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Effects of Fiber Weight Ratio, Structure and Fiber Modification onto Flexural Properties of Luffa-Polyester Composites

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Abstract

The effect of chemical modification, reinforcement structure and fiber weight ratio on the flexural proprieties of Luffa-polyester composites was studied. A unsaturated polyester matrix reinforced with a mat of Luffa external wall fibers (ComLEMat), a short Luffa external wall fibers (ComLEBC) and a short Luffa core fibers (ComLCBC) was fabricated under various conditions of fibers treatments (combined process, acetylating and cyanoethylating) and fiber weight ratio. It resorts that acetylating and cyanoethylating enhance the flexural strength and the flexural modulus. The fiber weight ratio influenced the flexural properties of composites. Indeed, a maximum value of strength and strain is observed over a 10% fiber weight ratio. The uses of various reinforcement structures were investigated. The enhancement of elongation at break and the strain values of the composite reinforced by natural mat was proved.

Keywords: Luffa Fibers, Composite, Flexural Properties, Fiber Weight Ratio

1. Introduction

A combination of properties of some natural fibers including low cost, low density, non-toxicity, no abrasion during processing and recyclability has arisen more interest for the manufacturing industry of low cost and low weight composites [1-2].

The composite materials reinforced with natural fibers are used in many fields such as automotive industry, aeronautics and naval [3].

Despite the advantages of cellulosic fibers reinforcing thermoplastics, the polymer-cellulose composites materials are criticized for their low permissible processing temperatures and highly hydrophilic property associated with a low compatibility of hydrophobic polymers as well as a loss of mechanical proprieties after moisture uptake [2-4].

Due to the poor compatibility, the surface of fibers must be treated to improve the adhesion between fiber and matrix. Beldzki *et al.* [1] reported many methods to modify the surface of natural fibers for their use in composite materials such as acetylation, alkali and isocyanates treatments. Saha *et al.* [5] studied the effect of cyanoethylation on the mechanical properties of jute fiber reinforced polyester composite. They noted that a better creep resistance at lower temperatures was obtained for the composite reinforced with cyanoethylated jute fibers. According to Saha *et al.* [6], it has been found that cyanoethylation of jute improved flexural strength and modulus by 62% and 39%, respectively.

Results published in the open literature have indicated the increase of flexural strength of the heigh-density polyethylene (HDPE)-henequen by 36% after silane treatment [2]. The contribution of acetylation, propionylation, malaeic anhydride and styrene on the mechanical properties of obtained composites was investigated [7,8]. An increase in the interfacial shear strength between acetylated fibers and hydrophobic resins was reported [9]. However, acetylation made the fibers more hydrophobic by reacting its hydroxyl groups with acetyl groups [10]. The alkali treatments changed the mechanical proprieties of Luffa [11] and sisal fibers [1] reinforced polyester resin. They showed that the flexural mechanical properties increased with alkali treatment and explained this enhancement by the increase of fiber roughness and contact area. These results were corresponding to those ob-

tained with bagasse and bamboo fibers composites [13, 14].

In addition, the mechanical properties of composites reinforced with natural fibers were influenced by the fibers ratio and the reinforcing structure. According to Rao *et al.* [15], for example, and the flexural strength of vakka, sisal, bamboo and banana reinforced composites increased with fiber volume fraction. Good mechanical properties of different fibers reinforced composites were obtained for various amount of fiber ratio [15-19].

In the case of Luffa fibers composites, Demir *et al.* [20] had studied the influence of chemical modification by silane coupling reagents on Luffa fiber reinforced polypropylene composites. Boynard *et al.* [11] studied also the flexural properties of Luffa fibers reinforced polyester composite. They noticed that the flexural modulus increased by 14% after fibers Alkali treatment. In another study, Tanobe *et al.* [21] characterized the chemically modified Luffa fibers with methacrylamide and NaOH. However, other chemical modifications, reinforcing structure and fiber weight ratio onto Luffa fibers composite were still not investigated. Therefore the aim of this study is to explain the variations of the flexural proprieties of the composites Luffa-polyester with those of fibers weight ratio, structure (short fibers and mat) and treatments (acetylation and cyanoetylation).

