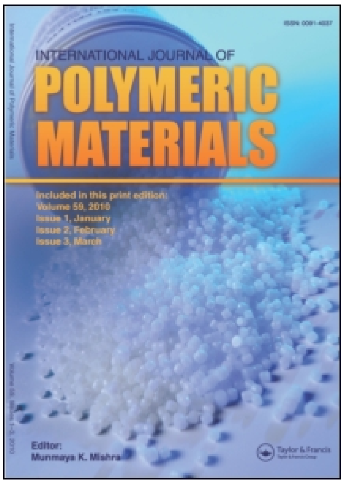

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Influence of Fiber Surface Modification on the Mechanical Performance of Isora-Polyester Composites

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Influence of Fiber Surface Modification on the Mechanical Performance of Isora-Polyester Composites

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This paper reports the effect of chemical treatment on the mechanical properties of a natural fiber, isora, as reinforcement in unsaturated polyester resin. Isora fiber is separated from the bark of the Helicteres isora plant by a retting process. The short isora fiber surface was modified chemically by acetylation, benzoylation, silane and triton treatments to bring about improved interfacial interaction between the fiber and the polyester matrix. The modified surfaces were characterized by IR spectroscopy and SEM. The SEM studies were carried out to investigate the fiber surface morphology, fiber pull-out and fiber-polyester interface bonding. They showed the changes occuring on the fiber surface during chemical treatment. Properties like tensile strength, flexural strength and impact strength have been studied. The chemical modification of fiber improved fiber/matrix interaction as evidenced by the enhanced tensile and flexural properties. The lower impact properties of the composites, except triton-treated fiber composite, further point to the improved fiber/matrix adhesion, compared to the untreated fiber composites.

Keywords: acetylation, benzoylation, composite, flexural strength, impact strength, isora fiber, polyester, silane treatment, tensile strength, triton treatment

INTRODUCTION

Cellulose-based natural fibers are a potential resource for making low-cost composite materials, where high strength and stiffness are not the first priority [1]. Although plant fibers are generally considered to be viable for use in composites, the problem of

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compatibility of the fiber with the matrix must be solved. Plant fibers, due to the abundance of hydroxyl groups, are incompatible with hydrophobic polyester resin. This incompatibility leads to a poor interfacial adhesion between the fiber and matrix, poor wetting of the fibers by the resin and a reduction in mechanical performance when the composite is exposed to moisture. Thus, in order to develop composites with good properties, it is necessary to improve the interface between the matrix and the lignocellulosic materials used as fillers. In order to improve interfacial bonding, modification of the fiber surface is essential [2–5].

When two materials are incompatible, it is often possible to bring about compatibility by introducing a third material that has properties intermediate between those of the other two by forming weak boundary layers, deformable layers or restrained layers. Coupling agents can improve the wettability between polymer and substrate or can form covalent bonds with both materials [1]. Chemical or irradiation-induced removal of some hydroxyl groups has been used to improve hydrophobicity and chemical bonding at the interface [6–8]. Silane interfacial coupling agents, widely used on glass fiber to form stable covalent bonds to both the mineral fiber surface and the resin, are potentially suitable for use on cellulosic fibers. Acetylation is an effective method of modifying the surface of natural fibers and making it more hydrophobic. In addition, acetylation is one of the most studied reactions of lignocellulosic materials [9]. Hill and Khalil [10] studied the effects of chemical modification of coir and oil palm fibers by acetylation, silane treatment and titanate coupling agent on the mechanical properties of the polyester composites. They found that the interfacial strength between the fiber and matrix was increased by acetylation. Rout et al. [11] observed that fiber surface modification by chemical treatment significantly increased the mechanical performance of coir—polyester composites. To improve the moisture resistance, Chand et al. acetylated sisal fiber and studied its tensile strength [12]. It was shown that acetylation could reduce the moisture content, but the tensile strength of the acetylated fiber was reduced due to the loss of hemicellulose in the fiber during acetylation. Zafeiropoulos et al. [9] studied the effect of acetylation on flax fibers and found that the presence of moisture is beneficial for the reaction and also acetylation increased the weight percent gain (WPG) of fibers at high levels. This implied that the treatment is affecting the bulk and not just the surface of the fibers. Uma Devi et al. [13] investigated the effect of silane A-172 treatment on the mechanical properties of pineapple leaf fiber (PALF)-reinforced polyester composites and observed significant improvement in the tensile strength for the treated fiber-composite.

EXPERIMENTAL

Materials

Isora Fiber

Isora fiber was separated from the bark of Helicteres isora plant by retting. It was dried in an air oven at 70° C for 24 h. The fibers were well-separated by hand, and cut into 30 mm length for composite fabrication.

Other Reagents

General purpose (GP) unsaturated polyester resin, methyl ethyl ketone peroxide (MEKP) and cobalt naphthenate were supplied by M/s. Sharon Engineering Enterprises, Kochi, Kerala. Chemicals used for the experiment are of laboratory reagent grade. All other ingredients used in the composite preparation were of commercial grade.

Chemical Modification of the Fiber

Acetylation

Alkali-treated fibers were soaked in glacial acetic acid for 1 h, decanted and then soaked for 5 min in acetic anhydride containing two drops of concentrated sulphuric acid. They were then filtered, washed and dried in an air oven at 70°C.

