

Influence of water saturation on fracture toughness in woven natural fiber reinforced composites

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Abstract—Woven sisal textile fiber reinforced composites were used to evaluate fracture toughness, tensile and three-point bending. The water absorption testing of all specimens was repeated five times in this study. All specimens were immersed in pure water during 9 days at room temperature, and dried in 1 day at 50°C. Two kinds of polymer matrices such as epoxy and vinyl-ester were used. Fractured surfaces were taken to study the failure mechanism and fiber/matrix interfacial adhesion. It is shown that it can be enhanced to improve their mechanical performance to reveal the relationship between fracture toughness and water absorption fatigue according to different polymer matrices. Water uptake of the epoxy composites was found to increase with cycle times. Mechanical properties are dramatically affected by the water absorption cycles. Water-absorbed samples showed poor mechanical properties, such as lower values of maximum strength and extreme elongation. The K_{IC} values demonstrated a decrease in inclination with increasing cyclic times of wetting and drying for the epoxy and vinyl-ester.

Keywords: Woven sisal textile fiber reinforced composites; water absorption fatigue; fracture toughness; deformation; mechanical property.

1. INTRODUCTION

In the past decades, growing environmental awareness has resulted in a renewed interest in the use of natural materials for different applications [1–3]. New and stricter environmental policies have forced industries like the automotive, packaging and construction industry to search for new materials that can substitute the traditional composite materials consisting of a plastic matrix and inorganic filler as reinforcement. The most used composites nowadays are the glass fiber

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filled thermoplastics, but also different inorganic fillers like aramid or carbon fibers are widely used. However, inorganic fibers present several disadvantages, for example their non-biodegradability, the abrasion that they produce in the processing equipment and the health problems that they cause to workers from skin irritations they cause during processing and handling. Sisal fiber is one of the most widely used natural fibers and is very easily cultivated. It has short renewal times and grows wild in the hedges of fields and railway tracks [2]. Nearly 4.5 million tons of sisal fibers are produced every year throughout the world. Tanzania and Brazil are the two main producing countries [2]. At present, sisal fiber is mainly used as ropes for the marine and agriculture industry [4, 5]. Other applications of sisal fibers include twines, cords, upholstery, padding and mat making, fishing nets, fancy articles such as purses, wall hangings, table mats, etc. A new potential application is for manufacture of corrugated roofing panels that are strong and cheap with good fire resistance [3].

Unfortunately, several disadvantages of natural fibers such as thermal and mechanical degradation during processing can make them undesirable for certain applications [6, 7]. Lack of good interfacial adhesion and poor resistance to moisture absorption make the use of natural fiber reinforced composites less attractive. This problem can be overcome by treating these fibers with suitable chemicals [8, 9]. However, several considerations have to be taken into account in the design of natural fibers composites. One of the most important issues is the degradation behavior of the composites exposed to environmental conditions such as humidity, sunlight or microorganisms. The poor resistance of the fibers to water absorption can have undesirable effects on the mechanical properties and the dimensional stability of the composites [10–12]. Therefore, it is important to study in detail the water absorption behavior in order to estimate not only the consequences that the water absorbed may have, but also the durability of natural fibers composites aged under water. Moisture penetration into composite materials is conducted by three different mechanisms [13]. The main process consists of diffusion of water molecules inside the microgaps between polymer chains. The other common mechanisms are capillary transport into the gaps and flaws at the interfaces between fibers and polymer, because of incomplete wettability and impregnation and transport by microcracks in the matrix, formed during the compounding process [14, 15]. In spite of the fact that all three mechanisms are active jointly in the case of moisture exposure of the composite materials, the overall effect can be modeled conveniently considering only the diffusional mechanism.

Fundamental considerations of design and applications for a composite material require a basic understanding of how the fracture process is initiated and progresses to final failure. As we know, sisal fiber reinforced polymer composite is a novel material. Therefore, the investigation of its fracture behavior is essential for its future application and design. Usually a composite is expected to sustain both static and dynamic loads without the danger of sudden catastrophic failure. The

local response of the fiber–matrix interface within the composite during fracture is particularly important.

This study is part of a series of work related to the effects of water absorption on mechanical properties of sisal fiber reinforced composites. This paper is concerned with the durability of sisal fiber composites aged under distilled water and exposed to controlled cycles of wetting and drying. The durability of sisal composites was measured as the loss of strength in cyclic time.

