

A STUDY ON THE EFFECT OF PLASMA TREATMENT ON MECHANICAL PROPERTIES OF JUTE FIBER/POLY (LACTIC ACID) BIODEGRADABLE COMPOSITES

D. W. Lee¹, G. B. Nam¹, C.Venkata Prasad¹ T. Fujii², B. S. Kim³, J. I. Song^{1*}

¹School of Mechatronics, Changwon National University, Changwon, South Korea- 641 773,

²Department of Mechanical Engineering & Systems, Doshisha University, Japan

³Korea Institute of Materials Science, Changwon, Korea

*jisong@changwon.ac.kr

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Abstract

Plasma treatment (PT) for various exposure timings (30, 60, 90 and 120 sec) was performed to study the mechanical properties of jute fiber and its composites using poly (lactic acid) (PLA) as the matrix. PT fiber composites showed superior properties compared to other treatments. Micro droplet test results showed that the interfacial shear strength (IFSS) of PT fiber composite is higher than that of AT fiber composites. Mechanical properties and hardness was increased on subjecting the fiber to plasma treatment. Tensile strength, young's modulus and flexural strength were increased in an order of 28%, 17%, and 20%, respectively for plasma polymerized jute fiber composites. Moreover, plasma polymerization leads to increase (>20%) in the flexural strength of the composites. Overall, PT is an eco-friendly method for surface modification of lingo cellulosic fiber to increase the compatibility between the matrix and fiber.

1. Introduction

Natural fibers show unique characteristics such as; low density, high mechanical properties, CO₂ neutralization, and bio-degradable nature, low cost, lightweight, biodegradable, and biocompatible features [1-7]. Furthermore, due to simple manufacturing technique natural fibers have found many applications Viz., infrastructure, automotive and packing sectors [8, 9]. All these characteristics motivate in finding the novel applications of the natural materials tending to improve standard living of the people, particularly in the rural sector.

In general, natural fibers have low interfacial shear strength (IFSS) with polymer matrix i.e. the former is highly polar and hydrophilic while the latter is non-polar and relatively hydrophobic in nature. This trend imposes the surface modification of natural fiber, with the aim of improving the fiber/polymer compatibility and their interfacial adhesion [5, 10]. Natural fibers without surface modification embedded in a polymeric matrix generate unstable interfaces and the stress applied to the fiber/polymer composite is not efficiently transferred from the matrix to the fiber and the beneficial reinforcement effect of the fiber remains under exploited. Likewise, the poor ability of the polymer to wet the fiber hinders the homogeneous dispersion of short fibers within the polymer matrix [11-14].

Hence, there is a need in improving the compatibility between fiber and matrix. Therefore, most of the composite researchers are working on this particular issue. The most common treatments to modify the surface of the fibers are; removing the superficial layer, changing the topography and the chemical nature by means of thermal, mechanical, and chemical treatments [15].

Physical treatment changes the structural and surface properties of the fiber and thereby influences the mechanical bonding in the matrix [16]. Among the physical methods; plasma polymerization is an effective and environmental concern method for surface modification of the natural fibers (hydrophilic to hydrophobic). Moreover, it is dry method and novel approach that reduces air, water and land pollution in comparison to conventional methods of wet chemistry [17-22]. Surface modification of natural fibers by plasma treatment involves either activation of the surface or grafting with suitable monomers.

In this study, the properties of plasma polymerized (surface activation) jute fibers (bundle) were studied and compared with NaOH treated and untreated fibers. This article also explains the effect of plasma treatment of jute fiber on the mechanical properties of jute/PLA bio-composites.

2. Experimental

2.1 Materials

Raw jute was obtained in the form of a long fiber from a domestic fiber company located in South Korea. Jute fibers were washed with detergent and then with distilled water repeatedly to remove greasy material adhered on the surface of the fiber and dried to constant weight. PLA (PL-1000) was purchased from Miyoshi Oil and Fat Co. in the form of pellets.