Benzoylation

Alkali-treated fibers were suspended in 10% sodium hydroxide solution and agitated with 50 ml benzoyl chloride for 1 h, filtered, washed with water and dried. They were then soaked in ethanol for 1 h to remove the unreacted benzoyl chloride, washed with water and finally dried in an air oven at 70°C.

Silane Treatment

Alkali-treated fibers were dipped in an alcohol/water mixture (60:40) containing 1% vinyltriethoxysilane coupling agent. The pH of the solution was maintained between 3.5 and 4. Fibers were washed in distilled water and dried in an air oven at 70°C.

Triton Treatment

Alkali-treated fibers were treated with 5% triton (isooctylphenoxypolyethoxyethanol) solution for 2 h. The fibers were then washed in water and dried in an air oven at 70°C.

Composite Preparation

Randomly oriented isora-polyester composites were prepared by hand layup method using a three-piece stainless steel mold having dimensions of 200 × 150 × 30 mm. Prior to the composite preparation, the mold surface was polished well and a mold-releasing agent (waxpol) was applied to the surface of the mold. Unsaturated polyester resin (GP) was mixed well with 1% w/w cobalt naphthenate accelerator and 1% w/w MEKP catalyst. The randomly oriented fiber mat prepared was placed in the mold and the resin mixture was poured evenly on it. Using a metallic roller, the air bubbles were carefully removed and the mat was allowed to wet completely. The mold was closed and the excess resin was allowed to flow out as “flash” by pressing in a hydraulic press. The pressure was held constant during the curing process at room temperature for 24 h. The composite sheet was post-cured at 80°C for 4 h. From the sheet, samples were cut for measuring tensile, flexural and impact properties according to ASTM standards.

Characterization of Treated Fibers and Testing of the Composites

The SEM photographs of the fibers and the fractured surface of the composites were taken using a JEOLJSM 35 C model scanning electron microscope. The IR spectra of the raw and chemically modified isora fibers were recorded using a Schimadzu IR 470 Infrared Spectrophotometer, by the KBr pellet technique. Tensile testing of the composite specimens was carried out according to ASTM D5083 on a Schimadzu Model AG1 50 kN universal testing machine at a cross head speed of 5 mm min^{-1} and a gauge length of 50 mm. Rectangular specimens of dimensions 150 × 15 × 3 mm were used for testing. The tensile strength and tensile modulus were determined from the stress-strain curves.

Flexural tests were performed according to ASTM D790 with rectangular strips of size 100 × 15 × 3 mm at a cross-head speed of 5 mm min^{-1}. Flexural strength and flexural modulus were determined from the stress-strain curves.

Izod impact strength of unnotched sample of the composite was determined according to ASTM D256 using a Tinius Olsen Model 503 instrument.

RESULTS AND DISCUSSION

Physical Changes: SEM Studies

The SEM photograph of untreated isora fiber surface is given in Figure 1(a) and that of chemically treated fiber surfaces in Figures 1(b–e). They give strong evidence for the physical and micro-structural changes occuring on the fiber surface during chemical treatment, compared to the untreated fiber. On modification, the pores became clearer and the fibers became thinner. This may be due to the dissolution and leaching out of the fatty acids and lignin components of the fiber. This roughened the fibers, enhancing the mechanical interlocking at the interface. As a result of the introduction of functional groups by chemical treatment, the fiber surface became irregular and rough as shown in the figure. Similar changes have been reported in grafted natural fibers such as cotton and jute [14].

The fibrils are well-exposed on chemical treatment. On triton treatment, the fiber fibrillation occurred to a great extent, thus reducing the fiber diameter and resulting in considerable increase in the fiber aspect ratio (Figure 1(e)). Indeed, with the use of solvents, these chemical treatments lead to a swelling effect, leaching out of the waxes, gums and partial removal of lignin and hemicellulose, which are the cementing materials in vegetable fibers [15–17].

Chemical Modifications: IR Spectroscopy

The IR spectra of the raw and treated isora fibers are given in Figure 2. The IR spectrum of raw isora fiber shows an absorption peak at 1730 cm^{-1}, which is the characteristic band for carbonyl stretching, associated with the carbonyl groups present in lignin and other cellulosic components. The strong broad peak at 3300–3320 cm^{-1} is the characteristic hydrogen-bonded -OH stretching vibration. The peaks at 2910 cm^{-1} and 750 cm^{-1} correspond to the C-H and C-O stretching vibrations, respectively. A band at 1600 cm^{-1} is due to the C-C stretching of the aromatic ring in the lignin components, while a strong peak at 950 cm^{-1} arises from the glycosidic linkages. The bands at 1370, 1330 and 1310 cm^{-1} are due to the -CH deformation, -OH in plane bending and -CH_2 wagging, respectively. The band near 1250 cm^{-1} is due to the –C-O-C bond in the cellulose chain. On chemical

