# Studies on Jute/Glass/Hybrid Composites of Polyurethane Based on Epoxy Resin of 9,9′-bis(4-hydroxy phenyl) Anthrone-10(EBAN) and PEG-200

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**ABSTRACT:** EBAN-PEG-200-TDI-Jute (EANPT-J), EBAN-PEG-200-TDI-Glass (EANPT-G), and EBAN-PEG-200-TDI-Glass-Jute-Glass (EANPT-GJG) composites have been prepared by hand lay up technique at 80°C under 15.4 MPa pressure for 2 h. EANPT-J, EANPT-G and EANPT-GJG possess 33–52 MPa tensile strength, 19–46 MPa flexural strength, 1.3–3.2 kV/mm electric strength and $3 \times 10^{11}$–$5 \times 10^{11}$ Ohm cm volume resistivity, respectively. Hydrolytic stability of the composites was tested against water, 10% aq. HCl and NaCl solutions at 35°C and also in boiling water. The percentage of water uptake, equilibrium time, and diffusivity of the composites have been determined and discussed their possible applications. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 118: 1469–1475, 2010

**Key words:** composites; tensile and flexural strengths; dielectric strength; volume resistivity; water absorption; diffusivity

## INTRODUCTION

The use of polymeric composites is common in industries. The most significant advantages of using polymeric composites of the machine design are that they are lighter, more resistant to chemical agents, easier to shape, and cheaper when compared with metals.[1] Fiber–reinforced polymer matrix composites are gaining potential applications in structural and nonstructural areas because of having their interesting properties like high specific stiffness and strength, good fatigue performance and damage tolerance, corrosion resistance, low thermal expansion, nonmagnetic properties, and low energy consumption during fabrication.[2–4] Natural fibers are gaining potential application as reinforcement because they are available easily, biodegradable, low-cost materials with attractive properties like low abrasive nature, which is beneficial in sense of processing techniques and recycling, etc. Besides having these advantages there are also some bottlenecks like poor compatibility with the hydrophobic polymer matrix, the tendency to form aggregates during processing and the low resistance to moisture related to these fibers greatly reduce the potential of natural fibers to be used as reinforcement for polymer matrix composites.[5] The most important problem is the fiber-matrix adhesion because load is transferred to stiff fibers through shear stresses at the interface and it requires a good bond between the polymeric matrix and the fibers. Because of the presence of pendant hydroxyl and polar groups in various constituents of the natural fibers resulting in poor wettability and also moisture absorption of the fibers is too high, giving rise to poor interfacial bonding with the hydrophobic matrix polymers.

Hybrid composites can be prepared by combining two or more different types of fibers in a common matrix. They offer a range of properties that can not be achieved with a single kind of reinforcement. With careful selection of reinforcing fibers, the cost of materials can be reduced substantially. The mechanical properties of the composites are dependent on the properties of the matrix; the amount, type, and arrangement of fibers within the composites; as well as on the interactions between matrix and reinforcing agent.

Epoxy resin is one of the important thermosetting resins with many desirable properties and is, therefore, widely used in various composites as a matrix. Because of having polar hydroxyl groups, the composites based on epoxy resin absorb moisture easily, resulting in the reduction of both mechanical and dielectric properties. Therefore, epoxy resin has to be modified. Polyurethanes based on diisocyanates and epoxy based polyols have been found their versatile applications as coatings, adhesives, filling compounds of high heat resistance, and chemical stability against aggressive agents.[6,7]

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To our knowledge no work has been reported on glass, jute, and their hybrid composites of 9,9′-bis(4-hydroxy phenyl) anthrone-10 based epoxy polyurethane resin. This work deals with the investigation of mechanical, electrical, and water absorption behavior of glass, jute, and glass-jute-glass composites of epoxy resin of 9,9′-bis(4-hydroxy phenyl) anthrone-10 and PEG-200 polyurethane resin(I).



