

## A REVIEW ON SISAL FIBER REINFORCED POLYMER COMPOSITES

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### ABSTRACT

The global demand for wood as a building material is steadily growing, while the availability of this natural resource is diminishing. This situation has led to the development of alternative materials. Of the various synthetic materials that have been explored and advocated, polymer composites claim a major participation as building materials. There has been a growing interest in utilizing natural fibres as reinforcement in polymer composite for making low cost construction materials in recent years. Natural fibres are prospective reinforcing materials and their use until now has been more traditional than technical. They have long served many useful purposes but the application of the material technology for the utilization of natural fibres as reinforcement in polymer matrix took place in comparatively recent years. Economic and other related factors in many developing countries where natural fibres are abundant, demand that scientists and engineers apply appropriate technology to utilize these natural fibres as effectively and economically as possible to produce good quality fibre reinforced polymer composites for housing and other needs. Among the various natural fibres, sisal is of particular interest in that its composites have high impact strength besides having moderate tensile and flexural properties compared to other lignocellulosic fibres. The present paper surveys the research work published in the field of sisal fibre reinforced polymer composites with special reference to the structure and properties of sisal fibre, processing techniques, and the physical and mechanical properties of the composites.

**Key words:** sisal fibre, polymer, composites, structure, properties

### COMPÓSITOS POLIMÉRICOS REFORÇADOS COM FIBRAS DE SISAL

### RESUMO

O uso da madeira como material de construção continua crescendo mundialmente enquanto a disponibilidade deste recurso natural está diminuindo. Esta situação tem conduzido ao desenvolvimento de materiais alternativos. Dentre os vários materiais sintéticos que têm sido explorados, os compósitos poliméricos reivindicam e buscam uma maior participação como material de construção. Nos últimos anos tem se observado um crescente interesse na utilização de fibras naturais como reforço de matrizes poliméricas para a produção de materiais de baixo custo. As fibras naturais são reforços com grande potencialidade e seu uso tem se dado de forma mais tradicional do que científica. Elas têm se prestado a inúmeras aplicações ao longo do tempo, mas a aplicação da tecnologia dos materiais

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visando a sua utilização como reforço de matrizes poliméricas é relativamente recente. As dificuldades econômicas e sociais observadas em muitos dos países em desenvolvimento, onde as fibras naturais são abundantes, requerem que cientistas e engenheiros apliquem tecnologias apropriadas para utilizar estas fibras da forma mais eficiente possível, de tal maneira que se possa produzir materiais compósitos poliméricos de boa qualidade visando a atender a demanda da população por habitações e componentes habitacionais. Dentre as várias fibras naturais, a fibra de sisal é de particular interesse uma vez que os compósitos que a utilizam como reforço apresentam alta resistência ao impacto além de possuírem moderada resistência à tração e à flexão. O presente artigo apresenta uma revisão dos trabalhos de pesquisa publicados no campo dos compósitos poliméricos ecológicos, especialmente no que se refere aos compósitos reforçados com fibras de sisal, visando seu uso como material de construção. Ênfase especial é dada à micro-estrutura e propriedades da fibra de sisal, às técnicas de processamento e às propriedades físicas e mecânicas dos compósitos poliméricos reforçados com fibras de sisal.

**Palavras-chave:** sisal, polímero, compósitos, estrutura, propriedades

## INTRODUCTION

In our everyday life timber plays a significant role. However timber resources are getting depleted continuously while the demand for the material is ever increasing. According to the literature, by the beginning of the next century the wood will be scarce for the whole world (Singh, 1982). This situation has led to the development of alternative material. Among the various synthetic materials that have been explored and advocated, plastics claim a major share as wood substitutes. Plastics are used for almost everything from the articles of daily use to the components of complicated engineering structures and heavy industrial applications (Rai & Jai Singh, 1986). Plastics find an extensive application in buildings as flooring material because they are resistant to abrasion, have a low heat conductivity and low water absorption, sufficient hardness and strength. They fail to swell when moistened, readily take on varnishes and paints. Hardware items like door and window frames, flushing cisterns, overhead water storage tanks and water fittings are commercially available and are finding acceptance in the building industry. Plastics are used to manufacture various sanitary wares, which include wash basins, bathtubs, sinks, shower cabins, washing racks and others. Plastic pipes are widely used in the installation of various industrial purposes, water supply etc.

However, during the last decade, the study of filled plastic composites has simulated immense interest in meeting the future shortage of plastic materials (Lightsey, 1983). In fact, synthetic fibres such as nylon, rayon, aramid, glass, polyester and carbon are extensively used for the reinforcement of plastics (Erich et al., 1984; Lawrence et al., 1995). Nevertheless, these materials are expensive and are non-renewable resources. Because of the uncertainties prevailing in the supply and price of petroleum based products, there is every need to use the naturally occurring alternatives. In many parts of the world, besides the agricultural purposes, different parts of plants and fruits of many crops have been found to be viable sources of raw material for industrial purpose. In recent years, polymer composites containing vegetable fibres have received considerable attention both in the literature and in industry. The interest in natural fibre reinforced polymer composites is growing rapidly due to the high performance in mechanical properties, significant processing advantages, low cost and low density (Satyanarayana et al., 1990a,b). Natural fibres are renewable resources in many developing countries of the world; they are

cheaper, pose no health hazards and, finally, provide a solution to environmental pollution by finding new uses for waste materials. Furthermore, natural fibre reinforced polymer composites form a new class of materials which seem to have good potential in the future as a substitute for scarce wood and wood based materials in structural applications.

Fibres obtained from the various parts of the plants are known as vegetable fibres. These fibres are classified into three categories depending on the part of the plant from which they are extracted.

1. Bast or Stem fibres (jute, mesta, banana etc.)
2. Leaf fibres (sisal, pineapple, screw pine etc.)
3. Fruit fibres (cotton, coir, oil palm etc.)

Many of the plant fibres such as coir, sisal, jute, banana, palmyra, pineapple, talipot, hemp, etc. find applications as a resource for industrial materials (Satyanarayana et al., 1990b; Thomas & Udo, 1997; Rowell et al., 1997). Table 1 presents the properties of some plant fibres. Properties of plant fibres depend mainly on the nature of the plant, locality in which it is grown, age of the plant, and the extraction method used. For example, coir is a hard and tough multicellular fibre with a central portion called "lacuna". Sisal is an important leaf fibre and is very strong. Pineapple leaf fibre is soft and has high cellulose content. Oil palm fibres are hard and tough, and show similarity to coir fibres in cellular structure. The elementary unit of a cellulose macromolecule is anhydro-d-glucose, which contains three alcohol hydroxyls (-OH) (Bledzki et al., 1996). These hydroxyls form hydrogen bonds inside the macromolecule itself (intramolecular) and between other cellulose macromolecules (intermolecular) as well as with hydroxyl groups from the air. Therefore, all plant fibres are of a hydrophilic nature; their moisture content reaches 8-13%.

