

Study of Mechanical Properties of Glutarylated Jute Fiber Reinforced Epoxy Composites

V. Manush Nandan, K. Lokdeep, R. Vimal, K. Hari Hara Subramanyan, C. Aswin, V. Logeswaran

Abstract—Natural fibers have attained the potential market in the composite industry because of the huge environmental impact caused by synthetic fibers. Among the natural fibers, jute fibers are the most abundant plant fibers which are manufactured mainly in countries like India. Even though there is a good motive to utilize the natural supplement, the strength of the natural fiber composites is still a topic of discussion. In recent days, many researchers are showing interest in the chemical modification of the natural fibers to increase various mechanical and thermal properties. In the present study, jute fibers have been modified chemically using glutaric anhydride at different concentrations of 5%, 10%, 20%, and 30%. The glutaric anhydride solution is prepared by dissolving the different quantity of glutaric anhydride in benzene and dimethyl-sulfoxide using sodium formate catalyst. The jute fiber mats have been treated by the method of retting at various time intervals of 3, 6, 12, 24, and 36 hours. The modification structure of the treated fibers has been confirmed with infrared spectroscopy. The degree of modification increases with an increase in retention time, but higher retention time has damaged the fiber structure. The unmodified fibers and glutarylated fibers at different retention times are reinforced with epoxy matrix under room temperature. The tensile strength and flexural strength of the composites are analyzed in detail. Among these, the composite made with glutarylated fiber has shown good mechanical properties when compared to those made of unmodified fiber.

Keywords—Flexural properties, glutarylation, glutaric anhydride, tensile properties.

I. INTRODUCTION

IN disquiet for environment, there is an increased focus in using natural fibers as the reinforcement material in polymer matrix composites. The main advantage of using natural fibers in manufacturing composite materials is that they are environment friendly. In addition to this they are abundant in nature, economic and easily processed. The natural fibers are resistant to the wear during the polymer processing and have no major reduction in size during processing [1]. Natural fibers are classified into plant and animal fibers, among which plant fibers are focused greatly because of the excellent wear resistant properties. Organic

cellulose-based fibers are hydrophilic in nature due to the presence of hydroxyl group which make them incompatible with hydrophobic polymers such as epoxy resin [1]. Weak fiber/matrix interface reduces the reinforcing efficiency of the fiber due to lack of stress transfer from the matrix to load bearing fibers [15]. Therefore, these fibers usually require chemical modification for the increased compatibility in polymer- fiber interface [2]. Many of the plant fibers such as neem, kenaf, wood, cotton, banana, bagasse, sisal, bamboo and hemp fibers were utilized in several forms for making composites [18], [19]. Among these fibers, jute fibers are largely manufactured in countries like India. Jute fiber is the crystallite cellulose consisting of parallel chain of number of glucose residues. Jute fiber is having length of 150 to 200 units of glucose and cross sections of 200 to 250 units. The density of the jute fiber was about 1.48 g/cm³ [20]. In the present study, jute fibers have been modified chemically using succinic anhydride and glutaric anhydride, its mechanical and thermal properties after reinforcing with epoxy resin were studied.

II. EXPERIMENTAL

A. Materials

Bi-directional jute fiber mats were purchased from Meena fibers, India. The fibers were placed in the oven for 3 hours at 45 °C. To prevent any excess absorbance, the oven-dried fiber is kept in the desiccator supported with Silica Gel. Glutaric anhydride was purchased from Avra chemicals, India. Dimethylsulfoxide, sodium formate, polyvinyl alcohol and ethanol were purchased from Fisher Scientific, India. Epoxy resin and ethyl methyl ketone peroxide were purchased from SVE Pvt Ltd, India.

B. Fiber Preparation

Jute fiber mats obtained were rinsed with double distilled water, followed by ethanol to remove any foreign impurities. The rinsed fiber mats were oven dried for about 12 hours at 45 °C. The jute mats of about 1 mm thickness with the side of 300 mm were used for further treatments.

C. Solution Preparation

The glutaric anhydride solution required for the fiber treatment has been prepared by dissolving required weight fraction of glutaric anhydride in 100 mL of dimethylsulfoxide and 10 mL of benzene with sodium formate as catalyst [5]-[14]. The weight fraction of the glutaric anhydride solutions used for the treatment were 5%, 10%, and 20%

V. Manush Nandan is an Independent Researcher graduated from Sri Sairam Institute of Technology, Chennai-600044, Tamil Nadu, India (phone: 9003491593; e-mail: manush926@gmail.com).