**(I)**
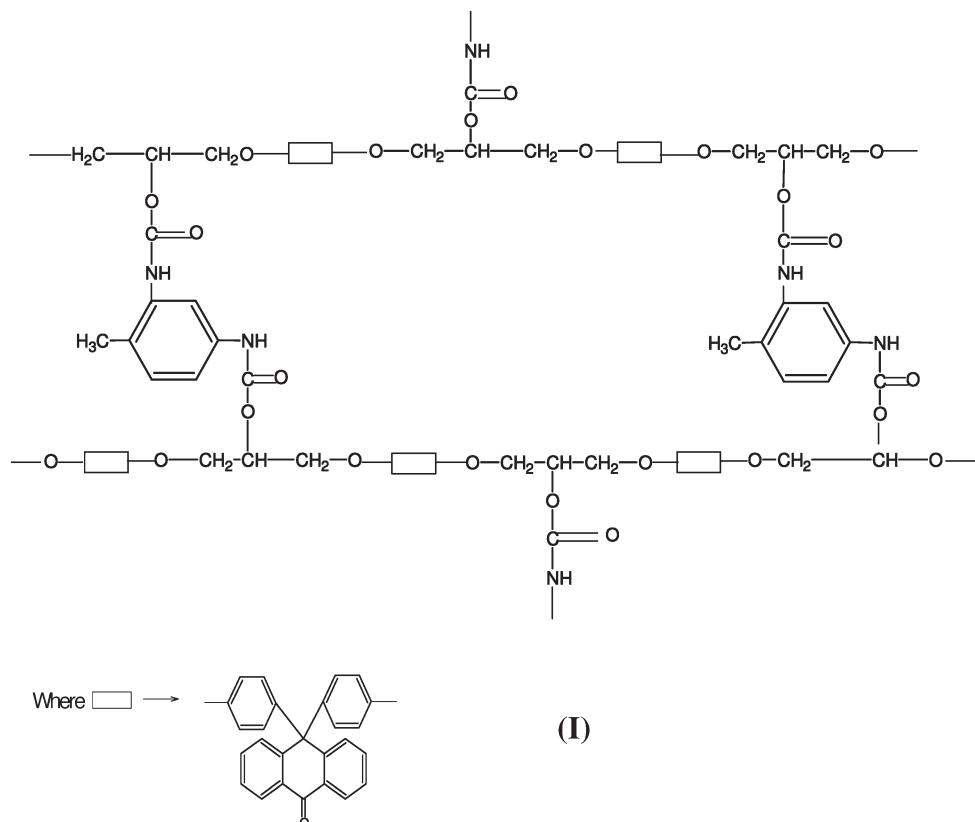
## EXPERIMENTAL

### Materials

Solvents and chemicals used were of laboratory grade and purified before use.[8] Woven jute fabric (Brown jute, *Corchorus capsularis*) used in this study was collected from local market. Silane treated E-glass fabric (7 mil) (Unnati Chemicals, India) was used for composite purpose. The toluene diisocyanate (2, 4-TDI: 2, 6-TDI, 80 : 20) was supplied by Narmada Chematur Petrochemicals, Bharuch, India. Polyethylene glycol (PEG-200) was supplied by Sd Fine-Chem, Mumbai.

### Synthesis of epoxy resin of BAN

The epoxy resin of 9,9′-bis(4-hydroxy phenyl) anthrone-10 (EBAN) was synthesized by condensing 0.01 mol BAN with 0.23 mol epichlorohydrin by using isopropanol (200 mL) as a solvent and sodium hydroxide (0.2 mol in 40 mL water) as a catalyst. The reaction mixture was stirred at reflux temperature for 4 h. The excess solvent was distilled off and the solid

resin was isolated from distilled water, filtered, washed well with water, and finally, with cold methanol and dried at room temperature. The resin was purified by extracting in chloroform and evaporating to dryness. The yield was 80%. Here after resin is designated as EBAN. The resin is soluble in chloroform, acetone, dimethyl formamide, THF, etc. The epoxy equivalent of BAN was determined by pyridine-pyridinium chloride method. The average of three measurements is 952.4.

### Preparation of composites

Glass, jute and their hybrid composites (15 × 15 cm) of EBAN and PEG-200 polyurethane resin were fabricated by hand layup technique. EBAN used for composite preparation was 50% of jute/glass fabrics. 2, 4-Toluene diisocyanate (TDI) and PEG-200 used were 30% of EBAN. The detail of fibers, EBAN, PEG-200, and TDI is described in Table I.

