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Surface treatment of natural fibers is one of the important methods to improve the mechanical properties of the composite material. In this paper, plasma treatment (PT) for various exposure timings (30, 60, 90, and 120 s) was performed to study the mechanical properties of jute fiber and its composites using poly (lactic acid) (PLA) as the matrix. The results were compared with alkali (AT) and plasma treated (PT) fiber composites. Bundle fiber test was carried out for untreated, AT, and PT jute fiber composites. 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 were 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 than untreated fiber composites. It is inferred that plasma treatment improves the interfacial adhesion between the jute fiber and PLA. These results were also confirmed by scanning electron microscopy observations of the fractured surfaces of the composites. Overall, plasma polymerization is an effective and eco-friendly method for the surface modification of the lingo cellulosic fiber to increase the compatibility between the matrix (hydrophobic) and fiber (hydrophilic).

Keywords: plasma treatment; jute fiber; alkali treatment; interfacial shear strength; poly lactic acid (PLA)

1. Introduction

Bio-composites are the materials composed of biodegradable polymer (matrix) and natural fibers (reinforcement). Development of bio-composites has attracted great interest due to their environmental concern (i.e. biodegradability) and improved properties.[1–4] Natural fibers show unique characteristics such as; low density, high mechanical properties, CO_2 neutralization, and biodegradable 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.

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[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 nonpolar 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.

Plasma surface treatment has several advantages over other processes e.g. it is an eco-friendly process, having the reduced number of chemicals involved and low treatment times. Plasma treatment of different natural fibers and composites has been reported in the literature.[21–26] These composites include combination of different matrix materials with wood flour, rice husk, sisal fibers, jute fibers, and different textile fibers. The authors have studied the effect of plasma treatment on different parameters ranging from surface morphology to mechanical, electrical, and optical properties of fibers and composites. The effect of plasma treatment on single jute fibers is available in literature,[22] which is the study of surface, optical, and electrical properties of fibers only.

This study is focused on effects of plasma treatment on mechanical properties of jute fiber as well as jute/PLA composites. The properties of plasma polymerized (surface activation) jute fibers (bundle) were studied and also compared with NaOH treated and untreated fibers. This study gives guidelines to the future researchers on how to improve the mechanical properties of their composite specimens by adopting a fiber treatment technique, which is more affective, eco-friendly, and takes less time as compared to other treatment procedures.

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. *Jute 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 3 kV and 20 kHz using helium and acrylic acid as a carrier gas and monomer, respectively, as per the procedure reported elsewhere. [13,14,19] The schematic representation of the plasma treatment apparatus is presented in Figure 1. Electron beam (E-beam) and E-beam/plasma treatment was carried for 30, 60, and 120 s on jute fibers.

2.3. *Compounding and sample preparation*

Prior to compounding, PLA and alkali treated (AT) jute short fibers were dried at 80 °C 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 175–1900 °C. After compounding, the compounds were palletized. The specimens for mechanical tests were prepared by injection molding at 180 °C. 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 were conducted as per the ASTM D 6389 standards using universal testing machine (Instron 3369, 165 × 13 × 3 mm) at a cross-head speed of 5 mm/min

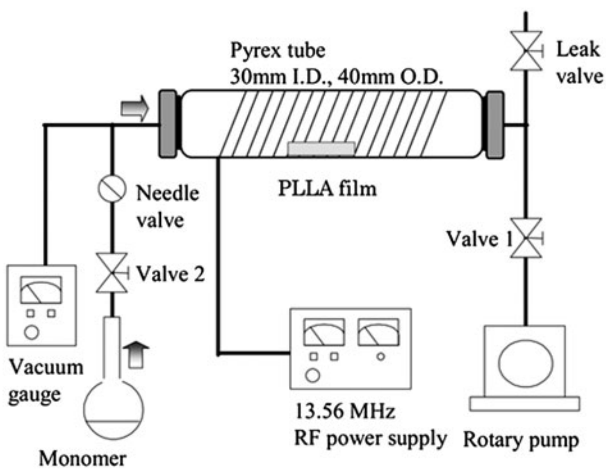

Figure 1. Schematic diagram of atmospheric-pressure plasma treatment apparatus.

and applied load of 10kN. In each case, at least five samples were 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 cross-head speed of 1 mm/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 cross-head speed of 1 mm/min for a period of 10 s.

