

# Effect of Surface Treatment on Performance of Tamarind Fiber–Epoxy Composites

C. Uma Maheswari, K. Obi Reddy, E. Muzenda and M. Shukla

**Abstract** - Randomly oriented short Tamarind fibre reinforced Epoxy composites were prepared by hand lay up process. To improve the surface, these fibers were treated with alkali (sodium hydroxide) and silane (3-aminopropyltriethoxy silane). The effects of alkali and silane treatment of fibers on tensile and flexural properties of the composites were investigated. Mechanical test results show that alkali and silane treatment significantly improves the tensile, flexural and impact properties of Tamarind fiber epoxy composites. Scanning electron microscopy (SEM) investigations show that surface modifications improve the fiber/matrix adhesion.

**Keywords** - Fiber composites, surface modification, mechanical properties, morphology

## I. INTRODUCTION

NATURAL fibers have become alternative reinforcing fillers in various areas of polymer composites due to their advantages over synthetic fibers, e.g. low density, less tool wear during processing, low cost, non-toxic, easy process, environmental friendly, and biodegradability [1, 2]. Recently, natural fiber reinforced polymer composites have been used in several fields such as automotive, construction, marine etc. [3, 4]. Many studies on mechanical properties of natural fibres and how these incorporate with various polymer matrices have been attempted [5]. All these studies emphasized that the properties of composites depend on the natural fibres' interfacial bonding with the polymer matrices. In other words, the adhesion between the reinforcing fibers and the matrix plays an important role in the final mechanical properties of the materials since the stress transfer between matrix and fibres determines reinforcement efficiency.

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However, an important problem for using fiber as reinforcement in plastic is poor adhesion between natural fiber and polymer matrix. Because fiber has low interaction with matrix, load transfer from matrix into fiber is not good. Many studies have been performed to improve the adhesion between the fiber and matrix by modification of the fiber and/or polymer matrix using physical and chemical methods [5-7]. These treatments leads to reduction in moisture gain, as well as changes in the fiber surface. The conditions of modification reactions affect the fiber properties, including the chemical composition, morphology and thermal stability. Chemical treatment is the most common procedure used to improve the adhesion with a polymer matrix as studied by many researchers. Alkali and silane treatments are the most common chemical treatment methods used in polymer composites [7, 8]. These methods are inexpensive and effective in modifying the fiber surface for enhanced interfacial adhesion between the natural fiber and the polymeric matrix. Many researchers have investigated the influence of alkali and silane treatment on the mechanical properties of natural fiber reinforced polymer composites.

In this study, tamarind fiber used as a reinforcing agent, with epoxy used as a matrix for the preparation of Tamarind fiber epoxy composite. The tamarind (*Tamarindus indica* L.) is a long-lived, bushy tree belonging to the Fabaceae family. It is widely distributed throughout the tropical belt from Africa to South Asia, Southeast Asia, northern Australia, and South America. The authors selected tamarind fruit fibers for the study because large quantities of this fiber are produced as waste in many food-processing industries. The authors have already reported preliminary studies on properties of tamarind fruit fibers and mentioned their suitability as reinforcement [9]. The possibility of finding use for Tamarind fibers in fibre composite will open a new market for what normally considered waste or used in low value products. The objective of this study is to investigate the effects of fiber surface treatment and fiber content on the mechanical properties, morphology and chemical resistance of Tamarind fiber epoxy composites based on renewable natural tamarind fiber.

## II. MATERIALS AND METHODS

### A. Materials

Extracted Tamarind fruit fibers, epoxy resins (Araldite LY 556) and ambient temperature hardener (HY 951) was

obtained from Ciba Geigy. Chemicals like acetic acid, sodium hydroxide and 3-aminopropyltriethoxysilane were obtained from Merck.

#### B. Extraction of Fibers

The shells of fully ripened tamarind fruits were broken and the fibers were separated. The extracted fibers were thoroughly washed with water with occasional shaking to remove any leftover pulp sticking to them. The cleaned fibers were dried at room temperature for a week and finally kept in a hot air oven at 100 °C until dry.

