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Analysis of the Effects of Atmospheric Helium Plasma Treatment on the Surface Structure of Jute Fibres and Resulting Composite Properties

Abdullah A. Kafi [a,*], **Christopher J. Hurren** [a], **Mickey G. Huson** [b] **and Bronwyn L. Fox** [a]

[a] Centre for Material and Fibre Innovation, Institute of Technology and Research Innovation, Deakin University, Geelong, VIC 3217, Australia

[b] CSIRO Material Science and Engineering, P. O. Box 21, Belmont, VIC 3216, Australia

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Abstract

This work investigates the mechanisms involved in the improvement of flexural properties of a jute/polyester composite when the reinforcement material has been atmospherically plasma treated using helium gas. All composites were laid-up by hand and cured using a Quickstep™ cure cycle. Surface characterization techniques including scanning probe microscopy (SPM), and surface wettability combined with fabric tensile strength, composite flexural strength and composite Mode-I properties have been used to quantify the effects of plasma modification. Flexural strength and modulus increased with plasma treatment time, reaching a maximum at 25 passes before decreasing. SPM topographical analysis showed that roughness of the fibre decreased as the plasma treatment time increased until 25 passes after which the roughness was found to increase again. The coefficient of friction increased rapidly after only a short plasma treatment time (5 passes) whilst wettability continued to increase until 25 passes after which it remained constant. The fabric tensile strength followed the same trend as the flexural properties of the composites. Decreasing fibre surface roughness is postulated as a reason for decreasing Mode-I interlaminar fracture toughness properties of the composites.

Keywords

Atmospheric helium plasma, flexural strength, jute, coefficient of friction, SPM

1. Introduction

Over the past decade, the stringent environmental legislations and fluctuating oil prices have motivated composite manufacturers to strongly consider renewable fi-

* To whom correspondence should be addressed. Tel.: +61 3 5227 1015; Fax: +61 3 5227 1103; e-mail: aakaf@deakin.edu.au

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bres as a reinforcement material. Many research articles have been published on the use of various natural fibres including jute, sisal, flax and hemp, mostly derived from the bast or stem of the plants. Among these, jute, the lignocellulosic bast fibre, appears to be an option for composite applications because of its low specific gravity (1–1.45), high specific modulus (19 GPa) and low cost [1].

Despite the relatively low cost, low specific gravity and high modulus of jute, there are some drawbacks. These include poor fibre–matrix adhesion, dimensional instability, and poor resistance to moisture. To overcome the present obstacles, various chemical [2] and physical [3–5] treatments or combinations of both [6] have been investigated to modify the jute fibre surface. The use of chemical treatments is effective for fibre property enhancement, however, the effluent produced can cause pollution problems as well as additional cost to the final product. Different physical surface modification techniques, such as UV irradiation [3], corona [4], low pressure plasma [7] and atmospheric plasma [5] have been available for many years as an alternative to chemical treatment.

Atmospheric plasma, a potential environmentally friendly technique, has been used to uniformly modify the fibre surface to a depth of 100 nm without affecting the bulk properties [7]. This technique offers an alternative to wet chemical treatments with consequent benefits to the environment. It is continuous in operation and does not require any vacuum systems; therefore, it saves time, space and energy over low pressure plasma [5]. Under an atmospheric plasma treatment two possible effects can take place, an etching process that removes the weakly bonded outermost pectin layer and the introduction of new functional groups [8]. The nature of these effects mostly depends on the feed gas composition and treatment time of the plasma process. The ultimate goal of exploiting atmospheric plasma is to improve the wettability of the natural fibre surface by removing non-cellulosic substances from the outermost surface which will play a major role in improving the composite behaviour.

Surface analysis of natural fibres is difficult to carry out as their surfaces may be hygroscopic, rough and chemically heterogeneous in nature. Scanning probe microscopy (SPM) is an effective technique for the quantification of a natural fibre surface with subnanometer resolution. Its major advantage over SEM is that it does not require a vacuum or conductive coating of the sample before imaging [9]. Additional information can be obtained from SPM experiments including surface roughness [10], friction [11] and adhesion force measurements [9, 12].