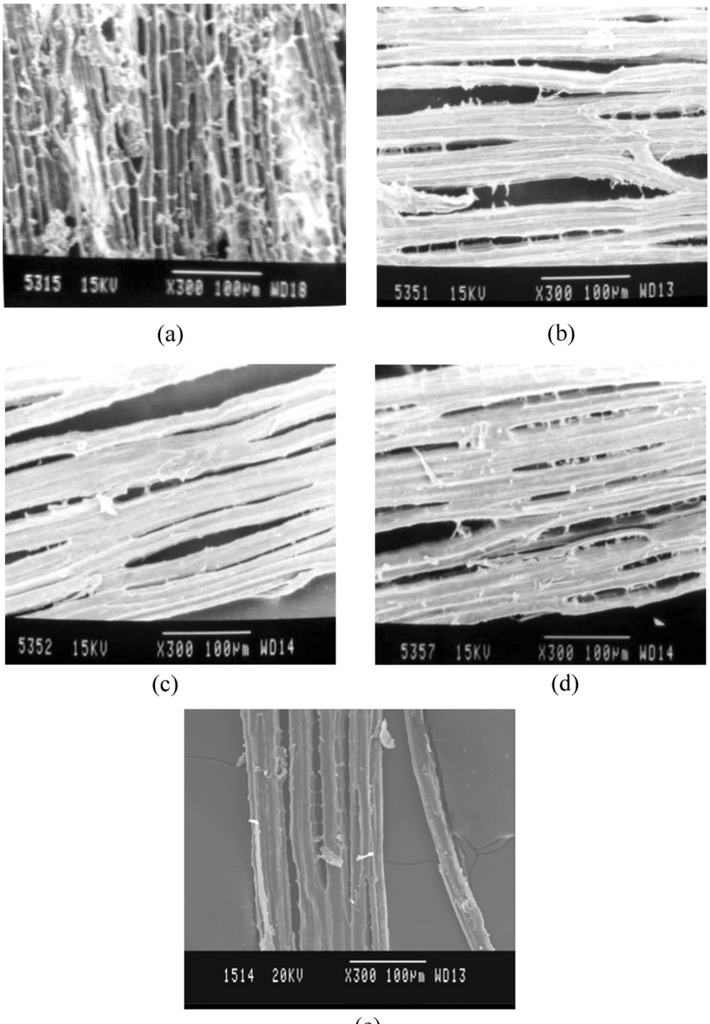


FIGURE 1 (a–e): SEM photographs of (a) untreated (b) acetylated (c) benzoylated (d) silane-treated and (e) triton-X-100-treated isora fiber.

modification of the fiber by acetylation, benzoylation and silane treatment, the peaks corresponding to the functional groups were observed in the IR spectra. For acetylated fiber a peak at $1700\,\mathrm{cm}^{-1}$ indicates the introduction of an acetyl group. As a result of esterification of the hydroxyl groups, the hydroxyl vibrational absorption at $3300\,\mathrm{cm}^{-1}$ decreased.

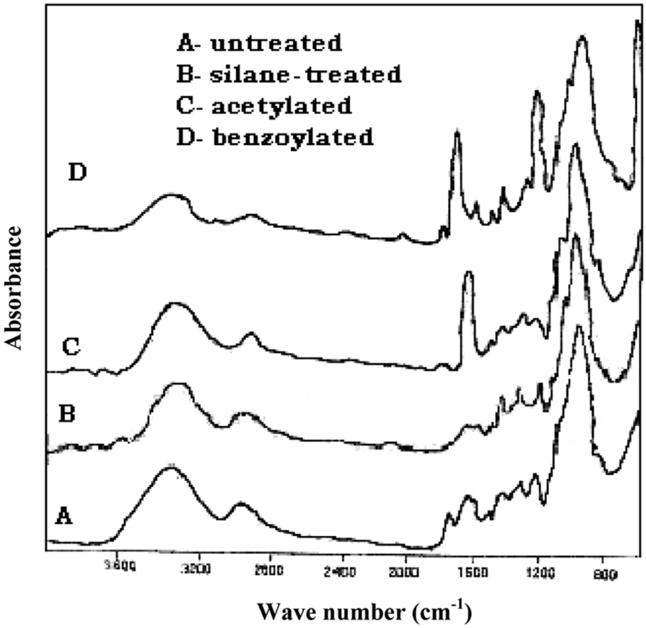


FIGURE 2 IR spectra of untreated and chemically modified isora fiber.

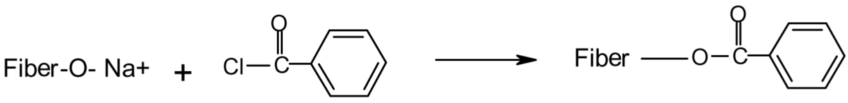
IR spectra of benzoylated fiber indicates an absorption in the range 1400–1600 cm^{-1} due to the C=C stretching of aromatic rings and a carbonyl group absorption in the range 1700 cm^{-1} is also observed. In the case of silane-treated fibers, an additional peak at 3500 cm^{-1} indicates intermolecular hydrogen bonding between the silanol -OH and cellulosic –OH of the fiber.