K. Lokdeep is an Independent Researcher graduated from Sri Sairam Institute of Technology, Chennai-600044, Tamil Nadu, India (phone: 9600129718, e-mail: lokdeepk@gmail.com).

R. Vimal is with Madras University, Chetpauk Campus, Chennai, Tamil Nadu, India, India (phone: 8807473156; e-mail: vimal.r.100@gmail.com).

K. Hari Hara Subramanyan, C. Aswin, and V. Logeswaran are graduated from Sri Sairam Engineering College, Chennai- 600044, India (e-mail: hhsallrounder@gmail.com, ashtmt@hotmail.com, logeswaranvaradharajen@gmail.com).

D. Fiber Treatment

The rinsed fiber mats were retted at room temperature at various retention times of 3, 6, 12, 24, 36 and 48 hours in the solution of glutaric anhydride at various weight fractions. The anhydride modified fibers were rinsed in double-distilled water, followed with ethanol to remove any remaining acids and impurities. The modified fiber mats were dried in oven at 45 °C for 6hr. The weight of the fibers before and after glutarylation was measured.

E. Sample Preparation

The treated jute fibers have been reinforced with epoxy matrix by means of hand moulding technique. For this purpose, a metallic mould of size $300 \times 300 \times 6 \text{ mm}^3$ has been made [4]. Polyvinyl alcohol solution has been coated over the mould surface, which facilitated the process as a release agent [3]. Now epoxy matrix was coated on the mould, above which jute fiber mat was placed. Alternate layers of jute fiber mats and epoxy matrix were used until the required thickness is obtained. Vacuum was created to remove the gases trapped in between the layers as a result of exothermic reactions [17]. To avoid the formation of voids and holes, the composite has been rolled with a steel roller. The composite was allowed to be cured at room temperature for nearly 36hours. The cured composite was analyzed to understand its various properties.

III. CHARACTERIZATION

A. Morphology

The Hitachi: S3400N type scanning electron microscope (SEM) with motorized 5- axis, eucentric specimen stage and Quad type detector was used for imaging with TMP/ RP based vacuum system. The SEM image of unmodified and glutarylated fibers were taken. The samples of standard size were coated with gold target using ion sputter coater and were placed in the eucentric specimen stage. The voltage of 30 kV was used to accelerate electron beam for imaging.

B. Infrared Spectroscopy

The chemical structure of the glutarylated jute fibers at 10% concentration and 24hours time interval was analyzed using Perkin Elmer FT-IR spectrometer in the transmittance mode as the function of wave number. The samples were grounded using ball mill and analyzed using KBr sampling technique. About 1mg of powdered sample was mixed with 100mg of KBr powder and the mixture was made into pellet and analyzed. The testing was done at the wave number range of 4000 cm^{-1} to 400 cm^{-1} .

C. Mechanical Testing

Uniaxial tensile testing of the standard and glutarylated fiber (GJF) reinforced composites were tested in accordance to the ASTM D638 procedure using Instron universal Testing Machine. The test specimen of standard size in dumbbell shape was cut using hydraulic cutter. The cross head speed of 5mm/ min was set and the test was conducted at room temperature. Three point flexural tests were also performed on the above mentioned samples in accordance with ASTM D790

procedure. The test specimen of standard size was cut using hydraulic cutter. The test was conducted using the Instron Universal Testing Machine with the load capacity of 20 kN. Both of the above tests were performed with six samples of respective dimensions.

IV. RESULTS AND DISCUSSIONS

A. Weight Gain

Fig. 1 shows the weight gain of the glutarylated jute fibers. The weight of the fibers before and after the treatment was examined. Weight gains of the fiber after treatment under various volume fractions were given in Table I. All the fiber samples used for the treatment were of size $300 \times 300 \text{ mm}^2$. The weight of the untreated fiber was found to be about 21g.