Natural fibre based composites have been manufactured by several methods, including hand lay-up [3], resin transfer molding (RTM) [13], compression moulding [13] and vacuum infusion [14]. A new approach to cure natural fibre composites called Quickstep™ process has been used to manufacture aerospace-grade composites for many years. Quickstep™ ensures better heat capacity to the panel because of its use of liquid instead of air or solid and also requires shorter cure cycle times in panel manufacturing [15]. Shorter cure cycle times, better heat control and improvement in resin dispersion over compression moulding enable this curing process to be suitable for natural fibre based composites manufacture.

Many studies have been devoted to investigate the Mode-I delamination behavior of high performance composites, as it is one of the most prevalent failure modes in laminate composites. The effect of the Quickstep™ process [16] and fibre surface treatment [17] on the fracture mechanics behavior of synthetic fibre based composites has been investigated. However, only limited literature exists on the study of fracture mechanics behavior of natural fibre based composites [18]. Detailed studies are required to understand the delamination behavior of composites when reinforcement material is subjected to either chemical or physical treatment.

The aim of this study is to develop a deep understanding of both physical and chemical surface characteristics of the jute fibre before and after helium plasma treatment. Various experimental parameters that affect the flexural and Mode-I interlaminar fracture toughness properties of composites were assessed and optimized. The ultimate goal is to correlate the surface characteristics of jute fibre with flexural and Mode-I interlaminar fracture toughness properties of composites.

2. Experimental

2.1. Materials

Unbleached woven jute fabrics (320 g/m^2) were obtained from Bangladesh Jute Mills Corporation, Dhaka. The yarns had an average fineness of 270 ($\pm$18) tex in the warp direction and 242 ($\pm$19) tex in the weft direction where the unit of tex is g/1000 m. The yarn twist was 191 tpm (turns per meter) in the warp direction and 167 tpm in the weft direction. Yarn twist was measured according to ASTM D1422-92 and yarn linear density was tested as per ISO 2060:1994(E).

The matrix polymer was a commercial unsaturated polyester resin ESCON 62-333 procured from Fiberglass International, Australia. 1.5% cobalt napthenate solution as promoter and 36% styrene as a cross-linker were premixed with ESCON. The curing agent was methyl ethyl ketone peroxide (MEKP) and the mould release agent poly(vinyl alcohol) (PVA) was supplied by the same company.

2.2. Plasma Treatment

A Sigma International APC 2000 atmospheric pressure glow discharge plasma machine (Sigma Technologies International, USA) was used as shown in Fig. 1. This device contains two aluminium electrodes mounted above a ceramic-coated roller. The fabric was attached to the roller and rotated past the plasma source. Treatment was completed for 5, 25, 35, 50 and 100 revolutions/passes of the treatment roller where each pass took 0.4 s. The power input was 970 W, and helium (14 l/min) was used as the carrier gas.

2.3. SPM Analysis

Topographic images of both untreated and plasma treated single fibre surfaces were collected with a Digital Instruments Dimension 3000 SPM. It was operated in con-