Reaction Schemes of Chemical Treatments

Benzoylation

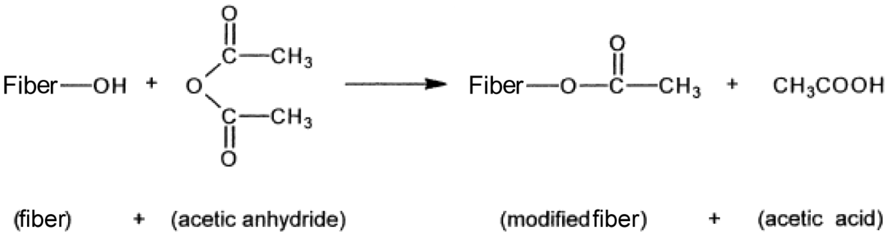
The reaction between the fiber surface and benzoyl chloride can be represented as

$$\text{Fiber}-\text{OH} + \text{NaOH} \rightarrow \text{Fiber}-\text{O}^{-}\text{Na}^{+}$$



Acetylation

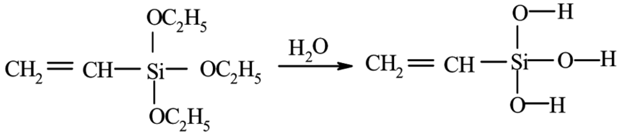
During acetylation, the typical reaction taking place on the fiber surface can be represented as

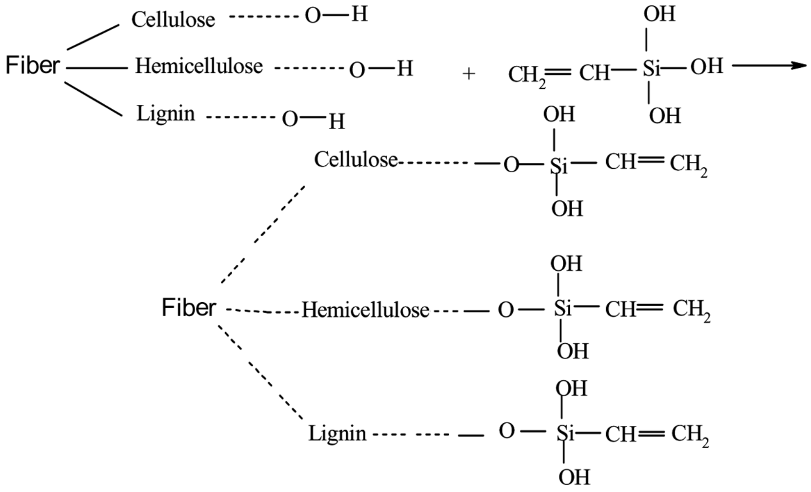


Silane (Vinyl Triethoxy) Treatment

The organo functional group in the silane coupling agent (here $CH_2{=}CH$-) causes the subsequent reaction with the polyester. This could be a copolymerization, and/or the formation of an interpenetrating network. The curing reaction of a silane-treated substrate enhances the wetting by the resin.

The general mechanism of formation of bonds of alkoxy silanes with the fiber surface [16], which contains hydroxyl groups, is as follows:

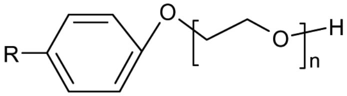




Alkoxy silanes undergo hydrolysis, condensation and bond formation stages, By base or acid-catalyzed mechanisms. In addition to these reactions of silanols with hydroxyls of the fiber surface, the formation of polysiloxane structures can take place.

Triton-X-100 Treatment

Chemically triton-X-100 is isooctylphenoxypolyethoxyethanol, with the structure shown below:



R = isooctyl, n = 10.

It is a non-ionic synthetic surfactant and has a hydrophilic head and hydrophobic tail. The main function of a surfactant is cleansing action. Non-ionic surfactants are most widely used for surface cleaning and have no charge in aqueous solutions. They are resistant to water hardness. The advantages of triton-X-100 are:

1. Excellent detergent and oil in water emulsifier for many applications
2. Excellent wetting agent and
3. Effective performance across a broad temperature range.

MECHANICAL PROPERTIES OF THE COMPOSITE

Tensile Properties

The effects of various chemical treatments on tensile strength, Young's modulus and elongation at break values of the chemically modified, randomly oriented isora-polyester composites are given in Figures 3–5, respectively.

The effect of chemical treatments on the tensile strength of the composites is given in Figure 3. It is observed that fiber modification improved the tensile strength of all composites and the improvement followed the order: triton-treated (TT) > acetylated (Ac) > silane treated (Si) > benzoylated (Bz) > untreated (UT). The maximum improvement in tensile strength was observed for triton-treated fiber composite. Compared to the untreated fiber composite, the percentage increase in tensile strength of treated fiber composites are TT–39%, Ac–27.8%, Si–16.7% and Bz–11%. The improvement in tensile

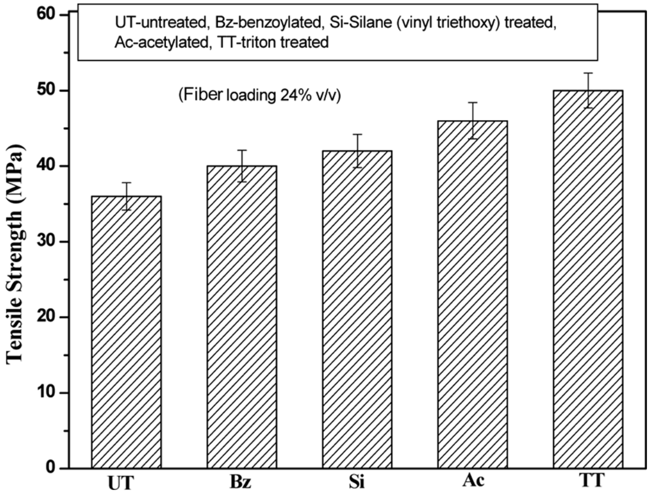


FIGURE 3 The effect of different chemical treatments on the tensile strength of randomly oriented isora-polyester composites.

strength can be attributed to the improved adhesion between the fiber and the matrix on chemical modification of the fiber.