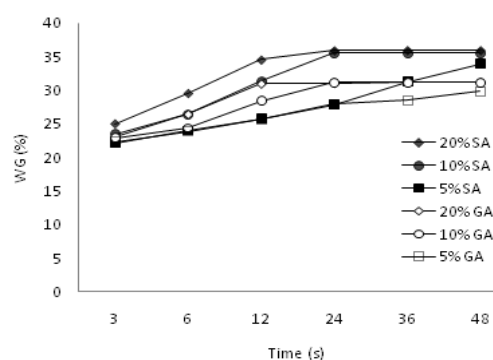


Fig. 1 Weight gain of glutarylated jute fibers at various weight fractions

The weight of the fiber at different weight fractions was increasing constantly which has been measured regularly. The saturation point on glutarylation was attained at the retention time period of 24 hours in the retention concentration of 5%, 10% and 20%. In the case of 30% and 40%, the fiber structure has been damaged and hence the fiber treated in those retention times were omitted for further processes. The modification of chemical structure of jute fiber on glutarylation, at the respective time interval was confirmed with infrared spectroscopy.

TABLE I
WEIGHT GAIN OF THE FIBER ON GLUTARYLATION PROCESS

Type	Glutarylation		
Time (hours)	20% WG (%)	10% WG (%)	5% WG (%)
3	22.971	22.856	22.352
6	26.428	24.327	23.921
12	30.946	28.476	25.672
24	31.038	31.213	27.985
36	31.103	31.174	28.532
48	31.148	31.246	29.891

Since the process of glutarylation involves complex chemical Structure, it is not that easy to mention the saturation point. The chain of the glutaric anhydride molecule is larger due to the presence of $-\text{CH}$ group between the two methylene groups. This was also confirmed with the broad band at 3247

cm^{-1} in Fig. 4 which represents the presence of $-\text{OH}$ group in the glutarilated fiber. At 30% and 40% weight fraction of the respective materials, fibers were almost damaged even at lesser time interval.

B. Morphological Assessment

Fig. 2 shows the typical microstructure of the surface of the untreated jute fiber. The waxy impurity over the surface of the jute fiber reduces the wettability of the fiber and hence the fiber had become less compactable with most of the polymer matrices. At higher magnification, the presence of waxy esters had been confirmed. The waxy surface of the fiber was due to the carboxylic ester compounds present in the fiber.

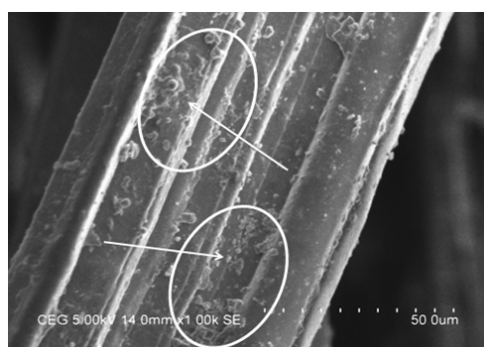


Fig. 2 Scanning electron micrograph of untreated fiber

Fig. 3 shows the microstructure of the jute fiber retted in glutaric anhydride solution. Glutarylation of jute fiber leads to the removal of waxy impurities from the fiber surface. The microstructure of the glutarilated fiber shows very smooth surface when compared to that of untreated fibers. This is because of the extended carbon chain of the glutaric compounds which reacts with $-\text{OH}$ of Jute fibers.

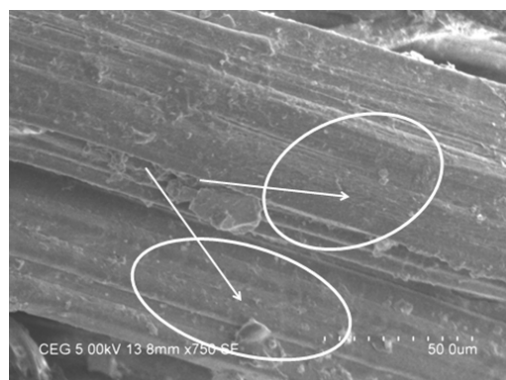


Fig. 3 Scanning electron micrograph of glutarilated fiber

C. Infrared Spectroscopy

Fig. 4 shows the infrared spectrum of glutarilated fibers. The chemical structure of the glutarilated fiber was confirmed with the strong peaks occurring at 1841 and 1734 cm^{-1} . This corresponds to the presence of two $\text{C}=\text{O}$ groups in the structure of glutarilated fiber. The high frequency band corresponds to the symmetrical vibrations and the lower

