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Effect of the Low and Radio Frequency Oxygen Plasma Treatment of Jute Fiber on Mechanical Properties of Jute Fiber/Polyester Composite

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Abstract: We investigated the surface modification of jute fiber by oxygen plasma treatments. Jute fibers were treated in different plasma reactors (radio frequency "RF" and low frequency "LF" plasma reactors) using O_2 for different plasma powers to increase the interface adhesion between jute fiber and polyester matrix. The influence of various plasma reactors on mechanical properties of jute fiber-reinforced polyester composites was reported. Tensile, flexure, short beam shear tests were used to determine the mechanical properties of the composites. The interlaminar shear strength increased from 11.5 MPa for the untreated jute fiber/polyester composite to 19.8 and 26.3 MPa for LF and RF oxygen plasma treated jute fiber/polyester composites, respectively. O_2 plasma treatment also improved the tensile and flexural strengths of jute fiber/polyester composites for both plasma systems. It is clear that O_2 plasma treatment of jute fibers by using RF plasma system instead of using LF plasma system brings about greater improvement on the mechanical properties of jute/polyester composites.

Keywords: Plasma treatment, Natural fiber, Polyester composite, Mechanical properties

Introduction

The interest in using natural fibers in composites has increased in recent years due to their lightweight, non-abrasive, combustible, non-toxic, low cost and biodegradable properties [1]. Particularly, natural fibers, such as flax, hemp, sisal, and jute, are interesting, environmentally friendly alternative to the use of glass fibers as reinforcement in polymer based engineering composites [2-4]. Composites manufactured using them find applications in diverse fields like automobile components, building materials, and furniture [5,6].

However, its hydrophilic character due to the high hydroxyl group content of cellulose is the main cause of poor compatibility between cellulose fibers and the polymers used as matrix, which leads to unsatisfactory mechanical properties for composites [7-9]. Namely, the low interfacial properties between fiber and polymer matrix often reduce their potential as reinforcing agents due to the hydrophilic character of natural fibers [10]. It is well known that the structure and properties of the interface play a very important role on the physical and mechanical properties of the composites due to the stress transfer between the matrix and the fibers [11]. Therefore, surface treatments are necessary to improve the interfacial interaction between fiber and polymer matrix. In the past, many attempts have been made to modify the surface properties of natural fibers to enhance adhesion with the matrix. Various chemical treatments such as alkali treatment [6,11], silane treatment [12], and acetylation [13] have been reported in relation to the compatibility in natural fiber composites. Although the chemical treatments of fiber surfaces have been somewhat successful in improving the interfacial bonding, there are problems related to the high cost of the treatment and the disposal of the chemicals after treatment [14,15]. In recent years, increasing concern about environmental pollution problems has limited wide industrial application of chemical surface treatments [15]. In contrast to chemical treatments, the cold plasma techniques are considered as dry and clean processes [13]. Plasma treatment improves fiber-matrix adhesion either by introducing polar or excited groups or even a new polymer layer that can form strong covalent bonds between the fiber and the matrix, or by roughening the surface of fibers to increase mechanical interlocking between the fiber and the matrix [16]. Also, cold plasma technique allows surface modification of fibers without affecting the bulk properties. In addition, no solvent is used, and the duration times of treatment are short [2,17].

In the present work, oxygen plasma treatments have been used to modify the fiber surfaces under suitable treatment parameters to improve the compatibility between jute fibers and polyester. Jute fibers were treated with different types of plasma generator (low frequency and radio frequency). The effects of oxygen plasma treatment of jute fibers on the mechanical properties of jute fiber and jute/polyester composites were investigated. Also, fractured surfaces of the composites were observed by scanning electron microscopy (SEM).

