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Effect of lac treatment on mechanical properties of jute fabric /polyester resin based biocomposite

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An attempt has been made to dissolve lac in methanol / sodium hydroxide solution and to use this lac solution as a coupling agent for jute fabric. Lac treated jute fabric has been used to reinforce the unsaturated polyester resin (USP). Flexural strength and inter-laminar shear strength (ILSS) of lac modified jute/USP biocomposite have been evaluated and then compared with sodium hydroxide treated jute as well as untreated jute based biocomposites. Lac treated jute fabric shows higher flexural properties of the biocomposite than that of untreated jute fabric, which infers that lac acts as a good compatibliser between jute fibre and USP. Lac treatment on jute fabric enhances the flexural properties of biocomposite better in alkaline medium than in solvent medium. It is concluded that lac treatment can be used to improve the flexural and ILSS properties of jute / thermoset resin based biocomposite.

Keywords: Biocomposite, Compression, Flexural strength, Inter-laminar shear strength, Jute fabric, Lac treatment, Polyester resin

1 Introduction

Jute is a natural lignocellulosic fibre having relatively high modulus, strength and low density. It is inexpensive natural fibre and available in different forms like woven and nonwoven fabrics, yarns, webs and slivers. These factors make jute textiles an attractive candidate for the reinforcement of polymer resin in the development of biocomposites. Studies on jute-unsaturated polyester resin based biocomposite has attracted various research groups[1,2]. Jute possesses high hydrophilicity due to the presence of hydroxyl groups and researchers are mainly focused on improving the compatibility between jute fibre and hydrophobic polymeric resin. Chemical modification has been preferred to improve the interfacial adhesion between jute fibre and resin. Sodium hydroxide treatment, graft copolymerization, coupling agent treatment, etherification and maleation are some of the important chemical modifications used for jute and allied fibres[3, 4]. Among them, coupling agents have been used to improve the compatibility of jute fibre and the resin[5]. Coupling agent treatment does not reduce the strength of jute fabric. However, literature information on application of natural compound for chemical modification of jute fibre is still scanty.

Lac is a scarlet resinous secretion of a tiny insect called *Laccifer lacca*. It is composed of natural lac resin (70-80%); sugars, proteins, and soluble salts (2-4%); colouring matter (1-2%); and wax (4-6%). Lac resin is acidic in nature due to the presence of aleuritic acid as major component. It has good adhesive strength, insulation property, water proof and UV protection property. On the other hand, it has brittleness and low softening point[6]. Since lac has low molecular weight acidic compound, it can be utilized for improvement in compatibility between jute fibre and resin. An attempt has been made to treat the jute fabric with lac both in alkaline and solvent medium and to study its effect on flexural and inter-laminar shear strength properties of jute/polyester resin based biocomposites.

2 Materials and Methods

2.1 Fabric

Plain woven jute fabric having the specifications 61 ends/dm (252 tex), 54 picks / dm (230 tex), 270 gm^{-2} area density and 0.93mm thickness was procured from the local market of Kolkata and used for this study.

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2.2 Chemicals

Lac was supplied by Indian Institute of Natural Resins and Gums, Namkum, Ranchi and used as such without any purification. Unsaturated polyester resin (USP) of commercial grade (FB-554) with density of 1.2 g/cc was procured from the local market of Kolkata. All other chemicals used elsewhere were of AR grade.

2.3 Treatments of Jute Fabric

2.3.1 Sodium Hydroxide Treatment on Jute Fabric

Jute fabric was treated with 1% NaOH (owf) in the presence of 0.1% non-ionic detergent at 1:10 material-to-liquor ratio (MLR) for 60 min at 25°C. After treatment, jute fabric was neutralized with acetic acid solution, washed with water, rinsed and finally dried.

2.3.2 Lac Treatment on Jute Fabric

Lac was separately dissolved in 1% NaOH solution / methanol and made into 2, 3 & 4% (w/v) stock solution and filtered. Jute fabric was treated with the above-mentioned lac solution separately with the material-to-liquor ratio of 1:10 for 60 min at 30°C. After treatment, jute fabric was neutralized with acetic acid solution, washed with water, rinsed and then dried.

