

# Synthesis and Physicochemical Study of Bisphenol-C-formaldehyde-toluene Diisocyanate Polyurethane–Jute and Jute–Rice Husk/Wheat Husk Composites

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**ABSTRACT:** Bisphenol-C-formaldehyde-toluene-2,4-diisocyanate polyurethane (PU) has been synthesized at room temperature and used for the fabrication of jute and jute–rice husk/wheat husk hybrid composites. PU–jute and PU–jute–RH/WH composites were prepared under pressure of 30.4 MPa at room temperature for 8 h, while PU–jute–RH/WH composites were prepared under same pressure at 110°C for 5 h. PU–jute composite has good tensile strength and flexural strength (50–53 MPa), while PU–jute–RH/WH hybrid composites have moderate tensile strength (9–11 MPa) and a fairly good flexural strength (15–31 MPa). Composites possess 1.1–2.2 kV electric strength and  $0.94\text{--}1.26 \times 10^{12}$  ohm cm volume resistivity. Water absorption in PU–jute composite is different in water (9.75%), 10% HCl (12.14%), and 10% NaCl (6.05%). Equilibrium water uptake time in

salt environment is observed 96 h, while in pure water and acidic environments it is 192 h. In boiling water equilibrium water content and equilibrium time are found to be 21.7% and 3 h, respectively. Water absorption increased 2.2 times in boiling water, whereas equilibrium time reduced 64 times. Thus, PU–jute composite has excellent hydrolytic stability against boiling water, 10% HCl, and 10% NaCl solutions. Fairly good mechanical and electrical properties and excellent hydrolytic stability of composites signify their usefulness for low cost housing units and in electrical and marine industries. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 101: 2363–2370, 2006

**Key words:** polyurethane; composites; mechanical and electrical properties; moisture uptake

## INTRODUCTION

Among reinforcing fibers, natural fibers<sup>1–5</sup> have gained their importance in making composites, especially for partial replacement of high cost synthetic fibers for low load-bearing applications. The incentives for producing the composites from natural fibers are their low cost, light weight, biodegradability, low density as compared to synthetic fibers. The low cost and moderate mechanical properties<sup>6,7</sup> of jute fibers make them suitable for composite fabrication. Various items such as school buildings,<sup>8</sup> food grain silos,<sup>8</sup> wood substitute, low cost housing units,<sup>9</sup> roofing,<sup>10</sup> pipes,<sup>11</sup> etc. have been made from jute fiber based composites.

Hybrid composites are materials made by combining two or more different types of fibers in a common matrix. They offer a range of properties that can not be obtained with a single kind of reinforcement. By careful selection of reinforcing fibers, the material cost can be substantially reduced.<sup>12</sup> Natural fiber composites are emerging as realistic alternatives to glass-reinforced

composites in many applications. Natural fibers are low cost, light weight, and apparently environmentally superior alternatives to glass fibers in composites.

Recently research is being directed in producing low cost composites by the use of biomass or agro waste in combination with jute. In India, rice husk (RH) and wheat husk (WH) are the biomass materials, which are byproducts from the crops. Many studies have been carried out on utilization of biomass such as particle board, medium density board, pulp, and composites.<sup>13–21</sup> In general, utilization of biomass in the fabrication of composites has several advantages such as greater deformability, low density, less abrasiveness to equipments, low cost, etc. However, in producing a good composite with biomass, the main obstacle to be resolved is the compatibility between the fiber, filler, and resin matrix.

Fiber-reinforced polymeric composites offer many potential advantages over the traditional construction materials namely steel and aluminum. The advantages of polymeric composites are low density, low thermal conductivity, excellent corrosion and chemical resistance, high strength to weight ratio, better design flexibility, cost effective, production of complex 3D structures, excellent fatigue and impact properties, improved acoustical performance, radar/sonar transparency, low maintenance, etc.

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PU have found their excellent applications in automotive parts, coatings, sealants, adhesives, and other infrastructure uses.<sup>22,23</sup> Nowadays polyurethanes (PUs) have found their versatile interest as composites because of increasing demand for light weight, durability, and cost effectiveness especially in automotive market.<sup>24</sup> PU-based composites are useful for seat frames, sun shades, door panels, package trays, and truck box panels.

