

## CAN THE OUTDOOR PROPERTIES OF NATURAL FIBER REINFORCED BIO-BASED COMPOSITES BE IMPROVED?

S. K. Ramamoorthy\*, Q. Di, K. Adekunle, M. Skrifvars

*School of Engineering, Polymer Technology, University of Borås, Sweden*

*\*Correspondence: sunil.kumar@hb.se*

**Keywords:** Natural fiber, Regenerated cellulose fiber, Composite, Acrylated epoxidized soybean oil.

### **Abstract**

*Natural fiber composites are known to absorb more moisture than glass or carbon fiber reinforced composites. The hybrid natural fiber composites prepared in this study have relatively less moisture absorption than natural fiber composites composed of a single fiber type. The composite laminates were manufactured by compression molding technique. A bio-based resin known as acrylated epoxidized soybean oil (AESO) was used as a matrix, while jute fiber, regenerated cellulose fiber and glass fiber were used as reinforcements. The alkali treatment effect of fibers on composite properties was investigated. The amount of water absorbed by the composites was also studied. To see the influence of water absorption on mechanical properties, specimens were immersed in distilled water before tensile and flexural testing. Impact testing was performed on the composite laminates in order to calculate the energy absorbed by specimen during fracture.*

### **1 Introduction**

#### *1.1 Description and Analysis*

Several environmental concerns like emission, biodegradability etc. poses threat to the living conditions. Due to this reason, synthetic materials are being replaced by biocomposites in various applications. Intense research is going on to produce lighter weight and more environmental friendly materials without compromising the strength. Cost of these materials has always been a hurdle for the producers to attract customers. Producing lighter materials for the automotive applications will reduce the fuel consumption. Several factors like these are taken into consideration while producing materials for different application.

Many studies have been conducted to investigate natural fiber reinforced composites made from annual plant fibers like flax, hemp, sisal etc. The large moisture absorption for the natural fibers is one of the several reasons which make them less attractive in composite production, [1] [2]. Regenerated cellulose fibers have also been used as reinforcement to produce environmentally friendly composites with good mechanical properties. The matrix resin, which acts as binding component, is commonly of synthetic origin Triglycerides or esters of glycerol and fatty acids, from plant oils have been extensively investigated as a biomass origin.

In this project, one natural fiber (jute) and two regenerated cellulose fibers (Lyocell and viscose) were used as reinforcements while a triglyceride from soybean oil was used as matrix. Some advantages like lightweight, cost effectiveness, renewable, non-abrasive process conditions etc. of jute fibers are of interest in structural composites, however jute fiber absorbs easily water [3]. This hydrophilic property of the jute fiber will induce the composites to absorb water, which restricts in their use in outdoor applications as it reduces the interfacial bonding between fiber and matrix. However, this could be reduced on pre-treatments. One of the most abundant resources on earth is cellulose, considering this fact; regenerated cellulose fibers were used as reinforcements. Nowadays, Lyocell fiber is produced in rather environmentally friendly way by direct dissolution of cellulose in N-methylmorpholine N-oxide (NMMO) [4] [5]. Lyocell has outstanding properties; stronger than any other cellulose fibers when wet, very stable in washing and drying, good thermal stability which makes it suitable to be used in hybrid composites [6] [7].

Biobased plant triglycerides thermoset resins have many applications including automotives, aerospace, marine etc., [8] [9] [10]. Several plants like linseed and rapeseed are good source of triglycerides, which could be used as matrix in composite preparation [11]. Various modifications are done to the triglycerides for it to be used as matrix, most common is epoxidation. In this study, epoxidized soybean oil is chemically modified by acrylic groups, to obtain resin an acrylated epoxidized soybean oil (AESO) resin. [12] [13]. Several other chemical modifications could also be used, such as methacrylated soybean oil (MSO) and methacrylic anhydride-modified soybean oil (MMSO) [14]. Recent developments in these resins have economic and environmental advantages.