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Experimental Details

Materials

Orthophtalic unsaturated polyester resin (C92N8 from Camelyaf, Turkey) was used as the polymer matrix. Methyl ethyl ketone peroxide (MEKP) as the catalyst and cobalt naphthanate as the accelerator was used. The quantities of the accelerator and the catalyst were 0.8 phr and 0.85 phr of the resin, respectively. Woven jute fabric (an areal mass density of 300 g/m^2) was purchased from Anil Limited (Turkey). Oxygen gas (99.995 % purity) and argon gas (Ar) (99.995 % purity) were provided from Habas (Turkey). Sodium hydroxide (NaOH) from Merck Corp. was utilized for the alkali surface treatment of jute fibers, as described in Sarikanat (in press) [18].

Fiber Surface Treatments by Using LF and RF Plasma Systems

Before plasma treatment procedure, jute fabrics were alkalized as indicated by Sarikanat (in press). Jute fibers were subjected to low pressure O_2 plasma treatment at plasma powers of 30, 60, and 90 W for 15 min. RF plasma generator (operating at 13.56 MHz with a maximum power of 100 W) and LF plasma generator (operating at 40 kHz with a maximum power of 200 W) was used for surface treatments of the fibers. Both plasma equipments were PICO type, Diener Electronics GmbH+Co (Germany). Plasma treatments were generated in the fully closed and semi-automatic systems. The jute fibers were placed on to the ground electrode in the middle of the reactor and the chamber was evacuated to a low pressure of 0.1 mbar. Then, the oxygen gas was let to flow into the chamber at 0.3 mbar and the surfaces of the fibers were exposed to glow discharge. When the treatment was completed, the plasma generator was turned off automatically and the gas valve was closed manually. Then, Argon gas was fed into the chamber for 10 min to reduce free radicals in the atmosphere. Finally, the chamber was evacuated to a low pressure of 0.1 mbar and the vacuum was applied for 15 min.

Composite Preparation

Hand lay-up technique was used to fabricate composite laminates. The polyester resin, the accelerator and the catalyst were mixed and then the mixture was applied onto the jute fabric surface. The volume fractions in composites are approximately 27 %. The laminate was compressed at a pressure of 100 bar in the mold, and the pressure was continued to apply to the laminate at room temperature for 150 min. After that, the jute/polyester composites were cured at room temperature.

Fiber Tensile Test

Tensile strengths of jute fibers were determined using a Shimadzu AUTOGRAPH AG-SI Series universal testing machine at contact speed 0.1 mm/min. A minimum of seven strands for each jute type were mounted on cardboard end tabs via a quick-setting polyester adhesive. The samples were mounted such that each specimen had a gauge length of 20 mm.

Mechanical Properties of Composites

Interlaminar Shear Test

Interlaminar shear strength values (ILSS) of jute/polyester composites were determined by conducting short-beam shear tests according to ASTM D-2344. A Shimadzu AUTOGRAPH AG-IS Series universal testing machine was used by attaching a 5 kN load cell. System control and data analyses were performed using Trapezium software. A span-to-depth ratio of 5:1 was chosen for specimen dimensioning while a cross-head speed of 1.3 mm/min was preferred for testing. At least five specimens were tested to ensure repeatability of the results.

Three Point Bending Test

Three-point flexural test method was used to determine the flexural properties of the composites according to ASTM D 790 standard. While the used span-to-dept ratio is 16:1, the crosshead speed is 1.3 mm/min. At least five specimens were used for testing and results were given averagely.

Tensile Test

Tensile testing (ASTM D-3039) was carried out using a Shimadzu universal testing machine having 5 kN load cell and video extensometer system (AUTOGRAPH AG-IS Series, Shimadzu Corp., Japan). At least five specimens, which were 197 mm long and 25 mm wide, were tested for each type of composite plate to check for repeatability at a cross head speed of 2 mm/min.

Scanning Electron Microscopy (SEM)

After tensile tests, fractured surfaces of fabricated composite were examined by scanning electron microscopy (SEM) using a JEOL-JSM-6060 (Jeol Ltd., Tokyo, Japan). Before SEM investigation, samples were coated with a thin layer of metallic gold using an automatic sputter coater (Polaron SC 7620, Thermo VG Scientific, West Sussex, England) in order to reduce the extent of sample arcing.

