CHAPTER 8:

TEXTILE COMPOSITES BASED ON NATURAL FIBERS

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1. INTRODUCTION

Developments in textile technologies such as weaving, knitting and braiding have resulted in the formation of textile composites that have superior mechanical properties. Woven fabrics are attractive as reinforcements since they provide excellent integrity and conformability for advanced structural applications. The driving force for the increased use of woven fabrics compared to their non-woven counterparts are excellent drapeability, reduced manufacturing costs and increased mechanical properties, especially the interlaminar or interfacial strength. The interconnectivity between adjacent fibers in the textile reinforcement provides additional interfacial strength to supplement the relatively weak fiber-resin interface. The non delamination characteristics of three-dimensional braided composites under ballistic impact also make them possess considerable potential in ballistic protection applications [1]. Formation of different textile preforms is an important stage in composite technology.

Plain, twill, satin, basket, leno and mock leno are some commonly used woven patterns for the textiles which are employed as reinforcements in making composites. The woven characteristics are shown in Figure 1 and their respective properties are summarized in Table 1.

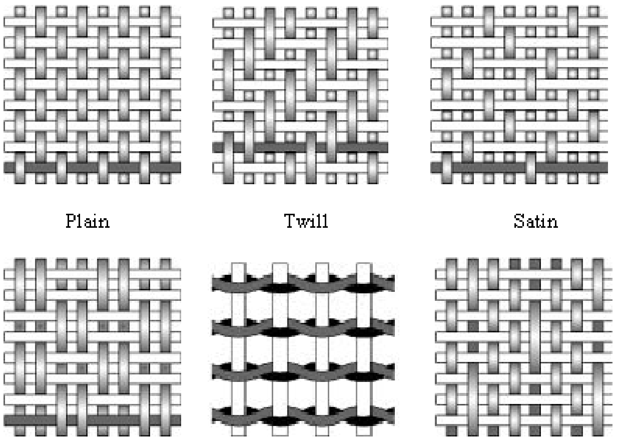
Textile structural composites are finding use in various high performance applications recently [2, 3]. Among them, three-dimensional fiber-reinforced polymer composites made by the textile processes of weaving, braiding, stitching and knitting were found to have tremendous potential for improving the performance of composite structures and reducing their cost of manufacture. The current applications of three-dimensional composites, including examples in the aerospace, maritime, automotive, civil infrastructure and biomedical fields are also enumerated.

In this chapter, textile composites based on natural fibers are investigated, which includes the manufacuring techniques, fracture and mechanical properties and other behaviors. Consolidation and permeability of the textiles based on natural fibers are specially addressed. Hybridization of different kinds of natural fabrics as reinforcement in making composites are also reported. Recent studies on biodegradable natural fiber composites are introduced.

Table 1. Properties comparison of the fabrics with different woven styles

Property	Plain	Twill	Satin	Basket	Leno	Mock leno
Stability	****	***	**	**	*****	***
Drape	**	****	*****	***	*	**
Porosity	***	****	*****	**	*	***
Smoothness	**	***	*****	**	*	**
Balance	****	****	**	****	**	****
Symmetrical	*****	***	*	***	*	****
Crimp	**	***	*****	**	**/*****	**

*****=excellent, ****=good, ***=acceptable, **=poor, *=very poor



Plain Twill Satin

Basket Leno Mock Leno

Figure 1. Some typical woven styles used as reinforcements in making composites.

2. TEXTILE COMPOSITES

2.1 Random Non-woven Mat

Random non-woven mats usually show good drape (the ability to adapt to complex geometries) and wet out characteristics and are easy to process, but they demonstrate only very limited mechanical properties in making composite laminates due to the short fiber length and the high resin content. Therefore, they are seldom used for high performance applications. However, mats are still used in applications requiring a high resin content layer without high mechanical stability, like the outer skin of boat hulls. Therefore, a brief investigation on natural fiber mats reinforced composites is conducted in this chapter.

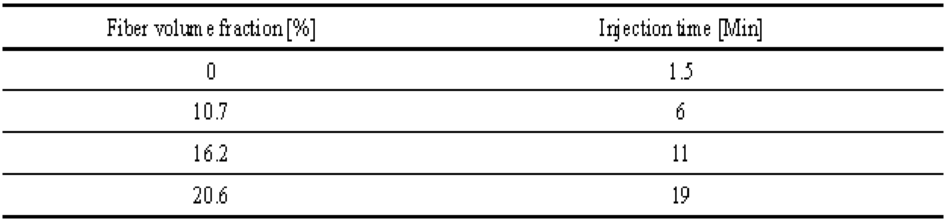
2.1.1 Processing of random non-woven mats and their composites

Random non-woven mats are also called chopped strand mats (CSM). They consist of chopped fiber strands with a length commonly varying between 25 mm and 300 mm according to the type of mat. The strands are randomly orientated and bound together either by a powder or an emulsion binder, or by stitching. The latter alternative is often selected to avoid chemical compatibility problems between the emulsion or the powder binder and the resin.

Recently, a new type of non-woven mat which is useful for a wide variety of purposes, including forming reinforcement of fiber reinforced composites was produced. It possesses some advantages over conventional chopped strand mats [4]. The mat is preferably made by the foam process (but may be made by the liquid process), and at speeds well in excess of 60 meters/min (normal production rate for conventional mats is between 20-30 meters/min). The construction is substantially uniform even at low density (e.g. 100 g/m^2 or less). At least 20% (preferably at least 85%) of the fibers are in fiber bundles with between 5-450 fibers per bundle. The fibers (typically at least 85%) have a length between 5-100 mm, preferably 7-50 mm, substantially the same as the length of the fiber bundle they are in. The fibers are preferably held in the bundles by substantially non-water soluble sizing, such as epoxy resin. The fibers in the bundles typically have diameters of approximately 7-500 μm, preferably about 7-35 μm. The bundles may comprise at least 10% reinforcing fibers. The mats made by this technique overcome the limitations of the conventional mats, such as the relative thick/dense distributions of reinforcement and uneven surface configurations.

Resin transfer molding (RTM) is the major processing method to make random non-woven natural fiber mats reinforced polymer composites. Rouison, Sain and Couturier reported making this kind of composites by RTM [5]. First, the surfaces of the mold were cleaned and coated with mold release agent. Once these coatings were cured, layers of natural fibers'mats having the mold's size were placed in the mold cavity. Then the mold was tightly closed and a vacuum of 725 mm of mercury was applied. At this point the fibers were dried for 2h by circulating water at 55^{o}C. This drying process before the resin's injection permitted to obtain a good wetting of the fibers as well as to avoid any formation of gas bubbles during curing. The mold was then cooled down with cold water. In the meantime the resin was mixed with the initiator and placed in the injection pot. From there the resin was injected in the mold with compressed air at a constant gauge pressure of 0.17MPa. Once the resin was observed at the outlet, the vent port was closed. A small flask was placed between the vent port and the tap for safety, to prevent any resin from flowing to the tap water. The resin was left flowing at the inlet for 5 min more to make sure that the mold was filled completely. Then the inlet ports were closed as well and hot water at constant temperature was circulated in the mold. The composite was cured under these conditions for 1 h. It has been seen that the wetting of the fibers in mat form was very good. The resin injection time was observed to increase dramatically at high fiber contents due to the low permeability of the mat (see Table 2). Keeping a constant mold temperature is the key to obtain fast and homogeneous curing of the part.