In addition to cellulose, plant fibres contain different natural substances. The most important of them is lignin. The distinct cells of hard plant fibres are bonded together by lignin, acting as a cementing material. The lignin content of plant fibres influences its structure, properties and morphology. An important characteristic of vegetable fibre is their degree of polymerization (DP). The cellulose molecules of each fibre differ in their DP and consequently, the fibre is a complex mixture of polymer homologue ( $C_6H_{10}O_5)_n$ . Bast fibres commonly show the highest DP among all the plant fibres (~10,000). Traditionally these fibres have been used for making twines, ropes, cords, as packaging material in sacks and gunny bags, as carpet-backing and more recently, as a geotextile material.

Table 1. Properties of some natural fibers (Mukherjee &amp; Satyanarayana, 1984)

Fiber Type	Diameter ( $\mu\text{m}$ )	Density ( $\text{g cm}^{-3}$ )	Cellulose (%)	Lignin (%)	l/d ratio*	Cell Wall Thickness ( $\mu\text{m}$ )	Microfibrillar Angle (deg)
Sisal	100-300	1.450	70	12	100	12.5	20-25
Banana	50-250	1.350	83	5	150	1.25	11-12
Coir	100-450	1.150	37	42	35	8.00	30.45

\*Cell length (l)/ Cell breadth (d) ratio

Vegetable fibres can be considered as naturally occurring composites consisting mainly of cellulose fibrils embedded in lignin matrix. These cellulose fibrils are aligned along the length of the fibre, irrespective of its origin, i.e. whether it is extracted from stem, leaf or fruit. It appears that such an alignment renders maximum tensile and flexural strengths, in addition to providing rigidity in that direction of the fibre as observed in the case of bamboo. Further, these fibres exhibit high electrical resistance in addition to being thermally and acoustically insulating. It can therefore be expected that when these fibres are incorporated in low modulus polymer matrices, they would yield materials with better properties suitable for various applications.

Since vegetable fibres are strong, light in weight, abundant, non-abrasive, non-hazardous and inexpensive, they can serve as an excellent reinforcing agent for plastics. Several cellulosic products and wastes such as shell flour, wood flour and pulp have been used as fillers in polymers, primarily to achieve cost savings and also to impart some desirable properties like decreasing shrinkage after molding, increasing elastic modulus and creep resistance (Lightsey, 1983; Prasad et al., 1983; Kokta, 1988; Maldas & Kokta, 1991). Cotton-polymer composites are reported to be the first fiber reinforced plastics used by the military for radar aircraft (Piggot, 1980; Lubin, 1982). However, over the past decade, cellulosic fillers of a fibrous nature have been of greater interest as they would give composites with improved mechanical properties compared to those containing non-fibrous fillers (Paramasivam & Abdulkalam, 1974; Joseph et al., 1993a,b; Carvalho, 1997; Pavithran et al., 1987, 1988). Vegetable fibres possess moderately high specific strength and stiffness and can be used as reinforcing materials in polymeric resin matrices to make useful structural composites materials.

Lignocellulosic fibres like jute, sisal, coir, and pineapple have been used as reinforcements in polymer matrices. Among these fibres, sisal is of particular interest in that its composites have high impact strength besides having moderate tensile and flexural properties compared to other lignocellulosic fibres (Pavithran et al., 1987). However, a large quantity of this renewable resource is being under-utilized. Sisal fibre is mainly used for the manufacture of ropes for use in marine industry and agriculture, for making twines, cords, padding, mat making, fishing nets, fancy articles such as purses, wall hangings, table mats etc. The use of sisal fibre as a textile fibre by mankind began with Weindling's work during forties (Weindling, 1947). Along with the study of agronomic and industrial aspects, a thorough and fundamental investigation on the sisal fibre was carried out by Wilson (1971). He also paid attention to the possibility of chemically modifying the fibre and put forward arguments for rejecting the idea due to the sacrifices that have to be allowed for the loss in strength as a result of chemical treatments. Over last few decades, several studies have been reported on the use of sisal fibres as reinforcements in polymer matrices (Barkakaty, 1976; Bisanda &

Ansell, 1991; Joseph et al., 1992, 1993ab, 1994; Mattoso et al., 1997). Therefore, a detailed and systematic survey has been carried out on the use of sisal fibre as reinforcement in polymer composites.

## STRUCTURE AND PROPERTIES OF SISAL FIBRE

Sisal fibre is obtained from the leaves of the plant *Agave sisalana*, which was originated from Mexico and is now mainly cultivated in East Africa, Brazil, Haiti, India and Indonesia (Nilsson, 1975; Mattoso et al., 1997). It is grouped under the broad heading of the "hard fibres" among which sisal is placed second to manila in durability and strength (Weindling, 1947). The name "sisal" comes from a harbor town in Yucatan, Maya, Mexico (Nilsson, 1975). It means cold water. Agave plants were grown by the Maya Indians before the arrival of the Europeans. They prepared the fibres by hand and used it for ropes, carpets and clothing. Some clothes were called "nequen", and this is where the present name of Mexican agave, henequen, probably originates. It is one of the most extensively cultivated hard fibre in the world and it accounts for half the total production of textile fibres (Lock, 1962; Wilson, 1971). The reason for this is due to the ease of cultivation of sisal plants, which have short renewing times, and is fairly easy to grow in all kinds of environments. A good sisal plant yields about 200 leaves with each leaf having a mass composition of 4% fibre, 0.75% cuticle, 8% other dry matter and 87.25% moisture. Thus a normal leaf weighing about 600g yields about 3% by weight of fibre with each leaf containing about 1000 fibres (Kallapur, 1962). The fibre is extracted from the leaf either by retting, by scraping or by retting followed by scraping or by mechanical means using decorticators (KVIC, 1980). The diameter of the fibre varied from 100 $\mu\text{m}$  to 300 $\mu\text{m}$  (Mukherjee & Satyanarayana, 1984).

The structure and properties of sisal fibre have been investigated by several researchers (Barkakaty, 1976; McLaughlin, 1980; Kulkarni et al., 1981; Gram, 1983; Mukherjee & Satyanarayana, 1984; Mattoso et al., 1997). Such understanding of structure-property relationship will not only help open up new avenues for these fibres, but also emphasize the importance of this agricultural material, which form one of the abundantly available renewable resources in the world.

