Studies on Jute-Reinforced Composites, Its Limitations, and Some Solutions Through Chemical Modifications of Fibers

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ABSTRACT: Jute, unlike other natural fibers, absorbs moisture and its moisture regain property is quite high. Water migration and subsequent degradation of jute-based composites can be a problem. Because jute is hydrophilic and the matrix resins are mostly hydrophobic, wetting of the fibers with resins is poor, for which high resin consumption may occur that would increase the cost of composites. To reduce the moisture regain property of jute fiber, it is essential to pretreat the jute fiber so that the moisture absorption is reduced and the wettability of the resin is improved. Jute fiber in the form of nonwoven jute has been pretreated with precondensate like phenol formaldehyde, melamine formaldehyde, cashew nut shell liquid-formaldehyde, and polymerized cashew nut shell liquid. The moisture content of the pretreated nonwoven jute has been determined by conventional methods and by a differential scanning calorimetric technique. Treatment of jute with precondensate causes the reduction of water regain property in jute. Pretreated nonwoven jute has been impregnated with phenol formaldehyde resin, and the composite board has been prepared therefrom. The jute composite board has been tested for bending strength, tensile strength, thickness swelling, and water absorption. Thermal analyses, such as differential scanning calorimetry and thermogravimetry, have also been conducted on jute and pretreated jute fibers. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci **67**: 1093–1100, 1998

Key words: jute fiber; pretreatment; composite; mechanical property; thermal stability

INTRODUCTION

There has been a tremendous growth in polymer matrix composites containing fibers. These composites provide unique a combination of properties (including high specific strengths and moduli) and therefore substitute other conventional materials.

Natural fibers, particularly vegetable fibers, do not appear much in the list of applications, because their use in reinforcement is minimal, compared with the consumption of glass and synthetic fibers. However, in recent times, many of the conventional uses of natural fibers are threatened by plastics. Thus, there is a need to develop new uses for these fibers. In addition, natural fibers like jute have attracted the attention of scientists due to their easy availability, renewable resource, low price, and low density leading to high specific strength. The potential strength and toughness of natural fibers like jute has not yet been exploited in true fiber-reinforced composites (equivalent to fiber reinforced plastics (FRP) composite based on glass fibers), because jute absorbs a great deal of moisture and has poor wettability with organic matrix resins.[1–10] There is a weak interfacial bonding between those fibers and commercially available resins (such as polyester resin, epoxy

resin, phenolic resin, amino resin, etc.). Surface modification is therefore necessary. Due to the presence of pendent hydroxyl and polar groups in the various constituents of fiber, moisture regain is very high. This leads to a poor interfacial bonding with the relatively more hydrophobic materials. The environmental performance of such composites is also poor due to delamination under humid conditions.[11] The high moisture regain of the fibers also leads to a reduction in properties, such as modulus and strength.

Jute, despite the dedicated work already conducted, is very much in its infancy in its use as a reinforcement.[1,12–19] Clearly, much work is to be done not only in using jute reinforcement in the most appropriate way and for the most appropriate products, but also to develop efficient chemical reagents for interfacial coupling.

Morphological studies of jute fibers show the outer sheath of lignin that develop the cellulosic ultimates. It is to this material that bonding must take place if an efficient composite is to be manufactured. Removal of this surface layer of lignin usually results in a better and more stable bond, but at the expense of a more complex operation and a higher priced product. The usual requirements of a coupling agent are that it should interact chemically with both the fiber surface and the matrix resin. This necessiates a structure that has lignin compatibility—a relatively unexplored area of research—and a reactive compatibility with the matrix resin. It is only with the development of effective couplers that the stiffness and strength inherent in the jute fiber and good water resistance of the composite can be achieved, and the way opened to the manufacture of a new range of jute-based products. One approach to improve the wet strength of jute fiber-reinforced compos-

ites has been to use interfacial agents having good water repellency and favorable hydrophobic-hydrophilic balance, thus ensuring good bonding of the cellulosic fiber with the resin matrix. Several workers have studied the efficiency of different interfacial agents that were expected to protect the fibers from the degrading effect of water, apart from improving the fiber–matrix bonding. Marginal to good improvements both in dry and wet strength have been reported with the application of these interfacial agents. However, improvement was not adequate enough for successful commercial applications.