Table 2. Injection time of hemp/kenaf/polyester at various fiber content under a fixed pressure [5]



Fiber volume fraction [%]	Injection time [Min]
0	1.5
10.7	6
16.2	11
20.6	19

Bos *et al*. reported their technique to make natural fiber mat thermoplastic (NMT) composites [6]. First, flax fibers and PP fibers were stirred together in a water/ethanol (1:1) mixture. In order to get a stable dispersion of flax and PP fibers, a mixture of water and ethanol with a density of about 0.9 g/cm^3 was used. This was because the PP fibers rose very quickly to the liquid surface in pure water. A good distribution of the flax fibers in the PP fibers would be hindered. For the compatibilised materials, the compatibiliser was added as a powder during the wet mixing process. After drying the circular flax/PP fiber sheet at 60^{o}C for 24 h, the fluffy mat of flax and PP fibers was consolidated in a hot-press at 200^{o}C and 40 bar pressure for 15 min. Composites with fiber volume fractions of 20 and 40% (which equals 28 and 51 wt%, assuming a fiber density of 1.4 g/cm^3) have been made.

2.1.2 Properties

- Stochastic properties of random natural non-woven mats

It is easy to understand the large variations of properties of natural fiber non-woven mats reinforced composites due to the normally non-uniform distribution of natural fibers in the composites. Therefore, it is difficult to describe the properties of natural fiber non-woven mats reinforced composites accurately. Kaveline et al. proposed a method which could quantitatively estimate the properties of this kind of composites quite well [7]. The technique was based on spectral analysis of the digital image of the mats. Because natural fiber non-woven mats reinforced composites are non-homogeneous materials, some definitions of heterogeneity parameters which include the size and intensity of fiber cluster/voids for non-woven mats were proposed. The properties of the stochastically distributed clusters/voids in the fiber mats were determined by subdividing them into groups, described by harmonics. The groups contained clusters/voids with a certain range of sizes. The ranking of clusters/voids by size was performed with the help of spectrogram of fibers mat image. An example of a spectrogram is shown in Figure 2. The mean and scatter parameters for dimensions and intensity of fibers clusters/voids within the groups were estimated. A correlation technique was applied in order to obtain the representative for a particular mat data series when the heterogeneity parameters of the mats based on dominant harmonics are given.

The developed procedure was successfully applied to a number of different types of natural fiber (flax, coir etc.) and glass fiber mats. It appeared to be invariant to the nature of the fiber mats under investigation. This means that the developed technique is not only suitable for natural fibers non-woven mats but also for synthetic randomly distributed fibers mats as well. The results of heterogeneity parameters analysis can be directly used for further simulation of natural and synthetic mats' heterogeneity, designing of experiments and finite element simulations of composite materials reinforced with randomly distributed fibers.

- Mechanical properties of natural fiber mats reinforced composites

When Rouison et al. studied the curing simulation of natural mats reinforced composites by RTM technique, some mechanical properties of hemp/kenaf fiber mats reinforced unsaturated polyester composites were also reported [5]. As expected, both tensile strength and flexural strength increased with the increase of fiber volume fraction. The results can be seen from Figure 3, which shows the changes of tensile strength and flexural strength with the fiber volume fraction.

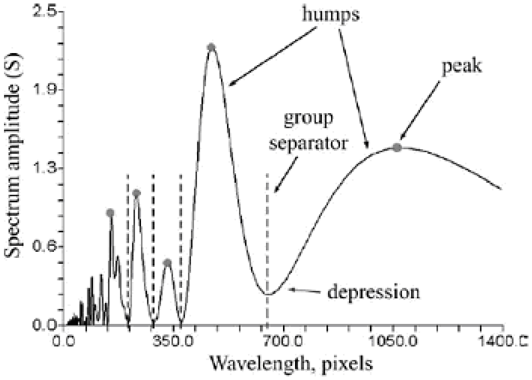


Figure 2. An example of a spectrogram [7]

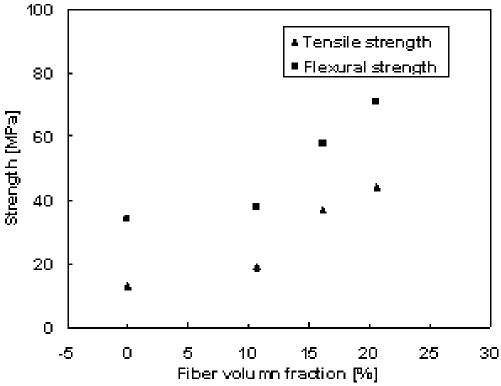


Figure 3. Mechanical properties of hemp/kenaf fiber mats reinforced unsaturated polyester composites [5]

2.2 Textile Forms

2.2.1 Textile manufacturing

In applications where more than one fiber orientation is required, a fabric combining different fiber orientations is useful. Woven fabrics are produced by the interlacing of warp fibers and weft fibers in a regular pattern or weave style. The fabric's integrity is maintained by the mechanical interlocking of the fibers. Drape (the ability of a fabric to conform to a complex surface), surface smoothness and stability of a fabric are controlled primarily by the weave style. An important criterion in determining the properties of textile composites is the weave pattern. Therefore, weaving natural fibers into different textile forms is important to their final properties.

Basically, the manufacturing of natural fiber textile can be summarized as the following steps: growing plants, harvesting, stem or leave collection (depending on where the fibers are extracted), water retting, fiber separation, fiber hackling (several times), fiber weaving. The detailed fiber weaving techniques have been fully developed by the textile or fabric industry [8, 9].

In this chapter, we take banana fiber textile manufacturing as an example. Sapuan, et. al. summarized the manufacturing of banana fiber textile as the following three steps [10]:

A) Abstracting natural continuous banana fibers.

The abstracting processes for the continuous natural banana fibers consist of two major procedures. Firstly, the banana fibre is abstracted from the fully grown trunk after the fruit has been plucked in order to avoid wasting of the fruits. Then, the banana trunks are placed under the sunlight for the drying process within two weeks. After the banana trunks are dried, they are soaked in the water for another two weeks. The drying time is quite important in this process. If less than two weeks, the fibre cannot be separated from the unused cell, whereas if the soaking time is more than two weeks, it is a waste of time.

B) Thinning fibers.

After banana fibres are fully dried, they must be cut horizontally in order to get the average width of fibre between 10 and 15 mm. The longest and the shortest fibre are 137 and 77 cm in length, respectively. A dried banana fibre should be thinned using a roller machine in order to ease weaving process.

C) Weaving the continuous fibers into fabrics.

In order to orientate the fiber in the composite material, dried banana fibers need to be made into monofilament fibers or be twisted before woven. The method for weaving the banana fiber for making a specimen is well detailed by Jones [11]. Sapuan and Maleque also reported making a household telephone stand with woven banana fibers [12].

2.2.2 Consolidation and permeability of natural fiber textile

Consolidation and permeability of woven fabrics are very important for obtaining good quality composites. Most of the fiber textile reinforced polymer composites are manufactured by RTM technology, which is a kind of liquid molding process. The key for liquid molding process in making fiber reinforced composites is to let the resin impregnate preform completely and before resin cures so that undesirable flaws, such as incomplete filling, non-uniform wetting and voids can be avoided. Permeability is a parameter used to describe the ease of liquid resin impregnating preforms. It is a complex function of woven pattern, tow structure, packing characteristics and intra-tow properties of the reinforcing fabrics [13-15]. Therefore, the studies of textile composites based on natural fibers would not be complete without considering the consolidation and permeability properties of natural fiber textiles.

Li studied the permeability properties of plain woven sisal textile by adopting linear flow of the liquid resin injected from a side inlet based on Darcy’s law to measure the permeability values [16]. The effects of fiber surface treatments and fiber volume fraction on the permeability of sisal textile were reported. The two kinds of fiber surface treatment methods were permanganate treatment and silane treatment. Vinyl ester was used as the matrix.

The permeability values of sisal textile before and after fiber surface treatments are listed in Table 3. Comparisons of sisal textile with synthetic preforms, such as woven glass and carbon fabrics, which have similar fiber volume fractions are also made.