2. EXPERIMENTAL

The natural sisal fibers used in this investigation were extracted from five years old sisal plant growing in southern China. The diameter of the fiber varied from 100 to 250 μm . The average diameter of the fiber, calculated from the fraction of fibers of different diameter in a sample, was about 140 μm . The sisal fiber contains 57% cellulose, 16% hemicelluloses, 11% lignin and others by weight. The tensile strength, modulus and failure strain of the fiber were 400–700 MPa, 9–20 GPa and 5–14%, respectively. The used materials were reinforced by two plies of woven roving sisal fiber and the fiber volume fraction was about 60%. The sisal textile used in this study is plain woven and has the same properties in the orthogonal direction. The used material is twisted 3 times per inch. An illustrative view on structure of the ply of woven sisal fiber is given in Fig. 1.

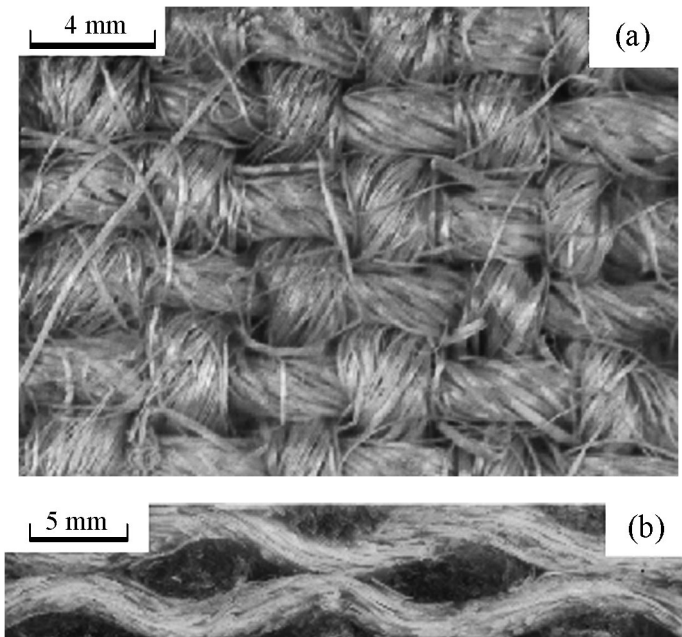


Figure 1. Photographs of woven sisal fiber composites. (a) Front view; (b) side view.

To produce a higher fiber–matrix adhesion, a fiber modification with silane was applied. For this procedure the fibers first had to be dewaxed in an alcoholic solution for 24 h by removing the weaving size; subsequently the fibers were washed with distilled water. The following silane treatment similarly was carried out in an alcoholic solution with a silane content of 2 wt% for 24 h at 23°C. The coupling agent used was a γ -glycidoxypropyltrimethoxysilane (SG6720, AP Resource Co.) with the chemical constitution of $\text{CH}_2\text{CHCH}_2\text{O}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$. The silane application was finished with a 4 h drying process in a vacuum oven at 75°C. For the preparation of composites, two different kinds of resin were used: an epoxy (BS-745A/B, DR Enterprise Co.) and vinyl-ester (R3791N/928CA, KK Chemical Co.) resin. The tested composites were made of woven roving fiber, which were embedded in a resin by using the resin transfer molding (RTM) technique. The sisal fibers were aligned in a Teflon mold, and then liquid resin mixture was poured into that mold. The curing was performed at 120°C for 2 h.

Rectangular specimens, 76.2 mm long, 25.4 mm wide and 3.2 mm thick, were cut from the composites to study the kinetics of water absorption. The specimens were then immersed in distilled water at $20 \pm 1^\circ\text{C}$. At regular intervals, the specimens were removed from distilled water, wiped with filter paper to remove surface water and weighed with a precision analytical balance (XT 320M, Precisa Inst. AG, Swiss) with 0.1 mg resolution. In this study, the results of tests on samples that had been exposed to cycles of wetting for 9 days until saturation and drying for 1 day at 50°C to simulate the natural extreme weather are presented. The durability of the composites is discussed using the results of tensile, bending and fracture toughness tests carried out before and after 5 cycles of wetting and drying, and observations of fracture surfaces in a scanning electron microscope (SEM, JSM 6400, Jeol Ltd., Japan). Two kinds of tests were performed in the dried samples in order to determine the influence of the water absorption cycles on the mechanical properties following the ASTM D 638-90, D790-90 and D 5045-96. The 1/4T compact tension (CT) specimens were used for the fracture toughness tests with 32.0 mm width. The standard dumbbell-shaped specimens were used for tensile tests with 25.0 mm gauge length and rectangular specimens of $40 \times 12.5 \times 3.2$ mm dimensions for 3-point bending tests as shown in Fig. 2. Tensile, bending and fracture toughness test were carried out crosshead speed of 0.05 mm/min using a universal testing machine (8516, Instron Co., USA). The woven fabrics of sample were tested in the direction of load ($0^\circ/90^\circ$ oriented).