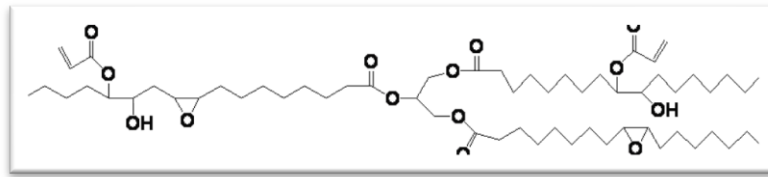
### *1.2 Results and Conclusions*

Jute fiber composites absorbed more water than Lyocell and glass fiber reinforced composites. Inclusion of Lyocell or glass in jute fiber composites reduced the water uptake. Although water absorption reduced the mechanical properties, hybridization of jute fiber composites improved the mechanical properties before and after water uptake. Alkali treatment of Lyocell was not necessary if the objective is to reduce the water absorption.

## **2 Materials and testing methods**

### *2.1 Materials*

Lyocell fibers were supplied by Lenzing AG (Austria); the linear density of the fiber is 0.17g/1000m (1.7 dtex) and the specific gravity is 1.5g/cm<sup>3</sup>, average length and diameter is 38mm and 13.4μm respectively. Viscose fibers were supplied by Suominen Nonwoven Ltd, Finland; the surface weight of the fiber is 60g/m<sup>2</sup> while thickness is 0.66mm. Woven glass fiber mats were supplied by Ahlstrom, Mikkeli, Finland with surface weight 500g/m<sup>2</sup> and woven jute mat was supplied by HP Johannesson Trading AB, Sweden. Acrylated epoxidized soybean oil (AESO, Figure 1) was supplied by Cognis GmbH, Germany while a free radical initiator (tert-butyl peroxybenzoate) was supplied by Aldrich Chemical Company, Wyoming, IL, USA. The carding and needling process of Lyocell fibers were done according to the work by Adekunle et al. [15]



**Figure 1.** Structure of Acrylated Epoxidized Soybean Oil (AESO)

## 2.2 Fiber Surface Treatment

Alkali treatment was done to the jute and Lyocell fibers by washing it with a 4 weight% NaOH solution for 1 hour. It was then rinsed with distilled water until neutral pH, checked by litmus paper. Then the fibers were dried at room temperature for 24 hours and further dried in oven at 105°C for 3 hours. These fibers were used to fabricate composites to see the effect of alkali treatment.

## 2.3 Composite Preparation

The resin viscosity was controlled as it has influence on the composite manufacturing. The viscosity of the resin was reduced by heating in oven at 60°C for 5 minutes. The AESO resin was then blended with 2 wt% initiator (tert-butyl peroxybenzoate) and mixed thoroughly to form homogeneous solution. The alkali treated fiber mats were straightened with an electric iron to keep the fabrics in longitudinal direction and the fiber mats were cut into 20x20cm. Sandwich type composites were prepared by alternate layers of fibers and matrix while the fiber-resin ratio was 60:40 wt%. Composites were manufactured with different weight fraction of natural fibers, glass fibers and regenerated cellulose fibers using compression molding. Heat (160-170°C) and pressure (40 bar) for 5 minutes were used for curing on hot press supplied by Rondol Technology, Staffordshire, UK. Laser machine, GCC LaserPro Spirit, was used to cut the specimens from the samples.

## 2.4 Characterization

### 2.4.1 Water Absorption Test

This test was carried out to find out the dimensional stability of the composites. Four specimens were tested for each sample and the average values were calculated. Before the testing, the specimens were dried in an oven for 24 hours at 60°C; and then the specimens were transferred to desiccators in order to cool down to room temperature. The weight of these specimens was noted as  $W_0$ . The specimens were immersed in distilled water at room temperature for 10 days and the amount of absorption was measured every 24 hours. The specimen was taken out of the water and the surface was wiped before weighing. This weight is denoted as  $W$  and the percentage absorption (WA%) was calculated by formula,  $WA\% = [(W - W_0) / W_0] \times 100$ . To see the influence of water absorption on mechanical properties, tensile and flexural tests specimens were immersed in distilled water in the same way before testing while dry specimens were tested for reference.

### 2.4.2 Tensile Test

This test was carried out according to the standard ISO 527 using Tinius Olsen H10KT universal testing machine and Qmat software. Ten specimens were tested for each composite sample along longitudinal direction. The load range was 5 kN and 10 kN while the rate of loading was 10 mm/min. Gauge length was 50 mm while the initial distance between the grips

was 115 mm. Strain was measured by extensometer. Tests were carried out before and after water absorption.