It is clearly shown that fiber surface treatments have great effect on the permeability of sisal textile, especially from silane treatment. The permeability value has been increased more than 2 times. It is easy to understand that the high permeability is beneficial to improve the quality of composite product and working efficiency. Table 3 also clearly indicates that sisal fiber has better processing properties than synthetic fibers by having larger permeability values.

Usually, the flow of resin through the preform includes inter-bundle flow and intra-bundle flow [17-19] which is illustrated in Figure 4. The higher permeability of sisal textile compared with glass and carbon textile was caused by the larger fiber diameters of sisal fibers. From the stacking theory, it is known that the larger the fiber diameters are, the more the cavities between fibers exist. Larger fiber diameters will facilitate the inter-bundle flow which is the predominant flow during the infiltration process because inter-bundle flow is faster than intra-bundle flow and more liquid resin flow through inter-bundles.

Table 3. Permeability of sisal and synthetic textiles [16]

Fiber	Fiber volume fraction [%]	Permeability [$10^{-10}m^2$]
Untreated sisal	32	6.38
Permanganate treated sisal	32	7.26
Silane treated sisal	30	15.4
Woven glass fabric	30	5.66
Woven carbon fabric	36	5.12

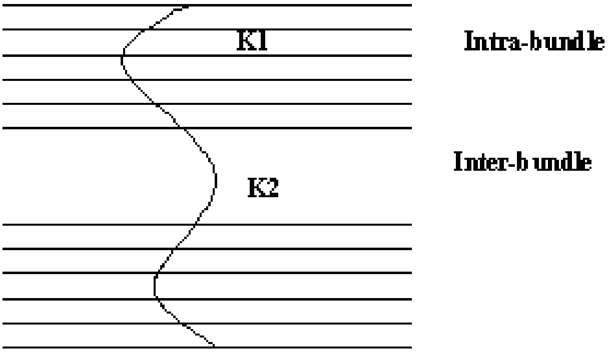


Figure 4. Intra-bundle and inter-bundle flows

As reported, permanganate, as an oxidant, can etch sisal fiber surface [20]. Scanning electronic micrograph of a permanganate treated sisal fiber shows that a fiber bundle was broken down into many sub-fibers, hence increasing the flow channels for the liquid resin (Figure 5). This enables the flow easier and faster, and leads to a higher permeability. But the increased flow channels belong to intra-bundle flow which would not affect the whole filling process a lot. So, the permeability of sisal textile was improved after permanganate treatment compared with untreated ones, but not too much.

Silane treatment method has different mechanism with permanganate treatment in improving interfacial properties between sisal fibers and vinyl ester resin [20]. Silane treatment is a kind of chemical way which introduces functional groups onto sisal fiber surface. It is known that sisal fiber is mainly made up of cellulose which has large amount of hydroxyl groups. No reactive functional groups attached on the fiber surface and this makes the surface non-polar. However, with the existence of moisture, silane can react with cellulose of sisal fiber. The resultant chemicals attached on sisal fibers after chemical reactions is:

$CH_2CH_3CCOO(CH_2)_3Si(OH)_2OG$ (here G stands for the molecular of cellulose). These chemical molecules resulted in a strong polarity of sisal fiber and increased the surface tension as well. The higher surface tension decreases the contact angels between solid fibers and liquid resin, which has been proved by the contact angle measurements [20]. As we know, the small contact angles means better wettability between solid and liquid. So the induced polarity greatly facilitates the filtration of vinyl ester through sisal textile. This explains why silane treatment can improve the permeability of sisal textile from $6.38x10^{-10}$ m^2 to $15.4x10^{-10}$ m^2.

Permanganate treated sisal textile reinforced composites with three different fiber volume fractions were used to study the effect of fiber volume fraction on the permeability of sisal textile. The results are shown in Figure 6. It shows that fiber volume fraction greatly affects the permeability of sisal textile. When fiber volume fraction was increased from around 17% to 33%, the permeability value decreased more than one order of magnitude. The reason for the significant change is quite obvious because increasing the fiber volume fraction dramatically decreases the cavity between and inside fiber bundles. Thus, the flow channels for both intra-bundle and inter-bundle flows within the preform were decreased. The smaller the fiber volume fraction is, the easier for the resin to infiltrate the preform, and the higher the permeability is.

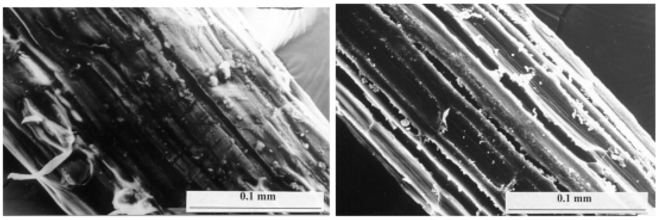


Figure 5. Sisal fiber surfaces (a)before permanganate treatment and (b) after permanganate treatment [21].

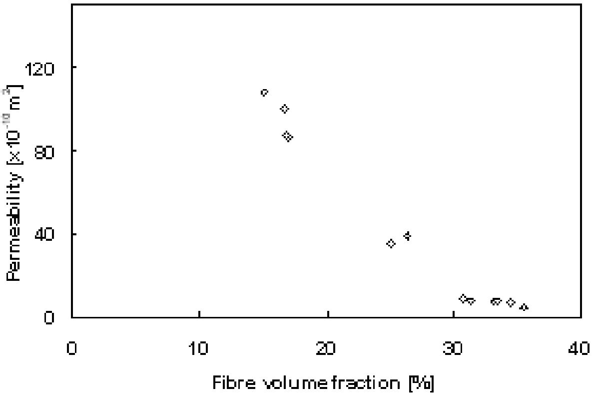


Figure 6. Permeability of sisal textile [16].

2.2.3 Manufacture of natural fiber textile reinforced composites

Compression molding is a conventional and simple method to make fiber reinforced composites. The prepreg is laid inside the mould by hand or robot and then hot pressed at a certain pressure by a compressive molding machine. With the aid of pressure and heat, the resin immerses into the reinforcement and cures inside the mould. Because the whole process involves a lot of human's efforts and controlling parameters, the quality of the final product is scattered a lot and the working efficiency is quite low. But it is still a popular method to make fiber reinforced polymer composites due to its extremely flexible, capable of making a wide variety of shapes.

RTM technology refers to a group of processes that inject resin into a fiber preform captured in a closed tool. Recent advances in textile processes have produced a variety of options for making very complex fiber preform by automatic means. Research interest has focused intently on the mould filling problem which is critical to the success of RTM. RTM appears to be best suited for medium volume, small to medium sized complex parts.

One of the major attractions of using natural fibers as reinforcements in making composites is their low cost. Finding an economic processing method to manufacture natural fiber reinforced composites is a key factor for the successful application of this kind of material. Based on the above discussion, it seems that both compression molding and RTM could be suitable for making natural textile reinforced polymer composites due to their low cost. Indeed, some research groups have reported making natural textile composites by these two processing techniques [22, 23]. Li also compared the mechanical properties of plain woven sisal textile reinforced vinyl ester composites made by these two methods. The quality of the final products was examined with the aid of optimal microscopy [24].

Figure 7 shows the tensile strength, flexural strength and impact energy of permanganate treated sisal textile reinforced vinyl ester composites made by RTM and compressive molding, respectively. It can be seen that composites made by RTM possessed higher tensile strength, flexural strength and Charpy impact energy than those of the composites made by compressive molding.