Thus, to a 250 mL beaker, required quantity of EBAN was dissolved in 90 mL chloroform at room temperature. To this solution required quantity of

TABLE I
Fiber-matrix, TDI, and PEG-200 Compositions for Epoxy Jute, Glass and Their
Hybrid Composites

| Type of Composite | Wt. of fabrics (g) | Wt. of resin (50% of fabrics) (g) | Wt. of TDI (30% of resin) (g) | Wt. of PEG-200 (30% of resin) (g) |
|---|---|---|---|---|
| EANPT-J | 82 | 41 | 12.3 | 12.3 |
| EANPT-G | 58 | 29 | 8.7 | 8.7 |
| EANPT-GJG | (22 + 42) | 32 | 9.7 | 9.7 |

TDI was dissolved in 15 mL chloroform and was added dropwise with stirring over a period of 10 min. To this reaction mixture required quantity of PEG-200 was added dropwise with stirring. The resultant reaction mixture was stirred for 20 min at room temperature and applied to jute/glass fabrics with the help of a smooth brush and prepregs were allowed to dry in the sunlight for about 15 min. Ten such prepregs were stacked one over the other and pressed between the two preheated stainless steel plates under the hydraulic pressure of 15.4 MPa at 80°C for 2 h and 12 h at room temperature. For hybrid composite the prepregs were stacked alternatively, i.e., glass-jute-glass. Silicone spray was used as a mold releasing agent. Here after composites are designated as EANPT-J, EANPT-G and EANPT-GJG. Similarly for water absorption study 3 × 3 cm composites were prepared and edges were sealed with the same matrix material.

## MEASUREMENTS

Tensile strength (IS6748:1994) measurements were made on a Shanta Engineering Tensile Testing Machine, Model No. SET/T/50 at a speed of 50 mm/min. Dielectric strength (IEC-60243-Pt-1–1998), and volume resistivity (ASTM D-257–99) measurements were made on a Universal 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. Water absorption study was carried out at 35°C in water, 10% aq. HCl and 10% aq. NaCl and in boiling water according to reported method.[9–11]

## RESULTS AND DISCUSSION

### Mechanical properties

Comparative tensile strength and flexural strength of EANPT-J, EANPT-G and EANPT-GJG are reported in Figures 1 and 2, respectively. From Figure 1, it is clear that tensile strength of EANPT-GJG (49 MPa) is intermediate of EANPT-J, (33 MPa) and EANPT-G (52 MPa). As glass fibers (1200–1800 MPa) is much stronger than that of jute fibers (300–700MPa). From
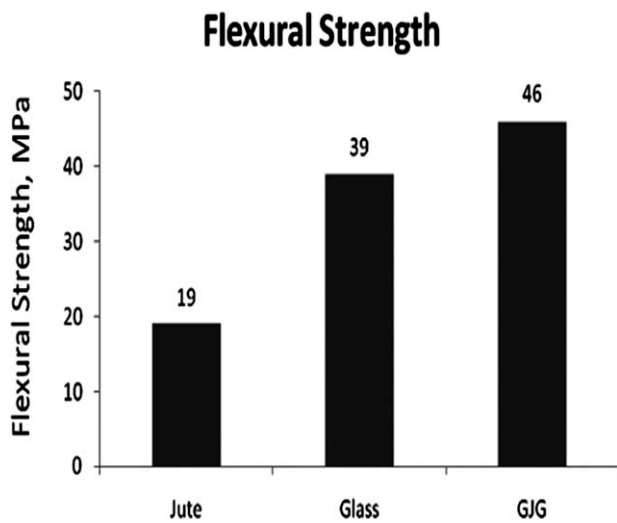
Figure 2, it is observed that flexural strength is improved to a considerable extent. Flexural strength of EANPT-GJG is improved by 142 and 18% when compared with EANPT-J and EANPT-G, respectively, because of strong and stiffer nature of glass fibers. Thus, moderate tensile and flexural properties of the composites signify their importance for low load bearing applications, especially for housing units. The mechanical properties of the composites depend on number of factors namely fiber content, fiber orientation, nature of fibers, fiber modification, interfacial bonding, additives like fillers, compatibilizer and impact modifier, humidity and mode of testing.[12]