The Young's modulus of the composites (Figure 4) also improved on chemical modification of fiber and followed the order: TT > Ac >

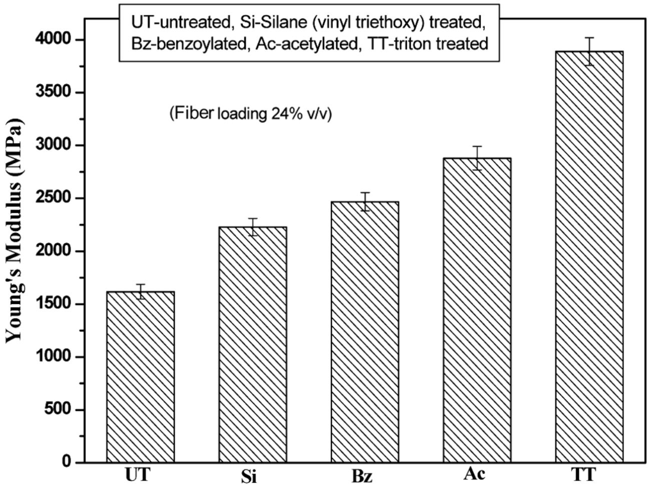


FIGURE 4 The effect of different chemical treatments on the Young's modulus of randomly oriented isora-polyester composites.

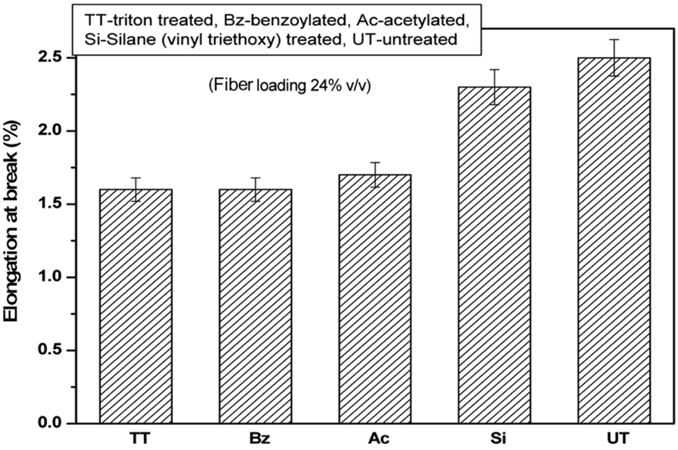


FIGURE 5 The effect of different chemical treatments on the elongation at break of randomly oriented isora-polyester composites.

Bz > Si > UT. Compared to the untreated fiber composite, the percentage increase in Young's modulus of treated fiber composites are TT-113, Ac-78, Bz-52.6, Si-37.8%. The improvement in Young's modulus is due to the improvement in adhesion between the fiber and the matrix, on chemical modification of the fiber.

The percentage elongation at break values of short isora-polyester composite varied with fiber treatment as shown in Figure 5. It is observed that on chemical modification of the fiber, elongation at break values of the composites decreased and followed the order: UT > Si > Ac > Bz > TT. As expected from the modulus values, TT fiber composite has the lowest and silane-treated fiber composite has the highest elongation at break values. When the fiber-matrix adhesion is higher, the composite will fail at a lower elongation. The reduced elongation at break values of the composites confirms the improved adhesion between the fiber and resin.

Now, we can examine the mechanism involved in the improvement of adhesion in each case. The improvement in tensile properties of benzoylated fiber composite is attributed to the presence of phenyl groups in the treated fiber which interacts with the benzene rings and methyl groups present in this polyester, which improves the compatibility between the fiber and polyester. Also, the reduction in hydrophilicity of the fiber on benzoylation makes the fiber more compatible with the hydrophobic polyester.

In addition, benzoylation makes the surface of the fiber very rough and provides better mechanical interlocking with the polyester matrix.

A hypothetical model of interface of benzoylated isora fiber-polyester composite is shown in Figure 6.

Acetylated fiber composites show improvement in properties, next to triton-treated fiber composites. Rong et al. (17) have reported such results for acetylated jute fiber-reinforced epoxy composites. They concluded that the improvement in interfacial bonding is due to the H-bonds between acetyl groups of the acetylated fiber and hydroxyl or amine groups in epoxy resin and mechanical interlocking between epoxy and the rough fiber surface due to alkali pretreatment. Since there is no possibility of H-bond formation between acetyl groups of the acetylated fiber and the unsaturated polyester resin, the improvement in tensile properties can be attributed to the second factor, which is the mechanical interlocking between the polyester resin and the fiber surface. This is supported by the rough surface morphology of the acetylated fiber evident from the SEM micrograph (Figure 1(b)). Also, the hydrophobic methyl groups attached to the fiber interacts with the methyl and benzene rings present in the polyester, thereby increasing the compatibility with the hydrophobic polyester matrix. A hypothetical model of the interface of acetylated isora fiber-polyester composite is shown in Figure 7. Another important contributing factor is the reduction in the hydrophilicity of the fiber as a result of acetylation, which makes the fiber more compatible with hydrophobic polyester.