Sisal textile used in the study shows a relatively loose woven pattern and larger fiber diameter compared to man-made fibers, like glass or carbon fibers. Therefore, much air can be trapped inside the sisal fiber bundles. As we know, the void content of the composites is a factor which affects their mechanical performances. High void content could reduce the mechanical properties of the composites. During RTM process, both the injection pressure and the vacuum could work together to draw the resin penetrating the reinforcements. With the flowing of the resin through the reinforcement, the air which was trapped inside sisal textile could be driven out through the vent. So the void content of the composites can be reduced and good mechanical properties can be expected. Compressive molding, however, drives the air out by compressive pressure between the two heating plates. The air bubbles caused by the matrix polymerization could be quenched by the applied pressure. But the small air bubbles inside the loose sisal bundles are hard to be driven out. The relatively higher void content led to the lower mechanical properties of sisal textile reinforced vinyl ester made by compressive molding. Microstructure analysis did indicate the presence of higher void in composites made by compressive molding than those made by RTM (Figure 8).

The above conclusions drawn from the study of sisal textile reinforced composites would be different if other kinds of natural fibers and woven styles were used.

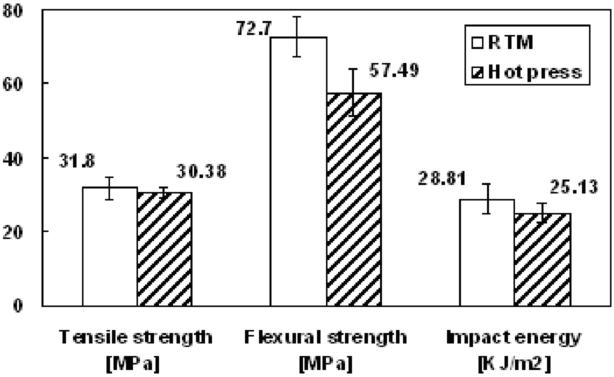


Figure 7. Comparisons of mechanical properties of sisal textile reinforced composites made by compressive molding and RTM, respectively [24].

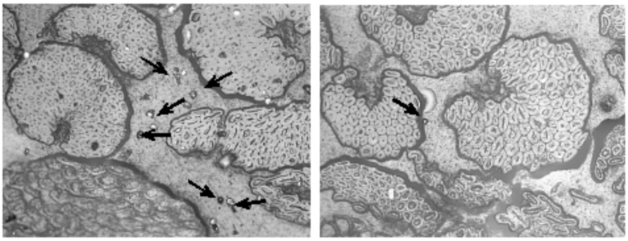


Figure 8. Cross sections of sisal fiber reinforced vinyl ester composites made by (a) compressive molding, (b) RTM. (The arrows indicate the voids/bubbles) [24]

2.2.4 Properties

- Mechanical properties of textile composites based on natural fibers

Pothen et al. conducted tensile and impact studies of woven sisal fabric reinforced polyester composites prepared by RTM technique [25]. Three different weaving patterns, i.e., plain, twill and matt woven were made by keeping sisal fiber yarns in both the warp and weft direction. The detailed weaving patterns for the three types of fabrics are shown in Figure 9. The results of tensile strength and impact strength of sisal fiber textile reinforced composites of different weaving patterns are summarized in Figure 10 and 11, respectively. It was found that weaving pattern was a crucial factor in determining the response of the composites. Both tensile and impact properties are found to be the maximum for composites made with twill woven fabric in the study where fiber bundles were used in the weft direction unlike in the other two cases where fiber yarns have been used. This was in turn found to be caused by the flow behavior of the resin and the permeability of the various fabric geometries. Increase in inter-tow flow channels led to a deterioration of the mechanical properties.

Jacob et al. studied the effects of different fiber surface treatments on the mechanical properties of sisal fabric reinforced natural rubber composites [26]. Sisal fabric was subjected to various chemical modifications like mercerization, silanation and heat treatments. The mechanical properties of the composites before and after these fiber surface treatments are shown in Figure 12. It can be seen that tensile strength and Young's modulus were observed to decrease with all chemical modifications except for the composites prepared with thermal-treated sisal fabric. Thermally treated composites exhibited superior mechanical properties because of increased crystallinity. The lower properties of the composites containing alkali treated fabric might be caused by the non-uniform penetration of alkali within the thick strands of the fabric. Chemically modified composites exhibited high hardness values. Swelling experiments confirmed strong bonding in composites containing thermally treated fabric.

Figure 9. Weaving architectures of three types of sisal fabrics [25].

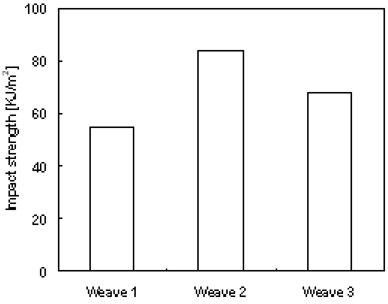


Figure 10. Effect of waving patterns on the impact strength [25]. Weave 1 is plain woven. Weave 2 is twill woven and weave 3 is matt woven.

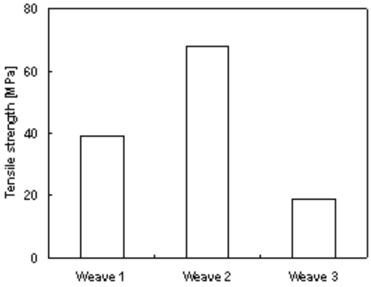


Figure 11. Effect of waving patterns on the tensile strength [25]. Weave 1 is plain woven. Weave 2 is twill woven and weave 3 is matt woven.

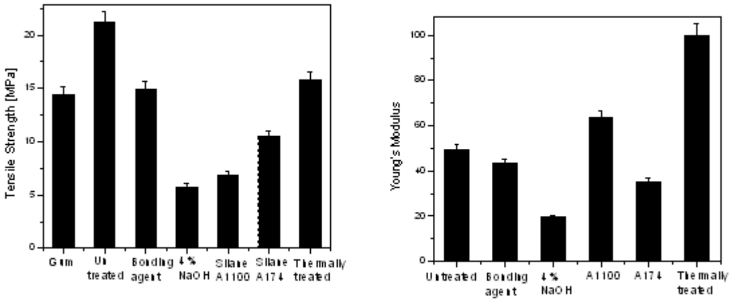


Figure. 12. Effect of fiber surface treatments on the tensile strength of sisal fiber reinforced rubber composites [26].

A complete study of the mechanical properties which includes tensile strength, compressive strength, flexural strength, impact strength, inplane shear strength, interlaminar shear strength and hardness of jute woven fabric reinforced polyester composites was conducted by Gowda et al. [27]. In this research, a type of woven fabric having a count of 20 x 12 (for yarns of 245–302 tex) was used. 20 x 12 indicates 20 in number larger yarns in the warp direction and 12 in number smaller yarns in the weft direction per inch are used. The composites were made by hand lay-up and the fiber volume fraction was around 45%. The mechanical properties of the composites are listed in Table 4.

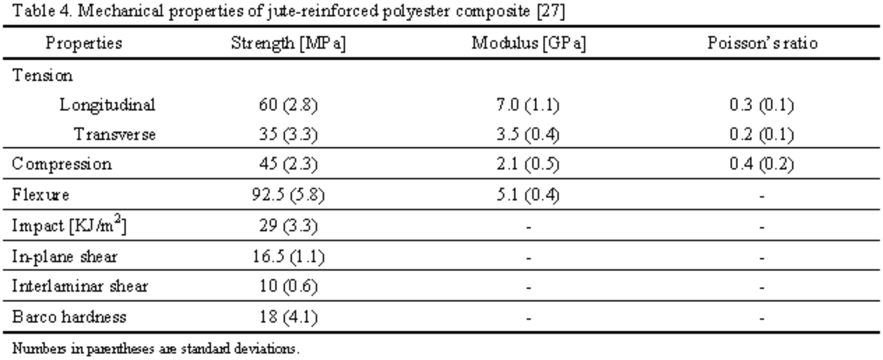


Table 4. Mechanical properties of jute-reinforced polyester composite [27]

Properties	Strength [MPa]	Modulus [GPa]	Poisson's ratio
Tension			
Longitudinal	60 (2.8)	7.0 (1.1)	0.3 (0.1)
Transverse	35 (3.3)	3.5 (0.4)	0.2 (0.1)
Compression	45 (2.3)	2.1 (0.5)	0.4 (0.2)
Flexure	92.5 (5.8)	5.1 (0.4)	-
Impact [KJ/m^2]	29 (3.3)	-	-
In-plane shear	16.5 (1.1)	-	-
Interlaminar shear	10 (0.6)	-	-
Barco hardness	18 (4.1)	-	-

Numbers in parentheses are standard deviations.