To our knowledge no work has been reported on PU-based bisphenol-C-formaldehyde resin (BCF) and 2,4-toluene diisocyanate as a matrix material for natural fiber reinforced composites. The main objectives of the present investigation are synthesis of PU, preparation of PU-jute, PU-jute-RH/WH composites and their characterization and also moisture absorption study in different environments

## EXPERIMENTAL

### Materials

Solvents and chemicals used in the present study were of laboratory grade and purified by appropriate methods prior to their use.<sup>25</sup> 1,1'-Bis(4-hydroxy phenyl) cyclohexane (BC) and bisphenol-C-formaldehyde resin (BCF) were synthesized according to our previous work.<sup>26–28</sup> BC was repeatedly recrystallized from benzene and methanol–water systems, while BCF was purified by methanol–water system. Woven jute fibers [brown jute, *Corchorus capsularis*; 61–64% cellulose, 21–23% hemicellulose, and 13–14% lignin] used in the present study was collected from the “Indian Jute Industries Research Association” (IJIRA), Kolkata, India. The agro wastes rice husk [*Oryza sativa*] (RH, 2–3 mm size; 35% cellulose, 25% hemicellulose, and 20% lignin) and wheat husk [*Triticum aestivum*] (WH, 6–8 mm size; 33–39% cellulose, and 16–23% lignin) were obtained from the local market. The toluene diisocyanate (TDI) (2,4-TDI: 2,6-TDI, 80:20) was supplied by Narmada Chematur Petrochemicals Ltd. (NCPL), Bharuch, India.

### Preparation of BCF-TDI based PU-jute composites

To investigate mechanical and electrical properties of composites, 15 × 15 cm<sup>2</sup> sheets were prepared by hand lay-up technique. For moisture absorption study 5 × 5 cm<sup>2</sup> sheets were prepared and edges were sealed with the matrix material.

Thus, to a 250-mL beaker, 0.023 mol (9 g)/0.007 mol (3 g) BCF was dissolved in 100/30 mL MEK at room temperature. To this solution 0.155 mol (27 g)/0.051 mol (9 g) TDI in 50/20 mL MEK was added portion wise over a period of 10 min and stirred manually for about 15 min. The resultant PU solution [Figure 1, 62.6% of jute fibers] was applied to 15 × 15 cm<sup>2</sup> (57.5

g)/5 × 5 cm<sup>2</sup> (19 g) woven jute fibers with a smooth brush. The prepregs were allowed to dry in sunlight for about 15 min. Ten such prepregs were stacked one over the other and pressed under hydraulic pressure of 30.4 MPa for 8 h at room temperature. Silicon spray was used as a mold releasing agent. The edges of 5 × 5 cm<sup>2</sup> composites were sealed with the same PU solution for water absorption study. Here after composite is designated as PU-jute.

### Preparation of BCF-TDI based PU-jute-RH/WH hybrid composites

The hybrid composites of jute-RH and jute-WH were produced by one shot process. BCF-TDI PU resin used for the preparation of hybrid composites was prepared according to method described in previous section. The resin (70.6% of jute and biomass) was applied to two 15 × 15 cm<sup>2</sup> woven 11 g jute fibers (21.57%) with the help of a smooth brush and remaining resin was mixed with 40 g RH/WH (78.43%) at room temperature. In hybrid composites the ratio of biomass filler to jute was kept 3.64. Resin-impregnated jute sheets and RH/WH filler were allowed to dry at room temperature for about 15 min. Resin impregnated RH/WH filler was sandwiched between two resin impregnated jute sheets and subjected to hydraulic pressure of 30.4 MPa at 110°C for 2.5 h. Silicon spray was used as a mold releasing agent. Here after composites are designated as PU-jute-RH and PU-jute-WH.