Vinyltriethoxysilane has two functional groups, a hydrolyzable group which can condense with the hydroxyls of the fiber and an organo functional group ($CH_2{=}CH$-) capable of interacting with the matrix. The hydrolyzed silanol forms strong covalent bonds or hydrogen bonds with-OH group of cellulose.

The individual coupling agent molecules attached to cellulose forms a continuous link. The long hydrophobic polymer chain can adhere to

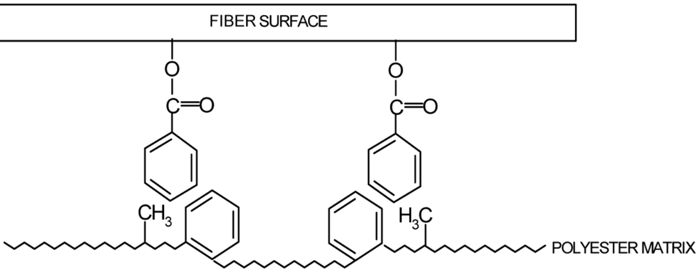


FIGURE 6 A hypothetical model of interface of benzoylated isora-polyester composite.

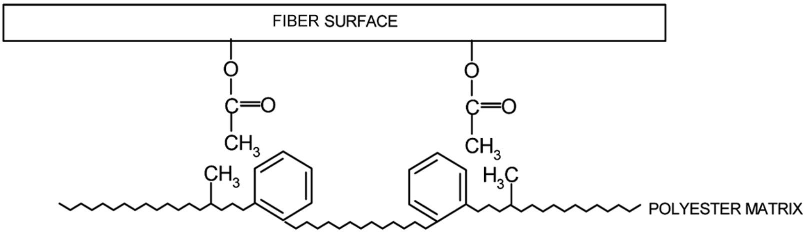


FIGURE 7 A hypothetical model of interface of acetylated isora-polyester composite.

the matrix mainly because of Van der Waals-type attractive force. As a result, silane-coupling agents form a bridge at the interface. The hydrophobic alkyl groups ($R = CH_2{=}CH$-) attached to the fiber as a result of silane treatment increases the compatibility with the hydrophobic polyester matrix and improves the mechanical properties of the composite. A hypothetical model of the interface of silane-treated fiber-polyester composite is shown in Figure 8.

On triton treatment of the alkali pre-treated fiber, they become more dispersable and well-exposed as evident from the SEM photograph in Figure 1(e). he aspect ratio of the fiber is also enhanced. The increased dispersability results in a larger area of contact and improved wetting of the fiber. This leads to greater mechanical interlocking between fiber and matrix, making the interfacial adhesion stronger and the mechanical properties higher. This explains the maximum values of tensile strength and Young's modulus for triton-treated fiber composite. Similar result was reported by D. Ray et al. [18] for alkali-treated jute fiber-vinylester resin composite. They found that alkali-treated fibers were dispersed more readily and uniformly

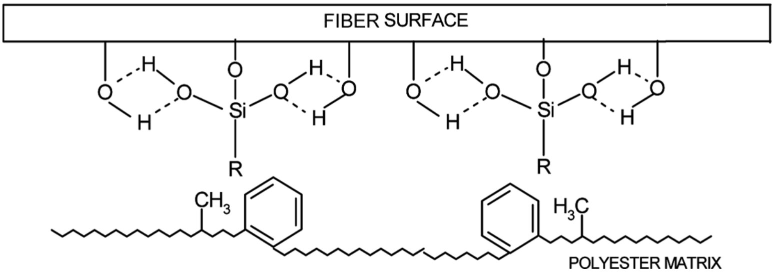


FIGURE 8 A hypothetical model of interface of silane-treated isora-polyester composite.

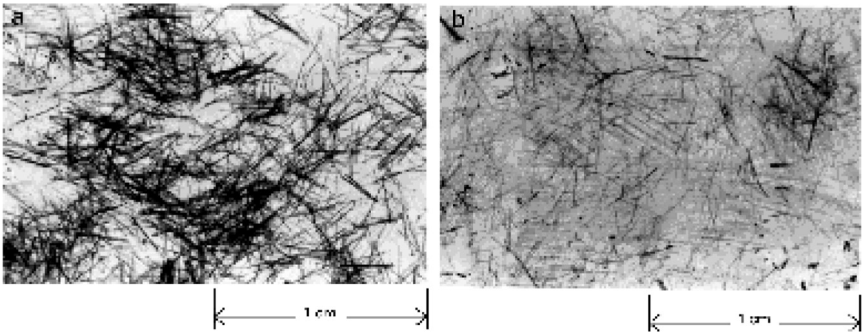


FIGURE 9 Dispersability of jute fibers in vinylester resin: **(a)** raw jute, **(b)** 2 h alkali-treated jute [18].

in the resin than the untreated ones. Treated fibers were seen to be finely separated from one another whereas the untreated fibers remained mostly in clusters (Figure 9).