frequency band corresponds to the asymmetrical vibrations. The splitting of the higher frequency band and lower frequency band is due to the occurrence of fermi resonance [16]. The peak at 1061 cm^{-1} represents the $\text{C}-\text{O}$ group of acid anhydrides, which confirms the bondage between jute fiber and Glutaric anhydride. The presence of broad band in the range of 3300 – 3100 cm^{-1} in the infrared spectrum of glutarilated fiber indicates the presence of hydroxyl group of the fiber. The presence of $-\text{CH}_2$ group was confirmed with the peak in the region of 1399 cm^{-1} for glutarilated fiber. The $\text{C}-\text{O}$ stretching vibration of the lignin group of the jute fiber was confirmed with the peak at 1273 cm^{-1} for glutarilated fiber. This represents the presence of ester compounds of the fiber which was not removed completely. Various observed spectra were listed out in Table II. The structures of the glutarilated fibers are as follows

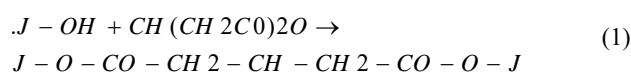


TABLE II
INFRARED SPECTRUM OF SUCCINYLATED AND GLUTARILATED FIBERS

Groups	IR Band (cm^{-1})	
	SJF	GJF
$-\text{OH}$ stretching	-	3247
$-\text{C}=\text{O}$ of anhydride	1848,1725	1841,1734
$-\text{C}-\text{O}$ of anhydride	1043	1061
$-\text{C}-\text{O}$ stretching vibration of lignin	1256	1273
$-\text{CH}$ asymmetric vibration	2873	2886
$-\text{CH}$ vibration of glutaric anhydride	-	2995
$-\text{CH}_2$ symmetric deformation	1406	1399

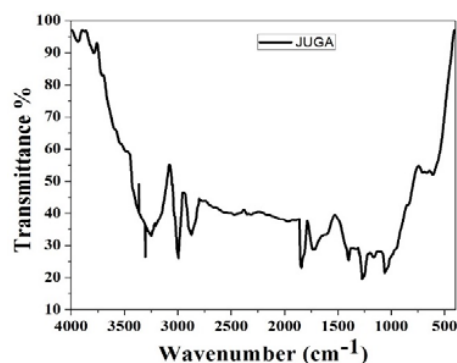


Fig. 4 Infrared spectrum of glutarilated fiber

D. Mechanical Testing

1. Tensile Properties

The tensile strength of the raw jute, 5% modified, 10% modified and 20% modified jute fiber reinforced epoxy composites are shown in Fig. 5. The tensile strength of the glutarilated fiber is found to be increasing with increased weight fractions, which is due to the presence of large carbon skeleton in the glutaric anhydride molecule. The tensile strength of the raw jute fiber reinforced composite is found to be 32.12 MPa, wherein the tensile strength of 10% modified jute fiber is found to be 46.12 MPa. The increment is by

69.6% in the tensile strength of 10% modified fiber reinforced composites to that of the raw fiber reinforced composite. The increment in the strength is because of the structural modification of the jute fiber, where the hydroxyl group of jute fiber is being replaced by the carbon chain of the glutaric anhydride. Epoxy resin, being more compatible with the carbon end chains is able to produce better bondability with the modified jute fiber, as a result of which increase in tensile strength is observed. The tensile strength of composites prepared with 5% modified fiber and 20% modified fiber. Even though the fiber have more hydroxyl group in its structure after treatments (confirmed with peak at 3247cm^{-1} in the infrared spectrum), there is a drop in tensile strength of 20% modified fiber reinforced composite than that of 10% modified fiber reinforced composite, the drop in the strength is confirmed with the rupture in the structure of fiber with increased concentration of Glutaric Anhydride (confirmed with the SEM Image).