3. RESULTS AND DISCUSSION

3.1. Water absorption behavior

The amount of water absorbed in the composites was calculated by the weight difference between the samples exposed to distilled water and the wetting and drying cycled samples. Figure 3 shows the percentage of water absorption plotted

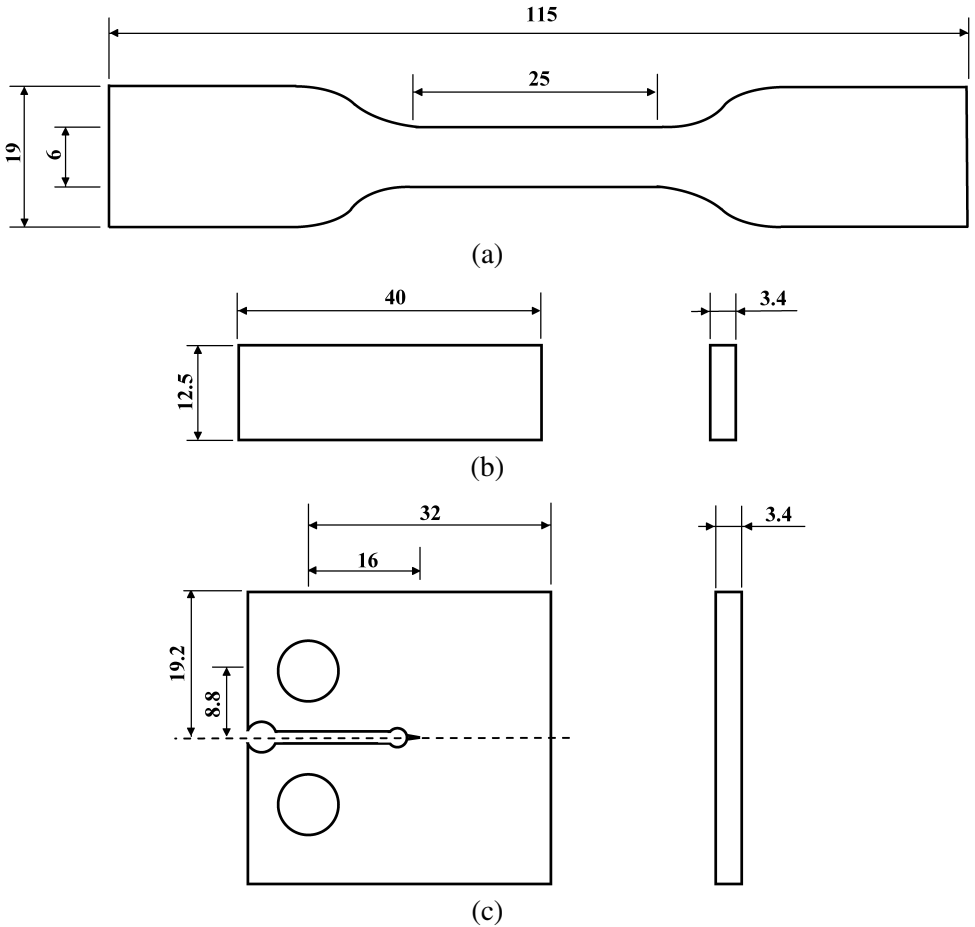


Figure 2. Specimen configuration for (a) tensile, (b) bending and (c) fracture toughness test (unit: mm).