### Electrical properties

Electrical properties of materials are useful to designers in predicting the relative insulation quality characteristic of material selection for specific properties in combination, to access the effects of material composition and environment. Comparative electric strength and volume resistivity of EANPT-J, EANPT-G, and EANPT-GJG are presented in Figures 3 and 4, respectively, from which it is clear that
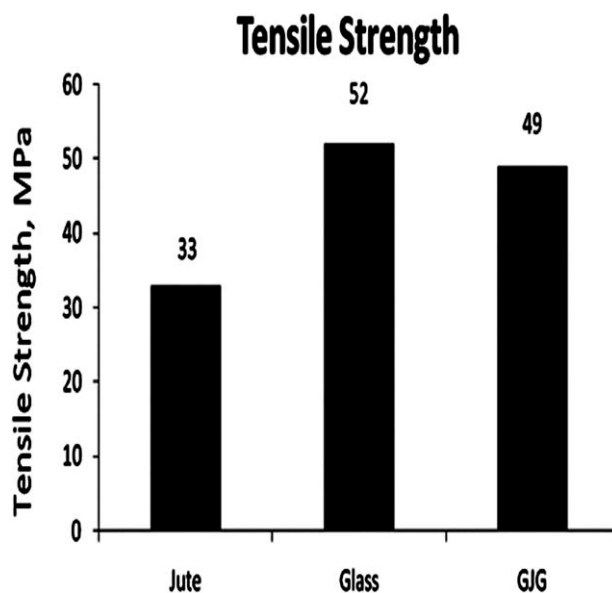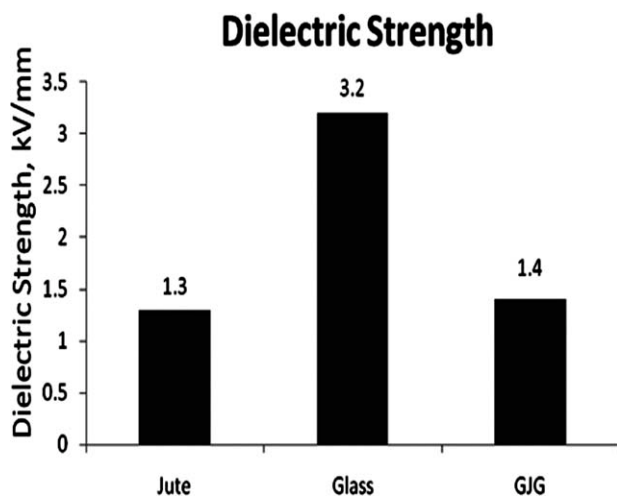


**Figure 1** A comparative tensile strength of EANPT-J, EANPT-G, and EANPT-GJG.

## Flexural Strength

**Figure 2** A comparative flexural strength of EANPT-J, EANPT-G, and EANPT-GJG.

practically no change in dielectric strength is observed but volume resistivity has decreased considerably upon hybridization when compared with EANPT-J (53%) and EANPT-G (71%). Relatively low volume resistivity of the composites is due to the presence of polar groups in fibers and polyurethane resin. Low volume resistivity of EANPT-J ($3 \times 10^{11}$ Ω cm) when compared with EANPT-G ($5 \times 10^{11}$ Ω cm) is due to polar nature of untreated jute fiber. Upon hybridization volume resistivity of EANPT-GJG ($1.4 \times 10^{11}$ Ω cm) is decreased as a consequence of charge accumulation between glass and jute layers. This charge accumulation is mainly because of free hydroxyl and urethane groups.