In our study,[7] it is thought that some water-thin, solvent-dilutable low molecular weight precondensate based on phenol formaldehyde (PF), cashew nut shell liquid (CNSL)-modified phenolic resin, modified melamine formaldehyde condensate, and CNSL have been examined to achieve the aforementioned objective to have the jute composite of higher strength and good water resistance.

EXPERIMENTAL

Materials

Jute fibers (nonwoven jute felt, needle-punched, 700–750 GSM, thickness of 7.5 mm) were obtained from Ludlow Jute Mills (Chengail, Howrah, India) and Birla Jute Mills (Birlapur, 24-Parganas, India).

Chemicals

Precondensate based on PF, CNSL-PF, water-soluble and methanol-soluble CNSL-formaldehyde

Table I   Results of Pretreatment of Nonwoven Jute with a Different Precondensate

Pretreatment Chemical Used	Nature of the Chemical	Concentration Used (%)	Moisture Regain[a] (%)	Moisture Content[b] (%)
PF precondensate (50%)	Water-soluble	5	8.98	9.4
Modified MF precondensate (80%)	Water- and methanol-soluble	5	8.84	7.8
CNSL-PF precondensate (50%)	Water-soluble	5	7.93	7.0
CNSL-PF precondensate (55%)	Methanol-soluble	5	8.10	9.3
Polymerized CNSL	Hydrocarbon-soluble	5	8.72	9.2
CNSL formaldehyde	Hydrocarbon-soluble	5	9.33	10.0
Control	—	5	10.52	10.4

MF = melamine formaldehyde.
[a] Dried and conditioned at 65% relative humidity.
[b] Determined by DSC.

Table II Physical Properties of the Jute Composite with Treated Jute Felt

Physical Properties	PF (5%)	MF (5%)	CNSL-PF (5%) (Water-Soluble)	CNSL-PF 5% (Methanol-Soluble)	CNSL-Formaldehyde (5%)	CNSL-Polymerized (5%)	Control (Laminated with 15% PF)
Density (g/mL)	1.13	1.13	1.12	1.20	1.16	1.14	1.16
Tensile strength (Mpa)	47.70	49.99	62.21	77.37	40.83	38.12	42.10
Flexural strength (Mpa)	72.32	73.97	90.03	100.03	59.98	49.73	68.24
Water absorption (%)							
After 2 h soaking	19.39	18.98	12.08	13.23	13.95	18.95	19.13
After 24 h soaking	26.98	25.93	20.72	21.13	22.00	26.11	25.71
Thickness swelling (%)							
After 2 h soaking	11.08	10.98	7.32	8.09	9.02	11.92	10.89
After 24 h soaking	14.89	14.00	11.97	12.01	13.72	15.13	14.12
After 2 h boiling with water							
Flexural strength (Mpa)	26.13	27.50	58.27	62.33	22.70	Delaminated	22.17
Thickness swelling (%)	27.03	26.91	16.96	17.00	26.63	—	25.62

Conditions: six layers of each pretreated jute felt were laminated with 10% PF resin at 160°C temperature under 500 psi pressure for 10 min. But, the control was laminated with 15% PF under the same conditions to make the resin percentage the same in all of the laminates. MF = melamine formaldehyde.

and polymerized CNSL (benzene-soluble), and PF resin for final bonding were all supplied by M/S Hindusthan Adhesive and Chemicals (Calcutta, India). Modified melamine resin was supplied by M/S Macrotech India (Calcutta, India).