The characteristics of the sisal fibres depend on the properties of the individual constituents, the fibrillar structure and the lamellae matrix. The fibre is composed of numerous elongated fusiform fibre cells that taper towards each end. The fibre cells are linked together by means of middle lamellae, which consist of hemicellulose, lignin and pectin. According to Gram (1983), a sisal fibre in cross-section is built up of about 100 fibre cells. Kulkarni et al. (1981) state that the number of cells in cross-section of a coconut fibre ranges from 260 to 584 depending on the diameter of the fibre.

Figures 1 and 2 show back scattered image and scanning electron micrographs of the microstructure of sisal respectively. As can be seen, the cross section of sisal fibres is neither circular nor fairly uniform in dimension. The lumen varies in size but is usually well defined. The longitudinal shape is approximately cylindrical. Physically, each fibre cell is made up of four main parts, namely the primary wall, the thick secondary wall, the tertiary wall and the lumen. Figure 3 shows a schematic sketch of a fibre cell. The cell walls consist of several layers of fibrillar structure consisting of fibrillae. In the primary wall, the fibrillae have a reticulated structure. In the outer secondary wall ( $S_1$ ), which is located inside the primary wall, the fibrillae are arranged in spirals with a spiral angle of  $40^\circ$  (for sisal fibre) in relation to the longitudinal axis of the cell. The fibrillae in the inner secondary wall ( $S_2$ ) of sisal fibres have a sharper slope, 18 to  $25^\circ$ . The thin, innermost, tertiary wall has a parallel fibrillar structure and encloses the lumen. The fibrillae are, in turn, built up of micro-fibrillae with a thickness of about 20 nm. The microfibrillae are composed of cellulose molecular chains with a thickness of 0.7 nm and a length of a few  $\mu\text{m}$  (Gram, 1983).

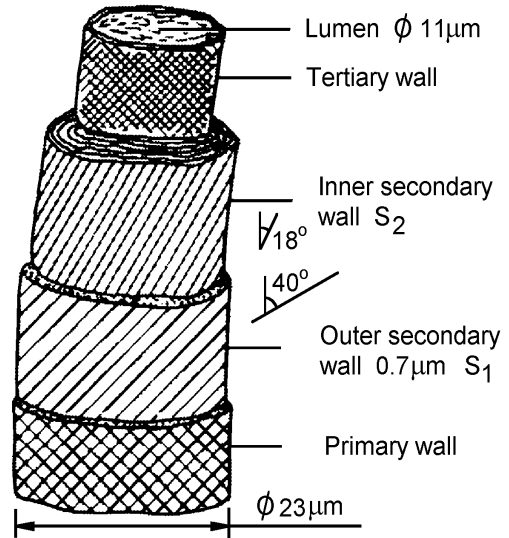


Figure 3. Schematic sketch of a sisal fiber cell with approximate dimensions (Gram, 1983)

According to Mukherjee & Satyanarayana (1986) the microfibrillar or spiral angle in the secondary walls of the cells of coconut fibres are of about  $30 - 45^\circ$ . Chemically the vegetable fibres comprise cellulose, hemicellulose, lignin, pectin and a small amount of waxes and fat. Dinwoodie (1981) summarizes the polymeric state, molecular derivatives and function of cellulose, hemicellulose, lignin and extractives as in Table 2. Barkakaty (1976) has reported the structural aspects of sisal fibre. He has studied the molecular structure of the paracrystalline cellulose, which forms the major constituent of the fibre by x-ray diffraction technique. He also studied the multicellular structure, surface topology, and fracture morphology and the effect of chemical treatment on sisal fibre. Mattoso et al. (1997) have reported the extraction methods, morphology and chemical modifications of sisal fibre and its application as reinforcement agents in polymer composites. Mukherjee & Satyanarayana (1984) have studied the mechanical properties of sisal fibre such as initial modulus (the extent to which the fibre resists the deformation in the low strain region is called the initial modulus of the fibre), ultimate tensile strength, average modulus and percent elongation as a function of fibre diameter, test length and the speed of testing. It was reported that tensile properties of fibre vary with test length of the fibre. Tables 3 and 4 list the observed variation of tensile properties with test lengths and speed of testing respectively. It can be seen from the Table 3 that both tensile

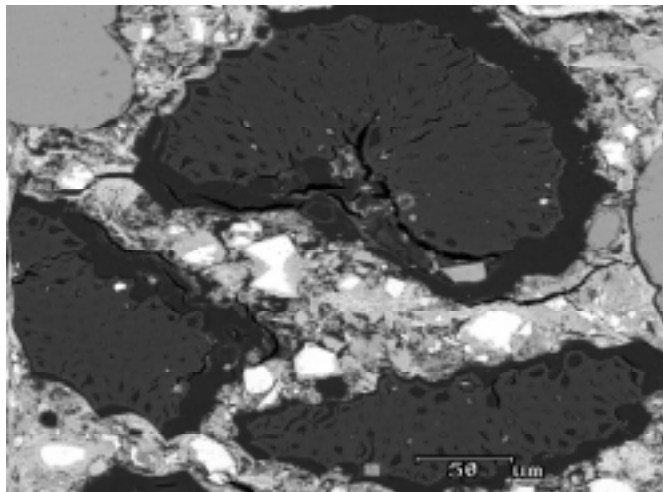


Figure 1. Back scattered image of the cross-section of sisal fiber embedded in cement matrix (Toledo Filho, 1997)

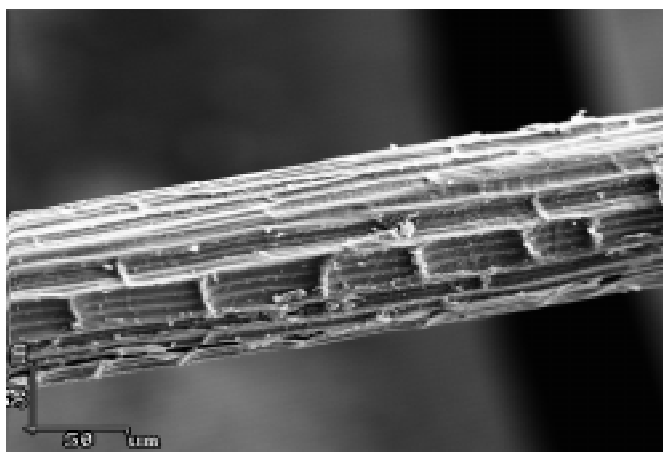


Figure 2. Scanning electron micrographs (SEM) of the of sisal fiber (Toledo Filho, 1997)

Table 2. Cellulose, hemicellulose, lignin and extractives, polymeric state, molecular derivatives and function (Dinwoodie, 1981)

Content	Polymeric Sstate	Molecular Derivatives	Function
Cellulose	Crystalline highly oriented large molecule	Glucose	"Fiber"
Hemicelluloses	Semi-crystalline smaller molecule	Galactose, Mannose, Xilose	"Matrix"
Lignin	Amorphous large 3-D molecule	Phenyl propane	"Matrix"
Extractives	Some polymeric; others nonpolymeric	e.g. Terpenes, Polyphenols	Extraneous

strength and percent elongation decrease with test length, whereas, Young's modulus and average modulus increase with test length. In natural fibres, since the flaws or weak links are irregularly spaced in the fibre, the strength will depend on the length of the fibre used for the tensile test (McLaughlin, 1980).