#### C. Alkali Treatment

The tamarind fibers were treated with 5% (w/v) alkali (sodium hydroxide) solution for 30 min at ambient temperature, maintaining a fiber-to-liquor ratio of 1:30 (w/v). The alkali treated fibers were washed several times with tap water to remove excess alkali and then with dilute acetic acid to remove any traces of alkali from the fiber surface and finally washed thoroughly with distilled water. Then the treated fibers were dried at room temperature for one week and finally kept in a hot air oven at 100 °C until dry.

#### D. Silane Treatment

A solution of 1% v/v 3-aminopropyltriethoxy silane was prepared in acetone. Acetone was used in preference to water to promote hydrolysis to take place with the moisture on the surface of the fibers rather than within the carrier. The pH of the solution was maintained at a value of 4 to bring about complete hydrolysis of the silane by the addition of acetic acid; the solution was then stirred continuously for 10 min. The dried alkali treated fibers were immersed in the solution for 1 h. After the treatment, fibers were separated from the solution and dried in a hot air oven at 60 °C until dry.

#### E. Preparation of Composites

For making the composite laminates by the hand lay up process, a glass mould covered with Teflon® sheet and having dimensions of 160 X 160 X 3 mm was used. Laminates were made from Epoxy and hardener taken in the ratio of 100 and 10 parts by weight, respectively. The mould was filled with a mixture of matrix and Tamarind fibers (chopped length 1.5 to 2 cm) in random orientation with varying fiber content (5, 10, 15, 20, 25 and 30 % by weight) and allowed to cure at room temperature for 24 h. The cured laminates were demoulded and post cured in a hot air oven at 100 °C for 3 h.

#### F. Mechanical Testing

Tensile tests were carried out according to ASTM D3039-78 standard using a Universal testing machine (INSTRON 3369) with a load cell of 10 kN, maintaining a crosshead speed of 5 mm/min. Flexural tests were performed as per ASTM D5943-96 standard utilizing the three point bending mode on the above machine with a crosshead speed of 2 mm/min and a support span length of

50 mm. Five specimens were used for each test and the average values recorded.

#### G. Scanning Electron Microscopy (SEM) Analysis

The morphology of the fractured samples was examined using the JEOL JSM 820 scanning electron microscope operating with secondary electron imaging at 5 kV. The composite samples were cryogenically cooled and brittle fractured. The fractured surfaces were coated with gold by electrodeposition to impart electrical conduction before recording the micrographs.

### III. RESULTS AND DISCUSSION

The tensile (modulus and strength) and flexural (modulus and strength) of the neat epoxy and untreated Tamarind fiber epoxy composites determined as a function of fiber loading, are presented in Table 1 and 2 the corresponding line graphs shown in Fig. 1 and 2. From Figures 1-2, for increased fiber content, the mechanical properties of composites increases up to 25 wt% fiber loading decreasing thereafter. Further increases cause the properties to decrease because of lower fiber–matrix adhesion and the quantity of fiber content being more than matrix. However, the tensile and flexural properties of the composites were found to be higher than that for the matrix. The increase in tensile (modulus and strength) and flexural (modulus and strength) of composites respectively (presented in parentheses) over the matrix were as follows: tensile (12.8 and 23.2%) and flexural (11.9% and 15.6%). The tensile and flexural properties of untreated fiber composites increased marginally when compared with the matrix. This is due to hydrophilic nature of the fibers leads to weak interfacial bonding, with consequent problems of poor stress transfer, void spaces and debonding in the resulting composites.

To overcome these problems, first the fibers were chemically treated with sodium hydroxide and silane. The tensile and flexural properties of chemically (alkali and silane) modified Tamarind fiber reinforced epoxy composites, as a function of fiber loading are presented in Table 1 and 2 the corresponding line graphs shown in Fig. 1 and 2. The tensile and flexural of the alkali treated fiber composites were found to be higher than those for the matrix and untreated fiber composites. Due to the alkali treatment can remove hemicellulose and surface (wax and pectin) substances create a rough surface topography and also alkali treatment leads to fiber fibrillation, i.e. breaking down the fiber bundle into smaller fibers [10]. This increases the effective surface area available for wetting by the matrix resin. Hence, increasing the fiber aspect ratio caused by reduced fiber diameter and producing a rough surface topography offer better fiber/matrix interfacial adhesion and increase in mechanical properties. The maximum increase in tensile (modulus and strength) and flexural (modulus and strength) properties of alkali treated fiber composites (presented in parentheses) over the matrix

were established to be as follows: tensile (19.4 and 31.4%) and flexural (18.0 and 21.3%).