### Measurements

The tensile strength (IS: 11,298-Pt-2-87) and flexural strength (ASTM D-790-92), electric strength (IEC-243-Pt-1-88), and volume resistivity (ASTM D-257-92) measurements were made on a Universal Tensile Testing Machine, Model No. 1185 at a speed of 50 mm/min, a high voltage tester (Automatic Electric-Mumbai) in air at 27°C by using 25/75 mm brass electrodes and a Hewlett-Packard high resistance meter at 500 V DC after charging for 60 s, respectively. Each test was carried out in duplicate and average value is used. Water absorption study of PU-jute composite was determined by a change in mass method. For this preweighed PU-jute samples were immersed in distilled water, 10% HCl, and 10% NaCl solutions at room temperature. Samples were periodically taken out from the solutions, wiped with tissue papers on both sides, reweighed, and reimmersed in the solutions. The process was carried out till equilibrium was reached. Water absorption study was also carried out in boiling water. For this a preweighed PU-jute sample was immersed in boiling water and change in mass was determined at the interval of 1 h till equilibrium reached.

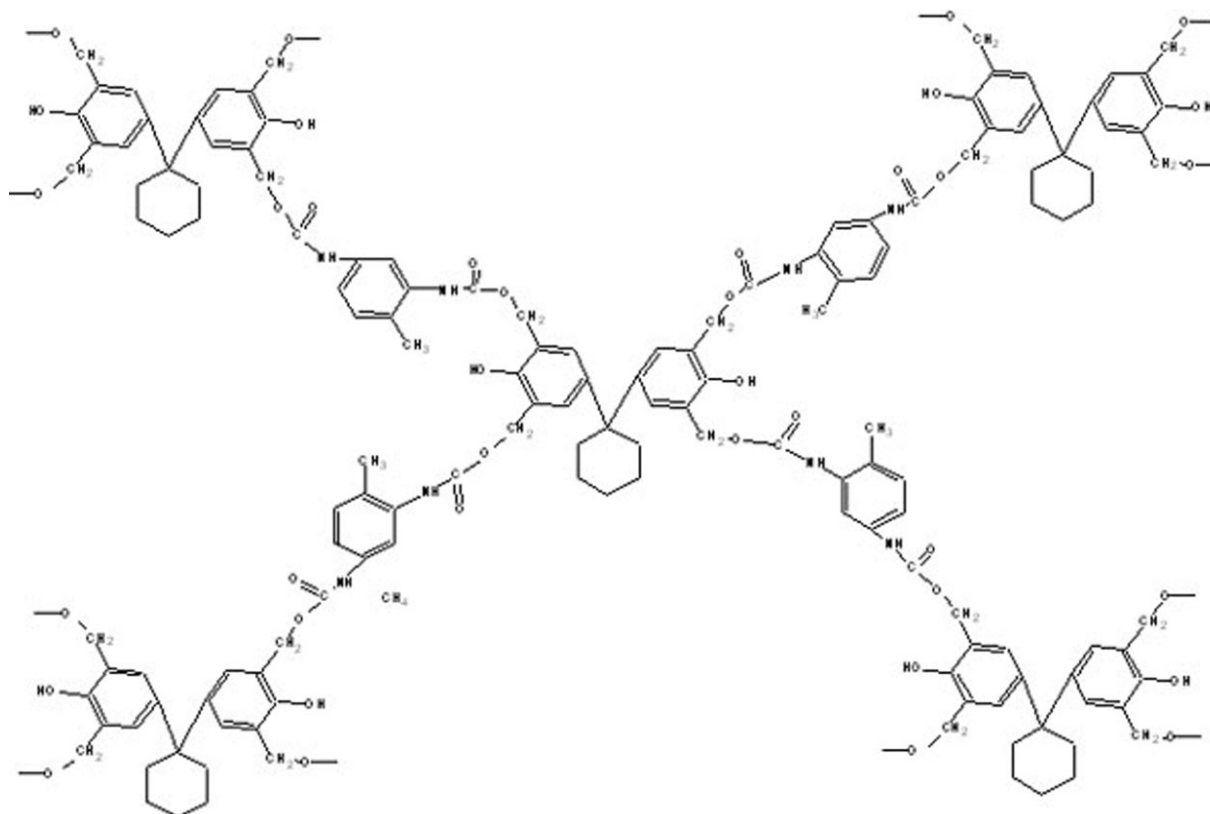


Figure 1 The structure of PU of BCF and TDI.