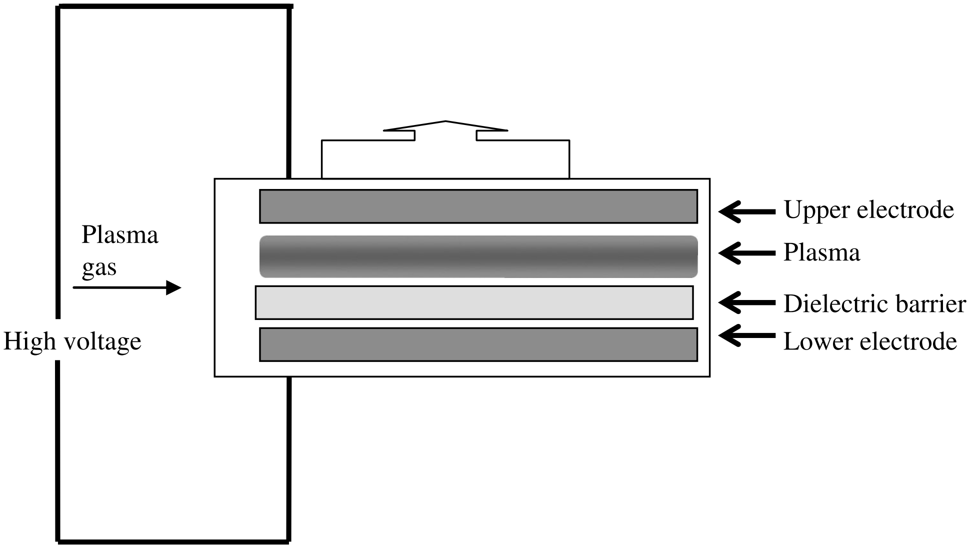

Figure 1. Schematic representation of atmospheric plasma.

tact mode using silicon nitride probes consisting of pyramidal tips on cantilevers with a low spring constant (0.12 N/m). At least three single fibres were pulled out from the yarn and fixed on a glass slide with a double sided tape. The images were acquired on a minimum of five different spots (5×5 µm) on each fibre to obtain a representative fibre surface topography. Measurements were carried out in air under ambient conditions. In contact mode, SPM operates by scanning a tip attached to the end of a cantilever across the sample surface. Cantilever deflection is monitored by a laser beam which reflects on a split photodiode detector, and the tip contacts the surface through the adsorbed fluid layer on the sample surface. A feedback loop maintains a constant deflection of the cantilever by vertically moving the scanner at each (x, y) data point to maintain a 'setpoint' deflection. Images were flattened (third order) and surface roughness was determined using Nanoscope (V5.31) software over the entire height image. The surface roughness was quantified in terms of two parameters *viz.* root-mean square roughness (R_{rms}) and average roughness (R_a). The first parameter represents the standard deviation of the Z values within a given area. The second parameter represents the arithmetic average of the deviations from the centre plane. Both R_{rms} and R_a were calculated using Nanoscope software following the equations given in the manual.

2.4. *Surface Wettability*

Six equal sized discs were cut from the jute fabric using a hole puncher and conditioned at 20 ± 2°C and 65 ± 2% relative humidity for 8 h prior to the test. Each sample disc was dropped into a 500 ml glass beaker half filled with distilled water and the time required for water to penetrate completely into the fabric disc was recorded for each sample.

2.5. Coefficient of Friction

In order to determine the coefficient of friction of fabric, a Lloyd LR30K universal tensile tester was fitted with a friction assembly following ASTM D1894 standard [19]. Prior to testing, jute fabric was stretched or pre-tensioned to obtain a flat surface. A Perspex block (721 g) with a contact area of 100 mm $\times$ 63 mm was pulled over the fabric at a constant speed of 1 mm/min by an inextensible thread attached to the load cell of the tensile tester. The static coefficient of friction was calculated from the ratio of maximum static frictional force and the weight of the Perspex block [20]. Five tests were carried out for each sample measured.

2.6. Fabric Tensile Strength

Fabric tensile strength tests were conducted following the Australian standard 2001.2.3.1 at a crosshead speed of 20 mm/min with a Lloyd LR30K universal tensile tester using the strip method. A preload of 5 N and a gauge length of 200 mm were used. Samples were conditioned at 20 ± 2°C and 65 ± 2% relative humidity for 24 h prior to the tests and cut into 300 mm long and 100 mm wide test specimens. An equal number of threads were removed from each side until the total number of threads in the middle was 24. Maximum force and elongation at maximum force were recorded for each specimen.

2.7. Composite Fabrication

Jute fabrics were conditioned for 24 hours at 20 ± 2°C and 65 ± 2% relative humidity before starting the wet lay-up process. Fabric samples of 200 mm^2 were cut out and a 3:1 resin to jute ratio, i.e., 33% fibre volume fraction, was maintained for each case. Twelve fabric plies were placed at a 0/90 orientation and vacuum bagged at −85 kPa. The mould surface was coated with a layer of PVA release agent at least 20 min prior to inserting a release film. It was cured in a clamshell style mould with flexible bladders *via* a Quickstep QS5 (Quickstep Technology Pty Ltd., Western Australia) process plant with a 30 min dwell time at 95°C. Two thermocouples were inserted, one on each side of the laminates, before vacuum bagging (Fig. 2) to record resin temperature and to control the cure cycle. Resultant composites were 6 mm thick and were stored in a fume hood overnight to remove uncured styrene.