Treatment and Preparation of Composite

Jute fibers in the form of nonwoven jute (750 GSM) were soaked in definite solution concentration (5–10% solid) of precondensate [phenolic, melamine, CNSL-modified phenolic, CNSL-modified phenolic (methanol-soluble), CNSL-formaldehyde resin, and polymerized CNSL (benzene-soluble)]. Jute fibers, after soaking, were then squeezed and air-dried. After the fibers were dried to a constant weight, nonwoven jute was then impregnated with 10% PF resin (as supplied) solution and similarly dried as per requirement. The impregnated nonwoven jute was then cut to size and pressed at 150–160°C for a period of 10 min, depending on the thickness of the composite board desired. It was then trimmed.

Testing of Pretreated Jute and Composite

Pretreated nonwoven jute was tested for moisture regain property at 65% relative humidity and tested for moisture content by differential scanning calorimetry (DSC) analysis.

The jute composite board made therefrom was tested for flexural strength, tensile strength, water absorption, and thickness swelling. Accelerated weather testing for water absorption by immersion in boiling water for 2 h was adopted. This procedure would account for aging due to humidity and thermal effects, but not photochemical effects (ASTM D-570-63).

Thermal Analysis

The thermogravimetry (TG) and DSC measurements were performed using a Mettler TG 50 and a DSC 25 module attached to a Mettler TC 11 4000 thermal analyzer. A heating rate of 10°C/min and a sample weight of 5–10 mg in an aluminium crucible with a pin hole were used in a nitrogen atmosphere (100 mL/min). During evaluation of differential thermogravimetry (DTG), the peak temperature and amount of char left at 600°C were measured. A computer model Epson PC AX 2 attached to TC 11 4000 was used for generating figures.

RESULTS AND DISCUSSION

Pretreated Jute

Results of pretreatment of jute fibers for moisture regain property and moisture content have been reported in Table I. It indicates that the reduction in moisture regain is quite considerable, particularly in the case of CNSL-modified phenolic resin in comparison with the other system. It may be due to two reasons. The resin used was water-soluble and water-thin, so that it was compatible with jute fiber very easily (i.e., its wettability was
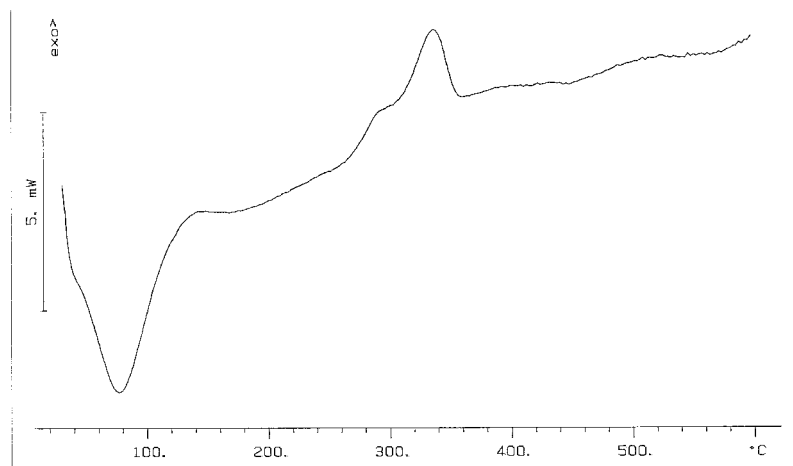

Figure 1   DSC curve of PF-treated jute fibers in a nitrogen atmosphere.
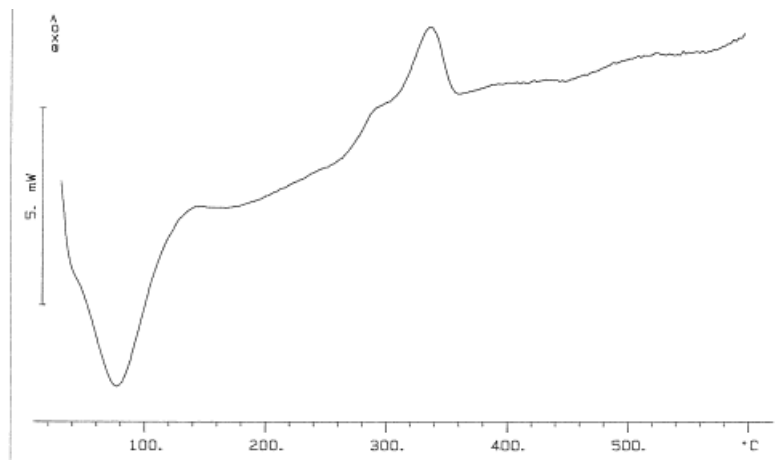
Figure 2 DSC curve of CNSL-PF-treated jute fibers in a nitrogen atmosphere.