### 2.4.3 Flexural Test

Flexural strength and modulus were determined by three point flexural test based on ISO 14125 using Tinius Olsen H10KT universal testing machine and Qmat software. Five specimens were tested for each sample along longitudinal direction. The load range was 5 kN and 10 kN while the rate of loading was 10 mm/min. The outer span and the displacement range were 64 mm and 10 mm respectively. Tests were carried out before and after water absorption.

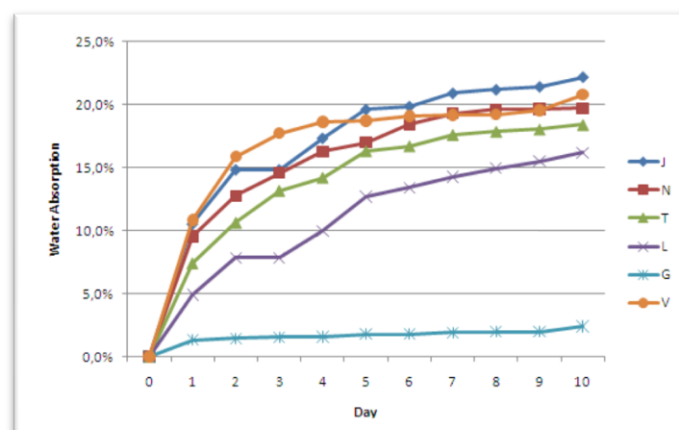
### 2.4.4 Impact Test

Charpy impact test was done to determine impact strength of un-notched specimens and mean impact resistance was calculated. Ten specimens along longitudinal direction were tested flatwise for each sample using Zwick test instrument. The Charpy impact strength was calculated as the ratio of the absorbed energy to the cross-sectional area.

## 2.5 Discussion of Results

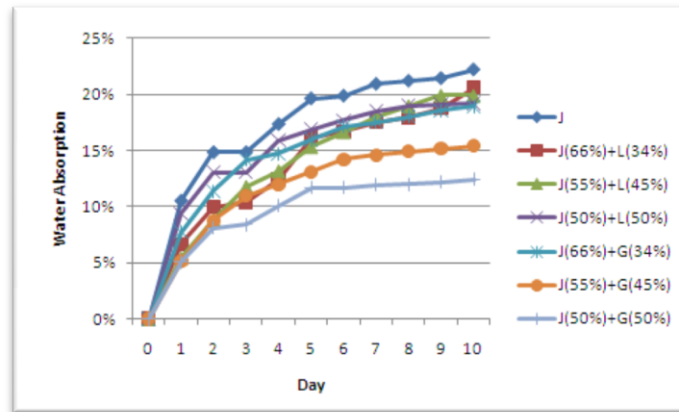
### 2.5.1 Water Absorption Test

Each material absorbed different amount water; 2.4% (glass, G) to 22.2% (woven jute, J) by weight at the end of 10 days exposure as shown in figure 2a. Composites made from Lyocell (L), alkali treated Lyocell (T), non-woven jute (N) and viscose (V) absorbed 16.2%, 18.4%, 19.7% and 20.8% by weight respectively. The neat resin absorbed negligible amount of water. Composites absorbed water consistently from the first day to the tenth day except glass fiber reinforced composites, which had no significant change. It is concluded from the results that alkali treatment to the Lyocell fibers was not essential when the objective is to reduce the water uptake and delamination was occurred in Lyocell-jute reinforced composites in wet condition due to incompatibility between fibers.

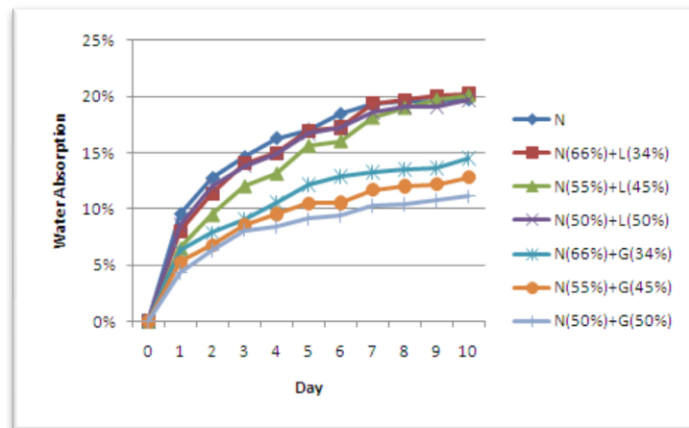


**Figure 2a.** Water absorption of individual fiber composites by weight% (J-woven jute; N-nonwoven jute; T-treated Lyocell; L-untreated Lyocell; G-glass; V-viscose)