Electrical properties of the composites rely on area and geometry of the electrodes, sample thickness, temperature, humidity, time of voltage application, extent of aging, frequency of current, impurities, degree of resin cure, nature of reinforcing agents,

## Dielectric Strength

**Figure 3** A comparative dielectric strength of EANPT-J, EANPT-G, and EANPT-GJG.

## Volume Resistivity
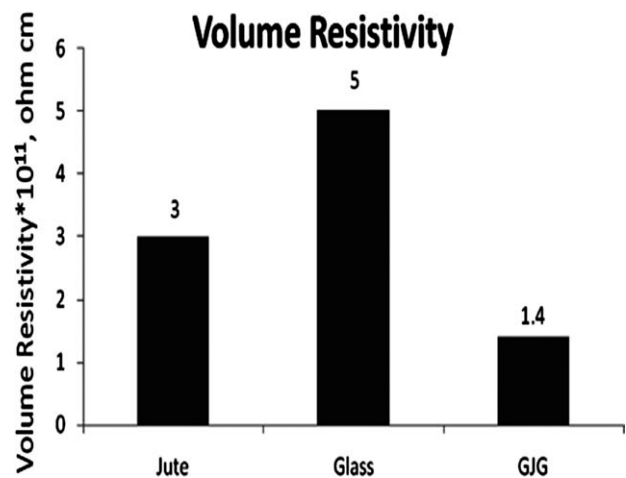
**Figure 4** A comparative volume resistivity of EANPT-J, EANPT-G, and EANPT-GJG.

etc. Moderate electric strength and volume resistivity of the composites may find their applications in electrical and electronic fields.

### Water absorption study

All polymers and composites absorb moisture in humid atmosphere and when immersed in water. Natural fibers absorb more water when compared with synthetic fibers. Jute and other natural fibers are highly hygroscopic and possess low wettability with hydrophobic resins mainly because of hydroxyl groups of cellulose, hemicellulose, and lignin.[13–16]

The percentage weight gained by the composites in water, 10% aq. HCl and 10% aq. NaCl solutions with the passage of time ($t^{1/2}$) is shown in Figures 5–7. The percentage weight gained by each composite increased, reached maximum and then practically remained constant when equilibrium was established in each of the environment. The equilibrium time and equilibrium water content for composites in water, 10% aq. NaCl, 10% aq. HCl environments are recorded in Table II. The observed trend in percentage equilibrium water content for EANPT-J, EANPT-G, and EANPT-GJG is HCl > $H_2O$ > NaCl, respectively. High equilibrium water content of EANPT-J is due to presence of hydrophilic OH groups in jute and EBAN and to some extent because of surface solvolysis. Glass composite showed a little water uptake tendency (6.46% in water, 9.67% in HCl and 4.89% in 10% NaCl) over a period of 96 h because of silane treatment of glass fibers.[17] For hybrid composite it is intermediate of EANPT-J and EANPT-G.

### Diffusivity (Dx)

With the consideration that absorbed water plays a significant role in influencing mechanical behavior, long-term durability of the polymer and polymer
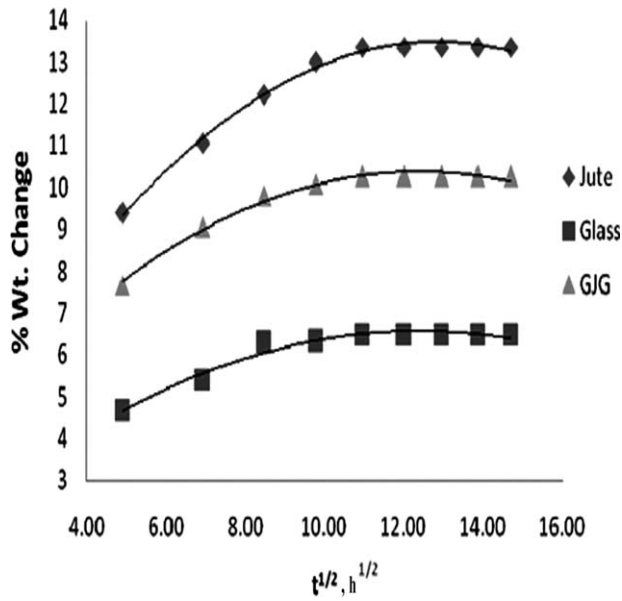
**Figure 5** The plots of percent weight gain against $t^{1/2}$ for EANPT-J, EANPT-G, and EANPT-GJG composites in water at 35°C.
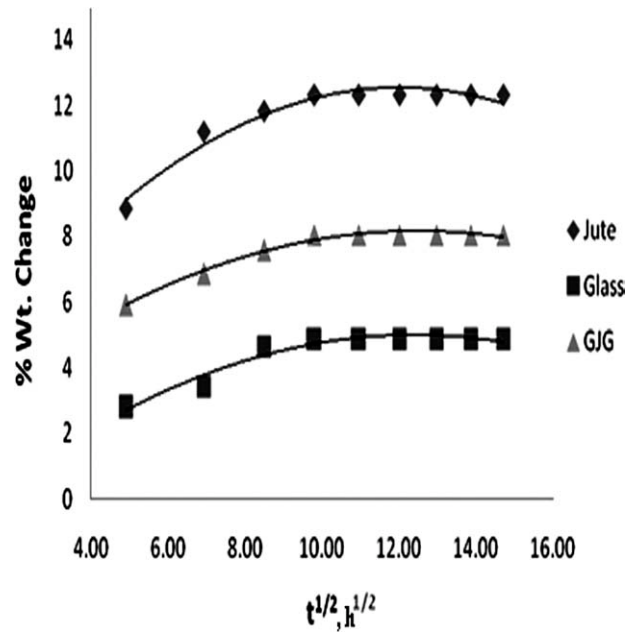
matrix composites. Water absorption in composites is proved to be Fickian as well as non-Fickian in character.[18] We have carried out water absorption study in different chemical environments by assuming one-dimensional diffusion only. For one-dimensional moisture absorption in semi-infinite plate exposed on