against time for the cycled samples. Figure 3 is for 5 cycled specimens that were immersed for 216 hours (9 days) and then dried for 24 h (1 day) in the epoxy and vinyl-ester composites. The same behaviour was observed: the samples absorbed water very rapidly during the first stages (0–260 h for epoxy and 0–150 h for vinyl-ester) reaching a certain value, the saturation points, where no more water was absorbed and the content of water in the composites remained the same. The hydrophilic character of natural fibers is responsible for the water absorption in the composites, and therefore a higher content on fibers leads to a higher amount of water absorbed. The matrix has little effect on the amount of water absorbed as pure resin composites like an epoxy and vinyl-ester [13]. The cycles of wetting and drying process also has an influence on the water absorption curves. It can be concluded that the cycling process increases the water uptake of the composite materials. When the wetting and drying process is increased, the amount of water

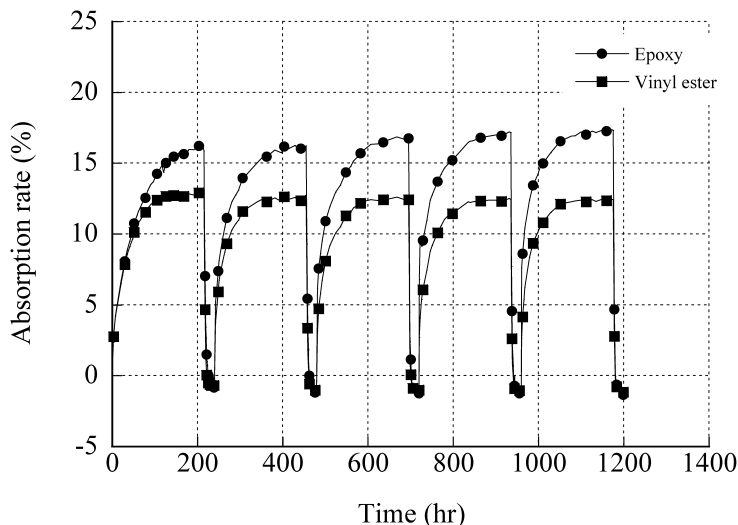


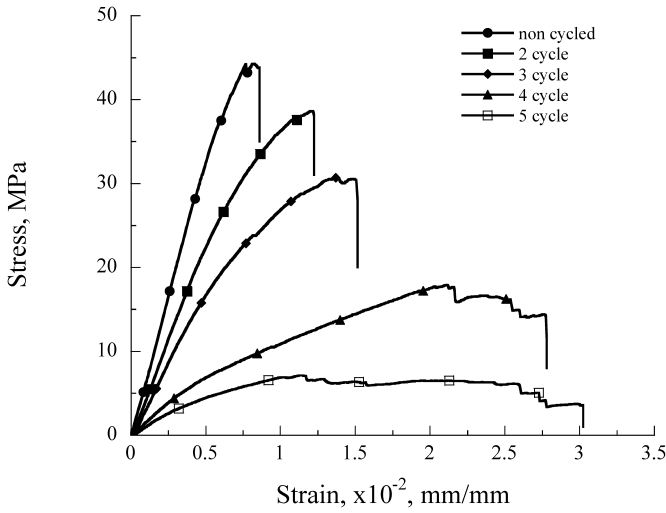
Figure 3. Water absorption curves for sisal composites at 20°C.

uptake is slightly increased in the same wetting period. For the epoxy composites, the absorption rate is increased from 16.2 wt% to 17.4 wt% as the cycling process increased from 1 to 5th wetting stage. But in vinyl-ester case, rate is almost same. When the drying process is increased from 1 to 5th drying stage, the absorption rate is decreased to from 0.62 wt% to -1.12 wt% for the composites with epoxy and vinyl-ester, as shown in Fig. 2. This weight reduction may be due to the extracted cellulosic fibers from the polymeric matrix [10].

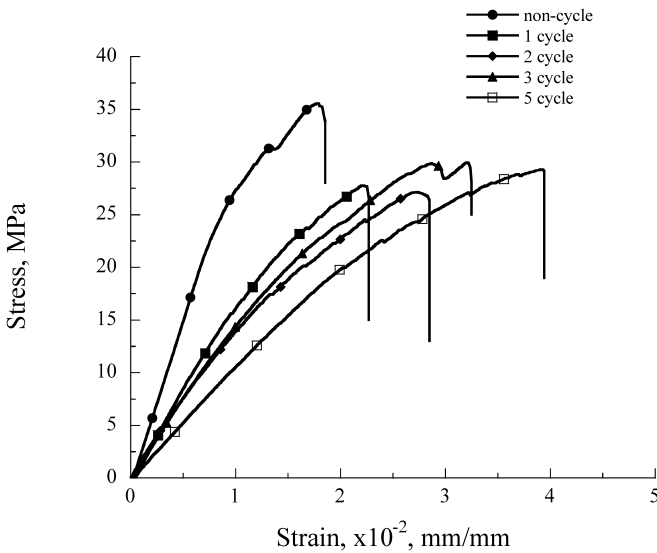
3.2. Effect of absorption cycles on mechanical properties