From the above results, it can be concluded that although the mechanical properties of jute woven fabric reinforced polyester composites do not possess strengths and moduli as high as those of conventional composites, they do have better strengths than wood composites and some plastics materials.

Ballistic properties of natural fabric reinforced composites were reported by Wambua, et. al. [28]. The composites they studied were flax, hemp and jute fabrics reinforced polypropylene (PP) composites and the composites were made by compression moulding under heat. It was observed that shear cut-out, delamination and fiber fracture were the major failure modes of these natural fiber textile reinforced PP composites. Among them, flax composites exhibited better energy absorption than hemp and jute composites. However, the ballistic properties of the hemp composites increased significantly when a mild steel plate was used as facing and backing.

- Fracture properties of textile composites based on natural fibers

Compact tension test was employed to study the fracture toughness of sisal textile reinforced vinyl ester composites by Li et al. [29]. Effects of fiber surface treatments were investigated. The fiber surface treatment methods and the matrix used in this study were the same as those when they studied the permeability properties [16].

Figure 13 shows the relationship between the crack resistance K_R and the crack length of treated and untreated sisal textile reinforced vinyl ester composites. With increase in crack length, there are more bridging fibers and the toughness of all the composites increases until the plateau is achieved. They defined the K_R values at the plateau as the fracture toughness, K_{mc}. The length when K_{mc} is reached was defined as the saturated fiber bridging length. Permanganate and silane treated sisal textile reinforced vinyl ester possessed a higher K_{mc} and a shorter saturated fiber bridging length compared with untreated composites. The results are summarized in Table 5.

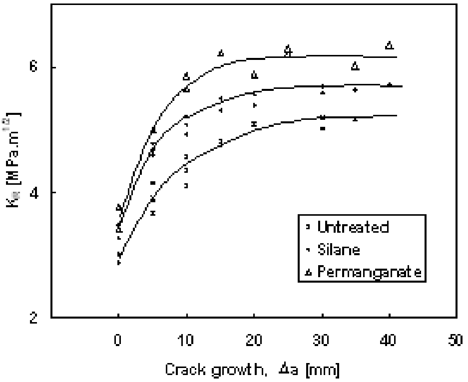
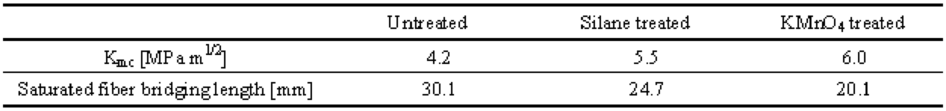


Figure 13. Effect of fiber surface treatments on the fracture toughness of sisal textile reinforced vinyl ester composites [29].

Table 5. Fracture toughness and fiber bridging length of sisal textile reinforced vinyl ester composite [29]



	Untreated	Silane treated	$KMnO_4$ treated
K_{mc} [MPa m$^{1/2}$]	4.2	5.5	6.0
Saturated fiber bridging length [mm]	30.1	24.7	20.1

The whole fracture process of pre-cracked sisal textile reinforced vinyl ester composites during compact tension tests was observed with the aid of microscopy.

With the increase of the applied load, the crack was initiated within vinyl ester resin due to its low fracture strain compared with sisal fibers. Then the load allowed the crack to pass around the fiber without breaking the interfacial bond. With increase in interfacial shearing and lateral contraction of the fiber, debonding between fiber and matrix and a further increment of crack extension were observed. That is, a fiber-bridging zone started to grow. At this stage, the load continued to increase until considerable debonding occurred and the fiber started to break at some weak spot within the matrix. During this period, the fracture toughness continued to rise. After the fiber-bridging zone was fully developed, the bridging fiber was either pulled-out or fractured. At this point, the fracture toughness reached a plateau. Figure 14 shows the whole fracture process of untreated sisal textile reinforced vinyl ester composite. In summary, the whole fracture process can be described as: matrix crack; matrix crack and fiber bridging; matrix crack, fiber bridging and matrix, fiber breakage. The existence of fiber bridging is favorable to the fracture performance of this kind of composite by the prevention of the catastrophic failure of the material.

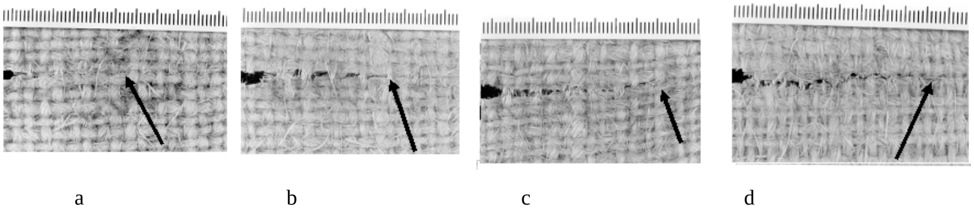


Figure 14. A crack initiation and propagation during CT test of untreated sisal textile reinforced vinyl ester [29] (arrow indicates the matrix crack tip). (a) Matrix crack (b) Matrix crack and fiber start to bridge (c) Fiber bridging reaches saturation length (d) Fully developed fiber bridging and fiber breakage

Figure 13 and Table 5 show that silane and permanganate treated sisal textile reinforced vinyl ester composites possessed higher fracture toughness and shorter saturated fiber bridging lengths compared with those of untreated counterparts. It indicates that for the composites which possess better interfacial bonding due to fiber surface treatments, their fracture toughness would be higher, while the bridging zone was shorter. Untreated sisal textile reinforced vinyl ester composites, which had poor interfacial bonding between the fiber and the matrix, showed a lower fracture toughness due to the fact that a lower load is needed to break and pull-out the fiber from the matrix. However the poor interfacial bonding property between the fiber and the matrix made it easier for the fiber to debond from the matrix. The crack was somewhat blunt and the composites showed ductile properties. This explains the longer fiber bridging length the untreated composites showed. For treated sisal textile reinforced vinyl ester composites, due to the improved interfacial bonding properties, the load used to pull-out the fiber from the matrix is high and a good fracture resistance property can be expected. The strong interfacial bonding made the composites brittle so that fewer fibers would be involved in the deformation and fiber bridging, leading to shortened fiber bridging lengths. Therefore, permanganate treated sisal textile reinforced vinyl ester showed the highest fracture toughness and the shortest fiber bridging length.

Silane treated composites possess medium fracture toughness and fiber bridging length. The untreated composites showed the lowest fracture toughness and the longest fiber bridging length.

- Moisture absorption behavior

Researchers have studied the micromechanics of moisture diffusion in woven composites [30]. The woven pattern of the fabric was found to have a profound effect on the water uptake of the composites. They observed that woven composites exhibited quicker diffusion than that of a unidirectional laminate with the same overall fiber volume fraction. Among woven composites, the plain weave with a lenticular tow and large waviness was observed to exhibit the quickest diffusion process.