2.4 Preparation of Biocomposite

For hand laying method, three layers of jute fabric (30 cm × 30 cm dimension) was taken for each biocomposite sample, dried in a hot air oven at 70°C for 1 h and weighed. The USP resin was taken in the ratio of 1:3 with respect to jute fabric and thoroughly mixed with 2% (over the weight of resin) of cobalt naphthenate as accelerator and 2% methyl ethyl ketone peroxide as catalyst. The mixed resin was applied on the individual jute fabric by brush dabbing followed by rolling technique. For compression moulding, resin coated jute fabrics were superimposed in the alternative direction (warp + weft + warp) and consolidated in a hydraulic press of M/s PEECO Hydraulic Pvt. Ltd., Howrah (capacity 30 tonnes) under a pressure of 1.2 $kg.cm^{-2}$ at room temperature for 30 min. After compression, the biocomposite sample was taken out and the border of the sample was trimmed and kept at room temperature for 3 days followed by curing in an oven at 80°C for 4 h. Biocomposites samples were conditioned for 24 h at 65±5% RH before evaluation of mechanical properties.

2.5 Immersion Test

Lac treated and untreated jute fabrics of 25mm × 25mm dimension were placed on top of the resin in a 250 mL beaker and the time required (s) for fabric to immerse in the resin was observed.

2.6 Dry and wet Sample Preparation for Testing

The biocomposite samples were dried at 100±5°C for 60 min in hot air oven , then kept in a desiccator for 60 min and conditioned for 24 h at 65±5% RH.

The biocomposite samples were immersed in a water bath at room temperature for 7 days. After seven days, the samples were taken out from the water bath and wiped with filter paper to remove surface water and conditioned for 24 h at 65±5% RH.

2.7 Evaluation of Mechanical Properties

2.7.1 Flexural Strength

The flexural strength and flexural modulus of the biocomposites in dry and wet conditions were determined using a three-point bending test method as per ASTM D790 and ASTM D7264 test methods. The test was conducted using a load cell of 30kN at 5 mm/min rate of loading on Instron 5967.

2.7.2 Inter-laminar Shear Strength

Inter-laminar shear strength (ILSS)testing was carried out as per ASTM D 2344 standard on Instron 5967 of capacity 30 KN. Five tests were done for each biocomposite sample and mean value of inter-laminar shear strength was calculated [7]

3 Results and Discussion

3.1 Immersion Time of Jute Fabric

The objective of this test is to evaluate the rate of wetting and spreading of hydrophobic polyester resin on untreated and treated jute fabrics.

Table 1 infers that there is no extensive variation in wetting behaviour of jute fabrics by different treatments. Sodium hydroxide treatment is used to

Table 1—Immersion time (s) in USP for jute fabric

Fabric type	Mean ± SE
Untreated	10±2
NaOH treated	7±1
Alkaline lac treated	
2%	8±2
3%	8±1
4%	7±1
Solvent lac treated	
2%	7±2
3%	6±1
4%	6±1

remove the hemicellulose portion that leads to decrease the hydrophilic nature of the jute fibre [8]. During lac treatment with sodium hydroxide (LIS) and methanol medium (LIM), lac resin may react with the functional groups of the jute fibre and then deposite as a thin film on the surface. This surface modification may lead to reduction in hydrophilicity of jute fibre, which can improve the adhesion of hydrophobic polyester resin on the jute reinforcement. Among all the treatments, LIM (3-4%) treatment shows lower wetting time than other treatments.

3.2 Flexural Strength

Figure 1 shows the effect of sodium hydroxide treatment and both lac treatments (LIS / LIM) on flexural strength of jute /USP based biocomposite in both dry and wet states. It is observed that the sodium hydroxide (SH) treated jute fabric has significantly improved (54%) flexural strength of biocomposites in comparison with untreated jute based biocomposite. However, the increase in flexural strength of 3% LIS and 2% LIM treated jute fabric based biocomposite are found 42% and 18% respectively.