2.8. Flexural Tests

Flexural strength (FS) and flexural modulus (FM) were determined using the 3-point bending method as per ASTM D790-84a at a crosshead speed of 2 mm/min and 16:1 span to thickness ratio using a Lloyd LR30K universal tensile tester. All tests were conducted at 20 ± 2°C and 65 ± 2% relative humidity allowing sample conditioning for 24 h prior to the tests. At least six specimens were tested for each type.

2.9. Mode-I Double Cantilever Beam (DCB) Tests

Mode-I DCB tests were performed in accordance with the protocol of the European Structural Integrity Society using a Lloyd LR30K universal testing machine

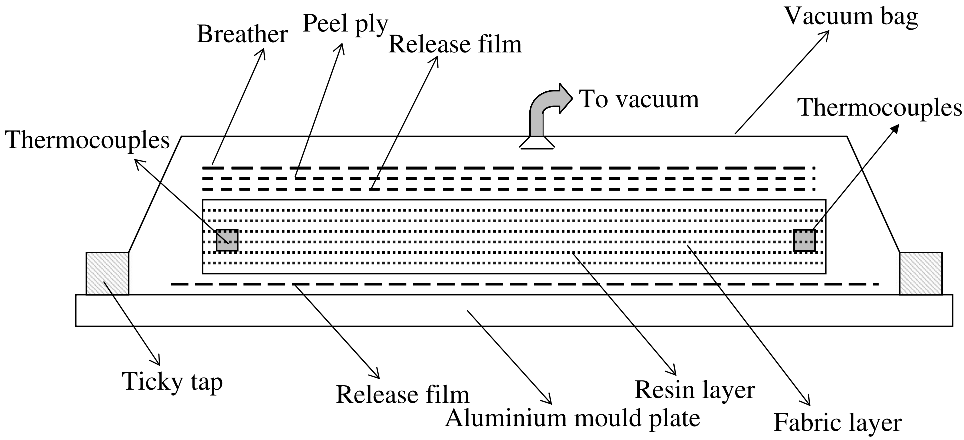

Figure 2. Schematic representation of vacuum bagging.

fitted with a 1 kN load cell [21]. A starter film of 65 mm width was inserted during the wet lay-up process to facilitate crack initiation during testing. DCB specimens were cut to an average size of 20 mm in width and 172 mm in length. The cut edges of each specimen were polished before one side of the specimen was coated with white correction fluid to aid observation of the propagating crack. Two rectangular shaped aluminum tabs were attached to the opposite sides of the specimen at the end containing the crack using an instant adhesive. All specimens were conditioned at 20 ± 2°C and 65 ± 2% relative humidity for 24 h prior to testing. The Mode-I critical energy release rate G_{Ic} was calculated from the corrected beam theory (CBT) [21]. G_{Ic}-initiation values were calculated when the pre-crack was visually observed to start. Values of G_{Ic}-propagation were calculated from the maximum or plateau of the delamination resistance curve, commonly known as the R-curve. Five specimens were tested from both untreated and 25 passes of plasma treated samples.

3. Results and Discussion

3.1. Surface Topography and Roughness

Figure 3(a–e) shows the flattened 3-D height images at three different spots on the untreated and treated jute fibres. It is observed that untreated jute fibre has a rough surface topography typical of the rough primary cell wall [9] (Fig. 3(a)). The surface roughness is reduced slightly after 5 passes (Fig. 3(b)) and continues to reduce until 25 passes (Fig. 3(c)). At 25 passes the surface has lost most surface roughness and is relatively smooth. Furthermore, for 50 and 100 passes destructive etching of the fibre is clearly visible (Fig. 3(d) and 3(e)). These observations are confirmed by measuring average roughness (R_{a}) and root-mean square (R_{rms}) roughness which were found to decrease from 51 to 15 nm (R_{rms}) and 37 to 12 nm (R_{a}) for 25 passes of plasma treated jute fibre before increasing again for 50 and 100 passes (Fig. 4(a)). Initially the plasma produces a surface cleaning effect, removing surface