TABLE I  
INFLUENCE OF CHEMICAL MODIFICATION ON THE TENSILE PROPERTIES OF TAMARIND FIBER REINFORCED EPOXY COMPOSITES

Fiber content (wt%)	Modulus (MPa)			Strength (MPa)		
	Unt	Alk	Sil	Unt	Alk	Sil
0	2560	2560	2560	40.2	40.2	40.2
5	2645	2701	2770	42.9	44.8	47.3
10	2710	2837	2958	45.7	48.4	52.8
15	2796	2954	3114	48.1	52.3	57.6
20	2858	3073	3266	50.6	55.7	62.1
25	2936	3176	3398	52.4	58.6	66.6
30	2843	3085	3348	49.1	56.3	63.9

Unt-Untreated; Alk-Alkali treated; Sil-Silane treated

TABLE II  
INFLUENCE OF CHEMICAL MODIFICATION ON THE FLEXURAL PROPERTIES OF TAMARIND FIBER REINFORCED EPOXY COMPOSITES

Fiber content (wt%)	Modulus (MPa)			Strength (MPa)		
	Unt	Alk	Sil	Unt	Alk	Sil
0	2875	2875	2875	45.7	45.7	45.7
5	2953	3009	3080	47.3	48.5	49.6
10	3024	3141	3255	49.1	50.9	53.7
15	3109	3266	3437	50.9	53.6	57.3
20	3193	3385	3595	52.7	56.1	60.4
25	3265	3506	3750	54.2	58.1	63.5
30	3176	3415	3678	53.1	56.7	62.3

Unt-Untreated; Alk-Alkali treated; Sil-Silane treated

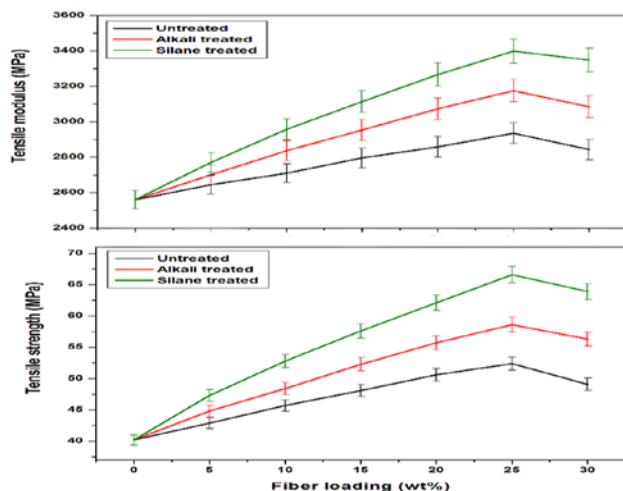


Fig. 1 Tensile properties of untreated and chemically treated tamarind fiber-epoxy composites

The tensile and flexural properties were found to be higher when silane treated tamarind fibers used in the composites. Silane agent may reduce the number of cellulose hydroxyl groups in the fiber-matrix interface [11]. In the presence of moisture, the hydrolysable alkoxy group leads to the formation of silanols. The silanol then reacts

with the hydroxyl group of the fiber, forming stable covalent bonds to the cell wall that are chemisorbed onto the fiber surface. Therefore, the hydrocarbon chains, provided by the application of silane, restrain the swelling of the fibers by creating an entangled network due to the diffusion of hydrocarbon chains into the matrix [11]. It was assumed that the hydrocarbon chains produced by the silane application influenced the wettability of the fibers, thus improving the chemical affinity of the polymer matrix. Thus, this factor is accountable to be responsible for an improvement in the mechanical properties of the composites under study when the fibers were being silane treated. The maximum increase in tensile (modulus and strength) and flexural (modulus and strength) properties of alkali treated fiber composites (presented in parentheses) over the matrix were established to be as follows: tensile (24.6 and 39.6%) and flexural (23.4 and 28.0%).