In wet state, the flexural strength values of all the samples are reduced in comparison with the respective dry state samples. Mechanical property of a fibre reinforced polymeric composite mainly depends on the amount of reinforcement, orientation of reinforcement, mechanical property of fibre, the interfacial adhesion between the fibre and the polymer matrix. If the interfacial bonding between the fibre and the polymer matrix is good, there will be uniform distribution and transferring of the applied load from matrix to fibre, which leads to increase in the mechanical property. In wet state of the biocomposite, the jute fibre reinforcement is plasticized by the water

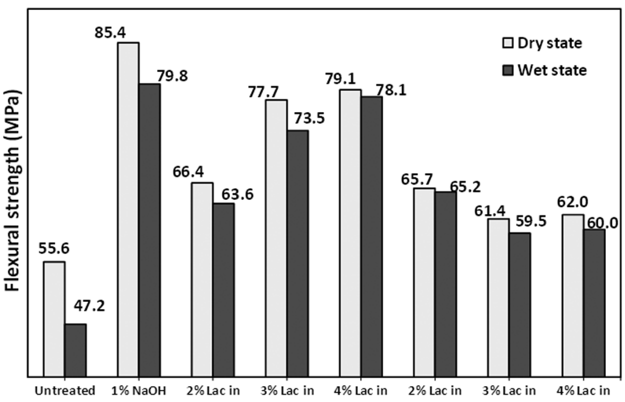

Fig.1—Effect of different treatments on flexural strength of jute based biocomposites in dry and wet states

film, which ultimately reduces the interfacial adhesion between fibre and polymer. This factor is responsible for the reduction in the flexural strength of the biocomposites in wet state [9,10].

The percentage reduction in flexural strength on untreated jute based biocomposites is 15%, while in SH, LIS and LIM treated jute fabric based biocomposite, the values are 8, 3 and 4% respectively, which indicate that these chemical modifications might improve the adhesion between jute fabric and polyester resin. Jute fibre is a hydrophilic fibre due to the presence of free hydroxyl groups (–OH) and can form hydrogen bonding between the macromolecules of the cellulose. During wetting in water, jute fibre absorbs water molecules through free hydroxyl groups that lead to swelling of the fibre [11]. Swelling of jute fibre in the biocomposite product reduces the interfacial bonding with matrix resin and reduces the mechanical property [12,13].

SH treated jute fabric based biocomposites show higher flexural modulus (3.9 GPa) in dry state than the other samples. However, in wet sate it shows more reduction (20%) in modulus than other samples (3-5%). SH treated jute fabric has more reactive groups and hence absorbs more moisture which leads to reduction in interfacial adhesion between jute fibre and resin. However, in LIS and LIM treated jute fabric based biocomposites, the lac resin masks the free hydroxyl groups of jute fibre and hence reduces the absorption of moisture. It is also apparent that water resistance of the biocomposite is improved by LIS and LIM treatments on jute fabric.

Flexural properties of a biocomposite is improved by increasing the proportion of fibre volume up to 50% and then it gradually reduces, due to inadequate adhesion/ spreading of polymer resin into the jute fabric, which also favours more fibre/fibre interaction than fibre/matrix interaction during preparation [14]. If the fibre load is at optimum level (30-50%), the transfer of load between the fibres and the polymer matrix remains uniform and efficient [15].

Table 2 shows the amount of jute reinforcement present in the biocomposites. It infers that lac modified jute based biocomposites have higher fibre load (40 - 42.9%) than untreated jute fabric based biocomposite (36.9%) due to better compatibility between modified jute fibre and polyester resin. Superior compatibility between fibre and resin might reduce the void formation among fibre, yarn, fabric layer, and interface between fibre and resin, which ultimately increases fibre load [16].

In hand-laying cum compression moulding method, biocomposite is prepared by taking reinforcement fabric: polyester resin in 1:3 ratio. When the untreated jute fabric is used, there will be less adhesion between reinforcement and resin, which leads to formation of void due to creation of space between resin and fibre. After chemical modifications, adhesion between polyester resin and jute fabric is increased, so that the flow of resin throughout the jute reinforcement becomes good, which reduces the void formation in between fibre and resin [17].

During compression, the removal of excess resin is higher in chemically modified jute fabric than in untreated jute fabric. These factors lead to decrease in resin percentage in the biocomposite, which ultimately increases the fibre load as well as mechanical properties.