Tensile and bending tests were performed in the epoxy and vinyl-ester samples before (0 cycles) and after water absorption with the cycled stages. Figures 4(a) and 4(b) present the results of tensile tests for the epoxy and vinyl-ester composites, respectively. In general, the tensile strengths of these materials decrease after the moisture uptake, due to the effect of the water molecules, which change the structure and properties of the fiber, matrix and the interface between them [14–16]. Once the moisture penetrates inside the composite materials, the fibers tend to swell. The matrix structure can also be affected by the water uptake by processes such as chain reorientation and shrinkage. The aging under conditions of cyclic wetting and drying may also lead to the degradation of natural fibers by a hydrolysis mechanism [12]. Water absorption and their resulting effects contribute to the loss of compatibilization between fibers and matrix, which results in debonding and weakening of the interface adhesion.

Concerning the stress at maximum load, their values are gradually reduced after cyclic water absorption for the epoxy as shown in Fig. 4(a). This effect is particularly evident for the composites cycled 4 and 5 times, in which the maximum



(a)



(b)

Figure 4. Stress and strain behaviors according to various cyclic times. (a) For sisal epoxy composites; (b) For sisal vinyl-ester composites.

stress gets drastically reduced. For the vinyl-ester, the values of maximum stress are decreased with cyclic water uptake as shown in Fig. 4(b), but the maximum stresses of cyclic composites are distributed within the limits of 27.0–29.8 MPa without wide dispersion.

The changes in the strain at failure with cyclic aging are more complex. It has been reported that water molecules act as a plasticizer agent in the composite material, which should lead to an increase of the maximum strain for the composites after

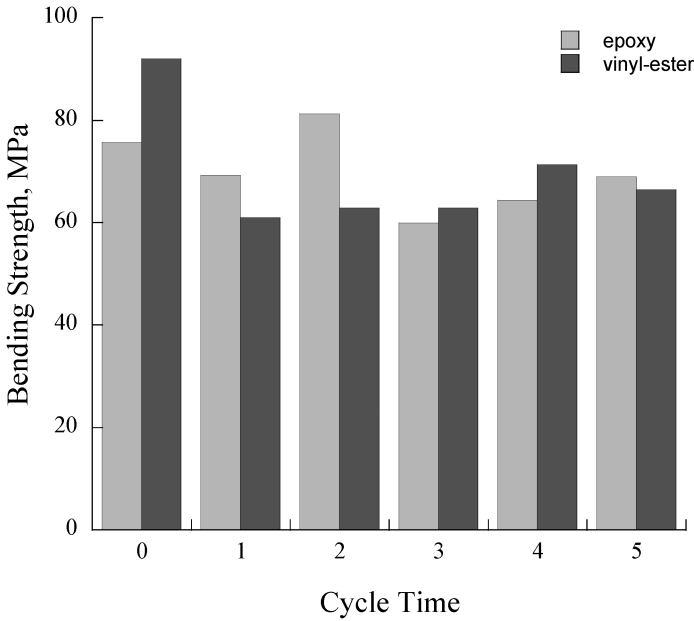


Figure 5. Bending strength behaviors according to various cyclic times.

water absorption [16]. Such an increase is shown for composite with the epoxy resin immersed in water at 4 and 5 times cycling as shown in Fig. 4(a); in particular, for the 5 times cyclic composite, the maximum strain is 3.1%. For the vinyl-ester composites, the maximum strain is also gradually increased after cyclic water absorption with the increase of cyclic times as shown in Fig. 4(b). In this case, the plasticizer effect of water in the composite structures is superior to the degradation effect, so the maximum strain is increased in spite of indistinct stress behavior of cyclic composites.

Figure 5 presents the results of bending test for the epoxy and vinyl-ester composites. The value of bending strength at maximum load increased in non-cyclic time, after immersion cycle time; however, the values were not different between immersion cycle time. In 1 and 2 times cycling, the value of bending strength for composite with epoxy resin was bigger than composite with vinyl-ester resin.