Table 3. Variation of tensile properties of sisal fiber with test length (diameter of fiber: 200 $\mu$ m), (Mukherjee & Satyanarayana, 1984)

Test Length (mm)	Initial Modulus (GNm <sup>-2</sup> )	Tensile Strength (MNm <sup>-2</sup> )	Elongation at Break (%)	Average Modulus (GNm <sup>-2</sup> )
15	14.15	793.80	8.15	9.74
25	17.26	757.10	5.70	13.28
35	19.71	728.10	4.65	15.64
50	22.52	630.10	3.98	15.83
65	25.36	620.81	3.50	17.87

Table 4. Variation of tensile properties of sisal fiber with speed of testing (diameter of fiber: 200 $\mu$ m; test length: 50mm) (Mukherjee & Satyanarayana, 1984)

Speed of Testing (mm min <sup>-1</sup> )	Initial Modulus (GN m <sup>-2</sup> )	Tensile Strength (MN m <sup>-2</sup> )
1	8.41	481.00
2	20.00	608.80
10	22.12	630.12
50	34.16	759.70
500	-	441.60

If a fibre having length  $L$  and strength  $\sigma$  is now changed in length by  $dL$  a corresponding change in strength  $d\sigma$  will be observed. The incremental change can be related by the equation given below:

$$d\sigma = \alpha \frac{dL}{L}$$

where,  $dL$  represents the probability of having an imperfection introduced or reduced,  $L$  represents the probability of already having an imperfection in  $L$  and  $\alpha$  represents a measure of the frequency of occurrence of weak links in the fibre. So with an increase in test length, the number of weak links or imperfections increases, thus resulting in reduction in tensile strength and percent elongation values. The stress-strain curve for sisal fibres was characterized by an initial linear region followed by a curvature indicating the viscoelastic nature of the fibre. The applied stress is shared between crystalline and non-crystalline components in a natural fibre, which is also basically a fibre-reinforced composite on a microscale. As the applied stress increases, the weak primary cell wall collapses and decohesion of cells begins following decohesion of cellulosic and non-cellulosic molecules mainly through weak links and imperfections. This leads to the curvature of the stress-strain curve. The applied stress also causes the uncoiling as well as extensions of the crystalline fibrils in the secondary walls of the cells.

Padmavathi & Naidu (1998) have studied the chemical resistance and tensile strength of sisal fibres (*Agave veracruz*). It was noted that sisal fibres were more resistant to concentrated

HCl compared to other acids. The fibres treated with 18% solution of NaOH showed more tensile load than the other chemically modified fibres. Edwards et al. (1997) have studied the application of FT-Raman microscopy to the non-destructive analysis of sisal fibres. Chand & Joshi (1995) have investigated the effect of gamma irradiation on structure and dc conductivity of this sisal fibre. It was found that exposure of sisal fibre to gamma-irradiation increased the dc conductivity, which has been explained on the basis of microstructure. Singh et al. (1998) have studied the adsorptive interaction between sisal fibre and coupling agents using contact angle measurements and Fourier transform infrared spectroscopy. It was found that high contact angle and reduced hydroxyl groups on titanate-treated fibres favor its better hydrophobicity over the other treatments. The presence of adsorbed layer of coupling agent on the fibre surface was ascertained by appearance, shifting, and decreased intensity of absorption bands. The lowest polar component of surface-free energy for N-substituted methacrylamide-treated fibre indicates the formation of ordered layers of its organofunctionality at the surface. The reason for enhanced interaction between sisal fibre and N-substituted methacrylamide is suggested by the formation of hydrogen bond, besides extracting a surface-active proton from the fibre surface by alkoxy group to form a covalent bond. An optimum treating condition of fibre for effective adsorptive interaction has been reported. The deposition of compound in the form of an aggregate on the fibre surface was also observed under scanning electron microscopy.

## SISAL FIBRE REINFORCED THERMOSET COMPOSITES

Incorporation of sisal fibre into thermosetting plastics have been reported by various workers (Paramasivam & Abdulkalam, 1974; Pavithran et al., 1987, 1988; Joseph et al., 1996a). Paramasivam & Abdulkalam (1974) have investigated the feasibility of developing polymer based composites using sisal fibres due to the low cost of production of composites and amenability of these fibres to winding, laminating and other fabrication processes. It was found that the fabrication of these composites was fairly easy and cost of production was quite low. Winding of cylinders with longitudinal or helical and hoop reinforcement was successfully carried out. Tensile strength of sisal epoxy composites was found to be 250-300 MPa, which was nearly half the strength of fibre glass-epoxy composites of the same composition. Because of the low density of the sisal fibre, however, the specific strength of sisal composites was comparable with that of glass composites. The unidirectional modulus of sisal-epoxy composites was found to be about 8.5 GPa. This study indicated the feasibility of developing composites incorporating one of the abundantly available natural fibres, to be used in the field of consumer goods, low cost housing and civil engineering structures. Satyanarayana et al. (1984) have studied the mechanical properties of chopped sisal fibre - polyester composites. Chopped sisal fibre-polyester composites were prepared by the compression molding technique. It was found that the specific modulus of the composite was 1.90 compared with 2.71 for glass fibre reinforced plastics, while the specific strength was of the same order as

that of polyester resins (34 - 41 MPa). The impact strength was  $30 \text{ J m}^{-2}$ , which is three times higher than that of polyester and 30% less than glass fibre reinforced plastics. Accelerated testing revealed little change in initial modulus, and reductions of 5% in ultimate tensile strength, 16% in flexural strength and 5.4% in water absorption.