considerably good while it was dried; it was more hydrophobic because of the presence of CNSL, which as such is hydrocarbon-soluble). This was further confirmed by the fact that, whereas simple phenol formaldehyde condensate was being used, it was reducing the moisture regain property not to the extent as in the case of CNSL-modified phenolic resin that is much more hydrophilic in nature. The same was the fact with CNSL-formaldehyde resin or polymerized CNSL resin that are hydrophobic in nature, even at the time of impregnation so that wettability with jute was poor. There should be a balance of a hydrophilicity and hydrophobicity in the chemicals to be treated.

Through DSC, the moisture content in treated jute was found to be considerably lower than that in control jute fiber.

Jute Composite

The composite had been made using treated jute fiber with 10% phenolic resin matrix, and their flexural strength and tensile strength had been determined. The thickness swelling and the percentage of water absorption had also been determined in each case, and the results have been reported in Table II. The wet strength of the composite had also been determined after boiling for 2 h according to ASTM-D-570-63 for estimating accelerated weathering. This procedure would account for aging due to humidity and thermal effects, but not photochemical effects. Elaborate weathering tests by subjecting the specimens to control environmental conditions in a chamber where almost all of the factors contributing to

Table III Results of DSC of Jute and Pretreated Jute in Nitrogen

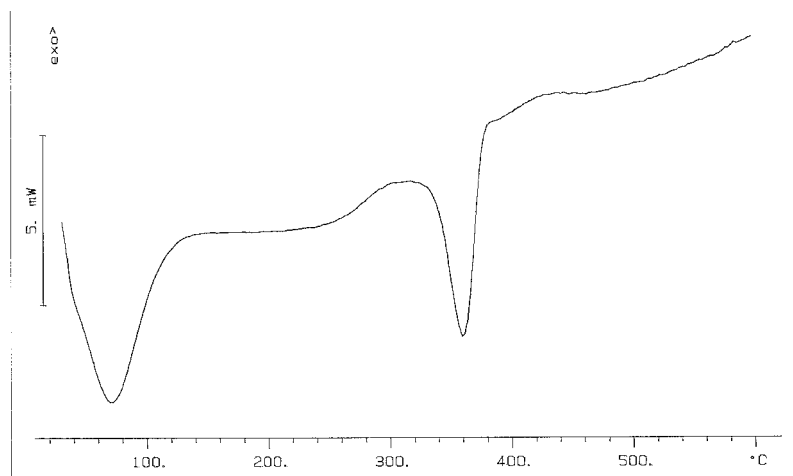| Sample	Peak Temperature (°C)	Nature of Peak	Moisture Content (%)	ΔH (J/g)
Jute fiber	68.8	Endo	10.4	239.0
	303.1	Exo		
	358.7	Endo		
	426.9	Exo		
PF-treated jute	75.9	Endo	9.4	217.0
	291.4	Exo		
	343.0	Exo		
	410.5	Exo		
CNSL-PF-treated jute	75.9	Endo	7.0	210.4
	289.1	Exo		
	336.0	Exo		
	391.8	Exo		

Figure 3 DSC curve of jute fibers in a nitrogen atmosphere.