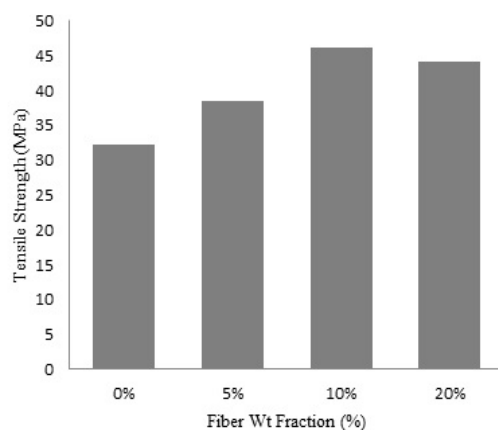


Fig. 5 Tensile strength of raw, 5%, 10% and 20% modified fiber reinforced composites

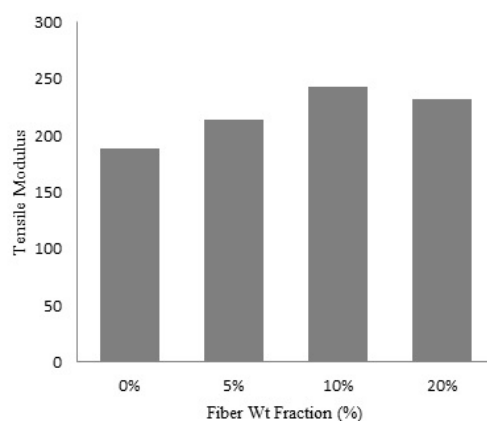


Fig. 6 Tensile modulus of raw, 5%, 10% and 20% modified fiber reinforced composites

Fig. 6 shows the tensile modulus of raw jute, 5% modified, 10% modified and 20% modified jute fiber reinforced epoxy composites. As in the case of tensile strength, tensile modulus is increased with increase in the weight fractions of Glutaric

anhydride. The tensile modulus of 10% modified fiber reinforced epoxy composite is 242.76 MPa, which is 77.8% increment when compared to the tensile modulus of raw jute fiber reinforced epoxy composite. The tensile modulus of 5% modified jute fiber reinforced composite and 20% modified fiber reinforced composite is found to be 213.76 MPa and 232.21 MPa, respectively.

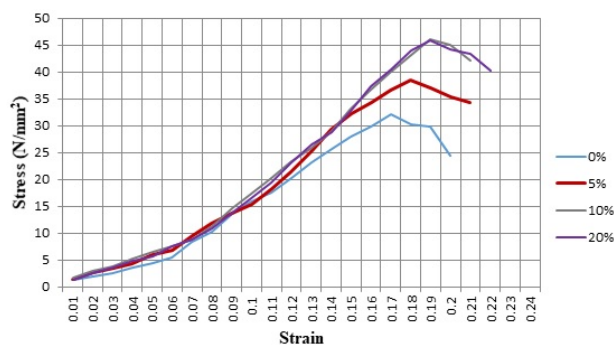


Fig. 7 Stress-strain curve of raw, 5%, 10% and 20% modified fiber reinforced composites

Fig. 7 shows the stress strain curve of raw, 5% modified, 10% modified and 20% modified jute fiber reinforced epoxy composite.

2. Flexural Properties

Flexural strength of raw, 5% modified, 10% modified and 20% modified jute fiber reinforced epoxy composite were shown in fig. 8. As in the case of tensile strength, flexural strength analysis too had given the same for of variations in its graph. The flexural strength has increased with increase in weight fractions of glutaric anhydride and there is a sudden drop in strength when 20% modified fiber reinforced composite was subjected to the load. The flexural strength of 10% modified jute fiber reinforced composite is 383.18MPa, which is 78.2% more than that raw jute fiber reinforced composite. The flexural strength of 20% modified jute fiber reinforced composite is found to be 366.52MPa.

Fig. 9 shows the flexural modulus of raw, 5% modified, 10% modified and 20% modified jute fiber reinforced epoxy composite. Even though, the flexural modulus of 5% modified jute fiber reinforced composite did not show any huge variation with that of raw jute fiber reinforced epoxy composite, the 10% modified jute fiber reinforced epoxy composite has shown gradual change in the flexural modulus of the composite. The flexural modulus of 10% modified fiber reinforced composite is around 1.77 GPa.

Fig. 10 shows the load displacement curve of raw, 5% modified, 10% modified and 20% modified jute fiber reinforced epoxy composite. The 10% modified jute fiber reinforced epoxy composite has attained the maximum load of 4.6 kN at the displacement of 14 mm.