## RESULTS AND DISCUSSION

The mechanical performance and durability of composite materials are mainly governed by three factors<sup>29</sup> namely reinforcement, the matrix, and interfacial bond strength. Strength, stiffness, and stability of fibers and matrix are very important for long term service of composites. Natural fibers are hygroscopic and possess low wettability with hydrophobic matrix material and therefore it is necessary to concentrate on fiber modification, compatibility of resin, and coupling agents.

The mechanical properties of natural fiber composites are dependent upon several factors namely (i) fiber strength, modulus, fiber length, and orientation, (ii) interfacial bond strength, (iii) fillers, (iv) compatibilizers and impact modifiers, (v) fiber content, (vi) degree of crosslinking, and (vii) the presence of soft and hard segments, fiber loading.<sup>29–33</sup> Good interfacial bond strength is necessary for effective stress transfer from matrix to fiber in the composite. Lignocellulosic fibers have better resistance to weathering and moisture uptake.

Tensile properties of materials are most widely useful for engineering design and understanding quality characteristics of polymeric materials. In tensile testing, samples can be prepared by different ways namely molding, compression molding, and punch-

ing. In present investigation samples were prepared by punching technique as per prescribed standard tests for tensile and flexural testing. The tensile strength ( $\sigma$ ) of the composites was determined according to eq. (1):

$$\sigma = \frac{W}{A} \quad (1)$$

where  $W$  = the load value at break and  $A$  = original cross-sectional area of the sample. A comparative tensile strength of PU-jute (53 MPa), PU-jute-RH (11 MPa), and PU-jute-WH (9 MPa) composites is shown in Figure 2. Upon hybridization tensile strength of PU-jute-RH (79%) and PU-jute-WH (83%) has decreased to a great extent mainly because of random orientation of sandwiched biomass fillers supporting discontinuous stress transfer from matrix to fiber. There are two types of interfacial interactions namely covalent and H-bonding. Hydroxyl groups of lignocelluloses serve as reaction sites with various functional groups present in the resin. Thus, RH and WH both can be used as fillers as well as reactive components (polyols). No much change in tensile strength is observed between PU-jute-RH and PU-jute-WH.

Flexural strength is the resistance of material under the bending mode. Flexural properties are useful for

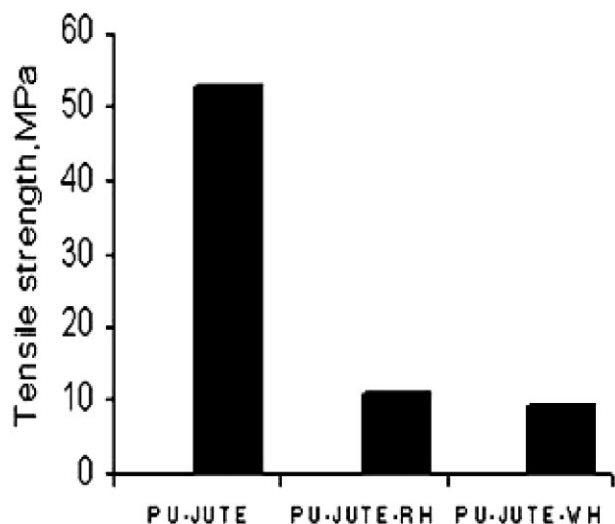


Figure 2 The comparative tensile strength of PU-jute, PU-jute-RH, and PU-jute-WH.

quality control and classification of materials with respect to bending strength and stiffness. They depend upon various factors namely (i) type and amount of additives, which can soften or reinforce the material, (ii) method of sample preparation e.g., molding or machining, (iii) temperature, (iv) surface roughness, sinks, voids, and other imperfections, (v) anisotropy, and (vi) accuracy in measured dimensions. Flexural strength of the composites was determined according to eq. (2):

$$\text{Flexural strength} = \frac{1.5FL}{wt^2} \quad (2)$$

where  $F$  = breaking load,  $L$  = span length (60–65 mm),  $w$  = sample width (mm), and  $t$  = sample thickness (mm)