2. Materials and Methods

2.1. Fibers Extraction

In this experimental study, many structures of Luffa fibers were used. These fibers can be extracted from the external wall of sponge (LE) or from its core (LC). The external wall Luffa fibers were used as a vascular fibrous network (LEMat) or shelled and cut fibers (LEBC). A Shirley analyser instrument was used to shell the fibrous network. The core Luffa fibers were also shelled and cut (LCBC) to be used as short fibers reinforced polyester matrix.

The Shirley Analyzer uses a cleaning technique combining mechanical and airflow actions as follows:

Luffa fibers are taken by a feed roller to be presented to the following parts of the machine;

The taker-in with the help of its needles opens the mass of the fibers so that it can be cleaned later in the machine;

A variable airflow separates used fibers and dust and trash by the use of performed roller.

Finally, the guard fibers and waste are collected in the trash tray and used fibers are recovered in the fibers box. The various forms of Luffa fibers were chemically extracted using an optimized solution containing 4% of sodium hydroxide and 10% of hydrogen peroxide at 100˚C during 2 hours. Samples were then washed and dried. This treatment is called a combined process [22]. It was used to remove waxy and gummy substances such as lignin and hemicellulose.

2.2. Surface Modification of Fibers

The first modification method of Luffa fibers is the acetylating. Samples of Luffa fibers treated with combined process were added to a round bottomed flask with sufficient acetic anhydride and brought to the desired reaction temperature of 100˚C for 3 hours [7]. The fibers were washed using acetone in ambient temperature to ensure a good removal of acetic anhydride.

The second method used in this study is the cyanoethylating. The Luffa fibers were dried for 4 h at 80˚C. A 2 g of Luffa fibers were impregnated with a sodium hydroxide solution for 2 min at ambient temperature. The fibers were hydro extracted to about 90% wet pickup. The alkali soaked fibers were then put in round bottomed flask containing acrylonitrile solution. The fiber weight to acrylonitrile solution ratio of 1:20 was maintained. In this study toluene was chosen as a diluent for a series of experiments in which the concentration of acrylonitrile was fixed at 50%. The alkali concentration, temperature and time reaction were kept unchanged, *i.e.*, 4%, 60˚C and 60 min were considered [23]. After a stipulated time of reaction, the fibers were thoroughly washed with 5% acetic acid solution and finally with distilled water. The fibers were then dried at 80˚C until constant weight was obtained.

To identify the chemical modifications at fiber surface, infrared spectroscopy analysis of treated and untreated fibers was conducted using an IR-840 SHIMADZU spectrometer. A mixture of 5 mg of dried fibers and 200 mg of KBr was pressed into a disk for FTIR measurements. 100 scans were collected for each measurement over the spectral range between 400 - 4000 cm^{-1}. All the IR spectra presented in this work were obtained in an absorbance mode.

2.3. Composites Preparation

The treated and untreated Luffa fibers were used to reinforce an unsaturated polyester matrix. The composites were manufactured manually by placing the mixture of fibers and resin inside a mould. In order to remove entrapped air bubbles and also enabled manufacturing all uniform thickness composites, the mould was closed and pressed.

2.4. Flexural Test

After composite material preparation, test samples hav-

ing dimensions of (80 × 15 × 4) mm^3 were cut for the mechanical characterization with the three-point bending flexure test according to the standard EN ISO 14125 (1998).

The flexural properties of polyester samples and polyester reinforced by treated and untreated Luffa fibers were made using a universal testing machine LLOYD with a 10 mm/mn test speed. The flexural strength (σ_f), the flexural modulus (E_f) and the surface elongation at break (ε) are calculated respectively by:

$$\sigma_f = \frac{3FL}{2bh^2} \quad (1)$$

$$E_f = \frac{L^3F}{4bh^3w} \quad (2)$$

$$\varepsilon = \frac{6wh}{L^2} \quad (3)$$

where F is the maximum load (N), L the range (mm), h the specimen thickness (mm), b the specimen width (mm) and w indicates the defection (mm).