Pavithran et al. (1987, 1988) have reported on the impact properties of oriented sisal fibre-polyester composites. Unidirectionally aligned sisal fibre-polyester composites containing  $\cong 0.5$  volume fraction of sisal fibre were prepared from unsaturated polyester pre-pregs. Impact strength of the composites was measured by Charpy test in a pendulum impact-testing machine using a pendulum load of 0.4 kgs. They have compared the work of fracture of sisal fibre polyester composites to those of composites containing other natural fibres. The work of fracture obtained in their study is presented in Table 5. It can be seen that sisal fibre composites have the maximum work of fracture followed by pineapple fibre composite. Banana and coir fibre composite have comparatively low work of fracture. It is a generally accepted fact that the toughness of a fibre reinforced composite is mainly depending on the fibre stress-strain behavior. Strong fibres with high failure strain impart high work of fracture on the composites. From the above table it is interesting to note that, among sisal, pineapple and banana fibre reinforced polymer composites, sisal fibre-polyester composites is likely to give high work of fracture because of the high toughness of sisal fibre which is found in agreement with the experimental results. However, the large difference observed between banana and pineapple fibres is not explained by taking into account of their comparative mechanical properties. Similarly, very low toughness cannot be expected for coir composites because of the high toughness of the fibre. They have also studied the variation in impact properties of various natural fibre composites with microfibrillar angle of the fibre. Figure 4 shows the influence of microfibrillar angle of the fibre on the work of fracture values of different natural fibre reinforced polymer composites. It can be seen that microfibrillar angle in plant fibres plays an important role in determining the impact behavior of these composites and this effect should be taken into account along with the other parameters while predicting the impact properties of natural fibre composites.

Pavithran et al. (1988) have compared the impact properties of unidirectionally oriented sisal fibre-polyester composites with those of composites having ultra high-density polyethylene [UHDPE] and glass fibres. It was observed that sisal composites shows work of fracture identical with that of ultra high-modulus polyethylene composites and the toughness of sisal fibre composites is only 25% less than that of glass fibre composites when the density of the latter is taken into account. The high

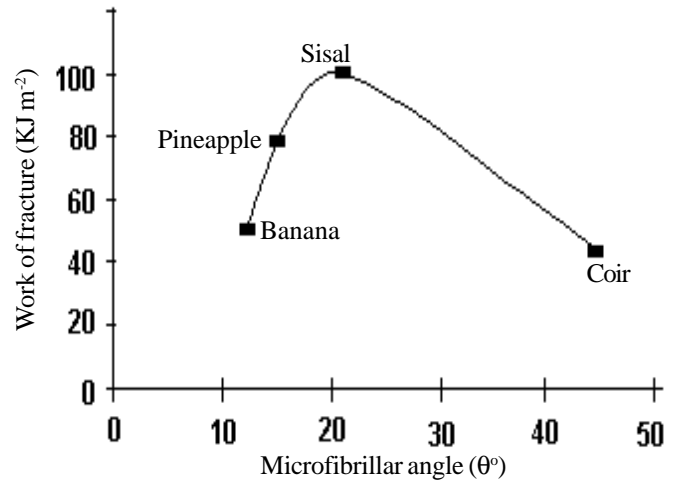


Figure 4. Work of fracture of natural fiber composites plotted against microfibrillar angle of the fibers (Pavithran et al., 1987)

work of fracture obtained for the sisal fibre composite, in spite of the fibre having low strength and modulus confirms their earlier argument that prediction of impact behavior of natural fibre composites will not be valid unless the contribution from the helically wound microfibrillar structure of the fibre is taken into account. It is a generally accepted fact that the toughness of a fibre composite is mainly dependent on the fibre stress-strain behavior. Strong fibres with high failure strain impart high work of fracture on the composites.

Bisanda & Ansell (1991) have studied the effect of silane treatment and alkali treatment on the mechanical and physical properties of sisal-epoxy composites. They have reported that incorporation of sisal fibres in an epoxy resin produces stiff and strong composite materials. The treatment of the sisal fibres with silane, preceded by mercerisation, provides improved wettability, mechanical properties and water resistance.

Joseph et al. (1996a) have studied the influence of interfacial adhesion on the mechanical and fracture behavior of short sisal fibre reinforced polymer composites of several thermoset resin matrices (polyester, epoxy, phenol formaldehyde) and a thermoplastic matrix (low density polyethylene) with respect to fibre length and fibre loading. They observed that all the composites showed a general trend of increasing properties with fibre loading. However, the optimum length of the fibre required to obtain an increase in properties varied with the type of matrix. It is well known that different degrees of reinforcement effects are achieved by the addition of hydrophilic fibres to different polymers, even though the flow limits and Young's moduli of most of the commercial plastics are relatively close. This may be due to the different adhesion strength between matrices and fibres. The adhesion is usually strongest in polar

Table 5. Mechanical properties of natural fibers and work of fracture of their composites (Pavithran et al., 1987)

Fiber Type	Fiber Properties			Composite Properties	
	Tensile Strength (MPa)	Elongation (%)	Toughness (MN m <sup>-2</sup> )	Fiber Pull-out Layer (mm)	Work of Fracture (KJ m <sup>-2</sup> )
Sisal	580	4.3	1250	3.5	98.7
Pineapple	640	2.4	970	2.2	79.5
Banana	540	3.0	816	1.9	51.6
Coir	140	25.0	3200	1.1	43.5

polymers capable of forming hydrogen bonds with hydroxyl groups available on the fibre surface. It was observed that the fibre pull out stress or debonding stress of sisal-polyester composites is only 166 MPa whereas, the debonding stress of sisal-epoxy matrix is about 226 MPa. It was also observed that, among polyester, epoxy and phenol - formaldehyde composites of sisal fibre, a phenolic type resin performed as a better matrix than epoxy and polyester resins with respect to tensile and flexural properties due to the high interfacial bonding in phenolic composites. They concluded that, compared to thermoset resin composites, sisal fibre-low density polyethylene (LDPE) composites showed a better reinforcing effect due to the high matrix ductility and high strength/modulus ratio of sisal fibres as compared to that of LDPE matrix.

Singh et al. (1996) have studied the effect of several chemical treatments, such as organotitanate, zirconate, silane, and N-substituted methacrylamide, on the physical and mechanical properties of sisal fibre reinforced unsaturated polyester resin composites. An improvement in the mechanical properties was observed when sisal fibres were modified with surface treatments. Under humid conditions, a decrease of 30 to 44% in tensile and 50 to 70% in flexural strength has been noted. The strength retention of surface-treated composites (except silane) is high compared with untreated composites. It was also observed that N-substituted methacrylamide-treated sisal composites exhibited better properties under dry as well as wet conditions.