A comparative flexural strength of PU-jute (50 MPa), PU-jute-RH (31 MPa), and PU-jute-WH (15 MPa) is shown in Figure 3. From Figure 3, it is evident that flexural strength of PU-jute-RH (38%) and PU-jute-WH (70%) has decreased considerably upon hybridization because of decrease in stiffness of the hybrid composites. In hybrid composites biomass fibers are not lined up in any direction as shown in Figure 4. They are just tangled mass. The composites can be made stronger by lining up all the fibers in the same direction. Oriented fibers are strong when pulled in fiber direction but they are weak at right angles to the fiber direction. The woven fibers give a composite good strength in many directions. Under tension, the strength of the composite is entirely due to reinforcement. The tensile strength (53 MPa) and flexural strength (50 MPa) of PU-jute are almost same but they decreased upon hybridization. The decrease in flexural strength is considerably lower than decrease in

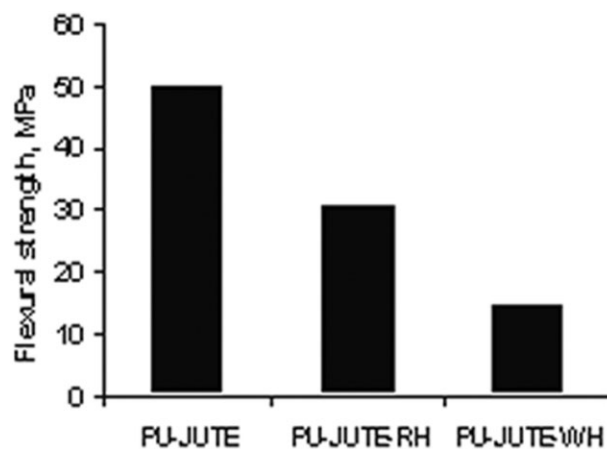


Figure 3 The comparative flexural strength of PU-jute, PU-jute-RH, and PU-jute-WH.

tensile strength. Thus, decrease in tensile strength and flexural strength of hybrid composites are due to random orientation of filler fibers, which are responsible for discontinuous stress transfer and decrease in strength and stiffness of PU. It is expected that the lone pairs of electrons of urethane linkages form H-bonding with OH groups of jute and RH/WH fibers and moreover unreacted TDI may form network structure with hydroxyl groups. The observed data indicate brittle nature of PU. In present case PU-jute possesses good tensile and flexural properties, but hybrid composites possess fairly good tensile and flexural properties indicating their usefulness as low load bearing applications especially for housing units and others.

### Electrical properties

Volume resistivity and electric strength data are very useful for comparing relative insulation quality of material selection, to evaluate the effects of material com-

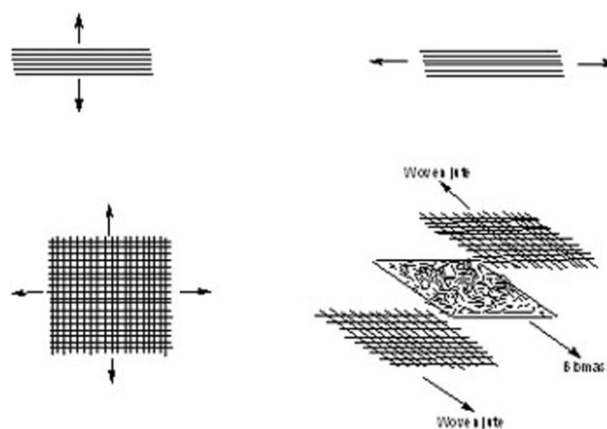


Figure 4 Arrangement of woven fibers and biomass fibers in composites.