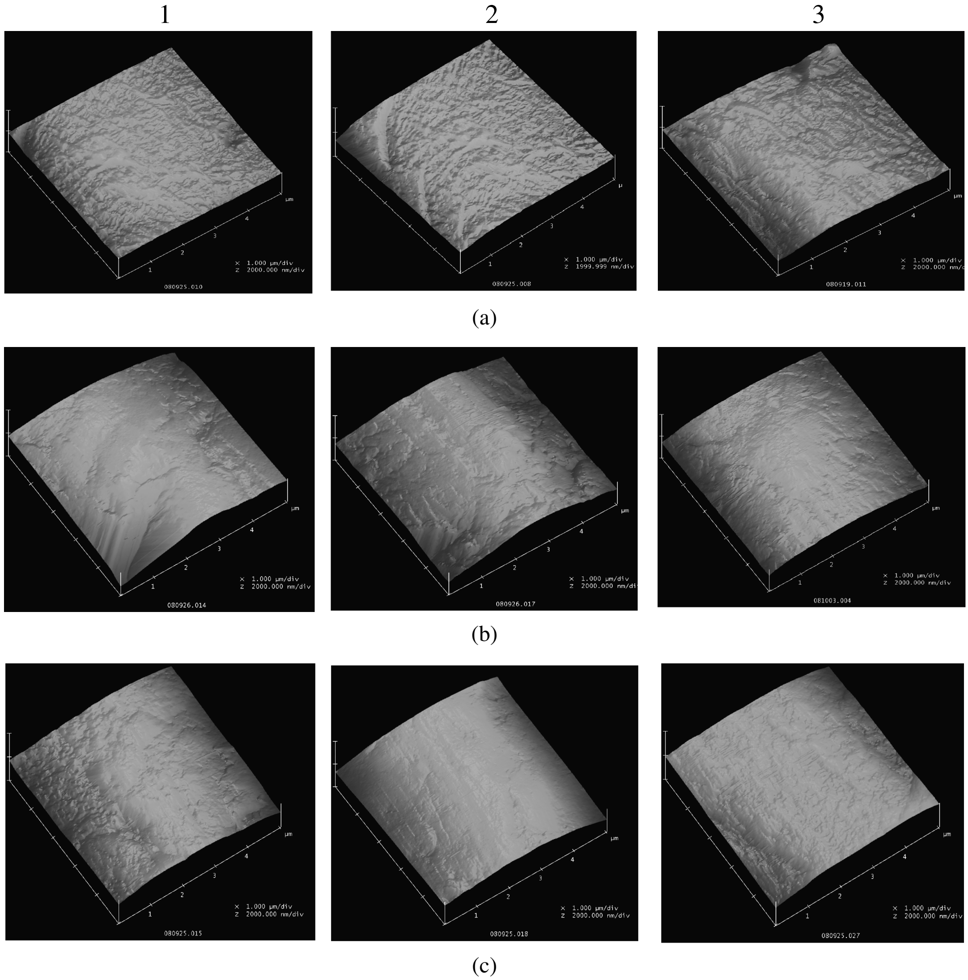

Figure 3. Typical SPM contact mode height images of plasma treated jute fibres after third-order flattening. In rows from top: (a) untreated, (b) 5 passes, (c) 25 passes, (d) 50 passes, (e) 100 passes. Numbers 1, 2 and 3 on top of height images show three representative spots.

contaminants and reducing surface roughness [22]. After prolonged exposure the plasma removes the outer layer [8] of the surface followed by destruction of micro or macrofibriller structure [23]. This affects the structural integrity of the jute fabric by introducing fibre breaking, fibre waviness and fabric distortion.