Figure 2b and 2c showed that the water absorption was reduced by inclusion of Lyocell or glass fiber to make hybrid composites.



**Figure 2b.** Water absorption of woven jute hybrid composites by weight% (J-woven jute; L-untreated Lyocell; G-glass)



**Figure 2c.** Water absorption of nonwoven jute hybrid composites by weight% (N-nonwoven jute; L-untreated Lyocell; G-glass)

### 2.5.2 Tensile Test

Tensile strength (Figure 3) and modulus (Figure 4) of the individual fiber composites and hybrid composites before and after water uptake were discussed. Tensile properties were reduced on water absorption but improved on hybridization. Lyocell and glass fiber composites have better properties and inclusion of these fibers to natural fiber composites improved properties. Unstable nature of alkali treated Lyocell after water absorption is less preferred over untreated Lyocell. Either 34% of Lyocell or glass improved the tensile strength in both dry and wet conditions.

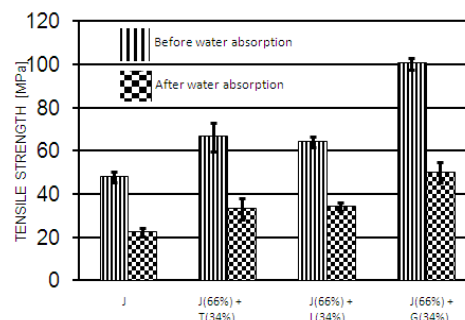
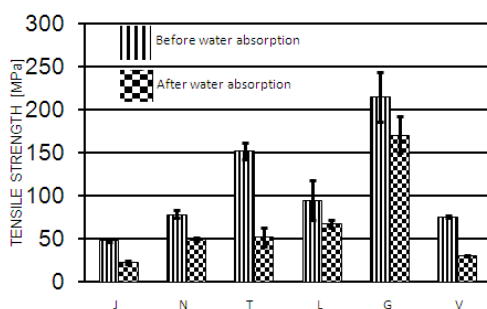


Figure 3. Tensile strength of composites before and after water absorption (J-woven jute; N-nonwoven jute; T-treated Lyocell; L-untreated Lyocell; G-glass; V-viscose)

Tensile modulus also followed the same trend. Nonwoven hybrid composites also had similar results.

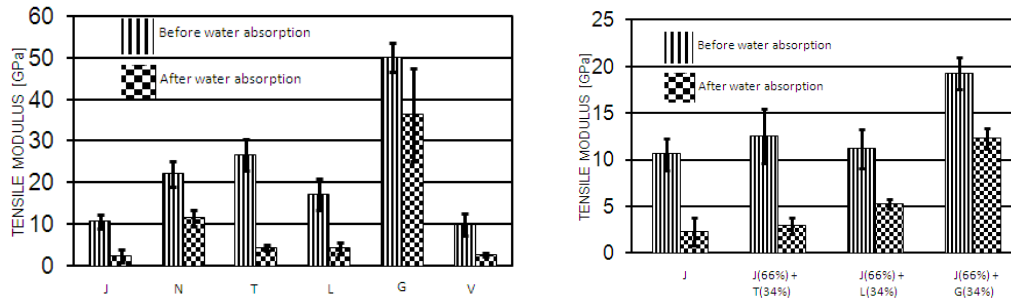


Figure 4. Tensile modulus of composites before and after water absorption (J-woven jute; N-nonwoven jute; T-treated Lyocell; L-untreated Lyocell; G-glass; V-viscose)

### 2.5.3 Flexural Test

Figure 5 and 6 shows the flexural properties (strength and modulus). Flexural properties were reduced by water uptake and improved by hybridization like tensile properties.