3. Results and Discussions

The three point bending flexure tests were carried out on pure polyester specimen (PES), polyester reinforced by untreated Luffa fibers, polyester reinforced by Luffa fibers treated with combined treatment, polyester reinforced by acetylated Luffa fibers and polyester reinforced with cyanoethylated Luffa fibers.

Table 1 shows the results of three points bending flexural test of polyester matrix reinforced with the external wall Luffa fibers.

It can be noted from **Table 1** that the reinforcement of polyester matrix with external wall Luffa fibers changes the flexural proprieties of composite material. In fact, the initial flexural modulus decreased from 3.4 GPa for the unreinforced polyester matrix to 2.45 GPa for the ComLEMat treated with combined process. The stress and deformation at break were increased with the addition of fibers mat. Thus, the flexural stress increased slightly from 41.3 MPa for unreinforced polyester matrix to 42.79 MPa for the untreated Luffa fibers reinforced polyester matrix. We can notice also the decrease of the mechanical properties of the composite reinforced with Luffa fibers treated with combined process. This decrease could be due to a degradation of fibers during the chemical treatment or to a low cohesion between fiber and matrix. In fact, the SEM micrographs of Luffa fibers treated with combined process showed an opened structure (**Figure 1**).

Table 1. Flexural proprieties of ComLEMat.

Specimen	Fiber treatment	Fiber weight ratio (%)	E_f (GPa)	σ_f (MPa)	ε (%)
ComLEMat	Combined process	5.03	2.45	34.17	1.47
ComLEMat	Acetylating	5.04	3.29	52.3	1.79
ComLEMat	Cyanoethylating	5.27	2.55	41.81	1.72
ComLEMat	Untreated	4.93	3.05	42.79	1.61
PES	-	-	3.4	41.3	1.38

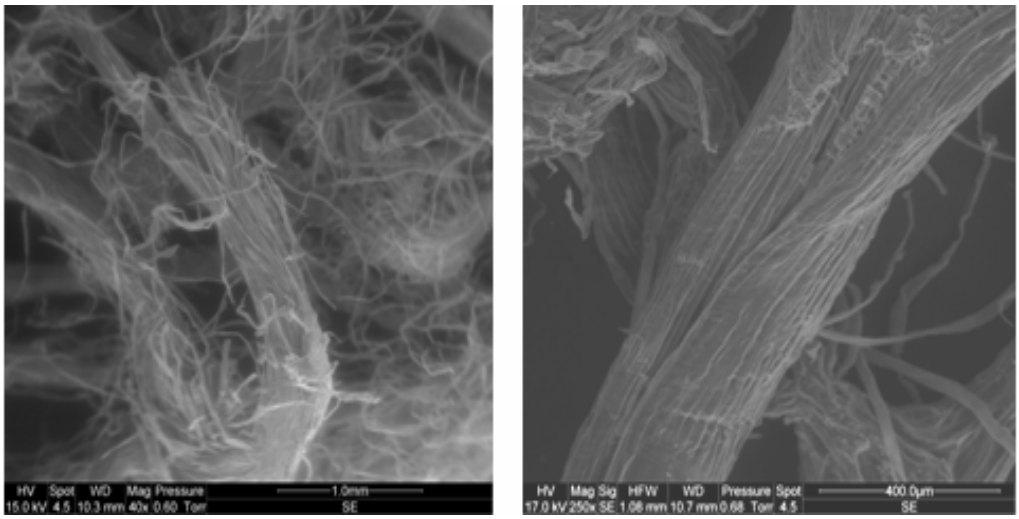

Figure 1. SEM micrographs of Luffa fibers extracted with combined process.