3.2. Surface Wettability

The effect of plasma treatment time on surface wettability is shown in Fig. 4(b). It is observed that wettability time decreases exponentially with an increase in plasma exposure until 25 passes. After 25 passes wettability was found to stabilize with only little further change. Plasma etching removed the weakly bonded hydrophobic layer from the fibre surface, thus increasing wettability as evidenced earlier [8]. This

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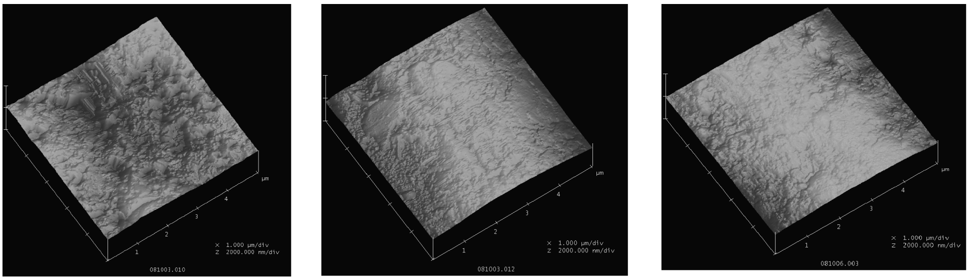
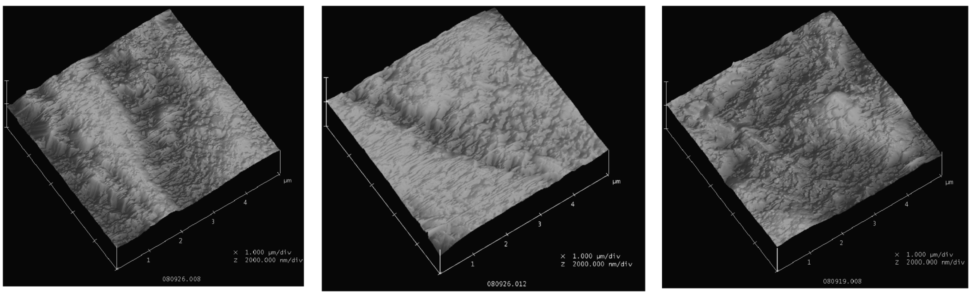


(d)



(e)

Figure 3. (Continued.)

would suggest that after 25 passes of plasma treatment outer hydrophobic layer was completely removed exposing the cellulose structure. Further treatment has only negligible effects on wettability as the cellulose structure simply is etched away leading to an increase in roughness as shown in Fig. 4(a).

3.3. Surface Coefficient of Friction

The effect of plasma treatment on the surface coefficient of friction (COF) is shown in Fig. 4(c). The surface COF increased about 15% after first 5 passes of plasma treatment. At this stage roughness has hardly changed (Fig. 4(a)) whilst wettability time has decreased by about 40% (Fig. 4(b)) suggesting that it is a chemical modification of the surface rather than a physical change that is responsible for the increased friction. Further plasma treatment up to 50 passes does not result in any additional increase in friction, however after 100 passes the friction goes up again, possibly due to the increased roughness shown in Fig. 4(a).

3.4. Fabric Tensile Properties

Plasma treatment resulted in an initial increase in the tensile force required to break the fabric, the force increasing up to 25 passes before dropping off (Fig. 4(d)). This initial increase is most likely due to the increase in fibre to fibre friction shown in Fig. 4(c). Increased fibre-to-fibre friction improves yarn tensile strength [24] and hence fabric strength [26] as it is harder for fibres to pull past each other in a staple