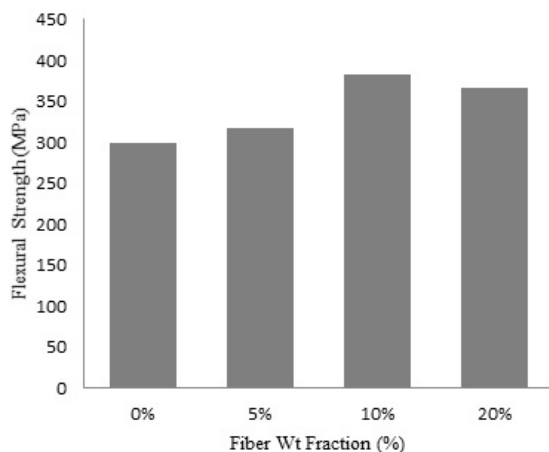


Fig. 8 Flexural strength of raw, 5%, 10% and 20% modified fiber reinforced composites

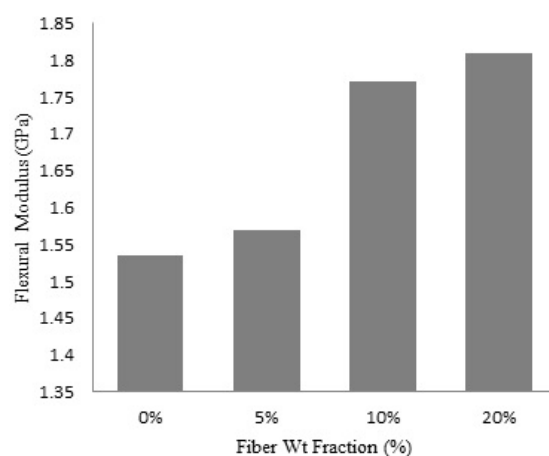


Fig. 9 Flexural modulus of raw, 5%, 10%, and 20% modified fiber reinforced composites

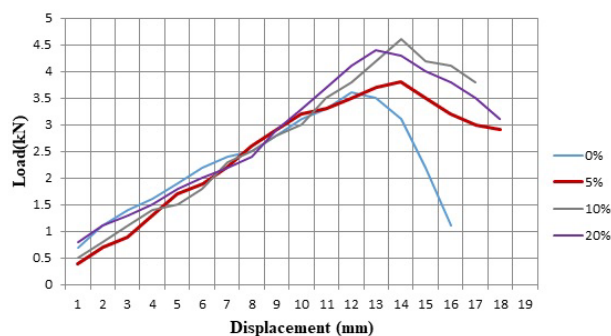


Fig. 10 Load-displacement curve (Bending Test) of raw, 5%, 10% and 20% modified fiber reinforced composites

V. CONCLUSIONS

The bidirectional jute fiber was treated chemically using glutaric anhydride by the process of retting under various concentrations to modify the chemical structure. The modified fibers were reinforced with epoxy matrix using hand molding process and the following conclusions were drawn:

- 1) In the case of glutarylation, all the hydroxyl groups were not bonded because of the reason that the chain length of the glutaric anhydride is larger which was confirmed by the observance of broad band in the range of 3300- 3100 cm^{-1} in the infrared spectroscopy. Also, the presence of hydroxyl group of the jute fiber has been confirmed with the strong peak at 3247 cm^{-1} .
- 2) The maximum weight gain was not observed during the process of glutarylation, still the stability in the process of weight gain has been confirmed. The stable weight of 31.03% was attained when the fiber is treated for 24 hours in the 20% glutaric anhydride solution. In the case of 10% solution, the stable weight of 31.21% has been attained at 24 hours.
- 3) The tensile strength of the composites was found to increase with increase in the glutaric anhydride weight fraction. The 10% modified jute fiber has shown good result on tensile testing. The reason for the drop in the tensile properties of 20% modified jute fiber reinforced epoxy composite is explained due to the fact that the fiber surfaces has been ruptured with increased weight fraction of glutaric anhydride.
- 4) The flexural properties too had shown similar variations as of tensile properties, where the maximum flexural properties have been attained, on reinforcing 10% modified jute fiber with epoxy matrix.

Based on the above results, considering various attributes and characteristics, we could conclude that jute fiber treated at 10% weight fraction of glutaric anhydride solution could be ideal for the purpose of reinforcement with epoxy matrix.

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