This opened structure showed the ultimate Luffa fibers. This individualization lost the technical Luffa fibers its cohesion therefore its strength.

The weak interface between fiber and matrix was improved by acetylation and cyanoethylation of Luffa fibers. Thus, the properties of the composite reinforced with surfaces treated fibers increased. The composites became more deformable and resistant. The infrared spectra examination presented by **Figures 2(b)-(c)** confirmed that the Luffa fibers extracted with combined process were modified by acetylation and cyanoethylation. Moreover, the treatment of Luffa fibers with acetic anhydride led to the appearance of an absorbance peak in the regions of 1743 and 1243 cm^{-1}. The peak observed at 2254 cm^{-1} confirmed the apparition of acrylonitrile groups on the surface of Luffa fibers.

It's important to indicate that the good cohesion between fibers and matrix is governed by many parameters such as the surface area, the roughness and the surface tensile of fibers. In fact, the acetylation and cyanoethylation made the fibers more hydrophobic by the substitution of hydroxyls groups with acetyls and cyanoethyls groups [6,7].

The SEM micrographs of the fracture surface of composite specimens reinforced with treated and untreated Luffa fibers presented in **Figure 3** showed that adhesion between fiber and matrix enhances with the chemical treatments. In fact, as shown in **Figure 3(a)**, before modification, the wettability between Luffa fibers and unsaturated polyester matrix seemed to be poor because of the presence of a gap between the two componements. This gap is much less visible in the case were the fibers were treated with combined process (**Figure 3(b)**).

In the other cases (**Figures 3(c)-(d)**) we can remark that the matrix surrounding the fibers led to a good adhesion. This result confirmed the enhancement of mechanical proprieties of composite reinforced with acetylated and cyanoethylated fibers. Thus, when the Luffa fibers were treated with acetic anhydride and acrylonitrile, the gap is much less pronounced and an improved interface corroborated well the enhancement of the previously mechanical properties observed in **Table 1**.

Figure 4 shows the variation of the flexural modulus and the flexural strength of the composite LCBC treated with combined process—polyester with the increase of the fiber weight ratio. The curves present two variation zones. In the first zone, the flexural modulus and the ultimate strength reached a maximum values at about 8.5% and 11% of Luffa fibers content, respectively. Thereafter, the two flexural proprieties appear to decrease steadily. Similar results were reported elsewhere [24], indicating this is a typical characteristic of fiber composites since higher fiber weight ratio often results in more defects owing to fiber contacts.

For the case of the composite LEBC fiber treated with combined process-polyester matrix (ComLEBC combined), the **Figure 5** shows the variation of flexural modulus and strength with the variation of fiber weight ratio. We can note that the flexural strength and modulus increase to reach a maximum at about 8% of Luffa fiber weight ratio. Thereafter, the flexural modulus decreased steadily and the flexural strength falls slightly.