The moisture uptake characteristics of woven sisal fabric reinforced natural rubber composites and thermodynamic parameters of the sorption process were investigated by Jacob et al [31]. Mercerization was seen to increase the water uptake in the composites while thermally treated fabric reinforced composites exhibited lower water uptake. This was attributed to the fact that the treatment with NaOH promotes the activation of hydroxyl groups of the cellulose units. As a result of this treatment, the hydrogen bonding among the layers of cellulose is partially cleaved and the chains undergo modification of their conformation. These activated hydroxyl groups can effectively form hydrogen bond with water which can increase the water uptake of mercerized composites.

- Other properties

The viscoelastic properties of sisal fabric reinforced rubber composites were investigated by Jacob et al [32]. Storage modulus was found to increase upon reinforcement of natural rubber with woven sisal fabric. Chemical modification of sisal fabric resulted in a decrease of storage modulus. The damping factor was found to decrease with chemical treatment and gum compound exhibited maximum damping characteristics. Thermogravimetric analysis revealed that composites containing chemically treated fabric were thermally more stable than untreated composite and among the composites containing chemically treated fabric, thermally treated composites were found to be more stable. This was substantiated in the increase of decomposition temperatures of hemicelluloses and α-cellulose for the treated composites.

2.3 Hybrid Textiles

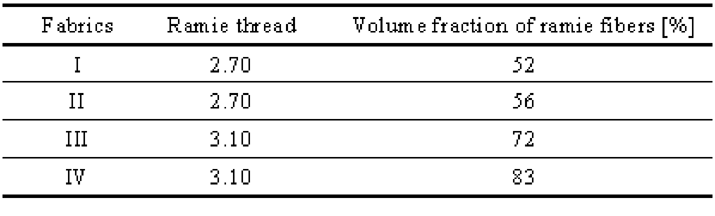
Researchers have looked into tensile strength of ramie-cotton hybrid fiber textile reinforced polyester composites [33]. The plain woven fabrics had ramie strings on the warp and cotton strings on the weft. Four kinds of fabrics with varying amounts of fibers were manufactured. The fibers used in this work were threads classified as 2.70 and 3.10. This classification stems from the textile industry, and the first figures refer to the number of filaments per thread. Therefore, the designation 2.70 means that two filaments of ramie are twisted along the thread length, and so on. The practical result for this work is that thread 3.10 is slightly thicker than thread 2.70. The details for these fabrics are shown in Table 6. Composites with varying volume fraction and orientation of ramie fibers were made by compressive molding. The results obtained are summarized in Table 7.

It is observed that tensile behavior was dominated by volume fraction of ramie fibers aligned in the test direction. The fabric and diameter of the thread did not play any role in tensile characteristics. Cotton fabric was found to have minor reinforcement effect due to weak cotton/polyester interface. Similar studies were performed by Mwaikambo and Bisanda [34] on kapok-cotton fiber reinforced polyester composites.

Novolac type phenolic composites reinforced with jute/cotton hybrid woven fabrics were fabricated and their properties were investigated as a function of fiber orientation and roving/fabric characteristics [35]. Results showed that the composite properties were strongly influenced by test direction and roving/fabric characteristics. The anisotropy degree was shown to increase with test angle and to strongly depend on the type of architecture of fabric used, i.e., jute roving diameter, relative fiber content, etc. The best overall mechanical properties were obtained for the composites tested along the jute roving direction. Composites tested at 45° and 90° with respect to the jute roving direction exhibited a controlled brittle failure combined with a successive fiber pullout, while those tested in the longitudinal direction (0°) exhibited a catastrophic failure mode. The researchers are of the opinion that jute fiber promotes a higher reinforcing effect and cotton fiber avoids catastrophic failure. Therefore, this combination of natural fibers is suitable to produce composites for lightweight structural applications.

The thermal diffusivity, thermal conductivity and specific heat of jute/cotton, sisal/cotton and ramie/cotton hybrid fabric-reinforced unsaturated polyester composites were investigated by Alsina et al [36]. These properties were measured both parallel and perpendicular to the plane of the fabrics. Thermal properties of hybrid fabrics, composites with as-received fabrics and composites with pre-dried fabrics were studied. The results obtained are shown in Table 8. It can be seen that higher values were obtained parallel to the plane of the fibers. Sisal/cotton composites showed a particular behavior, with thermal properties very close to those of the resin matrix. The thermal properties of the fabrics, i.e. without any resin, were also evaluated and were used to predict the properties of the composites from the theoretical series and parallel model equations. The effect of fabric pre-drying on the thermal properties of the composites was also evaluated. The results showed that the drying procedure used did not bring any relevant change in the properties evaluated.

Table 6. Configurations of ramie-cotton fabrics [33]



Fabrics	Ramie thread	Volume fraction of ramie fibers [%]
I	2.70	52
II	2.70	56
III	3.10	72
IV	3.10	83

Table 7. Tensile strength of ramie/cotton hybrid fabric reinforced composites . (σ_p, strength for the composites with ramie fibers disposed parallel to the tensile axis. σ_{orth}, strength for the composites with ramie fibers disposed orthogonal to the tensile axis.) [33].

Materials	Fiber volume fraction [%]	Ramie V_f [%]	Cotton V_f [%]	σ_p [MPa]	σ_{orth} [MPa]
Neat resin	0	0	0	24.2 (0.5)	24.2 (0.5)
Fabric I	49.7 (0.7)	25.8 (0.4)	23.7 (0.4)	75.8 (6.1)	20.2 (2.1)
	55.3 (1.1)	28.7 (0.6)	26.4 (0.5)	80.9 (4.9)	27.4 (2.7)
	57.2 (3.4)	29.7 (1.8)	27.3 (1.6)	74.2 (3.5)	23.1 (4.7)
Fabric II	45.3 (3.1)	25.5 (2.3)	20.5 (1.5)	62.0 (4.6)	24.0 (2.6)
	49.3 (3.3)	27.8 (2.5)	21.4 (1.5)	66.3 (6.8)	23.6 (3.5)
	50.9 (3.4)	28.6 (2.6)	23.7 (1.7)	89.2 (8.9)	23.1 (2.3)
Fabric III	54.1 (1.8)	39.0 (1.3)	14.9 (0.5)	111.8 (19.6)	9.4 (3.5)
	60.2 (1.4)	43.4 (1.0)	16.5 (0.4)	115.2 (14.3)	9.0 (4.0)
	60.9 (1.3)	43.8 (0.9)	16.7 (0.4)	105.3 (5.2)	6.3 (2.0)
Fabric IV	52.9 (0.6)	41.1 (0.4)	10.8 (0.2)	90.9 (12.7)	9.0 (1.6)
	58.5 (0.6)	45.5 (0.5)	11.9 (0.3)	117.3 (13.3)	9.8 (2.8)
	58.0 (0.6)	45.1 (0.5)	11.9 (0.3)	118.0 (6.5)	14.9 (1.3)

Numbers in parentheses are standard deviations.