3.3. Effect of absorption cycles on fracture toughness

The critical stress intensity factor in mode I fracture (K_{IC}) as a function of the wetting and drying cycle times is shown in Fig. 6 for the epoxy and vinyl-ester. Here, K_Q values were calculated according to ASTM D 5045-96 as shown in equations (1) and (2).

$$K_Q = \frac{P_Q}{BW^{1/2}} f\left(\frac{a}{W}\right), \quad (1)$$

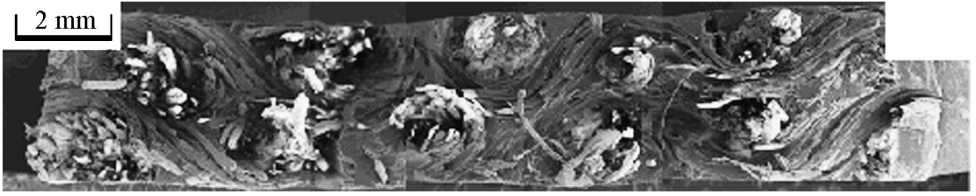


Figure 6. Fracture photographs of compact tension specimens.

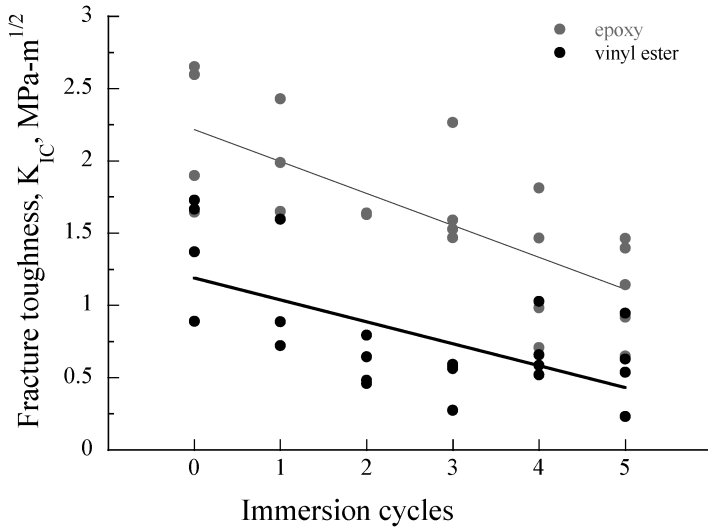


Figure 7. Distribution of critical stress intensity according to immersed cyclic times.

$$f\left(\frac{a}{W}\right) = \frac{(2+x)(0.886 + 4.64x - 13.32x^2 + 14.72x^3 - 5.6x^4)}{(1-x)^{3/2}}, \quad (2)$$

where P_Q is load corresponding to a certain crack length, B specimen thickness, W is the specimen width, a the initial notch length (produced by saw and razor blade; 16 mm), and $f(a/W)$ is the specimen geometry related correction factor. The notches were machined perpendicularly to the direction of woven fibers as shown in Fig. 6. The K_Q value determined by compact tension test characterizes the resistance of a material to fracture in a neutral environment in the presence of a sharp crack under severe tensile constraint, such that the state of stress near the crack front approaches plane strain, and the crack tip plastic region is small compared with the crack size and specimen dimensions in the constraint direction [17].

The K_{IC} values demonstrate a decrease in inclination with increasing cyclic times of wetting and drying for the epoxy and vinyl-ester. Two curves of the average K_{IC} values show the parallelism with each other in a downward direction with the increase of cyclic times as shown in Fig. 7. The effect of crack orientation on K_{IC} values is not clear. The K_{IC} values of the epoxy and vinyl-ester ranged from