2.2 Fiber Treatment

Alkali treatment of jute fiber was carried out using various concentrations (1-7 w/w%) of NaOH as per the procedure reported by Tapasi et al [3]. Plasma polymerization was carried out at a plasma power of 3kV and 20 kHz using helium and acrylic acid as a carrier gas and monomer, respectively as per the procedure reported elsewhere [13, 14, and 19].

2.3 Compounding and sample preparation

Prior to compounding, PLA and alkali treated (AT) jute short fibers were dried at 80⁰C for overnight in an oven. The jute fiber/PLA composition was kept constant at 20/80 w/w% in all formulations. The formulations were prepared in a twin-screw extruder at a screw speed of 100 rpm and the temperature of different zones was maintained in the range of 175-190⁰C. After compounding, the compounds were palletized. The specimens for mechanical tests were prepared by injection molding at 180⁰C. To study the effect of various treatments, composites were also prepared using neat PLA, untreated (UT), plasma treated (PT), and alkali/plasma treated (APT) fiber by repeating the above procedure.

2.4 Characterization

The tensile tests was conducted as per the ASTM D 6389 standards using universal testing machine (Instron 3369, 165X13X3 mm) at a cross-head speed of 5 mm/min and applied load

of 10kN. In each case at least five samples was tested and the values presented are in averages. The bundle fiber tests were carried out using a tensile test machine (Micro load system, R&B, Korea) at a crosshead speed of 1mm/min.

In order to study the IFSS of the composites, micro droplet test was carried out by micro indenter (EZ-S, Shimadzu, Japan) at a crosshead speed of 1mm/min for a period of 10sec.

The surface and tensile fractured surface morphology of the composites was analyzed using scanning electron microscope (JEOL, JSM Model 6360). In order to prevent arcing, sample surfaces were coated with gold.

Hardness of the samples was obtained using Vickers micro hardness tester with a contact load of 5 kg for 15 s (Dimensions: 25×25×3 mm). Five hardness values from different locations well distributed on the sample surface were measured for each sample, the average value of these measurements being considered the hardness of the sample.

The flexural tests of the specimens was performed as per the ASTM D standards (ASTM D 790-03) at a cross-head speed of 2 mm/min using three-point bending mode (80X 24.2X 3.2 mm)

3.0 Results and Discussion

3.1 Effect of surface treatment on tensile properties of bundle fiber

Figure 1 represents the tensile test results of untreated and treated (alkali, plasma, alkali/plasma, E-beam and E-beam/plasma) jute fiber (bundle fiber tensile test). The tensile strength of untreated (UT) fiber (285 MPa) is higher than those of alkali (AT) and plasma treated (PT) fibres (227 MPa), where an increase in tensile strength was observed in case of alkali/plasma treated (APT) fibers. The tensile strengths of APT fibers treated for 30, 60, 120 seconds were 282, 310 and 385 MPa, respectively. Whereas, tensile strength of E-beam treated (EBT) jute fiber was 261 MPa. The APT fibers treated for 120 sec showed higher tensile strength (>41%) than untreated fiber. The tensile results of E-beam/plasma treated (EBPT) fibers are lower than those of EBT fibers. Tensile strength of PT fibers treated for 30, 60, 120 sec are 244, 180 and 168 MPa, respectively. E-beam and plasma treatment did not show much effect on tensile properties of jute fibre but whereas, APT showed significant effect on the properties.

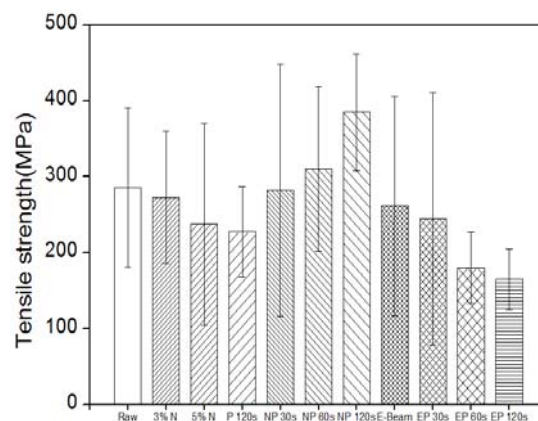


Figure 1. Tensile strength of jute fiber (bundle) of various treatments.