Table 8. Thermal properties of lignocellulosic fabrics and their composites [36]

Material	Direction of the heat flux	Thermal properties		
		Specific heat [$Jcm^{-3}\,°C^{-1}$]	Thermal diffusivity [mm^2s^{-1}]	Thermal conductivity [$Wm^{-1}\,°C^{-1}$]
Resin	-	0.987 (0.002)	0.153 (0.0004)	0.15
Sisal/cotton				
Fabrics	Parallel	1.037 (0.003)	0.178 (0.001)	0.185 (0.005)
	Perpendicular	0.94 (0.02)	0.20	0.19
Composites[1]	Parallel	1.236 (0.007)	0.200 (0.0006)	0.25
	Perpendicular	1.065 (0.021)	0.194 (0.002)	0.213 (0.006)
Composites[2]	Parallel	1.194 (0.006)	0.203 (0.001)	0.24
	Perpendicular	1.553 (0.071)	0.132 (0.004)	0.205 (0.007)
Ramie/cotton				
Fabrics	Parallel	1.128 (0.008)	0.510 (0.004)	0.575 (0.005)
	Perpendicular	0.640 (0.01)	0.648 (0.002)	0.415 (0.006)
Composites[1]	Parallel	0.894 (0.005)	0.251 (0.002)	0.22
	Perpendicular	0.839 (0.01)	0.220 (0.0007)	0.19
Composites[2]	Parallel	1.467 (0.019)	0.164 (0.002)	0.24
	Perpendicular	0.861 (0.014)	0.218 (0.002)	0.19
Jute/cotton				
Fabrics	Parallel	1.068 (0.076)	0.524 (0.037)	0.555 (0.006)
	Perpendicular	0.536 (0.04)	0.677 (0.006)	0.36 (0.02)
Composites[1]	Parallel	1.017 (0.017)	0.231 (0.003)	0.237 (0.006)
	Perpendicular	0.869 (0.015)	0.218 (0.003)	0.19
Composites[2]	Parallel	0.793 (0.027)	0.252 (0.007)	0.20
	Perpendicular	1.032 (0.005)	0.192 (0.0007)	0.20

Composites[1] is made by as-received fabrics; Composites[2] is made by pre-dried fabrics

Numbers in parentheses are standard deviations.

3. BIODEGRADABLE TEXTILE COMPOSITES BASED ON NATURAL FIBERS

The versatile natural fiber, ramie (Boehmeria nivea), a bast fiber belonging to the family Urticaceae or Nettle commonly referred to as china grass, white ramie, green ramie and rhea is one of the most valuable natural fibers. The main natural distribution of ramie lies in subtropical China, Japan, Southeast Asia and in Brazil. Ramie is characterized by its high length, great strength, which are much greater than that of cotton and silk. In terms of specific strength ramie fibers can compete with synthetic fibers. The specific strength value of ramie is almost the same as that of E-glass fibers and shows higher elongation [37]. They can be easily woven and are good candidate for textile composites. They are widely used in fabric industry due to their softness, bleachability and better dyeability. A fully biodegradable plastic (Ecoflex produced by BASF Japan) is reinforced by ramie fabric in order to develop cost effective and eco friendly 'Green' composite. Ramie fabric with plain woven structure of non-twisted ramie fibers, supplied from Tosco Co. Ltd, Japan, was used as reinforcement. Important mechanical properties of ramie fibers are given in Table 9 [38].

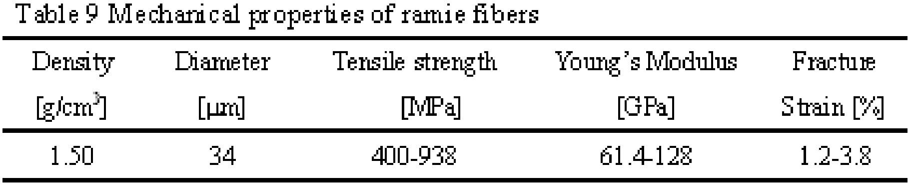


Table 9 Mechanical properties of ramie fibers

Density [g/cm^3]	Diameter [μm]	Tensile strength [MPa]	Young's Modulus [GPa]	Fracture Strain [%]
1.50	34	400-938	61.4-128	1.2-3.8

Composites were fabricated by compression molding technique at about 120^{0}C. A schematic model of the Ecoflex /Ramie fabric composite is given in Figure 15.

The tensile properties of the composites were analysed. It was obtained that the tensile strength of Ecoflex has been greatly improved (900% increment) by ramie textile ramie reinforcement. Neat Ecoflex sample exhibits very high extensibility. A strain value of 164% was observed for the neat Ecoflex. Upon ramie textile reinforcement in Ecoflex, high strength and high stiffness composite material could be resulted.

Sorption behavior of water, naphthenic oil, and diesel in neat Ecoflex and Ecoflex/Ramie fabric composites were also studied in detail. The equilibrium swelling, kinetic parameters, diffusion coefficient, solubility parameter and permeability upon water sorption were analyzed below.

3.1 Water Sorption Characteristics

A systematic study on the water diffusion characteristics of neat Ecoflex and Ecoflex/Ramie mat composite at room temperature were carried out. The mechanism of diffusion was analyzed. The mole percentage uptake of water with time by the neat Ecoflex and composite is shown in Figure 16.

It was found that water diffusion was more for Ecoflex/Ramie fabric composite than for neat Ecoflex. For the neat Ecoflex resin, there is only one phase and perfect polymer structure is observed. But for ramie textile reinforcement, the fibers interfere the three dimensional polymeric network in Ecoflex and the presence of fiber-matrix interface would affect the diffusion process. Water could penetrate and diffuse through the interface where as in the neat Ecoflex there is no possibility for this kind of process.

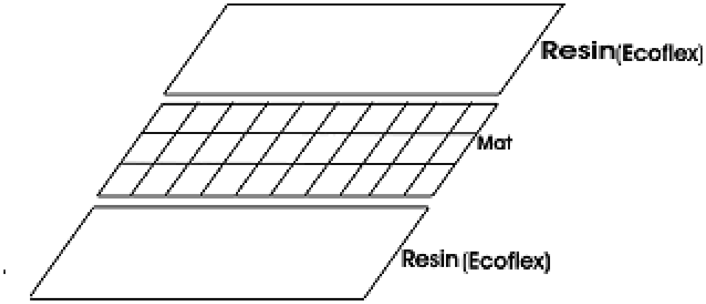


Figure 15. Schematic model of Ecoflex /Ramie fabric composite: one layer of ramie fabric is in between of two layers of Ecoflex.

Since ramie fiber is lignocellulosic, it is more susceptible to water. Ramie mat reinforced Ecoflex shows more affinity towards water due to its hydrophilicity arising from the exposed reinforcing fibers. This enables it to absorb considerable amount of water than neat Ecoflex by forming H-bonds between water and hydroxyl groups of cellulose, lignin and hemicellulose present in the cell wall. The fiber reinforcement causes subsequent reduction in the amount of matrix phase per unit volume of the composite and results in increase of uptake of polar solvent. In the Ecoflex/Ramie mat composite, fast water sorption is observed in the initial stage than for the neat Ecoflex (Figure 16).

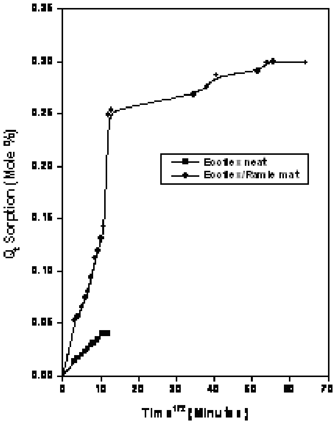


Figure 16. The water sorption behavior of neat Ecoflex and Ecoflex/Ramie mat composite.

The difference was attributed to the interface properties and presence of hydrophilic natural fibers in the Ecoflex/Ramie mat composite system. After the initial sorption a slow increase in water sorption was observed for a long duration in the Ecoflex/Ramie mat composite. This was due to the good interface properties in the composite. The attainment of the equilibrium sorption occurred in long duration for the textile composite than that for neat Ecoflex resin in which the equilibrium was resulted at an early stage. Kinetic parameters of water diffusion process were analyzed and were given in Table 10.

From Table 10, it is found that the value of n for neat Ecoflex and Ecoflex/Ramie mat composite is below 0.5. Hence transport behavior slightly deviate from Fickian value. Deviation from Fickian behavior is attributed to processes such as desorption, surface crazing, osmotic cracking, micro crack formation, moisture diffusion etc. The value of k is more for neat Ecoflex than for mat reinforced Ecoflex. The factor k is a constant that vary with each polymer. In neat Ecoflex, the water absorption is less due to close packing of chains in this polymer and the water cannot penetrate easily in to the polymer.