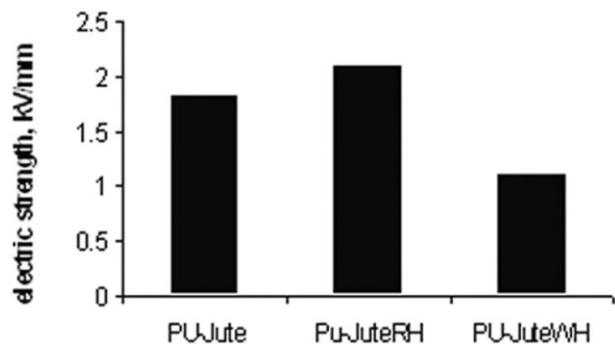


Figure 5 The comparative electric strength of PU-jute, PU-jute-RH, and PU-jute-WH.

position and environment and for material selection. They are useful to material scientists to design specific properties in combination. Electrical properties of the polymeric composites are affected by several factors namely (i) humidity, (ii) impurities, (iii) degree of resin cure, (iv) temperature, (v) nature of polymers, (vi) nature of fillers and additives, (vii) geometry, electrode area, and electrode material, (viii) sample thickness, (ix) time of voltage application, (x) current frequency, and (xi) extent of aging. Electric strength and volume resistivity of composites were determined according to eqs. (3) and (4), respectively:

$$\text{Electric strength} = \frac{V}{t} \quad (3)$$

where  $V$  = puncture voltage (volts) and  $t$  = sample thickness (mm)

$$\text{Volume resistivity} = \frac{R_v A}{l} \quad (4)$$

where  $R_v$  = volume resistance (ohms),  $A$  = area of electrodes ( $19.6 \text{ cm}^2$ ), and  $t$  = sample thickness (cm)

The comparative electric strength and volume resistivity data of PU-jute, PU-jute-RH, and PU-jute-WH composites are presented in Figures 5 and 6, respectively. From Figure 5, it is evident that the electric strength of PU-jute (1.82 kV/mm) and PU-jute-RH (2.1 kV/mm) is comparable, but for PU-jute-WH (1.11 kV/mm) it is almost half of later composites. Similarly, volume resistivity (Fig. 6) of PU-jute ( $9.38 \times 10^{11}$  ohm cm), PU-jute-RH ( $1.14 \times 10^{12}$  ohm cm), and PU-jute-WH ( $1.26 \times 10^{12}$  ohm cm) is comparable. Upon hybridization it is improved to 22–34%. The lone pairs of electrons of urethane linkages form H-bonds with unreacted OH groups resulting in neutralization of partial charges present on polar groups and hence increase in electric strength and volume resistivity. The interfacial bond strength is weaker in PU-jute-WH composite, which is also supported by com-

paratively low tensile and flexural properties of this composite. Thus composites possess fairly good electric strength and volume resistivity signifying their importance as insulating materials and can be used in electrical industries.

### Water absorption study

All polymers and composites absorb moisture in humid atmosphere and when immersed in water. Natural fibers absorb more water as compared to synthetic fibers. Jute and other natural fibers are highly hygroscopic and possess low wettability with hydrophobic resins mainly due to hydroxyl groups of cellulose, hemicellulose, and lignin.<sup>30,34–36</sup> Hemicellulose is mainly responsible for water absorption, although noncrystalline cellulose and lignin also play an important role in this process. The effect of absorbed water is to degrade tensile strength.<sup>37</sup> Absorbed water causes swelling of fibers and it continues till the cell walls are saturated with water. Beyond saturation point water exists as free water in the void structure leading to composite delamination or void formation.<sup>34,35</sup> Absorbed water leads to weakening of interface and accelerates delamination and hence decreases the strength of the composites.<sup>36</sup> Absorbed water causes hydrolytic degradation of both matrix and interface during service.<sup>38,39</sup>

Water absorption in polymeric composites is shown to be Fickian as well as non-Fickian in character.<sup>40–44</sup> The viscoelastic nature of polymers and cracks are responsible for non-Fickian diffusion. Water absorption in composites depend upon fraction of fibers, void volume, additives, humidity, and temperature. In present case assuming one dimensional Fickian diffusion water absorption by PU-jute in pure water, 10% HCl, and 10% NaCl is carried out at room temperature by change in weight method. The study is also carried out in boiling water.