3.3 Flexural Modulus

Figure 2 shows the effect of different treatments on flexural modulus (GPa) of jute fabric based biocomposites in dry and wet state. Similar to flexural strength, flexural modulus of all biocomposite samples in dry state is increased after NaOH and lac treatment. The percentage increase in flexural modulus of SH, LIS and LIM treated jute fabric based biocomposites in comparison with untreated jute based biocomposite are 88, 42 and 12% respectively. The reduction of flexural modulus of biocomposite samples in wet state as compared to that in dry state is higher in untreated jute based biocomposite (29%) than in chemically modified jute based biocomposites (2 - 19%). The percentage reduction in flexural modulus is lower for LIS treated sample than for SH and LIM treated jute based biocomposites, which indicates that lac treatment enhances the stiffness of jute fibre.

Table 2—Weight percentage of jute fabric in biocomposite

Treatment	Wt %	% increase
Untreated	36.5	--
1% NaOH	41.1	13
2% lac in NaOH	42.3	16
3% lac in NaOH	42.7	17
4% lac in NaOH	42.9	18
2% lac in methanol	42.6	17
3% lac in methanol	41.4	13
4% lac in methanol	40.0	10

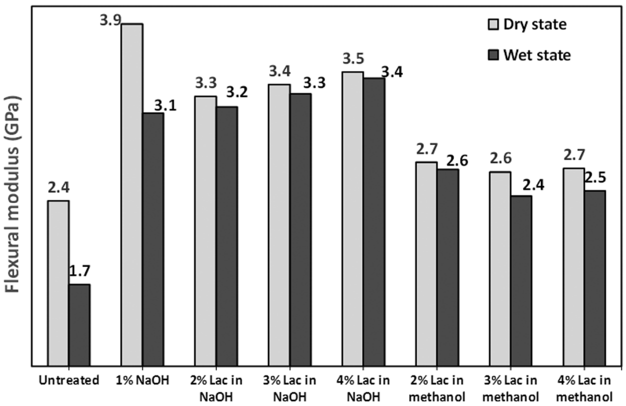

Fig.2—Effect of different treatments on flexural modulus of jute fabric based biocomposites in dry and wet states

Diffusion of moisture in natural fibre based biocomposite mainly depends on amount of fibre reinforcement and voids[18]. If the compatibility between fibre and resin is not good, there will be less interfacial adhesion, which leads to the formation of voids. Water molecule can easily penetrate inside the biocomposite through cellulose network. The entrapped water molecules swell the fibrils, create the space and reduce the bound areas of the fibrils with resin. Ultimately the rigidity of the jute fibre will reduce and permits the cellulose polymers to move freely. It can also lead to moisture induced interfacial cracks followed by degradation in the fibre–matrix interface region[19]. During load, there will be an accumulation of stress in a particular crack with high elongation and hence reduction in flexural modulus. The formation of interfacial cracks may be more in untreated jute based biocomposite in wet state than in chemically modified jute fabric based biocomposites[20,21]

3.4 Inter Laminar Shear Strength

Figure 3 shows the effect of different treatments on inter-laminar shear strength (ILSS) of jute fabric

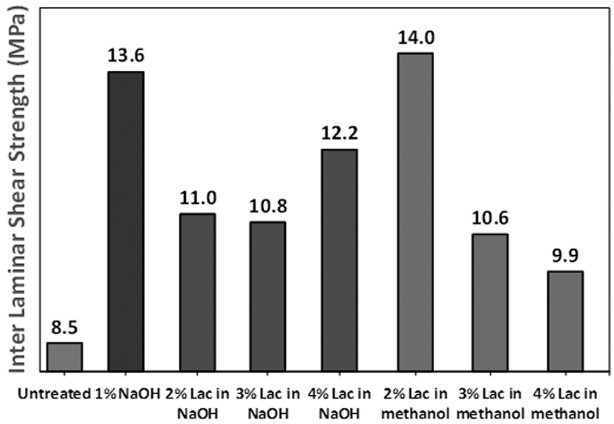

Fig.3—Effect of different treatments on inter laminar shear strength of jute fabric biocomposites