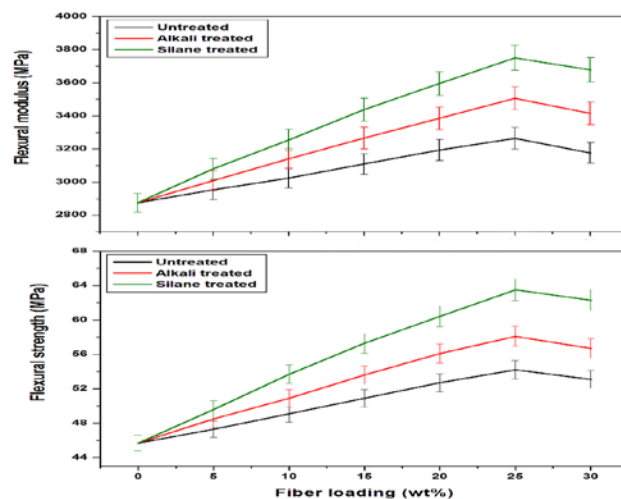


Fig. 2 Flexural properties of untreated and chemically treated tamarind fiber-epoxy composites

In order to probe the effect chemical modification on the interfacial bonding in the composites under study, their fractographs were recorded. The scanning electron micrographs of the fractured surfaces untreated fiber epoxy composites are shown in Fig. 3(a-b). From these micrographs, it is the clear that extensive fiber full out was observed as indicated by the protruding fibers and holes on the matrix surface. These two observations clearly indicate a poor adhesion between the fiber and matrix due to the smooth surface of the fibers, leading to inferior mechanical properties. However, where alkali treated tamarind fiber epoxy composite is concerned, the fiber full out and holes cannot be seen properly. This may support better adhesion between fiber and epoxy matrix. The SEM micrographs in Fig. 3(c-d) clearly indicate that the interfacial adhesion between the fiber (alkali treated) and the matrix. On the contrary, silane treated fiber is covered (skin formation) by the matrix that considerably reduce the gaps between them (Fig. 3e) and fiber was even found to break without any pullout (Fig. 3f), which indicates the improvement of

interfacial adhesion between the fiber and the matrix. This suggests the strongest interfacial interaction between fiber and matrix.

#### IV. CONCLUSION

In this study, the effect of chemical modified (alkali and silane) Tamarind fiber loading on the mechanical properties of the epoxy composites was investigated. The mechanical properties of epoxy composites were greatly increased by two kinds of surface treatment proposed in this study. The morphological observation supports the improved mechanical properties of the Tamarind epoxy by the alkali and silane treatments. This study will provide valuable contributions to the development of high-value-added fiber reinforced composites based on natural Tamarind fibers as one of sustainable resources and epoxy resin.

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#### REFERENCES

- [1] S.V. Joshi, L.T. Drzal, A.K. Mohanty and S. Arora, "Are natural fiber composites environmentally superior to glass fiber reinforced composites," *Compos Part A.*, Vol. 35, pp. 371-376, 2004.
- [2] M. J. John and S. Thomas, "Biofibres and biocomposites," *Carbohydr Polym.*, Vol. 71, pp. 343-364, 2008.
- [3] J. Holbery and D. Houston, "Natural fiber reinforced polymer composites in automotive applications," *J Miner Meta Mater Soc.*, Vol. 58, pp. 80-86, 2006.
- [4] J. K. Pandey, S. H. Ahn, C. S. Lee, A. K. Mohanty and M. Misra' "Recent advances in the application of natural fiber based composites," *Macr Mater Eng.*, Vol. 295, pp. 975-989, 2010.
- [5] M. J. John and R. D. Anandjiwala, "Recent developments in chemical modification and characterization of natural fiber-reinforced composites," *Polym Compos.*, Vol. 29, pp.187-207, 2008.
- [6] M. N.Belgacem, P. Bataille and S. Sapiaha, "Effect of corona modification on the mechanical properties of polypropylene/cellulose composites," *J Appl Polym Sci.*, Vol. 53, pp. 379-385, 1994.
- [7] Y. Xie , C. A. S. Hill, Z. Xiao, H. Militz, and C. Mai, "Silane coupling agents used for natural fiber/polymer composites: A review," *Compos Part A.*, 41, pp.806-819, 2010.
- [8] M. Das and D. Chakraborty, "Evaluation of improvement of physical and mechanical properties of bamboo fibers due to alkali treatment," *J Appl Polym Sci.*, Vol. 107, pp. 522-527, 2008.
- [9] C. U. Maheswari, B. R. Guduri and A. V. Rajulu, "Properties of lignocellulose tamarind fruit fibers," *J Appl Polym Sci.*, Vol.110, pp.1986-1989, 2008.
- [10] X. Li, L.G. Tabil and S. Panigrahi, "Chemical treatments of natural fiber for use in natural fiber-reinforced composites: A review," *J Environ Polym Degrad.*, Vol.15, pp. 25-33, 2007.
- [11] H.Dhakal, Z. Zhang and M. Richardson, "Effect of water absorption on the mechanical properties of hemp fibre reinforced unsaturated polyester composites," *Compos Sci Technol.*, Vol. 67, pp.1674-1683, 2007.