A novel composite material of tamarind seed gum and the cellulosic rich sisal plant fibre was prepared and techniques were developed to increase the strength of the prepared composite material by a process of humidification and compression (Veluraja et al., 1997). It was reported that the prepared composite material have potential industrial applications such as false roofing and room partitioning. Sisal fibre reinforced rigid foam system based on plant polyols has been developed by Dahlke et al. (1998). They have reported that the properties of the polyurethane-sisal fibre system were comparable to polyether-based standard systems. Gupta et al. (1998) have studied the nature of interfacial adhesion between chemically modified sisal fibre and polyester resin in composites. Recently Bai et al. (1999) have studied the failure mechanisms of continuous sisal fibre reinforced epoxy matrix composites. They have examined the micro-failure behavior and interfacial debonding of sisal fibre bundle/epoxy matrix using scanning electron microscopy after four points bend tests. It was reported that sisal fibre bundle-epoxy interface had a moderate high strength, but the adhesive strength between the micro-tubular fibre and the bonding material appeared to be small.

## **SISAL FIBRE-REINFORCED THERMOPLASTIC COMPOSITES**

Thermoplastic polymers constitute an important class of materials with a wide variety of applications. Because of its increasing use combined with the high demand, the cost of the polymer has increased rapidly over the past decade. This situation made it necessary to use low cost fillers as means of reducing the cost of the end product. However, the widely used inorganic fillers, such as glass fibre and mica are very expensive compared to wood fibres. Several cellulosic products and wastes such as shell flour, wood flour and pulp have been used as fillers in thermoplastics (Lightsey, 1983; Kokta, 1988; Maldas &

Kokta, 1991). The influence of wood flour on the mechanical properties of polypropylene was studied by Raj et al. (1989) and they found that the cost of material could be reduced without too much loss of elastic modulus. However, fibrous fillers are now gaining more importance over particulate fillers due to their high performance in mechanical properties. Published data show that various commercial wood fibres have good potential as reinforcements in thermoplastics. Wood fibres are non-abrasive so that relatively large concentrations of fibres can be incorporated into polyolefins without causing serious machine wear during mixing and processing. Raj & Kokta (1989) have studied the mechanical properties of wood fibre filled medium density polyethylene (MDPE) composites. They observed a significant increase in modulus with increase in filler content. However, very limited studies have been reported in the literature on the use of sisal fibre as a reinforcing agent in thermoplastic matrices.

Joseph et al. (1992, 1993ab, 1994) have investigated the mechanical, rheological, electrical and viscoelastic properties of short sisal fibre reinforced LDPE composites as a function of processing method, fibre content, fibre length and fibre orientation. They have reported that the fibre damage normally occurs during blending of fibre and the polymer by the melt mixing method can be avoided by adopting a solution mixing procedure. They have also reported that unidirectional alignment of the short fibres achieved by an extrusion process enhanced the tensile strength and modulus of the composites along the axis of the fibre alignment by more than two fold compared to randomly oriented fibre composites. They have compared the experimentally observed tensile properties (tensile strength and modulus) of short sisal fibre reinforced-LDPE composites with the existing theories of reinforcement such as Parallel and Series, Hiesch, Cox, Halpin-Tsai, Modified Halpin-Tsai and modified Bowyer and Bader models Kalaprasad et al. (1997a). They concluded that tensile properties of short fibre reinforced composites strongly depend on fibre length, fibre loading, fibre dispersion, fibre orientation and fibre matrix interfacial bond strength. Influence of short glass fibre addition on the mechanical properties of short sisal fibre reinforced LDPE composites have been reported by Kalaprasad et al. (1997b). They observed that by the addition of a small volume fraction of ( $\cong 0.03$ ) short glass fibre into the above system enhanced the tensile strength of longitudinally oriented composites by more than 80%. It was also observed that water absorption tendency of the composite decreases with the process of hybridization. The viscoelastic properties, and rheological properties of LDPE filled with short sisal fibre as a function fibre length, fibre content and fibre orientation have been investigated by Joseph et al. (1992, 1993 b, 1994). They have reported that longitudinally oriented composites showed maximum storage moduli and a critical fibre length of 6 mm is necessary to obtain maximum dynamic moduli. The electrical properties of coir fibre and sisal fibre reinforced LDPE composites have been studied by Paul & Thomas (1997) and Paul et al. (1997). They have noticed that dielectric constant of sisal-LDPE and coir-LDPE progressively increases with increase of fibre loading in all frequencies ranging from 1 to 107 Hz. Selzer (1995) have studied the effect of environmental influences on the mechanical properties of sisal fibre reinforced polymer composites. Dependencies between moisture, acid as well as alkali attacks were determined and

mechanical properties of sisal fibre-polypropylene were evaluated. They have concluded that the bending properties of sisal fibre-polypropylene composites were sensitive to environmental attack.

The major drawbacks associated with the use of natural fibres as reinforcements in thermoplastics matrix to achieve composite material with improved mechanical properties and dimensional stability are the poor wettability and weak interfacial bonding with the polymer due to the inherently poor compatibility as well as dispersability of the hydrophilic cellulose fibres with the hydrophobic thermoplastics (Carvalho, 1997; Marcovich et al., 1997). So in order to improve the fibre-matrix adhesion a pre-treatment of the fibre surface or the incorporation of surface modifier during processing is required. Several studies have been reported based on the influence of various type of chemical modification on the physical and mechanical properties of sisal fibre filled thermoplastic composites (Bisanda & Ansell, 1991; Joseph et al., 1995b, 1996b; Paul et al., 1997). Graft copolymerization of methyl methacrylate onto sisal fibres using potassium persulfate initiator was studied by Sabaa et al. (1995). They have investigated the effect of the initiator concentration, monomer concentration, reaction time, reaction temperature and pH, oil grafting percentage, grafting efficiency and total conversion. The surface topology, as well as the x-ray diffraction patterns of the modified fibres were also studied.

Joseph et al. (1995a, 1996b) have studied the effect of chemical treatment on the tensile, dynamic mechanical, electrical and ageing properties of short sisal fibre reinforced LDPE composites. The effects of various chemical treatments on the tensile properties of sisal-polyethylene composites are presented in Table 6. Treatments using chemicals such as sodium hydroxide, isocyanate, permanganate and peroxide were carried out to improve the bonding at the fibre-polymer interface. It was observed that the treatments enhanced the tensile properties of the composites considerably, but to varying degrees. Figure 5 clearly indicates the extent of fibre-matrix adhesion after peroxide treatments on sisal fibre.