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.

The flexural tests of the specimens were 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 ($80 \times 24.2 \times 3.2$ mm)

3. Results and discussion

3.1. Effect of surface treatment on tensile properties of bundle fiber

Figure 2 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) fibers (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, and 120 s 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 s showed higher tensile strength (>41%) than untreated fiber. The tensile results of E-beam/plasma treated fibers are lower than those of EBT fibers. Tensile strength of PT fibers treated for 30, 60, and 120 s are 244, 180, and 168 MPa, respectively. E-beam and plasma treatment did not show much effect on tensile properties of jute fiber but whereas, APT showed significant effect on the properties.

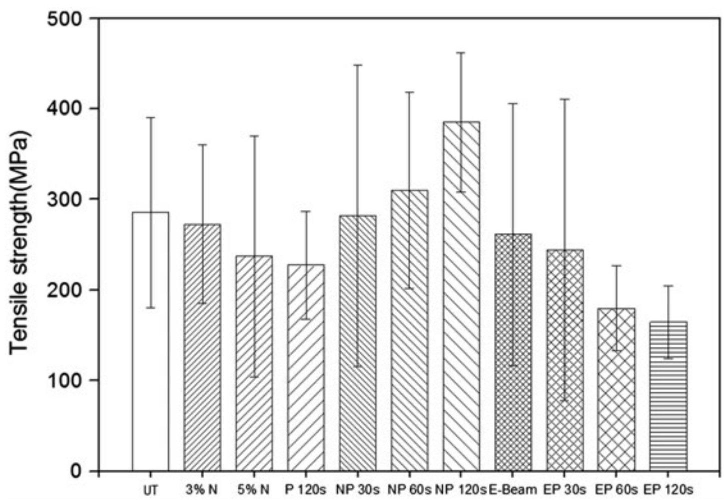

Figure 2. Tensile strength of jute fiber (bundle) of various treatments.
Note: UT: Untreated; N: NaOH; P: Plasma; NP: NaOH/Plasma; EP: Ebeam/Plasma.

3.2. Surface morphology of jute fiber

The long jute fibers isolated from the plant by breaking and scotching processes, are usually called fiber bundles.[27] The fiber bundles have an irregular cross-section unlike the synthetic fibers. These isolated fibers are formed by fibrils, bonded together with weak pectin and lignin interphase. Alkali treatment helps in the removal of weak structures viz. lignin, waxes, and oils on the external surface of the fibrils, depolymerizes cellulose, and exposes the short length crystallites.[28]