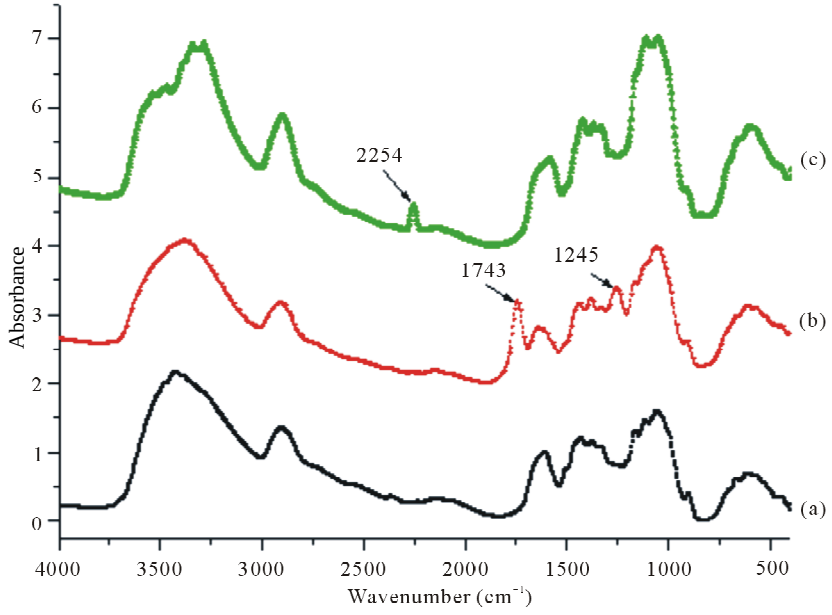

Figure 2. Infrared spectra of Luffa fibers (a) treated with combined process; (b) acetylated; and (c) cyanoethylated.

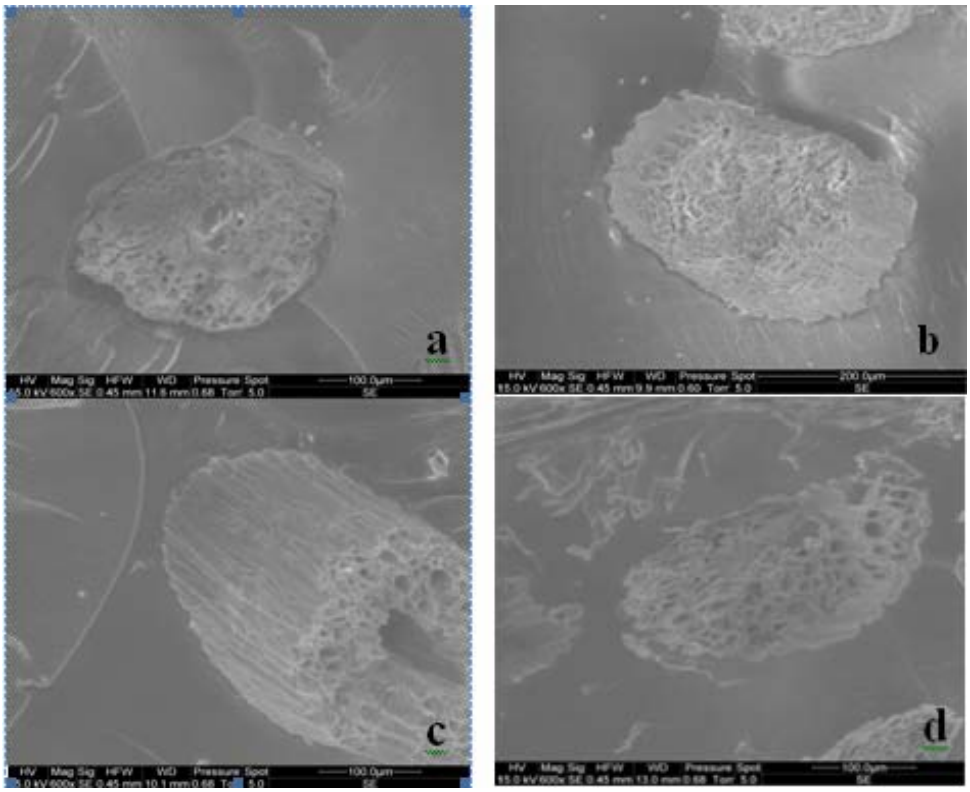

Figure 3. SEM micrographs of fracture surface of: (a) ComLEBC untreated; (b) ComLEBC combined; (c) ComLEBC acetylating; (d) ComLEBC cyanoethylating.