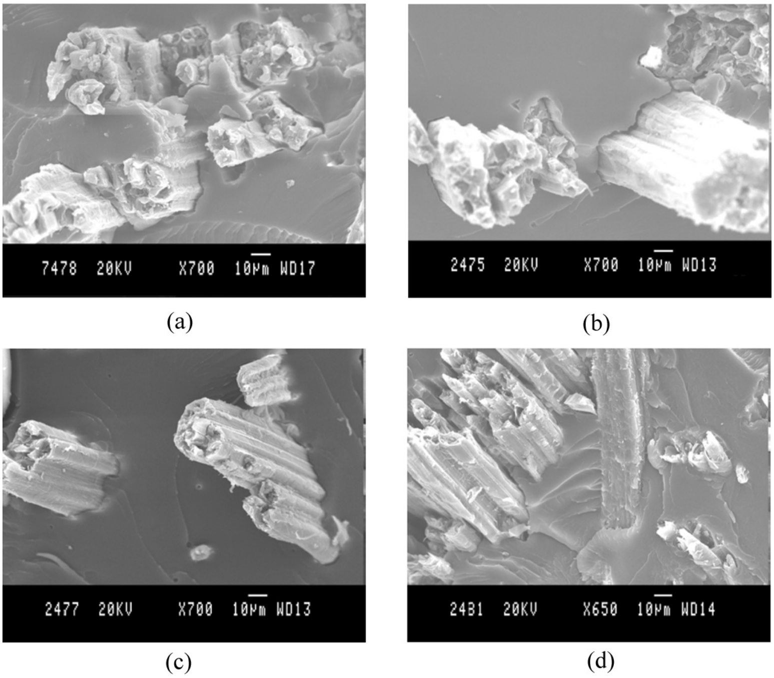
The improvement in adhesion between the treated fiber and polyester resin can be understood from the SEM photographs of the fracture surface of untreated isora fiber-polyester composite and that of treated fiber composites given in Figures 10(a–d). While the fracture surface of untreated fiber composite shows holes and fiber ends indicating poor adhesion between the fiber and matrix, the fracture surface of treated fiber composites shows fiber breakage rather than pull-out, indicating better interfacial strength.

Flexural Properties

The effects of fiber modification on flexural strength, flexural modulus and flexural strain of short isora fiber-reinforced polyester composites are shown in Figures 11–13, respectively. All fiber treatments resulted in improvement in flexural strength and flexural modulus of the composites. The improvement in flexural strength of the composites followed the order: TT > Ac > Si > Bz > UT. The percentage increase in flexural strength compared to untreated fiber composite is TT-50, Ac-27.3, Si-10.6, and Bz-4.5%.

In the case of composites, the flexural properties are controlled by the resistance to interlaminar failure. Therefore, high flexural strength and modulus of treated fiber composite is due to better interfacial adhesion in the composite.

The improvement in flexural modulus followed the order TT > Ac > Bz > Si > UT. The percentage increase in flexural modulus compared to untreated fiber composite is- TT-88.4, Ac-63.2, Bz-23 and Si-18.8%.



(a) (b) (c) (d)

FIGURE 10 SEM photographs of the tensile fracture surface of (a) triton-treated (b) acetylated (c) silane-treated and (d) benzoylated randomly oriented isora-polyester composites (magnification ×700).

The improved flexural properties of the treated fiber composite can be attributed to the physical and chemical changes on the fiber surface effected by the treatments, which enhanced the adhesion between the fiber and matrix as in the case of tensile properties.

Flexural strain values of all treated fiber composites were lower than that of untreated fiber composite, and followed the order: UT > Si > Ac > TT > Bz. This is due to improved bonding between the treated fiber and resin.

Impact Properties

The impact strength of a composite is influenced by many factors such as the toughness properties of the reinforcement, the nature of interfacial region and the frictional work involved in pulling the fibers from the matrix [10]. The nature of interface region is of extreme importance in determining the toughness of the composite. The lowering of adhesion between fiber and matrix leads to improvement of

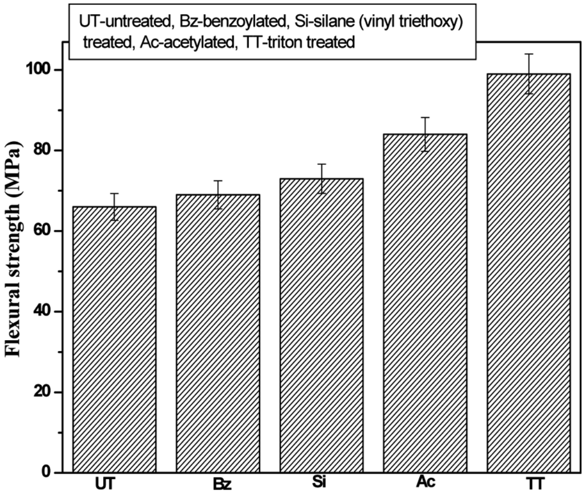


FIGURE 11 The effect of different chemical treatments on the flexural strength of randomly oriented isora-polyester composites.

toughness. However, very low adhesion efficiency may result in the lowering of toughness.

The variation of impact strength with different chemical treatments is given in Figure 14. It is observed that the impact strength decreased as the interfacial bond strength increased except for the case of

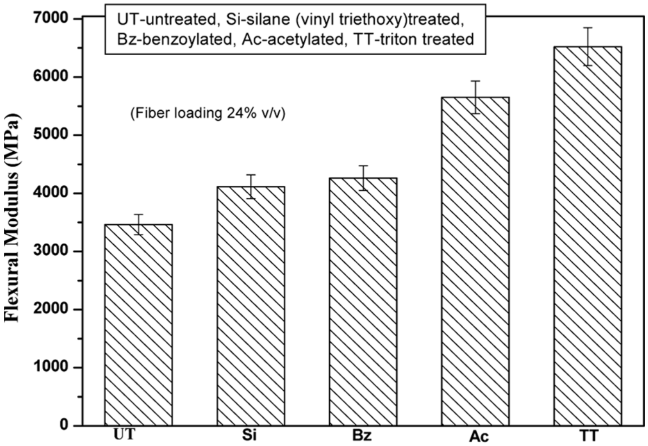


FIGURE 12 The effect of different chemical treatments on the flexural modulus of randomly oriented isora-polyester composites.