Ecoflex/Ramie mat composite has solubility parameter more than that for neat Ecoflex due to the presence of fibers. Percentage swelling index is also more for Ecoflex/Ramie mat composite due to increase in number of hydroxyl group of cellulose fiber which make hydrogen bonding possible with water molecules. D and P parameters also increased for neat Ecoflex. D and P parameters varied with polymer and solvent. Reinforcment of textile ramie in the polymer, fibers helped to retain water for long time in composite and penetrating into the polymer. Hence sorption is high for Ecoflex/Ramie mat composite.

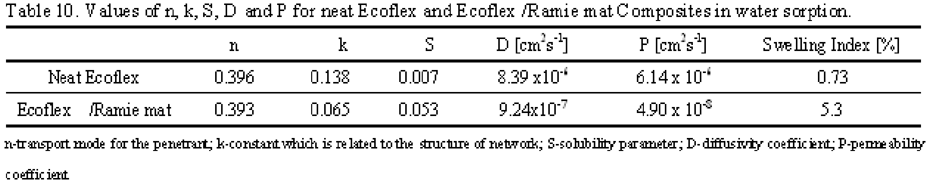


Table 10. Values of n, k, S, D and P for neat Ecoflex and Ecoflex /Ramie mat Composites in water sorption.

	n	k	S	D [cm^2s^{-1}]	P [cm^2s^{-1}]	Swelling Index [%]
Neat Ecoflex	0.396	0.138	0.007	8.39×10^{-6}	6.14×10^{-6}	0.73
Ecoflex /Ramie mat	0.393	0.065	0.053	9.24×10^{-7}	4.90×10^{-8}	5.3

n-transport mode for the penetrant; k-constant which is related to the structure of network; S-solubility parameter; D-diffusivity coefficient; P-permeability coefficient

3.2 Oil Sorption Characteristics

In case of oil diffusion, Ecoflex/Ramie mat composite has more sorption than neat Ecoflex resin. This is due to the presence of ramie fabric. Percentage swelling index and solubility parameter S, of the sorption process were calculated. Solubility parameter and swelling index are higher for Ecoflex/Ramie mat composite than for neat Ecoflex. The presence of ramie fiber is the reason for the increase of the swelling index. Increased percentage swelling index for Ecoflex/Ramie mat composite is due to the increased number of hydroxyl groups available from cellulose fibers.

3.3 Diesel Sorption Characteristics

The neat Ecoflex showed more absorption of diesel than Ecoflex/Ramie mat composite. The low molecular weight and molecular hydrocarbons present in diesel made it penetrate into neat polymer easily. Diesel molecules would have more interaction with the neat Ecoflex than the textile composite due to the aromatic hydrocarbons present in it. In Ecoflex/Ramie fabric composite hydrocarbons diffused through the fiber- matrix interfaces of the composite. The hindrance exerted by the fibers restricted the movement of diesel within the composite and hence possibility of liquid diffusion decreased. In Ecoflex/Ramie mat composite there was a decreased hydrophilicity of cellulose as a result of chemical interaction of fibers and hydrocarbon. Also enhanced interfacial bonding between fiber and matrix prevent the fiber from absorbing diesel. One peculiarity observed in diesel sorption is the fast rate of initial absorption in both neat Ecoflex and mat composite. This was due to the increased affinity of diesel to the matrix Ecoflex.

Swelling index and solubility parameter, S was higher for textile composite than neat Ecoflex. This was due to the more interaction of neat Ecoflex and solvent. High penetrating power of hydrocarbon contributed to the higher sorption of neat Ecoflex. But in Ecoflex/Ramie mat composite, the hydrocarbon in diesel diffused into the interfaces and voids if any in the composite and the fibers restricted the movement of diesel. Hence percentage swelling index and solubility parameter decreased in this case. The decreased amount of matrix due to fabric content and the good fiber matrix interaction and effective binding of fiber in the composite contributed to the lower diesel uptake in the textile composite sample. Studies on the diesel uptake of natural fibers (isora) reinforced natural rubber composite were also reported elsewhere [39]. The study reported the effect of natural fiber content upon the sorption behavior. They reported that the percentage swelling index and swelling coefficient of the composite were found to decrease with increase in fiber content. This was due to the increased hindrance exerted by the fibers at higher fiber content and also due to the good interactions between fibers and rubber [39].

The resultant fully biodegradable ‘Green Composite’ of Ecoflex and ramie fabric is cost effective and reducing health hazardness. They possessed excellent mechanical and barrier properties. The textile composite exhibit lowered diesel sorption and reduced wear to the processing tools. Green composite can find versatile applications due to its better properties.

4. COMPARATIVE STUDY OF SHORT FIBER COMPOSITES AND TEXTILE COMPOSITES

For better understanding of the advantages for the composites made from natural textile, the properties of the composites made from short fibers and fabrics were compared.

Silva et. al. studied the fracture toughness of natural fibers reinforced castor oil polyurethane composites by conducting compact tension test [40]. Short sisal fiber, coconut fiber and sisal fabrics were used to make the composites. Alkaline was selected to treat natural fiber surfaces. The fracture toughness of these composites is shown in Figure 17. Note that the volume fraction of the treated fabric composites was higher than the corresponding untreated ones. This was due to the fabric shrinkage during the drying stage of the alkaline treatment that promoted a more closed woven and consequently larger volume fraction. From the results, it can be seen that the best fracture toughness performance was displayed by the sisal fabric composite, which clearly indicated that natural fiber reinforced composites in textile form would possess better fracture properties than the composites made by short fibers. The alkaline treatment showed to be harmful for fracture toughness of sisal fiber composites since the improved interfacial adhesion impaired the main energy absorption mechanisms. On the other hand, an enhancement on the fracture toughness of coconut fiber composites was observed, which was caused by the fibrillation process occurring under the severest condition of the alkaline treatment, which created additional fracture mechanisms.

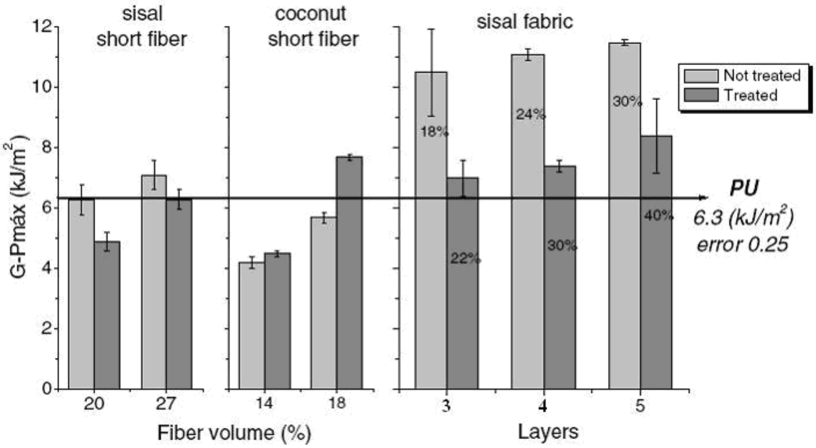


Figure 17. Fracture toughness for all short fiber and fabric composites tested at 0.5 mm/min. The percentile values in the columns correspond to the fabric volume fraction. (G-Pmax here is the fracture toughness which was characterized by critical energy release rate (G) concept, determined at the maximum load point) [40].

5. CONCLUSION

Textile reinforced composites based on natural fibers have been studied by many research groups in recent years due to their good mechanical performances, easy to handle, excellent integrity and reduced manufacturing cost. It can be concluded that properties of the composites made by fiber textiles are better than the composites made by short fibers. Permeability, mechanical and fracture properties are all affected by the weaving architectures of the reinforcing fabrics.

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