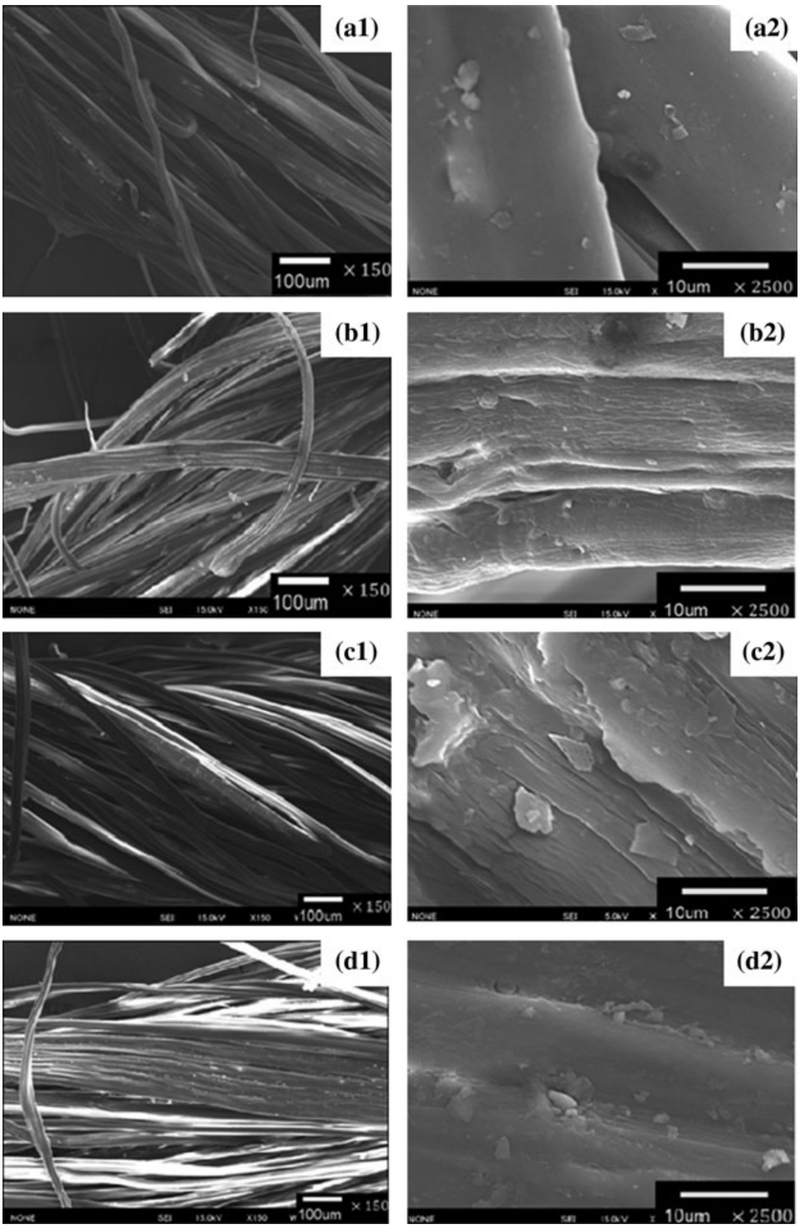

Figure 3. Surface morphology of jute fiber: Untreated (a), 3% AT (b), 5% AT (c), and Plasma treated (d) at 150 (left) and 2500 (right) magnifications.

The SEM micrographs of jute fibers are shown in Figure 3 at different magnifications (a × 150 and b × 2500). In addition to the removal of impurities, alkali treatment also provides cracks on the surface of the jute fiber leading to the rough surface of the fiber (Figure 3(b1) and (b2)); this might be due to change in the crystalline structure of the fibers (e.g. removal of amorphous and partially crystalline impurities, lignin, and hemicelluloses). It can be observed from Figure 3, as the concentration of NaOH increases from 3 to 5%, decrease in fiber diameter can be observed. This can be attributed to the increased extent of removal of non-cellulosic material out of the jute fibrils. From Figure 3 (d1) and (d2), it can also be observed that plasma treatment tends to increase in the surface roughness of the jute fiber which helps for better compatibility between fiber and matrix.

3.3. *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.[29–31] The mechanical property of the interface was characterized by IFSS method. Figure 4 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 fiber 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 is comparatively less than those of UT fiber composites. 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.

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 fiber. 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. [32]. The IFSS results can also supported by SEM analysis described in the forth coming section.

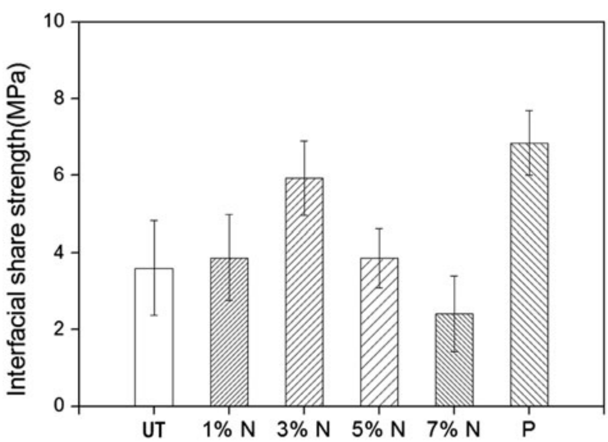

Figure 4. Micro-droplet test results of various jute fibers.
Note: UT: Untreated; N: NaOH; P: Plasma; NP: NaOH/Plasma.