weathering were incorporated had not been considered necessary at this stage of work. However, the results indicate that the strength (tensile and flexural) had been considerably improved in all of the cases. The maximum strength was observed in the case of CNSL-modified phenolic resin (spirit-soluble and water-soluble). The thickness swelling and water absorption (after 2 and 24 h, and after 2 h boiling) had also been found to be decreased, in comparison with blank as expected. In the case of CNSL-modified resin treatment, the thickness swelling was reduced considerably. As discussed previously, the accelerated weathering test (2 h boiling water test) as per ASTM D-570-63 could be a test for dimensional stability of the composites. The dimensional stability of the jute composites thus achieved may be a guideline for

its success in structural application. The process found seems to be easier for commercial exploitation in the sense that it is easy to apply, and minimum energy is required for application.

Thermal Analysis of Treated Jute Fiber Vis-à-Vis Untreated Jute

DSC curves of PF-treated jute fiber and CNSL-modified PF-treated jute fiber (Figs. 1 and 2; Table III) reveal water desorption endotherm below 100°C. It shows that the enthalpy changes (ΔH) associated with the moisture desorption peak of the treated jute (both phenolic and the CNSL-modified phenolic system) decrease when compared with untreated jute (Fig. 3; Table III). The moisture content of the treated jute also reduces. These results indicate that, due to pretreatment with both the resin sepa-
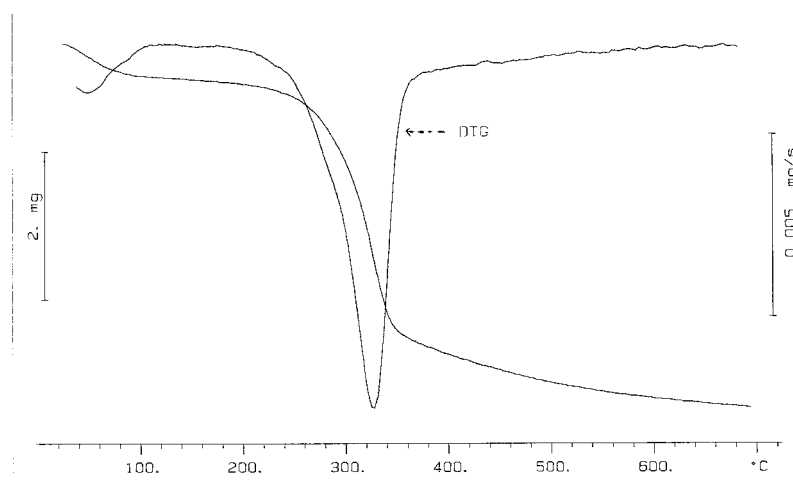

Figure 4 TG and DTG curves of PF-treated jute fibers in a nitrogen atmosphere.
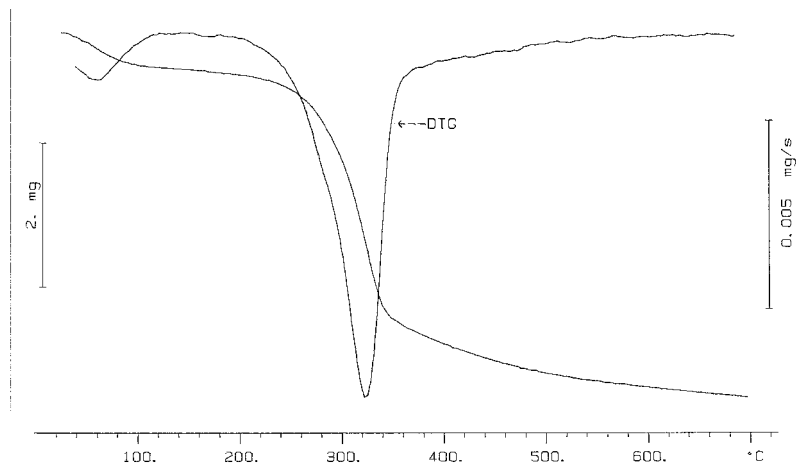
Figure 5   TG and DTG curves of CNSL-PF-treated jute fibers in a nitrogen atmosphere.

rately, water repellancy to the fiber is imparted. The nature of the main decomposition peak temperature due to α-cellulose shifted from endothermic to exothermic to a lower temperature value. The peak temperature due to hemicellulose also shifted to a lower temperature. These results clearly show that the thermal stability of the jute fiber when treated with PF resin/CNSL-modified PF resin, respectively, decreases.