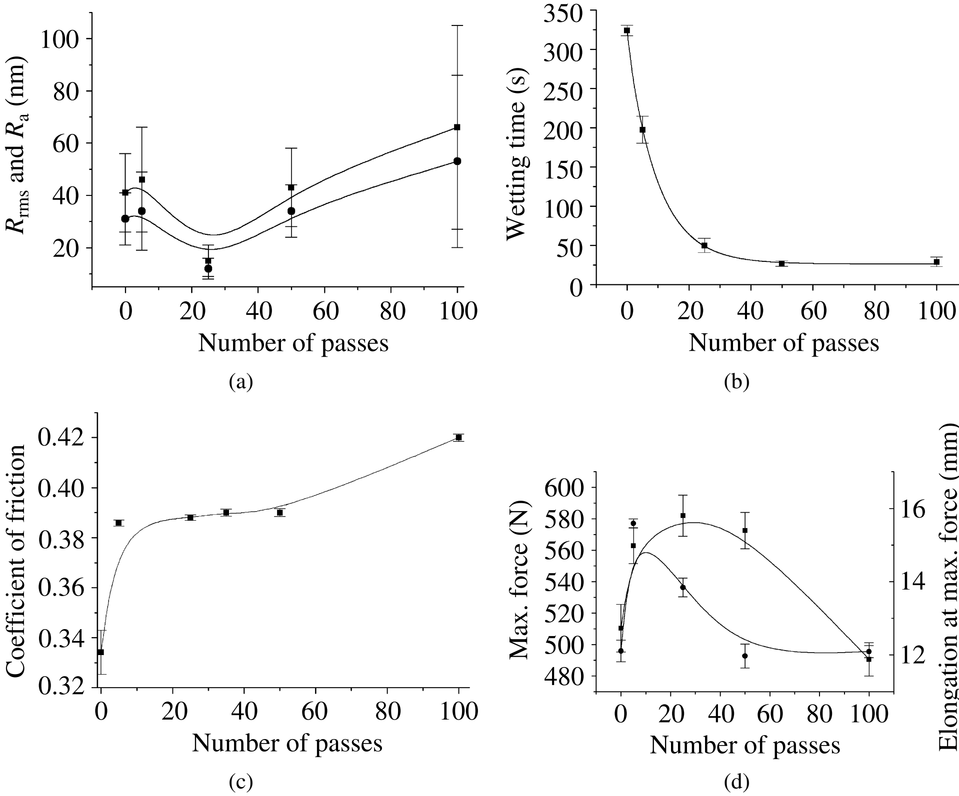


Figure 4. Effect of plasma treatment time (represented in terms of number of passes) on (a) fibre surface roughness, R_{rms} (■) and R_a (●), (b) fabric wetting time (■), (c) fabric coefficient of friction (■) and (d) fabric tensile properties, maximum breaking force (■) and elongation at maximum force (●).

spun yarn during fabric strain. Prolonged treatment by plasma causes fibre damage related strength losses in the fabric which outweigh the friction related improvements after 25 passes. The elongation at maximum force showed an initial increase for 5 passes before decreasing with further treatment.

3.5. Flexural Properties of Composites

The effects of plasma treatment time on the flexural strength (FS) and flexural modulus (FM) of the jute/polyester composites are shown in Fig. 5(a). A 45% increase in FS and 141% increase in FM were seen in composites manufactured with 25 passes of plasma treated fabric. The FS and FM followed similar trends to fabric strength (Fig. 4(d)). The majority of the improvements in FS and FM can be directly related to the strength improvement in the fabric reinforcement material. Based on optimum flexural strength and fabric strength, 25 passes was selected as an optimum plasma treatment time for DCB testing.