3.2 Effect of surface treatment on interfacial strength

It is well known that mechanical properties of composites are largely controlled by the interface, which is usually required to be strong in polymer matrix composites, thus transferring load from the matrix to the fibers efficiently [23-25]. The mechanical property of the interface was characterized by IFSS method. Figure 2 shows the effect of various surface treatments on IFSS of jute fiber/PLA composites. The IFSS of untreated jute fiber/PLA composite is 3.59 mPa, and for 3% AT fibre composites increase in IFSS was observed (5.93 mPa). Beyond 3% alkali treatment (5 and 7%) decrease in shear strength (3.82 and 2.41 mPa) was observed which less than those of UT fiber composites are comparably. On the other hand, marginally increase in IFSS was observed in case of PT fiber composites (90% more than UT) with a value of 6.84 MPa.

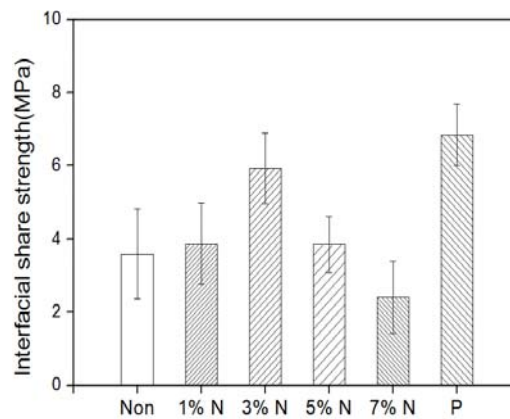


Figure 2. Micro droplet test results of Jute/PLA composites.

The IFSS result indicates that plasma treatment tends to improve the interfacial adhesion between the fiber (hydrophilic) and PLA matrix (hydrophilic). Surface friction coefficient value was increased on PT; this is because of the etching effect of plasma treatment. It is well known that surface friction value indicates the surface roughness character of the fibre. Therefore, increase in IFSS can be explained on the basis of increase in surface roughness of jute fiber tending to better mechanical interlocking. Similar results were also reported by Demir et al [26]. The IFSS results can also supported by SEM analysis described in the forth coming section.

3.4 Morphology of the tensile fractured surface

Figure 3 shows the SEM images of tensile fractured surface of untreated (a1, a2), alkali (3 and 5 %) (b1, b2 and c1, c2) and plasma treated (d1, d2) specimens. Fractured surfaces of UT and AT fiber composites (3%) shows fiber pull out holes in the PLA matrix, indicating the poor interfacial interaction between PLA and UT, 3% AT fiber (not enough concentration to remove the amorphous material). Whereas, in case of 5% AT (optimum concentration to remove the amorphous material) and PT fiber composites, the fibers are good in contact with PLA matrix further leading to increase in tensile strength. These results were also further confirmed by interfacial shear strength results (previous section). Similar results were also reported by Demir et al [26].

It is evident from the comparison of results, that plasma treatment (physical method)

improves the composite's performance more favorably than alkali treatment (chemical method). One advantage of plasma treatment over other wet chemical methods is that material integrity is maintained while there is a large possibility of fiber degradation during chemical treatment. Plasma treatment also has the advantage of being a clean and dry process without affecting the atmosphere associated with chemical modification and also cost effective.