Results and Discussion

Fiber Tensile Test

The effects of O_2 plasma treatment at different plasma powers on tensile strength of jute fibers for the LF and RF

Table 1. Tensile strength of untreated and O_2 plasma treated jute fibers (MPa)

		LF	RF
	Untreated	324.4±9.9	
Plasma power (W)	30	319.6±12.5	321.1±9.7
	60	311.7±10.6	317.2±8.8
	90	306.6±7.7	314.6±10.2

Table 2. Mechanical properties of jute fiber/polyester composites after O_2 plasma treatment of jute fibers

Plasma power/ Plasma system	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	ILSS (MPa)
Untreated	52.6±2.0	4.0±0.1	81.3±4.6	5.5±0.2	11.5±1.5
30 W/LF	61.9±3.5	4.1±0.1	93.4±6.5	5.7±0.2	15.3±1.4
60 W/LF	71.9±4.3	4.4±0.2	102.4±6.8	5.8±0.2	19.3±1.5
90 W/LF	73.7±6.3	4.5±0.2	110.0±7.7	5.8±0.2	19.8±1.7
30 W/RF	67.6±4.8	4.1±0.1	99.5±5.4	5.6±0.2	17.5±1.2
60 W/RF	77.5±5.8	4.4±0.1	110.3±4.5	5.7±0.2	21.6±1.5
90 W/RF	85.7±5.1	4.6±0.2	143.6±8.7	5.9±0.2	26.3±1.8

plasma systems are presented in Table 1. It can be seen that all of the tensile strengths of the oxygen plasma-treated jute fibers are slightly lower than that of alkali treated jute fiber. Plasma treatment may change the surface topography [16,19]. Micro pits and voids may be generated. The tensile strength of single fiber decreases because of more flaws and irregularities. Plasma treatment may lead damages on the fiber surface, resulting in decrease in fiber strength [19].

The Mechanical Properties of the Composites

The mechanical properties of untreated and O_2 plasma treated jute fiber reinforced polyester composites are given in Table 2. ILSS values of untreated jute/polyester and O_2 plasma treated jute/polyester composites are presented in Figure 1. It can be seen that composites reinforced with O_2 plasma treated jute fibers have higher ILSS values than that of the untreated one. In LF plasma systems, O_2 plasma treatments of jute fibers resulted in obvious increases for plasma powers of 30 and 60 W, while the ILSS increased only marginally at the plasma power of 90 W when compared to power of 60 W. ILSS values of O_2 plasma treated jute/polyester at plasma powers of 30, 60, and 90 W increased by about 33, 68, and 72 %, respectively, when compared to that of untreated jute/polyester composite, as given in Table 2. In RF plasma system, significant increases in ILSS values of the composites were recorded for all plasma powers (Figure 1). That is, fiber/matrix adhesion improved continuously with increasing of plasma power. When compared to that of untreated jute/polyester, ILSS values of O_2 plasma treated jute/polyester composites at plasma powers of 30, 60, and 90 W increased by approximately 52, 88, and 129 %, respectively (Table 2). Therefore, the value obtained at the plasma power of 90 W was more than twice than that of untreated one. These results indicate that O_2 plasma treatment of jute fiber improves jute fiber/polyester interfacial adhesion.

The effect of O_2 plasma treatment of jute fibers on flexural strength values of jute/polyester composites were shown in Figure 2. The flexural strength of untreated jute/polyester composite was determined to be 81.3 MPa. Considering LF plasma system, flexural strength values of O_2 plasma treated jute/polyester composites were obtained to be 93.4, 102.4, and 110.0 MPa, once the jute fibers were O_2 plasma treated at plasma powers of 30, 60, and 90 W, respectively. As a result of O_2 plasma treatment of jute fiber at 30, 60, and 90 W, the flexural strength values of jute/polyester increased by 15, 26, and 35 %, respectively. Based on RF plasma system, the flexural strengths of the composites after O_2