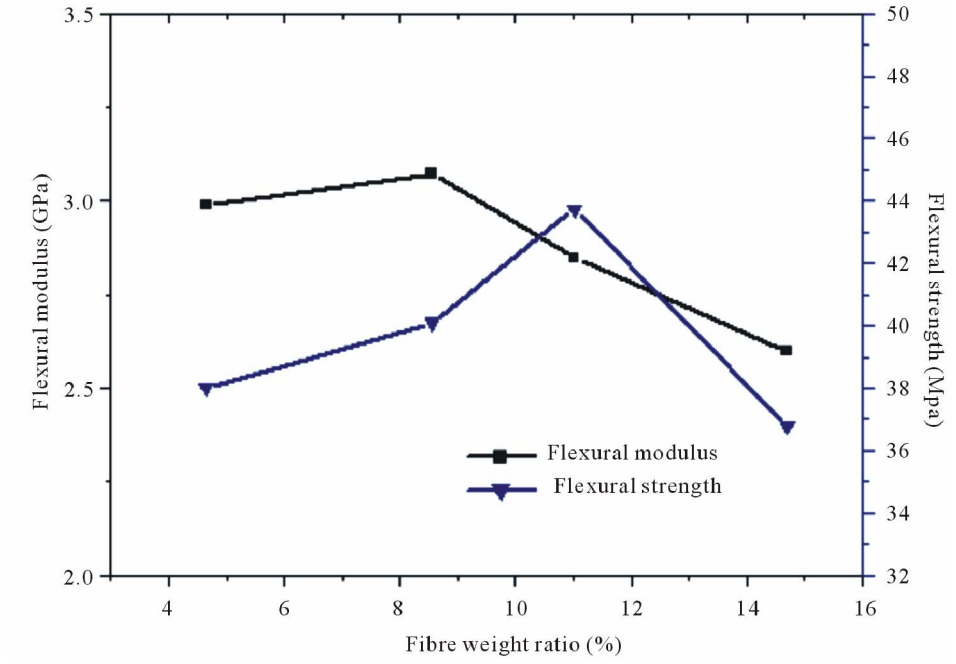

Figure 4. Influence of Luffa fibers weight ratio onto mechanical proprieties of ComLCBC combined.

The effect of fiber weight ratio on the flexural elongation at break of various Luffa composites is shown in **Figure 6**. The elongation at break values of the composite ComLEMat are higher than the results obtained with composites reinforced with short Luffa fibers (ComLEBC and ComLCBC).

This result indicates the variation of the mechanical behaviour with the change of the reinforcement structure. In the first step, the flexural elongation at break increased and reached a maximum for a fiber weight ratio at about 10% for the composite reinforced with mat of Luffa fibers. In the second step, the elongation at break dropped off steadily. For the polyester composites reinforced with short Luffa fibers, the elongation at break increases to maxima at about 11% of fiber weight ratio. Thereafter, the elongation at break decreases slightly.

As shown also in **Figure 5** that the two curves of elongation at break for the polyester composites reinforced with short Luffa fibers have the same shape. The elongations at break values of ComLCBC combined are higher