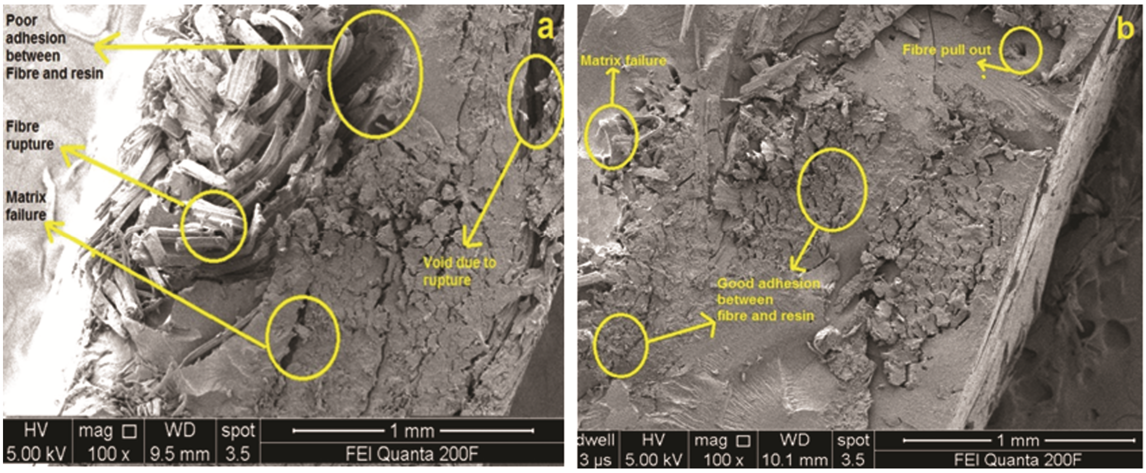

Fig.4—SEM photographs of transverse portion of untreated and lac treated jute fabric based biocomposites

based biocomposites. ILSS of natural fibre based composites is an important factor in the structural applications and it gives information about the extent of fibre-matrix adhesion. Adhesion between the fibre and resin may be improved either by removal of hydrophilic groups or by adding hydrophobic component on the jute fibre like maleation and acetylation. It is observed that thermoset polymer resin, such as epoxy, polyester and phenol formaldehyde shows better interfacial adhesion with sodium hydroxide modified natural fibre than with untreated fibre [3, 22]. Partial removal of hemicellulose portion by sodium hydroxide treatment apparently lowers hydrophilic property of jute fibre, while lac resin possibly forms a thin hydrophobic filmy layer on the surface of the jute fibre. These chemical modifications lead to increase in compatibility between jute fibre and polyester resin, so that ILSS value of modified jute fabric based biocomposites is higher than that of untreated jute based biocomposite.

The ILSS value of chemically modified jute based biocomposite is higher for 2% LIM treatment than for other treatments. The percentage improvement in ILSS value of SH and LIC/LIM treated jute based biocomposites in comparison with untreated jute based biocomposite are 60, and 64% respectively, which confirms the improvement in adhesion between resin and modified jute fabric. However, on increasing the lac concentration in the methanol medium (≥3%), lac resin may form a separate polymer layer between polyester resin and jute fibre, so that there is a reduction in ILSS values of the lac treated jute based composites.

3.5 Fracture Morphology

The improvement in the mechanical properties of the lac treated jute fabric based biocomposites might be due to superior bonding between unsaturated polyester resin matrix and jute fibre with less void content (Fig. 4b). On the other hand, in untreated jute fabric biocomposites, inferior bonding between jute fibre and matrix is observed (Fig. 4a). Such inferior adhesion might also lead to stress accumulation in the interface between the matrix and the reinforcement and it forms cracks throughout the biocomposites, which leads to reduction in mechanical properties[21].

4 Conclusion

The findings show that chemical modifications on jute fabric improve the flexural strength and flexural modulus of the biocomposite significantly as compared to untreated jute based biocomposite. Lac reacts with the hydroxyl groups of the cellulose polymer and forms a hydrophobic thin polymer film on the surface of the jute fibre, which improves the adhesion between polyester resin and jute fibre. Improvement in adhesion between unsaturated polyester resin and modified jute fabric possibly enhances the fibre loading, flexural strength and inter-laminar shear strength of the biocomposites. It is also observed that the strength of lac in sodium hydroxide medium treated jute fabric based biocomposites is higher than the respective lac in methanol medium treated biocomposites. It is concluded that lac treatment in sodium hydroxide medium on jute fabric can be used to improve the mechanical properties of jute / thermoset resin based biocomposite by the hand-laying followed by compression moulding method.

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