DTG thermograms of treated jute (Figs. 4 and 5; Table IV) and untreated jute (Fig. 6; Table IV) show initial moisture loss peak below 100°C and a main decomposition peak at 355.7°C for untreated jute, 328.2°C for phenolic resin-treated, and 323.7°C for CNSL-modified phenolic resin. In the cases of treated jute, the decomposition step due to hemicellulose, which is observed in the case of untreated

jute, is almost merged with α-cellulose degradation. Table IV shows that the main decomposition peak temperature of the treated jute fiber is shifted to the lower side (328.2°C for PF-treated jute and 323.7°C for CNSL-modified PF resin) with respect to untreated jute (355.7°C). Weight loss at this step (combination of hemicellulose and α-cellulose) decreases, whereas char left (%) at 600°C enhances that of untreated jute. It was reported previously that flame retardancy in a sample is imparted when the main decomposition temperature and the weight loss at that temperature reduce, whereas the percentage of char left is enhanced.[20] Treatment of jute with either PF resin or CNSL-modified resin reduces the pyrolysis temperature, decreases weight loss during pyrolysis in comparison with untreated jute, and enhances the char formation,
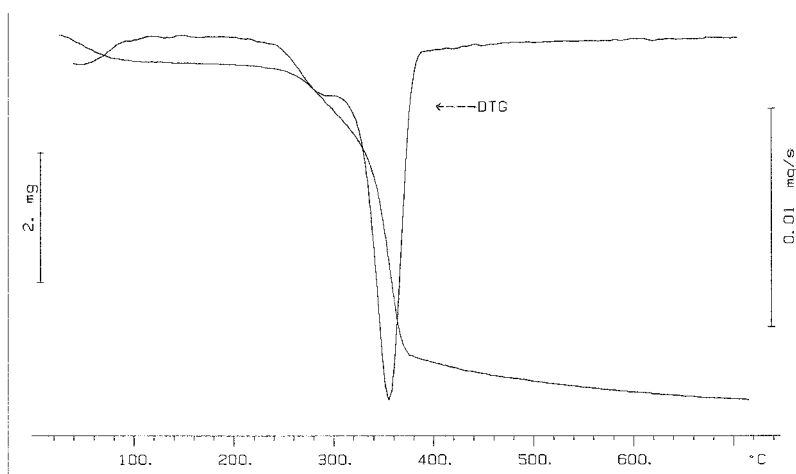

Figure 6   TG and DTG curves of jute fibers in a nitrogen atmosphere.

Table IV   Results of Derivative TG Analysis of Jute and Pretreated Jute Fiber in Nitrogen

Sample	Degradation Temperature (°C)	Weight Loss (%)	Residual at 600°C (%)
Jute fiber	43.7	8.97	12.79
	293.0	10.29	
	355.7	67.65	
PF-treated jute	50.5	7.24	25.48
	—	—	
	328.2	65.81	
CNSL-PF-treated jute	54.3	6.97	25.55
	—	—	
	323.7	65.62	

thereby facilitating the formation of a lower percentage of flammable volatiles. Due to treatment, the thermal stability is decreased to some extent—perhaps because of the strength loss due to treatment, as observed in the case of jute fiber for any wet processing.

CONCLUSIONS

It is observed that the moisture regain property is reduced through pretreatment, which is our main goal to make the composite more water resistant. In addition, this pretreatment also imparts flame retardancy. In comparison with the two types of treatments, CNSL-modified phenolic resin behaves better during reduction in moisture regain property and strength of composite in respect of tensile strength, three-point bending.

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