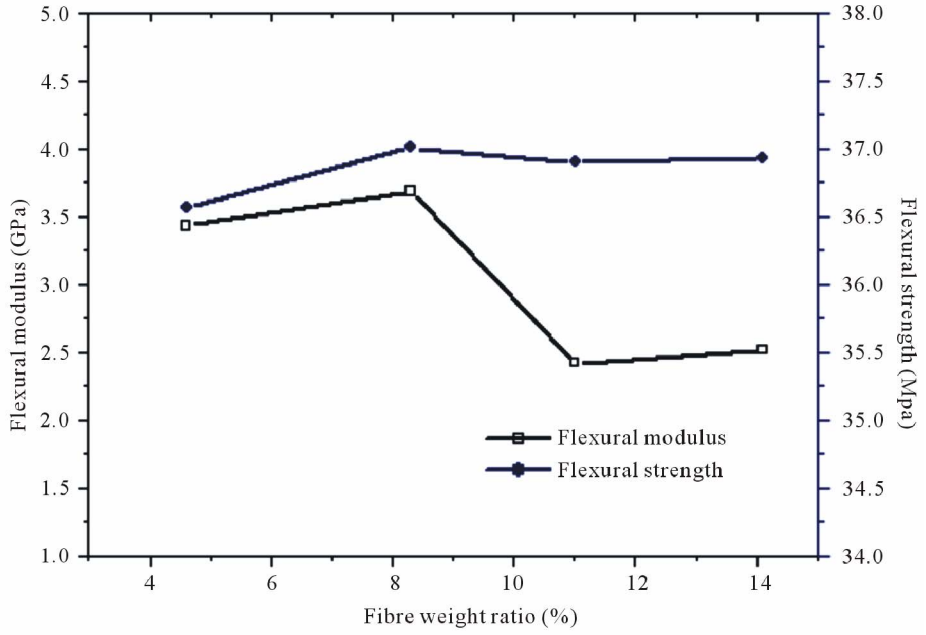

Figure 5. Influence of Luffa fibers weight ratio onto mechanical proprieties of ComLEBC combined.

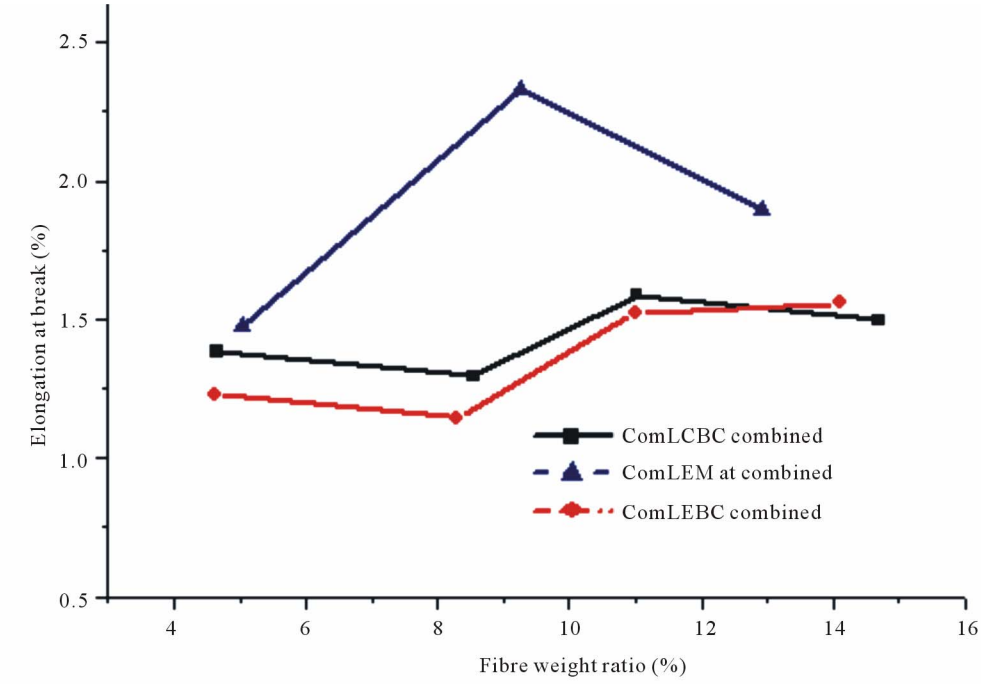

Figure 6. Relationship between the reinforcement structures and elongation at break of Luffa-polyester composite.

than those of ComLEBC combined.

4. Conclusions

In this study the influence of fiber modification and the reinforcement structure on the flexural proprieties of Luffa-polyester composite were investigated. The variation of these properties with the rise of fiber weight ratio was also studied. It is clear that the acetylating and cyanoethylating treatments of Luffa fibers enhanced the adhesion between fiber and matrix. Those treatments replaced the free hydroxyl groups of cellulose structure by acetyl and cyanoethyl groups respectively. Nevertheless the combined process treatment decreased the flexural proprieties of composite compared to the composite reinforced with untreated fibers. This result is due to the fiber degradation under several chemical conditions.

The uses of various reinforcement structures were investigated. The enhancement of elongation at break of the composite reinforced by natural mat was proved.

The study of the relationship between fiber weight ratio and the flexural proprieties of Luffa-polyester composites is confirmed by many research reported elsewhere.

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