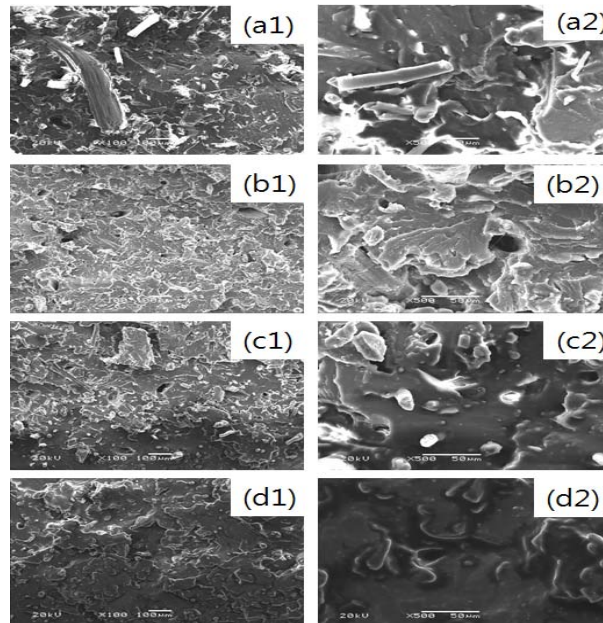


Figure 3. Fracture surface morphology of jute/PLA composites: Untreated (a), 3% AT (b), 5% AT (c) and plasma treated (d) at 100 (left) and 500 (right) magnifications.

3.5 Tensile Properties

The variation of tensile strength and young's modulus with various treatments are seen in figure 4. The composites comprising of plasma treated fibre possessed greater tensile strength (51.8 mPa) and young's modulus (4.07 GPa) followed by alkali treated (5 >3%) and untreated fibre composites. It was found from the SEM analysis that plasma treatment tends to form rough surface which further helps in better interlocking there by promoting good interfacial adhesion between fibre and matrix. The tensile strength of PT fibre composites increased by 28% compared to UT fiber composites. Similar observations were also reported by Moon and Jang [27] in case of polyethylene fiber/vinyl ester composites by the argon plasma treatment.

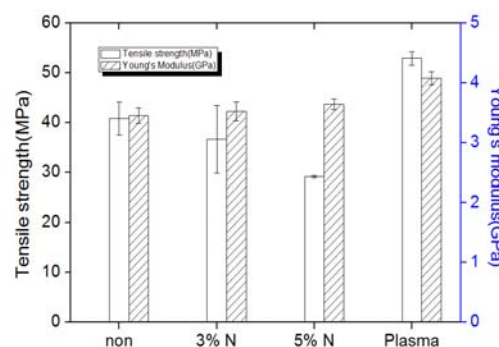


Figure 4. Tensile strength and modulus results of Jute/PLA composite

3.6 Hardness Test

The variation of hardness with PT is presented in figure 5. Plasma treated fibre composites exhibit maximum hardness (21.1 HV) followed by AT (5>3%) (20.7>20.37), UT (18.91) fibre composites and plain PLA matrix (17.22). Untreated jute fiber reinforced composite decreased by an order of 7% hardness pure PLA matrix. The PT method tends to increase the hardness of jute/PLA composites compare to untreated composites. The hardness of 3% and 5% AT jute fiber reinforced composites are 20.37 HV and 20.7 HV, respectively. The Hardness of plasma polymerized composite increased (21.1HV) in an order of 8% than UT jute fiber reinforced composite and 16% higher than pure PLA.

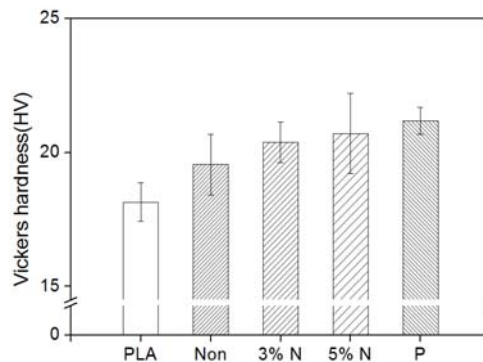


Figure 5. Vickers hardness results of Jute/PLA composites

3.7 Flexural Strength

Flexural properties of jute/PLA composites are presented in figure 7. It is obviously seen that the flexural strength of the composites marginally increases on surface modification compare to untreated fibre composites. Flexural strength of UT jute fiber reinforced composite is 37.7MPa while for AT jute fiber reinforced composites the values are 33.5 MPa and 34.2 MPa for 3 and 5% AT, respectively. Plasma polymerized jute fiber reinforced composite exhibited a flexural strength of 45.3MPa, which is comparably higher than UT and AT fibre composites. The increase in flexural strength of PT fiber composites is due to etching away the surface of fibre by plasma treatment, leaving more non polar lignin on the fiber surface, which contributes to the improve the interfacial adhesion. These results were also supported by SEM analysis of the fiber surface (earlier section). Similar observations were also supported by Yuan et al [28].