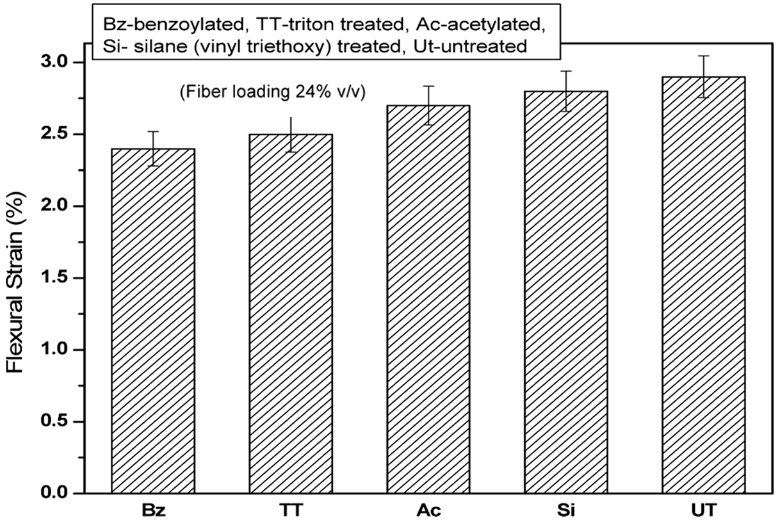


FIGURE 13 The effect of different chemical treatments on the flexural strain of randomly oriented isora-polyester composites.

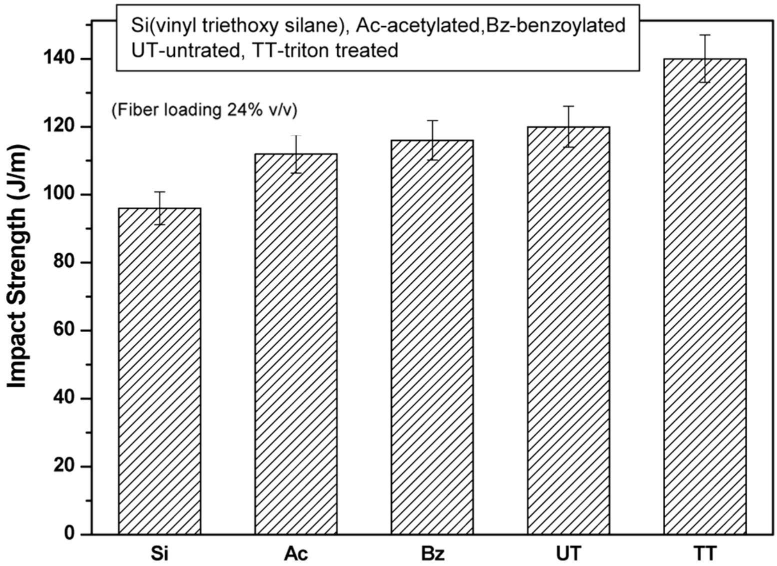


FIGURE 14 The effect of different chemical treatments on the impact strength of randomly oriented isora-polyester composites.

triton-treated fiber composite. The impact strength followed the order: TT > UT > Bz > Ac > Si. While silane treatment, acetylation and benzoylation of the fiber resulted in reduction in the impact strength of the composites, triton treatment of the fiber enhanced the impact strength of the composite, relative to untreated fiber version. It is an established fact that a strong interface between the fiber and the matrix reduces the impact strength of the composites [19–21]. At high levels of adhesion, the failure mode is brittle and relatively little energy is absorbed. In the case of a weak interface the triaxial stresses at the tip of an advancing crack cause debonding to occur and a crack bunting mechanism takes place and improves the toughness of the material [22].

In the case of triton-treated fibers, the treatment has improved the dispersion of the fibers in the resin. In addition, in this case the adhesion between the matrix and the fibers may be intermediate and leads to progressive delamination which requires additional energy and hence an improved impact strength. Hill and Abdul Khalil [10] also got an improvement in impact strength for acetylated oil palm fiber-reinforced polyester. In acetylated fiber the improved impact strength is assumed to be due to the improved toughness of the fiber. Similar results have been reported earlier [23]. When the fiber-matrix adhesion is strong, the mechanism of failure changes from fiber debonding and pull-out to brittle failure and reduces the impact strength.

CONCLUSION

Isora fibers were modified chemically by acetylation, benzoylation, silane treatment and triton treatment. All fiber modifications improved the tensile and flexural properties of the composite. The decrease in hydrophilicity and increase in compatibility of the treated fiber with the polymer matrix are responsible for the improvement in the mechanical properties. The maximum improvement in tensile properties was observed for triton treatment. SEM studies also suggest strong fiber-matrix adhesion in treated randomly oriented isora-polyester composites. In all cases except triton treatment, fiber modification decreases the impact strength of the composites.

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