It has been observed that the CTDIC (cardanol derivative of toluene diisocyanate) treatment reduced the hydrophilic nature of the sisal fibre and thereby enhanced the tensile properties of the sisal-LDPE composites. They found that peroxide treated composites showed an enhancement in tensile properties due to the peroxide induced grafting. It was also found that permanganate treated composites also showed a similar trend

Table 6. Variation of tensile properties of longitudinally oriented LDPE-sisal composites with different fiber treatment (fiber length 5.8 mm, fiber content 30%) (Joseph et al., 1996b)

Composites	Tensile Strength (MPa)	Modulus (GPa)	Elongation at Break (%)
Untreated	31.12	3086	2
Alkali treated	34.27	3328	1
Isocyanate treated	41.50	4066	4
BP treated	40.90	4018	3
DCP treated	41.80	4156	4
KMnO <sub>4</sub> treated	38.80	3816	3

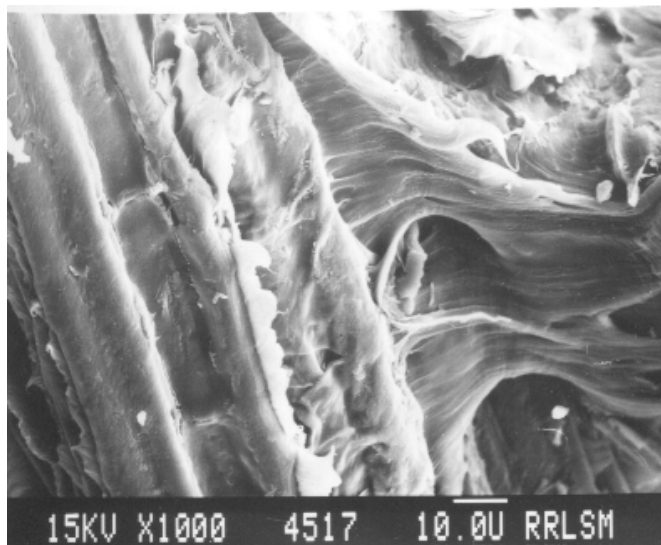


Figure 5. Scanning Electron Micrograph of peroxide treated sisal fiber-LDPE composites showing better fiber-matrix adhesion (Joseph et al., 1996b)

due to the permanganate induced grafting. It was observed that the dielectric constant values of the composites were found to have decreased as a result of chemical treatments Paul et al. (1997). This is due to the fact that the hydrophilic nature of natural fibre decreases with chemical treatment. It was shown that volume resistivity values of treated sisal fibre-LDPE composites were found to be greater than those of raw sisal fibre/LDPE composites.

They have also reported that the effects of fibre-matrix adhesion on the dynamic mechanical properties of the composites and found that the increase in adhesion increases the storage modulus. The effects of fibre length, fibre orientation, and fibre loading on the viscoelastic properties have also been investigated. It was found that in all cases, storage moduli ( $E'$ ) and loss moduli ( $E''$ ) decrease with temperature and increase with fibre loading. They concluded that among the various types of treatments they employed, CTDIC and dicumyl peroxide treated composites showed maximum physical and mechanical properties.

The effects of aging on the mechanical properties and dimensional stability of CTDIC treated and untreated sisal fibre reinforced LDPE composites have been studied by Joseph et al. (1995b). The ageing properties of sisal fibre composites were compared with those of glass fibre composites aged under identical conditions. Their results showed that CTDIC treated composites exhibited superior mechanical properties and better dimensional stability compared to untreated composites under identical aging conditions due to the existence of an efficient interfacial bond between fibre and the polymer matrix. The authors also reported that the better dimensional stability offered by glass/LDPE composite was due to the hydrophobic nature of glass fibre. They concluded that with a suitable fibre surface treatment, the mechanical properties as well as dimensional stability of sisal-LDPE composites could be improved.

LeThi et al. (1996) have studied the mechanical properties of sisal fibre reinforced polypropylene composites prepared by a reactive extrusion. It was reported that the grafting of the fibres by PP-graft-MA enhanced both the Impact strength and the



breaking stress of the composites. Manikandan Nair et al. (1996) have reported on the tensile properties short sisal fibre reinforced polystyrene composites. The influence of fibre length, fibre content, fibre orientation, and benzoylation on the tensile properties of the composites were evaluated. It has been reported that benzoylation on the fibre improves the fibre matrix adhesion and thereby increased the strength considerably. The properties were found to be almost independent of fibre length although the ultimate tensile strength shows marginal improvement at 10 mm fibre length. Effect of fibre surface treatment on the fibre-matrix bond strength of sisal fibre reinforced polyethylene composites was reported by Valadez-Gonzalez et al. (1999). It was observed that the interfacial shear strength (IFSS) between sisal fibres and polyethylene matrix has been improved by the morphological and silane chemical modification of the fibre surface. They have used an alkaline treatment to enhance both the matrix-fibre wetting and the chemical surface modification in order to improve the physicochemical interactions at the fibre-matrix interphase. It was found that both surface modifications, and pre-impregnation, improves the fibre-matrix IFSS. It was also observed that, results obtained from the single fibre fragmentation test seem to be better agreeing with the effective mechanical properties measured for the laminated material than those obtained with the pull out test.

Table 7 shows the tensile properties of solution mixed sisal fibre reinforced polypropylene (PP), low density polyethylene and polystyrene composites (Joseph et al., 1999). It is clear from the table that in the case both PP-sisal and LDPE-sisal composites, the tensile strength and modulus go on increasing as the percentage of fibre content increases from 0 to 30%, whereas, the values change in an irregular manner in the case of sisal – polystyrene composites. Since PP is more crystalline than LDPE, the increase in tensile strength by the addition of sisal fibre, is less in the case of PP compared to LDPE. But the strength of the composite formed by the addition of fibre is more in the case of PP compared to LDPE. In the case of polystyrene at 10% fibre loading, the tensile strength is decreased by 40% but in the case of PP, it is increased by 3%. However, at high fibre loading, the tensile strength values are comparable for both PP and polystyrene. Thus, PP is found to be a good matrix for sisal polyolefin composites.