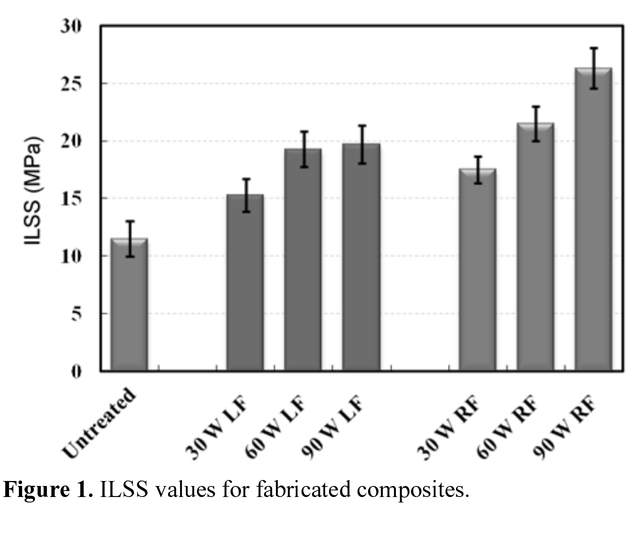

Figure 1. ILSS values for fabricated composites.

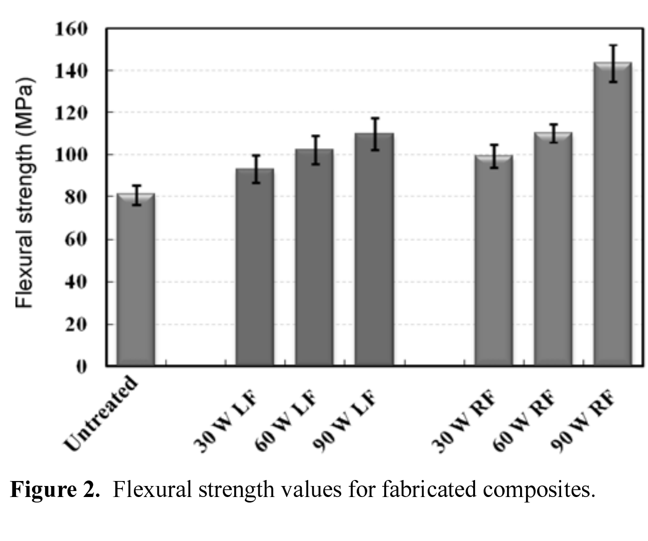

Figure 2. Flexural strength values for fabricated composites.

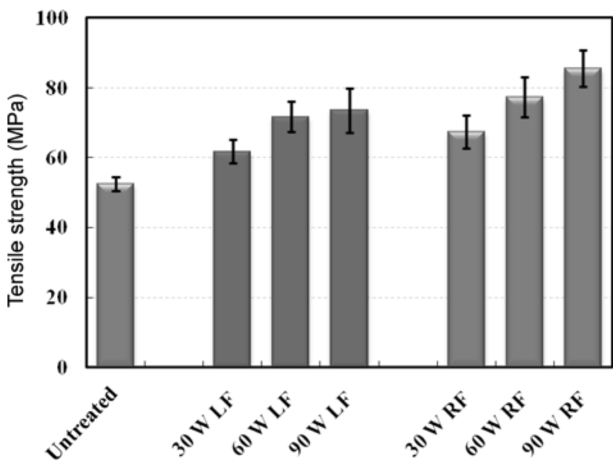

Figure 3. Tensile strength values for fabricated composites.

plasma treatment of jute fibers were obtained to be 99.5, 110.3 and 143.6 MPa at plasma powers of 30, 60, and 90 W, respectively. This means that using RF plasma system for O_2 plasma treatment of jute fibers causes 22, 36, and 62 % increases in terms of the flexural strengths of jute/polyester composites. The flexural modulus values were given in Table 2. The flexural modulus values seem to be compatible with flexural strength values. But, the increases in flexural modulus are not as high as in flexural strength. The greatest increase in flexural modulus was obtained to be 7 % as a result of treatment of jute fiber with RF plasma system at 90 W.