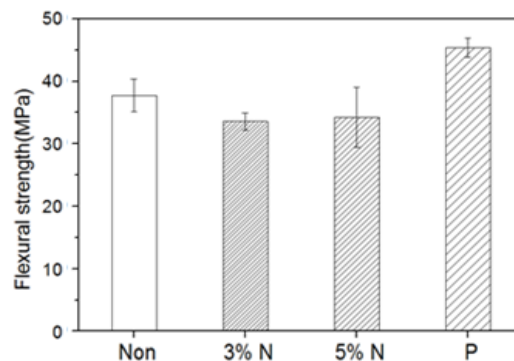


Figure 6. Flexural strength of jute/PLA composites.

4.0 Conclusions

The authors reported on the effect of plasma treatment of jute fibre on the mechanical, Morphological and interfacial shear strength of Jute/PLA (20/80 w/w%) composites. The mechanical properties of bundle fiber and its composites were performed as per ASTM D standards. Plasma treatment induces the changes on the morphology of jute fibre for and fracture surface of the composites with PLA. The tensile, modulus and flexural strength of the plasma treated jute fiber composites was found to be marginally increased compared to UT and AT. The increment in the mechanical properties is due to the heat and etching effect of the plasma treatment which makes the rough surface of the fiber, enabling good interlocking between fiber and matrix. The interfacial shear strength and hardness increases on plasma polymerization. As a result of this study it is concluded that the plasma treatment is an effective and environmental concern method compare to chemical methods.

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References

- [1] Joshi S. V., Drzal L. T., Mohanthy A.K., Arora S. Are natural fiber composites environmentally superior to glass fiber reinforced composites. *Comp Part A*, **35**, pp. 371-376 (2004).
- [2] Mohanty A. K., Misra M., Drzal L. T. Sustainable bio- composites from renewable resources: Opportunities and challenges in the green materials world, *Journal of polymers and the environment*. *J Polym Environ*, **10**, pp.19-26 (2002).
- [3] Tapasi M., Nhol K. PLA Based Biopolymer Reinforced with Natural Fibre: A Review. *J Polym Environ*, **19**, pp.714-725 (2011).
- [4] Mirza, F. A., Afsar, A. M., Kim B. S., Song, J. I. Recent developments in Natural Fiber Reinforced Composites. *J of Korea Society Comp Mater*, **22**, pp.41-49 (2009).
- [5] Sever K., Sarikanat M. Seki Y., Erkan G., Erdogan U., Erden S. Surface treatment of jute fabric: the influence of surface characteristics on jute fabrics and mechanical properties of jute/polyester composites. *Industrial crops and products*, **35**, pp.22-30 (2012).
- [6] Rasel S. M., Nam G. B., Byeon J. M., Kim B. S., Song J. I. Effect of coupling agent and fiber loading on mechanical behavior of chopped jute fiber reinforced polypropylene composites. *Elastomer*, **46**, pp.204-210 (2011).
- [7] Bledzki A. K., Gassan J. Composites reinforced with cellulose based fibres. *J. Prog Polym Sci*, **24**, pp.221-274 (1999).
- [8] Khondker A., Ishiaku U. S., Nakai A., Hamada H. A novel processing technique for thermoplastic manufacturing of unidirectional composites reinforced with jute yarns. *Comp Part A*, **37**, pp.2274-2284 (2006).
- [9] Doan T. T. L., Shang L. G., Edith M. Jute/polypropylene composites: Effect of matrix modification. *J Comp Sci Tech*, **66**, pp.952-963 (2006).
- [10] Ji S. G., Cho D., Park W. H., Lee B. C. Electron Beam Effect on the Tensile Properties and Topology of Jute Fibers and the Interfacial Strength of Jute-PLA Green Composites. *Macromol Res*, **18**, pp. 919-922 (2010).
- [11] Huda M. S., Drzal, L. T., Mohanty A. K., Misra M. Effect of fiber surface-treatments on the properties of laminated biocomposites from poly(lactic acid)(PLA) and kenaf

