mat. Tech. Vol. 9, No. 1/2, 1994, pp. 12-20.

[3] Jian, J., Okubo, K., and Fujii, T., *"Fabrication of bio-decomposable composites using natural fibers and their strength properties,"* Pro. of the 31st International SAMPE Technical Conference, Oct. 26-30, 1999, pp. 355-366, Long Beach, CA. U.S.A.

[4] V. Weyenberg, et al, *"Influence of processing and chemical treatment of flax fibers on their composites,"* Composite Sci. and Tech., Vol. 63, 2003, pp. 1241-1246.

[5] V. Gonzalez, et al, *" Effect of fiber surface treatment on the fiber-matrix bond strength of natural fiber reinforced composites,"* Composites: Part B, Vol. 30, 1999, pp. 309-320.

[6] Augustine, P. et al, *"Effect of surface treatments on the electrical Properties of low-density polyethylene composites reinforced with short sial fibers,"* Composite Sci. and Tech., V 57, ‘97, pp. 67-79.

[7] Kuruvilla, J. et al, *"Effect of chemical treatment on the tensile properties of short sisal fiber-reinforced polyethylene composites,"* Polymer, V 37, N 23, 1996, pp. 5139-5149.

[8] Belgacem, M. N., Batille, P., and Sapieha, S., *"Effect of corona modification on the mechanical properties of polypropylene/cellulose composites,"* J. Appl. Ploym. Sci., Vol. 53, 1994, pp. 379-385.

[9] Snijder, M. H. and Bos, H., *"Reinforcement of polypropylene by annual pant fibers: optimization of the coupling agent efficiency,"* Composite Interfaces, Vol. 7, No. 2, 2000, pp. 69-75.

[10] Felix J. M. and Gatenholm, P., *"The Nature of adhesion in composites of modified cellulose fibers and polypropylene,"* J. Appl. Polym. Sci. Vol. 42, 1991, pp. 609-620.

[11] Lu, J. Z., Wu, Q., and Negulescu, I. I., *"The Influence of maleation on polymer absorption and fixation, wood surface wettability, and interfacial bonding strength in wood-PVC composites,"* Wood and Fiber Science, Vol. 34, No. 3, 2001, pp. 434-459.

[12] Godavarti, S., *"Optimization of coupling agent characteristics for maximizing performance of wood fiber thermoplastic composites,"* ANTEC 2003-Proceedings of the 61st Annual Technical Conference and Exhibition, Vol. XLIX, Nashville, TN, May 4-8, 2003, S. of Plastics Engineers, p. 2047.

[13] Son T. Q., Park, J. M., and Hwang, B. S., *"Interfacial evaluation of modified jute and hemp fibers/polypropylene-maleic anhydride polypropylene copolymers (MAPP) composites using micro-mechanical test and nondestructive acoustic emission,"* The Korea Society for Composite Materials, Proceedings on Fall National Conference, Kyungpook National U., Nov. 18, ‘05, pp. 157-160.

[14] Keener, T. J. et al, *“Maleated coupling agents for natural fiber composites,”* Composites: Part A No 25. 2004, PP 357-362.

[15] Maldas D. and Kokta B. V., *"Improving adhesion of wood fiber with polystyrene by the chemical treatment of fiber with a coupling agent and the influence on the mechanical properties of composites," Adhesion Sci. Technol*, Vol. 3, No. 7, 1989, pp. 529-539.