Tensile strength values of untreated and O_2 plasma treated jute fiber/polyester composites were given in Figure 3. It can be seen from this table that the O_2 plasma treatment improves the tensile strength of jute fiber/polyester composites for both plasma systems. The tensile strength and modulus values of untreated jute fiber/polyester composite were obtained to be 52.6 and 4023 MPa, respectively. In LF plasma system, the tensile strength of untreated jute fiber/polyester composite was enhanced by approximately 18, 37, and 40 % as a result of fiber surface treatment at plasma powers of 30, 60, and 90 W, respectively. Based on RF plasma system, the tensile strength of O_2 plasma treated jute fiber/polyester composites was increased by approximately 29 % at a plasma power of 30 W, 47 % at a plasma power of 60 W and 62 % at a plasma power of 90 W in comparison with that of one without treatment.

When the increasing trends of the ILSS and tensile strength values are evaluated, it is seen that in LF plasma system rate of increment decreases beyond the plasma power of 60 W. Contrary to this, in RF plasma system similar trend was not observed for these strength values. It is clear that O_2 plasma treatment of jute fibers by using of RF plasma system brings about greater improvement on the mechanical properties of jute/polyester composites when compared to use of LF plasma system.

O_2 plasma treatment of the jute fibers causes improvement of interfacial adhesion between the fiber surface and polyester. It is known that cellulose and hemicellulose are more reactive to plasma; they are more easily etched away by plasma treatments, leaving more non-polar lignin on the fiber surface, which may contribute to the improvement of interfacial adhesion [16]. Although the fiber strength decreases slightly, as shown in Table 1, the tensile and flexural properties increases with O_2 plasma treatment of jute fibers due to improved fiber/matrix adhesion. In other words, increase in adhesion between fiber and matrix as a result of O_2 plasma treatment of jute fibers is dominant factor rather than fiber strength determining the tensile and flexural properties of composites in this study. The reason why adhesion between fiber and matrix increase may be attributed to increased surface roughness. Because, Yuan et al. reported that the increased surface roughness in natural fibers after plasma treatment leads to higher interfacial contact, possibly better mechanical interlocking and better viscoelastic dissipation at the interface [16].

As was shown in Figures 2, 3, and 4, O_2 plasma treated jute fiber reinforced polyester composites demonstrate superior mechanical properties providing that jute fibers are treated with RF plasma system instead of LF plasma system.

Fractographic Investigation

SEM analysis was conducted on the fracture surfaces of the tensile tested jute/polyester composites to evaluate the fracture morphology depending on the fiber surface modification. Figure 4 shows the fracture surface of untreated jute, LF and RF O_2 plasma treated jute/polyester composites.

SEM images demonstrated clean fiber surfaces for the untreated jute fiber reinforced polyester composite. This can be considered as the evidence of an adhesive failure, indicating a poor fiber/matrix interfacial adhesion.

For the LF O_2 plasma treated jute/polyester composites, remains of polymer were observed on fiber surfaces as shown in Figure 4. O_2 plasma treatment of jute fibers at 30 W causes deposition of very little amount of polymer particles on fiber surface. After O_2 plasma treatment at 60 W, fiber surfaces were deposited with more polyester, which partially at some regions resembled layers. Similarly, SEM images of LF oxygen plasma treated jute fibers at 90 W showed that they were almost covered with layers of polymer. This leads to an expectation for better fiber/matrix adhesion.

RF oxygen plasma treatment of jute fibers also seemed to enhance their compatibility with polyester matrix as can be seen in Figure 4. Polymer deposits were observed on jute fibers after plasma treatment at plasma power of 30 W. SEM image shows more polymer amount on jute fibers which were treated at 60 W. Plasma treatment of jute fibers at 90 W exhibited improved fiber/matrix interaction. Again, jute fiber surfaces were deposited with more polyester, which almost