- fibers. *J Comp Sci Tech*, **68**, pp.424-432 (2007).
- [12]Duigou L., Davies P., Baley C. Interfacial bonding of Flax fibre fibre/Poly(L-lactide) bio-composites. *Comp Sci Tech*, **70**, pp. 231-239 (2010).
- [13]Kim M. M.; Kim, B. S.; Ha, J. R.; Kim, S. K.; Yi, J. W.; Lim, J. Y. Interfacial optimization of lyocell fabric/PLA with silane treatments. *Adv Mater Res*, 2010, 123-125,1155-1158.
- [14]Hai, N. M.; Kim, B. S.; Lee, S. Effect of NaOH treatments on jute and coir fiber PP composite. *Adv Comp Mater*, 2009,18,197-208.
- [15]Bledzki, A. K.; Sperber, V. E.; Faruk, O. *Rapra Rev. Rep* 2002, 13, 15.
- [16]Bledzki, A. K.; Reihmane, S.; Gassan, J. Properties and Modification Methods for Vegetable Fibers for Natural Fiber Composites. *J Appl Polym Sci* 1996, 59, 1329-1336.
- [17]Gassan, J.; Bledzki, A. K. The influence of fiber-surface treatment on the mechanical properties of jute-polypropylene composites. *Compos A: Appl Sci Manuf* 1997, 28, 1001-1005.
- [18]Corrales, F.; Vilaseca, F.; Llop, M.; Giron`es, J.; M´endez, J. A; Mutj`e, P. Chemical modification of jute fibers for the production of green-composites, *J. Hazard Mater* 2007, 144, 730-735.
- [19]Ha, J.R.; Kurniawan, D.; Kim, B. S.; Song. J. I. Mechanical properties of plasma polymerized Cellulose Fabric/Poly(lactic Acid). *KSCM*, 2011, 2, 29-30.
- [20]Kim, B. S.; Chun, B. H.; Lee W. I.; Hwang, B. S. Effect of plasma treatment on the wood flour for wood flour/PP composites. *Thermoplastic Comp Mater*, 2009, 22, 21-28
- [21]Hai, N. M.; Kim, B. S.; Ha, J. R.; Song, J. I. *Adv comp mater*, 2011, 20, 435.
- [22]Khan, M. A.; Khan, R. A.; Zaman, H. U. Alam, M. N.; Hoque, M. A. Effect of surface modification of jute with acrylic monomers on the performance of polypropylene composites. *J Reinf Plastics Comp*, 2009, 29, 1195-1205.
- [23]Coffey, A. B.; O'Bradaigh, C. M.; Young, R. J. Interfacial stress transfer in an aramid reinforced thermoplastic elastomer. *J Mater Sci*, 2007, 42, 8053-8061.
- [24]Park, S. J.; Seo, M. K.; Lee, J. R. Relationship between surface characteristics and interlaminar shear strength of oxyfluorinated carbon fibers in a composite system. *J Colloid Interf Sci* 2003, 268, 127-132.
- [25]Chen, P.; Wang, J.; Wang, B.; Li, W.; Zhang, C.; Li, H.; Sun, B. Improvement of interfacial adhesion for plasma-treated aramid fiber-reinforced poly(phthalazinone ether sulfone ketone) composite and fiber surface aging effects. *Surf Interf Anal*, 2009, 41, 38-43.
- [26]Demir, A.; Seki, Y.; Bozaci, E.; Sarikanat, M.; Erden, S.; Sever, K.; Ozdogan, E. Effect of the atmospheric plasma treatment parameters on jute fabric: The effect on mechanical properties of jute fabric/polyester composites. *J Appl Polym Sci*, 2011, 121, 634-638.
- [27]Moon, S. I.; Jang, J. The mechanical interlocking and wetting at the interface between argon plasma treated UHMPE fiber and vinylester resin. *J Mater Sci* , 1999, 34, 4219-4224.
- [28]Yuan, X. W.; Jayaraman, K.; Bhattacharyya, D. Effects of plasma treatment in enhancing the performance of woodfibre-polypropylene composites. *Compos A: Appl Sci Manuf* 2004, 35, 1363-1374.