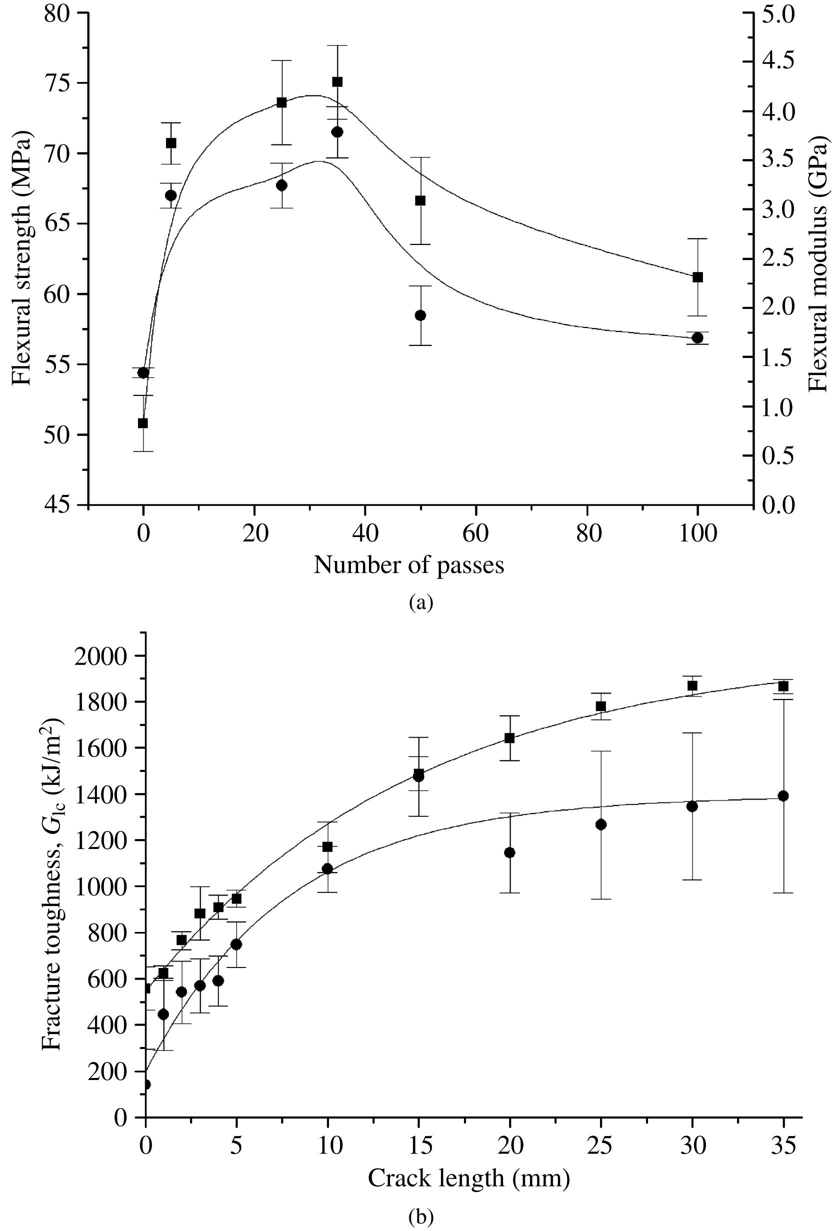


Figure 5. (a) Effect of plasma treatment time (represented in terms of number of passes) on flexural strength (■) and flexural modulus (●) of jute based composites. (b) Delamination resistance curve of untreated (■) and 25 pass plasma treated (●) jute based composites.

3.6. Mode-I Properties of Composites

Plasma treatment (25 passes) of the reinforcing jute fabric resulted in a decrease in the fracture toughness (in terms of both G_{Ic}-initiation and G_{Ic}-propagation) of the composite material cured using the Quickstep™ process, as shown in the delamination resistance curves (Fig. 5(b)). This may have occurred as a result of the observed decrease in the fibre roughness after 25 passes (Fig. 3(c)) of plasma treatment. It was demonstrated by Baley *et al.* [25] that the observed improvement in fracture toughness of chemically treated flax fibres in polyester matrix composites was the result of increased fibre surface roughness. Conversely, a smooth fibre surface may result in poor mechanical interlocking between the fibre and matrix, which could potentially explain the decrease in G_{Ic}-propagation value. It is also noted that the standard deviations of the G_{Ic}-initiation and G_{Ic}-propagation values for the plasma treated samples are higher than that of the untreated sample. This is mostly likely due to the difficulty in obtaining a consistent plasma treatment on the inhomogeneous and inherently rough natural fibre surface. The overall outcomes of the present study are still promising as our materials achieved significant improvement in flexural properties even though the fracture toughness properties were decreased.

4. Conclusion

The plasma treatment of jute fabric improves the flexural properties of jute/polyester composites. Flexural properties were improved due to a plasma induced increase in the strength of the fabric reinforcement material. Fabric strength improvements can be directly attributed to an increase in the fibre-to-fibre friction. Optimum properties are achieved after 25 passes as further plasma treatment results in strength deteriorating fibre damage. On the other hand, 25 passes of plasma treatment resulted in a decrease in Mode-I interlaminar fracture toughness properties of composites due to a decrease in surface roughness.

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