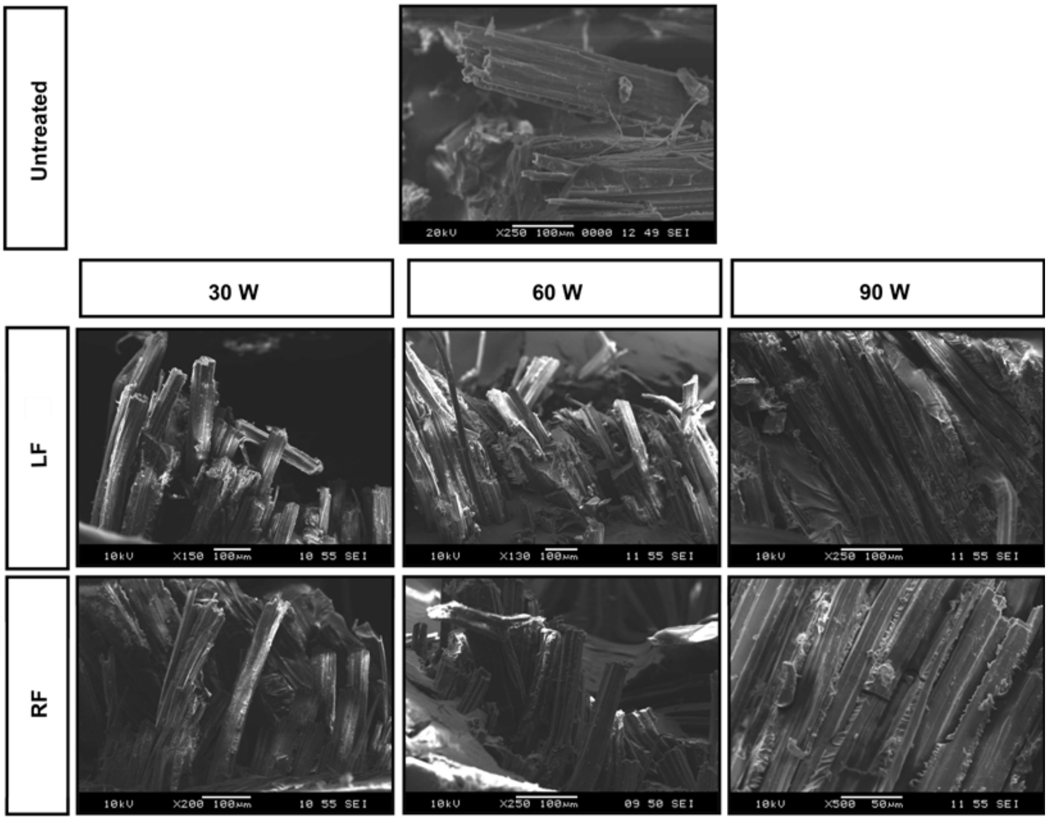

Figure 4. Fracture surfaces of the RF and RF oxygen plasma treated jute/polyester composites.

formed layers on fibers Therefore; it is evident by the SEM observations performed on the fractured tensile test specimens that there is a positive effect of surface modification of the fibers by O_2 plasma treatment on interfacial adhesion between jute fibers and polyester matrix.

Conclusion

The effects of low temperature O_2 plasma treatment using different plasma systems on interfacial adhesion and mechanical properties of jute fiber/polyester composites were investigated. The mechanical properties of the composites improved with increasing of plasma power for both radio and low frequency systems. The interlaminar shear strength was enhanced by 72 and 129 % at 90 W for LF and RF plasma systems, respectively, in comparison with that of the untreated sample. When compared to the O_2 plasma treatment in the LF plasma system, the surface treatment in the RF plasma system is more effective in improving the interfacial adhesion between the jute fiber and polyester. Fiber tensile strengths after O_2 plasma treatments are slightly lower than that of alkali treated jute fiber. Although the fiber strength decreased slightly, the tensile and flexural properties increased with O_2 plasma treatment of jute fibers because of the fact that the fiber/matrix adhesion improved. Improved fiber/matrix adhesion is able to be seen in SEM micrographs of fracture surfaces of the jute fiber/polyester composite.

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