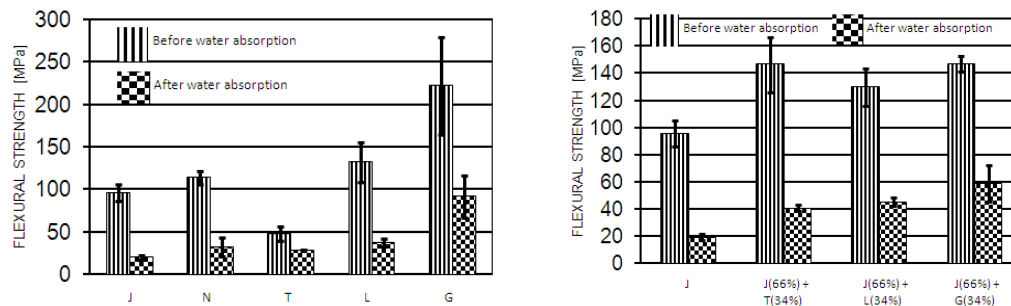


Figure 5. Flexural strength of composites before and after water absorption (J-woven jute; N-nonwoven jute; T-treated Lyocell; L-untreated Lyocell; G-glass)

Lyocell fiber reinforced composites had better properties than jute fiber composites. Inclusion of Lyocell or glass fiber improved the flexural strength. Flexural modulus did not considerable increase upon Lyocell hybridization before and after water absorption while a significant increase was seen when jute fiber was hybridized with glass fiber. However, viscose hybridization did not show appreciable results.

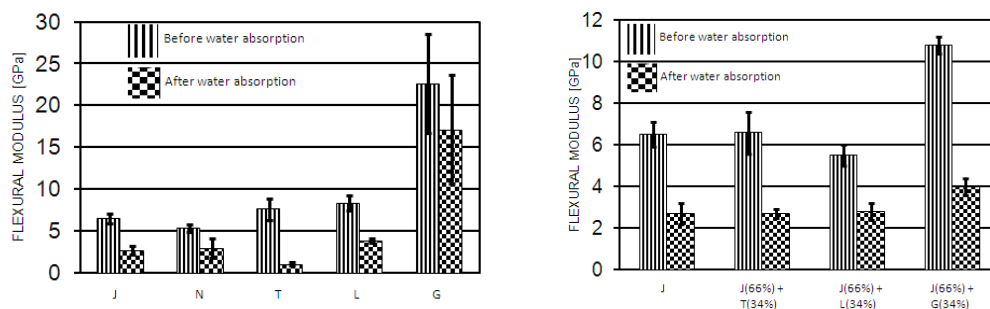


Figure 6. Flexural modulus of composites before and after water absorption (J-woven jute; N-nonwoven jute; T-treated Lyocell; L-untreated Lyocell; G-glass)

### 2.5.4 Impact Testing

Figure 7 show the steady increase in impact strength on hybridization. Impact strength of Lyocell and glass fiber composites was better than for the jute fiber composites. On increasing the amount of Lyocell in hybrid woven jute composites from 34% to 50% of total fiber content, the strength increased from 30 kJ/m<sup>2</sup> to 40 kJ/m<sup>2</sup>. Similar results were observed with hybrid nonwoven jute composites.

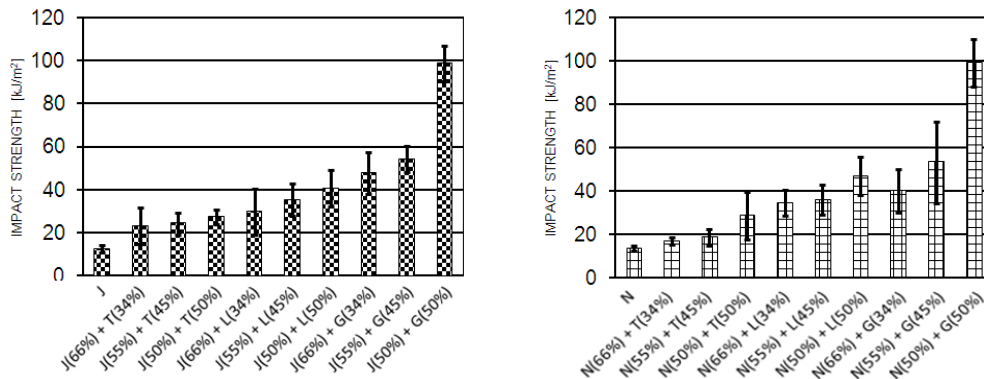


Figure 7. Impact strength of composites (J-woven jute; N-nonwoven jute; T-treated Lyocell; L-untreated Lyocell; G-glass)