3.4. Surface morphology of the tensile fractured composites

Figure 5 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

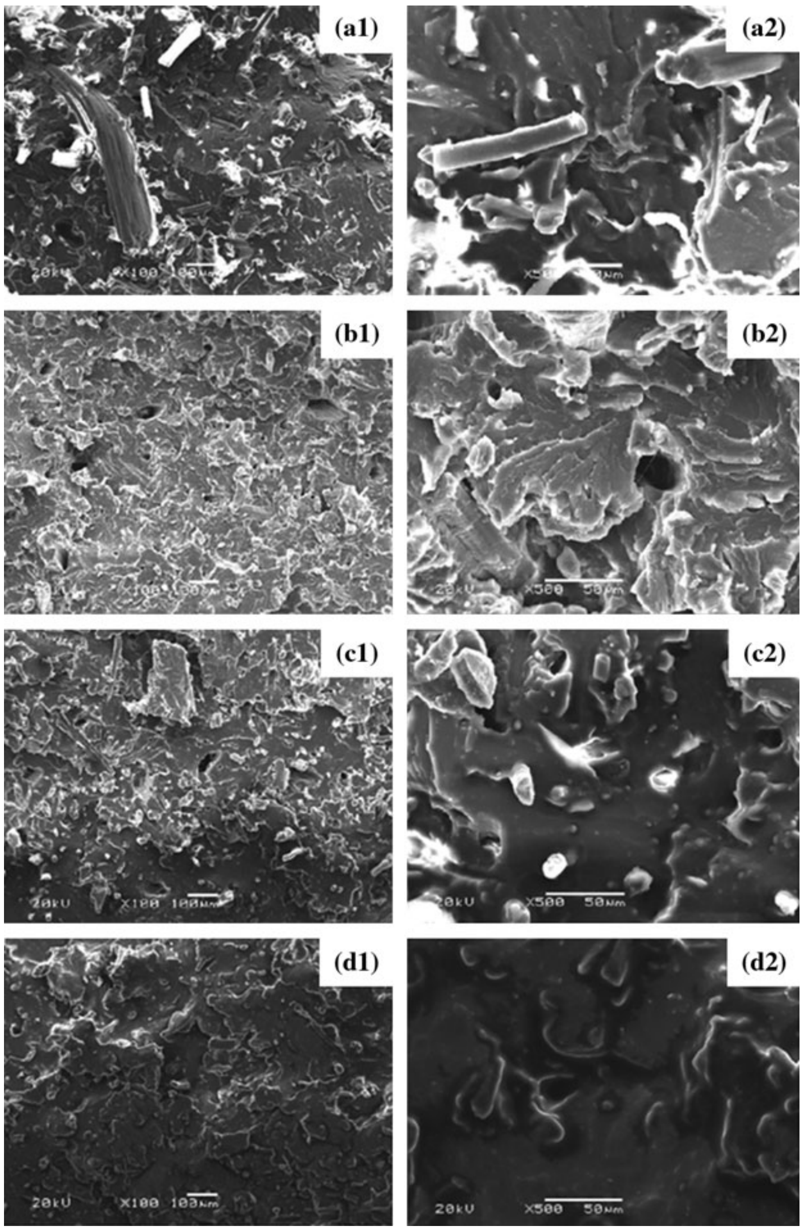

Figure 5. Fracture surface morphology of jute/PLA composites: Untreated (a), 3% AT (b), 5% AT (c), and plasma treated (d) at 100 (left) and 500 (right) magnifications.

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 IFSS results (previous section). Similar results were also reported by Demir et al. [30].

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.

3.5. Tensile properties

The variation of tensile strength and young's modulus with various treatments are seen in Figure 6. The composites comprising of plasma treated fiber possessed greater tensile strength (51.8 MPa) and young's modulus (4.07 GPa) followed by alkali treated (5 > 3%) and untreated fiber 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 fiber and matrix. The tensile strength of PT fiber composites increased by 28% compared to UT fiber composites. Similar observations were also reported by Moon and Jang [33] in case of polyethylene fiber/vinyl ester composites by the argon plasma treatment. The results of stress–strain curves are presented in Figure 7.