**Figure 6** The plots of percent weight gain against $t^{1/2}$ for EANPT-J, EANPT-G, and EANPT-GJG composites in 10% aq. HCl at 35°C.



**Figure 7** The plots of percent weight gain against $t^{1/2}$ for EANPT-J, EANPT-G, and EANPT-GJG composites in 10% aq. NaCl at 35°C.

both sides to same water environment, the total amount of water absorption is given by eq. (1):

$$\% \, M_t = \frac{W_m - W_d}{W_d} \times 100 \tag{1}$$

where $M_t$ = % water content at time $t$, $W_m$ = weight of moist material and $W_d$ = weight of dry material.

Diffusivity is related with water uptake with the passage of time according to eq. (2):

$$M = \frac{4M_m}{h} \sqrt{\frac{t}{\pi}} \sqrt{D_x} \tag{2}$$

where $M_m$ = equilibrium water content in the sample, $D_x$ = diffusivity and $t$ = time.

Diffusivity in a given environment can be determined from initial slope of the plot of M against square root of time:

$$D_x = \pi \left( \frac{h}{4M_m} \right)^2 (\text{slope})^2 \tag{3}$$

Diffusivity in different environments was determined according to eqs. (2) and (3) and is reported in Table II from which it is clear that diffusivity in EANPT-GJG is intermediate of jute and glass composites. Observed diffusivity order is NaCl > H₂O > HCl. Water absorption tendency of jute composite is more than that of glass composite because of the presence of hydrophilic hydroxyl groups in jute fibers. The presence of strong electrolytes in water affects water structure and hence diffusivity. The bigger the size of the solvated ions small is the
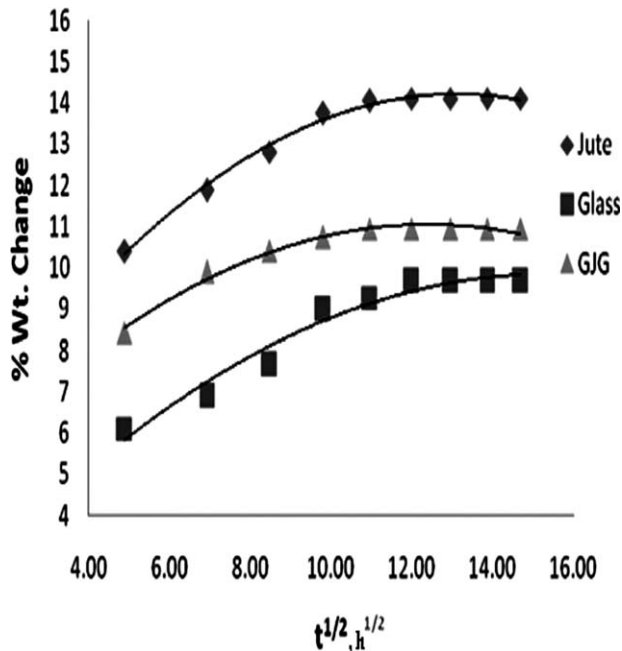
**Equilibrium Water Content and Diffusivity Data of Epoxy Jute, Glass and Their Hybrid Composites in Different Environments at 35°C and in Boiling Water**