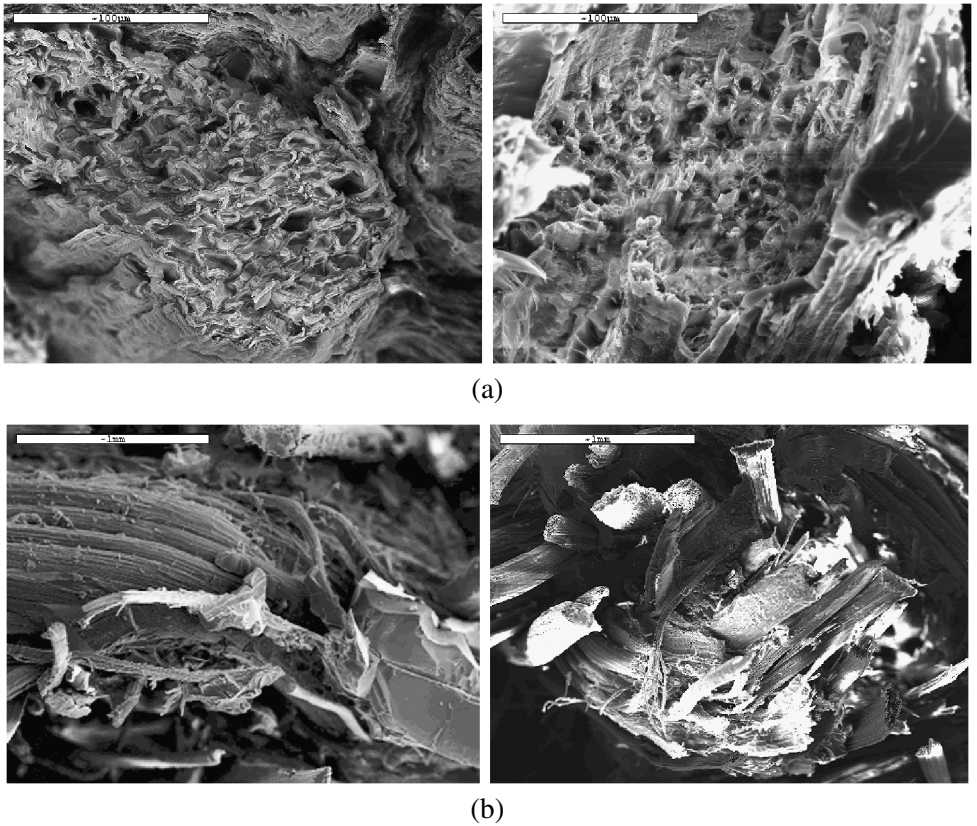


Figure 8. SEM photographs for sisal composites before (a) and after (b) cycles of wetting and drying.

0.64 to 2.66 MPa m^{1/2} and from 0.22 to 1.75 MPa m^{1/2}, respectively, in the cyclic composites.

3.4. Fracture mechanism in absorption cycles

SEM photographs clearly show the degradation of the fibers by the water absorption cycles. Figure 8 shows the composite structure before and after water absorption cycles, where the loss of adhesion between fiber and matrix, characterized by the appearance of voids can be noticed. The fibers appear seriously degraded with complete lack of resin layer and the microfibrils can be clearly observed.

The ruptured ends of the sisal fibers were not plane with some parts pulled-out of sisal fibers. This is the feature of microcell multi-cracking followed by interfacial debonding. From Fig. 8(a) it may be seen that the fiber section is neither circular nor regular. A bundle of microcells was pulled away, which left a hole. A pulled-out microcell can be seen. The fiber and resin interface seems to be moderate because the interfacial debonding took place at the sisal fiber contour. A microcell was covered with a layer of cuticle which had a thickness. The cuticle was very flexible and very deformed, and its adhesion to the microcell was not strong. The

microcell was ruptured by decoiling, splitting, torsion, etc. The decoiling is a specific characteristic of sisal fibers. In fact, inside the wall of microcells, there exist many short aligned microfibrils, which are oriented in a direction different from that of sisal fiber [18]. Therefore, the microcell wall is also a composite structure of lignocellulosic material reinforced by helical microfibrillar bands of cellulose. It was observed that some microcells, particularly near the fiber and resin interface, were compressed transversely and so split longitudinally along its length.

4. CONCLUSIONS

This study presented here described the influence of wetting and drying cycles on the mechanical properties of woven sisal fiber reinforced epoxy and vinyl-ester composites. The water absorption characteristics of sisal composites were studied with cycled conditions. Water uptake of the epoxy composites was found to increase with cycle times. Mechanical properties are dramatically affected by the water absorption cycles. Water-absorbed samples showed poor mechanical properties, such as lower values of maximum strength and extreme elongation. The K_{IC} values demonstrate a decrease in inclination with increasing cyclic times of wetting and drying for the epoxy and vinyl-ester. The fibers appear seriously degraded with complete lack of resin layer and the microfibrils can be clearly observed.

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