3.6. Flexural strength

Flexural properties of jute/PLA composites are presented in Figure 8. It is obviously seen that the flexural strength of the composites marginally increases on surface modification compare to untreated fiber composites. Flexural strength of UT jute fiber

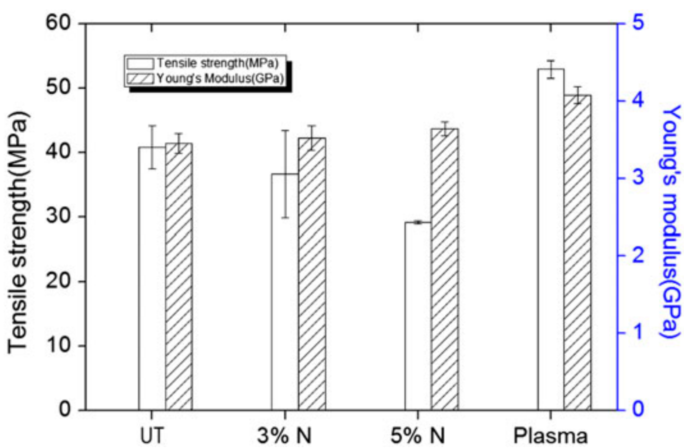

Figure 6. Tensile strength and tensile modulus of jute/PLA composite.
Note: UT: Untreated; N: NaOH.

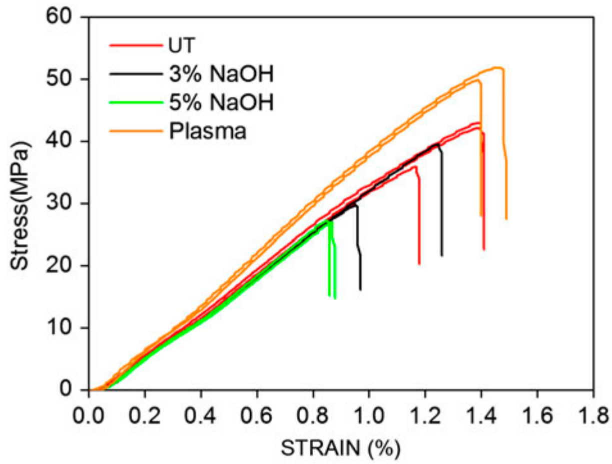

Figure 7. Stress–strain curve of tensile test.
Note: UT: Untreated.

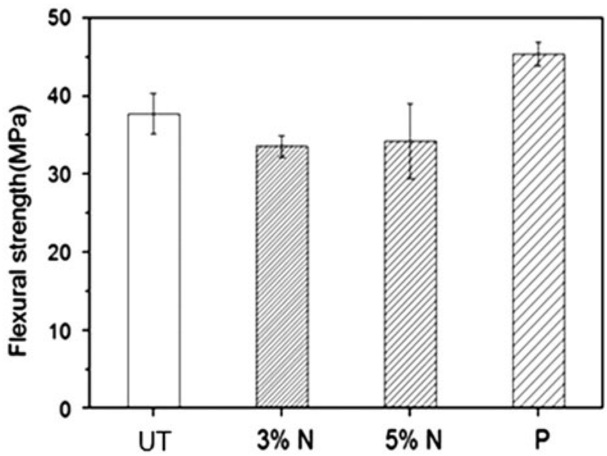

Figure 8. Flexural strength of jute/PLA composites.
Note: UT: Untreated; N: NaOH; P: Plasma.

reinforced composite is 37.7 MPa while for AT jute fiber reinforced composites the values are 33.5 and 34.2 MPa for 3 and 5% AT, respectively. Plasma polymerized jute fiber reinforced composite exhibited a flexural strength of 45.3 MPa, which is comparably higher than UT and AT fiber composites. The increase in flexural strength of PT fiber composites is due to etching away the surface of fiber by plasma treatment, leaving more nonpolar lignin on the fiber surface, which contributes to 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. [34].

4. Conclusions

The authors reported on the effect of plasma treatment of jute fiber on the mechanical, morphological, and IFSS 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 fiber for and fracture surface of the composites with PLA. The tensile, modulus, and flexural strength of the plasma treated jute fiber composites were 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 IFSS 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 compare to chemical methods.

Acknowledgments

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