| Composite | % Equilibrium water content at 35 °C | | | Equilibrium time, h | | | Diffusivity $(D_x)$, $10^{-11}$, $m^2/s$ | | | % Equilibrium water content in boiling water |
|---|---|---|---|---|---|---|---|---|---|---|
| | $H_2O$ | 10% aq. NaCl | 10% aq. HCl | $H_2O$ | 10% aq. NaCl | 10% aq. HCl | $H_2O$ | 10% aq. NaCl | 10% aq. HCl | |
| EANPT-J | 13.4 | 12.3 | 14.1 | 120 | 96 | 144 | 2.4 | 2.7 | 1.7 | 16.1 |
| EANPT-G | 6.5 | 4.9 | 9.7 | 96 | 96 | 120 | 0.2 | 0.5 | 0.2 | 9.2 |
| EANPT-GJG | 10.3 | 8.1 | 10.9 | 120 | 96 | 120 | 0.7 | 0.8 | 0.6 | 13.4 |

diffusivity. Thus, NaCl is found to break more water structure and hence high diffusivity. It is expected that the diffusion of hydrated ions will be more when compared with 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.[19] Absorption of water in composites causes swelling of fibers till the cell walls are saturated with water and beyond that water exists as free water in the void structure leading to composites delamination or void formation. Absorbed water causes weakening of interfacial adhesion and hydrolytic degradation of both matrix and fibers.[16,20,21] Cracking and blistering of fibers cause

high water absorption, while degradation causes leaching of small molecules.[22]

## Water absorption in boiling water

Water absorption in composites mainly depends on temperature besides presence of hydrophilic groups in composites. The effect of boiling water on pecentage of water absorption with time is shown in Figure 8 from which it is evident that the water absorption in composites is the maximum after 6 h and remained almost constant (Table II). From Table II and Figure 8, it is evident that equilibrium water absorption time is reduced drastically without any damage to the composites. Thus, composites possess excellent hydrolytic stability against boiling water and even in harsh acidic and saline environments.
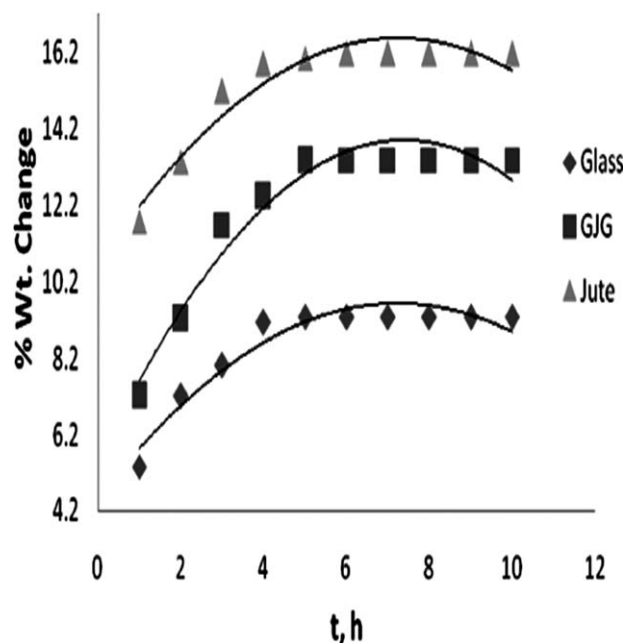
## CONCLUSIONS

Composites possess fairly good mechanical and electrical properties and excellent hydrolytic stability against harsh environmental conditions which signify their usage as low load bearing housing units and in electrical and electronics and in marine field.

**Figure 8** The plots of percent weight gain against time for EANPT-J, EANPT-G, and EANPT-GJG composites in boiling water.

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