The percent water absorbed by PU-jute in different environments (water, 10% HCl, and 10% NaCl) at

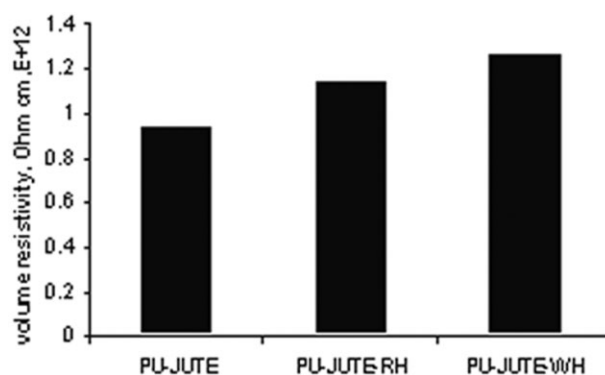
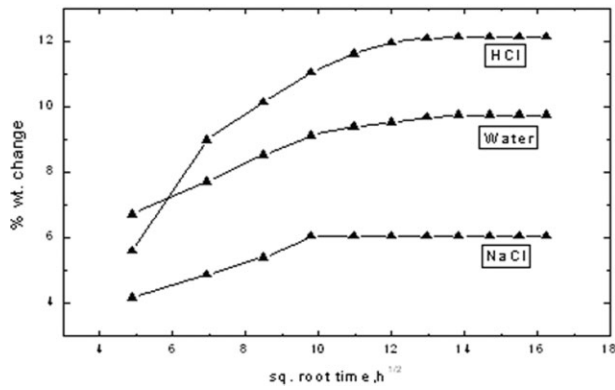


Figure 6 The comparative volume resistivity of PU-jute, -jute-RH, and PU-jute-WH.



**Figure 7** The plots of percent weight change against square root of time for PU-jute in different environments at room temperature.

room temperature with the passage of time is presented in Figure 7. From Figure 7, it is evident that water absorption behavior is quite different in different environments supporting different diffusion behavior. Equilibrium water content and equilibrium time data are reported in Table I. From Table I, it is clear that equilibrium time (96 h) and equilibrium water content (6.05%) in salt environment are almost half of water (192 h and 9.75%) and acid (192 h and 12.14%) environments. The observed trend in absorbed water in PU-jute is HCl > water > NaCl. Water penetration into composite is conducted mainly by diffusion mechanism. The penetration of water in composites also occurs through capillary flow along the fiber/matrix interface followed by diffusion from the interface in to the bulk resin and transported by micro cracks. Absorbed water may induce irreversible changes like chemical degradation, cracking, and debonding. Cracking and blistering cause high water absorption, while leaching of small molecules results in decrease in weight.<sup>45</sup>

### Diffusivity

Various models have been proposed for water absorption in composites. It is well established that absorbed water in polymers and composites plays a significant role in mechanical behavior and long term durability. Assuming one-dimensional Fickian diffusion in composite, an attempt has been made to determine diffusivity ( $D_x$ ) in different environments. Diffusivity is assumed to depend only on temperature and independent of the moisture content as well as stress levels in composites. For one-dimensional water absorption in semi-infinite plate exposed on both sides to same environment, the amount of water absorbed<sup>46</sup> is given by eq. (5):

$$G = 1 - \frac{8}{\pi^2} \sum_{j=0}^{\infty} \frac{1}{(2j+1)^2} \exp\left[-\frac{(2j+1)^2 \pi^2 D_x t}{h^2}\right] \quad (5)$$

where  $D_x$  = diffusivity,  $t$  = time (second), and  $h$  = sample thickness (m)

The water content in the sample can be determined according to eq. (6):

$$M = \frac{W_m - W_d}{W_d} \times 100 \quad (6)$$

where  $M$  = % water absorbed,  $W_m$  = weight of moist sample, and  $W_d$  = weight of dry sample. The solution of diffusion equation in terms of percent water absorption is given by eq. (7):