# Chromatographic Detectors and Column Hold-up for Organics–PDMS Systems

Edison Muzenda

**Abstract**—Gas chromatographic detector types and application, as well carrier gas flow rate and column hold-up measurements are discussed in this paper. The flame ionization and hot wire detectors are discussed in detail. The measurement of carrier gas flow rate using a soap film bubble flow meter is explained. Column hold-ups for 4 poly(dimethylsiloxane) packed columns were measured using the homologous series method.

**Keywords**—Column hold-up, detector, homologous, poly(dimethylsiloxane), retention, thermodynamic function

## I. DETECTORS

**D**ETECTORS may be divided into main classes (i) Those which respond to the total mass of material emerging from the column for example the automatic titration system and these are called integral detectors (ii) Those, which respond to the concentration of the vapour in the carrier gas at any instant for example the Katharometer and these are called differential detectors. Detectors of the type have obvious advantage in quantitative analysis since this only requires the measurement of step heights.

However the two types of detectors are limited in their application. Several types of detectors are used in gas chromatography such as the hot wire detectors (HWDs), flame ionisation detectors (FIDs), flame photometric detectors (FPDs) and the electron capture detectors (ECDs). The main purpose of the detector is to identify the components by means of their retention times and to measure their concentrations. Selection of detectors depends on many factors, for example, sensitivity (minimum detection), stability (linearity), selectivity (signal to noise ratio), response time and the components involved. In this work, the flame ionisation and the hot wire detectors are discussed in detail while other detectors are discussed briefly.

Thermal conductivity detectors have been widely used in the chromatographic determination of thermodynamic data. They are simple in construction and permit direct determination of the gas hold-up of the chromatographic column by the timing

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of an air peak. However, since their response depends on the difference between the thermal conductivity of the organic solute and that of the carrier gas, this response is reduced considerably when there is a constant bleed of pure volatile organic solvents into the detector. For volatile solvents, thermal conductivity detectors are expected to be limited in applicability to systems where thermal conductivities of solvent and solute vapours are different. A more suitable detector would be the flame ionisation detector. One drawback in the use of flame ionisation detectors in the accurate determination of retention data has been the elaborate and indirect method of measuring the column gas hold-up. This is because the detector is insensitive to air under the conditions prevailing in a conventional column. The other drawback associated with flame ionisation detectors when volatile solvents are used is the decrease in sensitivity of the detector due to the increase in ion concentration between the electrodes. This eventually leads to complete insensitivity when the condition of ion saturation is reached. However, since a non-volatile solvent (PDMS) was used in this work, this drawback was eliminated. FIDs are very useful in the detection of volatile organic compounds in very low concentrations, say down to the 1ppm range. Therefore, the flame ionisation detector was used in this work as the focus is in very dilute regions.

### A. The Flame Ionization Detector (FID)

#### - Principle of operation

This detector was developed by McWilliam and Dewar (1958) [1] and it is based on the principle that the electrical conductivity of a flame burning hydrogen and oxygen mixture increases when the organic molecules enter the flame. This is mainly as a result of the difference in the ionisation potentials of the gas (15.8 – 24.5ev) and organic molecules (9-12ev). However, the ionisation of the organic molecules that takes place in the flame is not well understood. The increase in conductivity of the flame is much more than would be expected from the normal ionisation potential of the organic compounds. The potentials of organic molecules are in the range 9-12ev while the effective ionisation potential is about 5ev. One explanation as postulated by Stern (1951) [2] in [3] could be the formation of carbon aggregates within the flame and their subsequent ionisation. This explanation is strengthened by the fact that the ionisation potential of solid carbon is 4.6ev and the detector response is essentially proportional to the number of carbon atoms in the organic