### 3. Conclusions

It is concluded that the natural fiber composites could replace several materials in outdoor applications on reducing the water absorption by hybridization. Further research can improve the mechanical properties and also reduce the water uptake. Tensile, flexural and impact tests showed the improvisation of properties on inclusion of Lyocell or glass. The potential of Lyocell is considered to be huge in field of composites due to its potential properties.

### References

- [1] Carrillo F., Colom X., Canavate X. Properties of regenerated cellulose Lyocell fiber-reinforced composites. *Journal of Reinforced Plastics and Composites*, **29**, pp. 359-371 (2010).
- [2] Van de Velde K., Kiekens P. Thermoplastic pultrusion of natural fiber reinforced composites. *Composite Structures*, **54**, pp. 355-360 (2001).
- [3] Bledzki A.K., Gassan J. Composites reinforced with cellulose based fibres. *Progress in Polymer Science*, **24**, pp. 221-274 (1999).
- [4] McCorsley C.C. Progress for Shaped Cellulose Article Prepared from a Solution Containing Cellulose Dissolved in a Tertiary Amine N-oxide Solvent. U.S. patent no. 4,246,221, January 20, 1981.
- [5] Berger W. Possibilities and Limitations of Alternative Processes for the Dissolution and Forming of Cellulose. *Lenzinger Berichte*, Rudolstat, Germany, 1994; 11-18, FW16-FW19 (1992).
- [6] Borbély É. Lyocell, The new generation of regenerated cellulose. *Acta Polytechnica Hungarica*, **5**, pp. 11-18 (2008).
- [7] Carrillo F., Colom X., Canavate X. Properties of regenerated cellulose Lyocell fiber-reinforced composites. *Journal of Reinforced Plastics and Composites*, **29**, pp. 359-371 (2010).
- [8] Khot S.N., Lascala J.J., Can E., Morye S.S., Williams G.I., Palmese G.R., Kusefoglou S.H., Wool R.P. Development and application of triglyceride-based polymers and composites. *Journal of Applied Polymer Science*, **82**, pp. 703-723 (2001).

- [9] Kolot V., Grindberg S. Vernonia oil-based acrylate and methacrylate polymers and interpenetrating polymer networks with epoxy resins. *Journal of Applied Polymer Science*, **91**, pp. 3835-3843 (2004).
- [10] Zhu J., Chandrashekhara K., Flagnigan V., Kapila S. Curing and mechanical characterization of a soy-based epoxy resin system. *Journal of Applied Polymer Science*, **91**, pp. 3513-3518 (2004).
- [11] Meier M.A.R., Metzger J.O., Schubert U.S. Plant oil renewable resources as green alternatives in polymer science. *Chemical society review*, **36**, pp. 1788-1802 (2007).
- [12] Falk B., Crivello J.V. Synthesis and modification of epoxy- and hydroxyl-functional microspheres. *Journal of Applied Polymer Science*, **97**, pp. 1574-1585 (2005).
- [13] Becquart F., Taha M., Zerroukhi A., Kaczun J., Stebani U. Functionalization of a poly(vinyl alcohol) in the solid state with a swelling agent by methacrylic anhydride. *Journal of Polymer Science Part A: Polymer Chemistry*, **42**, pp. 1618-1629 (2004).
- [14] Adekunle K., Åkesson D., Skrifvars M. Synthesis of reactive soybean oils for use as a biobased thermoset resins in structural natural fiber composites. *Journal of Applied Polymer Science*, **115**, pp. 3137-3145 (2009).
- [15] Adekunle K., Patzelt C., Kalantar A., Skrifvars M. Mechanical and Viscoelastic Properties of Soybean Oil Thermoset Reinforced with Jute Fabrics and Carded Lyocell Fiber. *Journal of Applied Polymer Science*, **122**, pp. 2855-2863 (2011).