$$M = \frac{4M_m}{h} \sqrt{\frac{t}{\pi}} \sqrt{D_x} \quad (7)$$

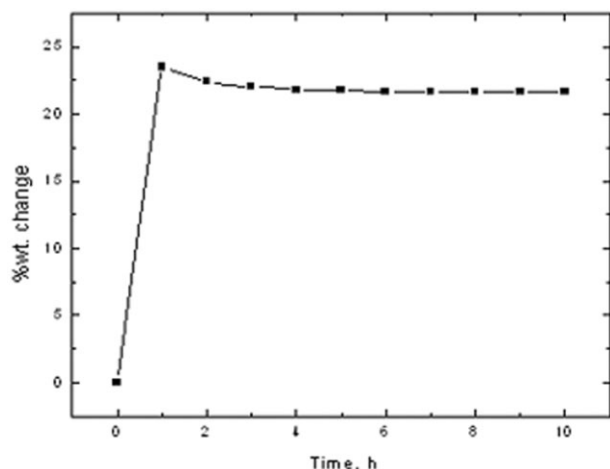
Where  $M_m$  = equilibrium water content. Diffusivity in a given environment can be determined from the initial slope of the plot of %M against  $\sqrt{t}$  (Fig. 7) according to eq. (8):

$$D_x = \pi \left( \frac{h}{4M_m} \right)^2 (\text{slope})^2 \quad (8)$$

The diffusivity in PU-jute composite in water, 10% HCl, and 10% NaCl solutions was determined using Figure 7 and eq. (8), and it is reported in Table I. From Table I, it is clear that diffusivity is different in different environments. The diffusivity is 2.64 times higher in salt solution and 5.62 times in acidic solution. The presence of electrolyte in water breaks water structure and hence increases diffusivity. High diffusivity in acid solution supports more structure breaking tendency as compared to salt solution. It is expected that the diffusion of hydrated ions will be more as compared to polymeric and clustered water molecules. Moreover, hydrated ions also undergo surface solvolysis because of the presence of polar groups in composite, which are responsible for high water uptake. The low water content in salt solution might be due to electrostatic repulsive forces acting among electronegative groups present in PU-jute composite. Ionic size has also affected water structure in diffusion process.  $H^+$  ions have high tendency to break water structure as compared to  $Na^+$  ions. The size of the hydrated  $Na^+$  ions is greater than that of  $H_3O^+$  ions leading to

**TABLE I**  
Water Uptake and Diffusivity Data of PU-Jute Composite at Room Temperature

| Properties                                   | Water | 10% NaCl | 10% HCl |
|--|-------|----------|---------|
| Equilibrium time (h)                         | 192   | 96       | 192     |
| Equilibrium water content (%)                | 9.75  | 6.05     | 12.14   |
| Diffusivity $D_x$ ( $10^{-12}$ ) ( $m^2/s$ ) | 0.47  | 1.24     | 2.64    |



**Figure 8** The plot of percent weight change against time for PU-jute in boiling water

low diffusivity. Thus, smaller is the size greater is the diffusivity.

### Water absorption in boiling water

Water absorption in composites mainly depends on temperature and it is independent on water content. The effect of boiling water on water absorption in PU-jute composite with time is shown in Figure 8. From Figure 8, it is evident that water absorption is maximum (23.5%) within an hour and then it decreased to 21.7% and reached an equilibrium within 3 h. The decrease in water uptake after 1 h is due to leaching of small components. From Table I and Figure 8, it is evident that equilibrium water absorption in boiling water is increased two times, but equilibrium time is reduced 64 times without damaging PU-jute composite supporting excellent hydrolytic stability of PU-jute composite, which indicates its usefulness in humid environment.

### CONCLUSIONS

1. The tensile and flexural properties of PU-jute-RH and PU-jute-WH composites have decreased due to random orientation of filler fibers and decrease in strength and stiffness of hybrid composites.
2. Electric strength except PU-jute-WH and volume resistivity of hybrid composites are improved.
3. Equilibrium time (96 h) and equilibrium water content (6.05%) in salt solution are almost half of water (192 h and 9.75%) and acid (192 h and 12.14%) environments.
4. Diffusivity is 2.64 times higher in salt solution and 5.62 times in acidic solution. The acid and

salt have tendency of breaking water structure and increased the diffusivity.

5. PU-jute possesses excellent hydrolytic stability in water, acid, and salt solutions and even in boiling water. In boiling water, water absorption increased two times, while equilibrium time shortens 64 times.
6. Fairly good mechanical and electrical properties and excellent hydrolytic stability of composites signify their usefulness for low load bearing applications in construction industry and marine industry.

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