## SISAL FIBRE REINFORCED RUBBER COMPOSITES

In recent years, short fibre reinforced elastomers have gained wide importance due to the advantages in processing and low cost coupled with high strength. Many researchers have used short glass fibres for reinforcing rubbers because of their high modulus, high strength and low creep. Moreover, reinforcement with short fibres offers some attractive features such as high modulus, tear strength etc. Major factors which affect the performance of rubber-fibre composites are fibre loading, fibre dispersion, fibre orientation, fibre to matrix adhesion and the aspect ratio of the fibre. These materials bridge the gap between conventional elastomers and fibres by combining the stiffness of short fibres with the elasticity of rubber. The major applications of these composites are in tyre treads, roofing's, hoses, sheeting's, V-belts, industrial rubber products and complex shaped articles. However, studies on composites containing plant fibres are important because of their renewable nature, low cost and amenability to chemical and mechanical modifications. A considerable amount of research work has been reported on plant fibre reinforced elastomer composites (Bhagavan et al., 1987; Varghese et al., 1992, 1994; Geethamma et al., 1995). Coran et al. (1974) have studied the properties of cellulosic fibre-elastomer composites and found that aspect ratio of the fibre has a major role on composite properties. The effects of particulate fillers on these composites have also been reported. It was found that fibre-matrix adhesion in this system could be promoted by the addition of definite proportions of silica/resorcinol/hexamethylene tetramine and that the addition of either carbon black alone or both silica and carbon black to a rubber compound containing resorcinol and hexa was associated to the achievement of a good adhesion between fibre and rubber-matrix and that the silica carbon black system exhibit an improved adhesion. It was also reported that processing properties like green strength and mill shrinkage were improved by the addition of fibre and that fibre addition also improved the tear strength by obstructing the development of the tear path.

Table 7. Comparison of the tensile properties of longitudinally and randomly oriented solution mixed sisal fiber reinforced polypropylene (PP), polystyrene (PS), and low-density polyethylene (LDPE) composites (fiber length 6 mm) (Joseph et al., 1999)

Fiber Content (%)	Composite Type	Tensile Strength (MPa)		Young's Modulus (MPa)		Elongation at Break (%)	
		L	R	L	R	L	R
0	PP	35.00	35.00	498	489	15.00	15.00
	PS	34.90	34.90	390	390	9.00	9.00
	PE	9.20	9.20	140	140	200.00	200.00
10	PP	36.00	29.00	730	605	7.82	8.00
	PS	21.30	18.16	629	516	9.00	7.00
	PE	15.61	10.80	1429	324	4.00	27.00
20	PP	39.10	31.14	971	798	7.11	7.33
	PS	43.20	25.98	999	553	8.00	6.00
	PE	21.66	12.50	2008	453	3.00	10.00
30	PP	44.40	33.84	1040	940	8.33	8.50
	PS	45.06	20.42	9998	624	7.00	4.00
	PE	31.12	14.70	3086	781	2.00	7.00

L = longitudinal and R = Random

O'Connor (1977) compared the mechanical properties of composites reinforced with five kinds of fibres and found that their mechanical properties depend on the type, volume loading, aspect ratio, orientation and dispersion of fibre and fibre-matrix adhesion. He also reported that for cellulosic fibres, a dicomponent dry bonding system consisting of hexamethylene tetramine and resorcinol is sufficient for getting good fibre - rubber adhesion, instead of the normal tricomponent dry bonding system consisting of hexa, resorcinol and silica. Geethamma et al. (1995) have studied the effects of fibre length, orientation and alkali treatment on short coir fibre reinforced natural rubber composites. The vulcanization parameters, processability characteristics and stress-strain properties of these composites were analyzed. They concluded that in general, the mechanical properties of the composites in the longitudinal direction were superior to those in the transverse direction, the optimum length for coir fibre in natural rubber system was found to be 10 mm in order to achieve good reinforcement in natural rubber composites; in order to achieve maximum tensile properties, coir fibre should be immersed in 5% sodium hydroxide solution for 48 h; anisotropic swelling studies indicated the poor adhesion between untreated coir fibre and natural rubber; the swelling was found smaller in composites containing alkali treated coir fibre along with the resorcinol-hexamethylene tetramine bonding agent.

Varghese et al. (1992) have studied the mechanical properties of acetylated and untreated short sisal fibre reinforced natural rubber composites and found that acetylation improves the adhesion between the rubber and the fibre. They have investigated the effect of different bonding agents on the physical and mechanical properties of sisal fibre reinforced natural rubber composites. The treatments employed included alkali immersion at high temperature and the use of bonding agent based on phenol-formaldehyde and resorcinol formaldehyde precipitated silica at different concentrations. They concluded that, the alkali treated fibre imparts better physical properties to the rubber mixes than the untreated fibre; sisal fibre acts as a reinforcing agent only when added above a volume loading of 10 phr (parts per hundred); the bonding between sisal fibre and rubber matrix is generally very poor but can be enhanced by resorcinol-formaldehyde pre-treated silica; the ageing resistance of the rubber - coir composites is excellent for a fibre loading of 30 phr with bonding agents and mechanical anisotropy is observed at fibre loading in excess of 10 phr. Prasantha & Thomas (1995ab) have investigated the processing behavior and mechanical properties of short sisal fibre reinforced styrene butadiene rubber (SBR) composites. Tear strength was examined with special reference to the effects of fibre length, fibre orientation, fibre concentration and bonding agent. It was observed that an increase in the concentration of fibre increases the tear strength in both longitudinal and transverse directions. It was also found that the tear strength values were almost three to four times higher than those of the unfilled vulcanizates under similar conditions. They have determined the green strength, mill shrinkage and Mooney viscosity of the composites to analyze the processing behavior.

The effect of adhesion on the equilibrium swelling of short sisal fibre reinforced natural rubber composites in a series of normal alkanes such as pentane; hexane, heptane and octane have been studied by Varghese et al. (1995). Their result showed that increased fibre content and the adhesion of bonding agent reduced the swelling considerably. It was also found that with improved adhesion between short fibre and rubber, the factor  $(V_r - V_r)/V_r$  decreases, where  $V_r$  and  $V_r$  are the volume fraction of rubber in dry and swollen samples respectively.

The increasing use of short fibre composites in static and dynamic applications led to the importance of stress relaxation measurements. Since the behavior of the rubber-fibre interface can be easily detected by stress relaxation studies. Vulcanized rubbers when subjected to constant deformation undergo a marked relaxation of stress both at low and high temperature. The stress under a constant deformation decays by an amount substantially proportional to the logarithm of the period in the deformed state. The stress relaxation behavior of short jute fibre reinforced - nitrile rubber composites with respect to the effect of strain level, bonding agent, fibre content, fibre concentration, temperature and pre-strain on the relaxation behavior has been studied in detail by Bhagawan et al. (1987). They concluded that in general, short fibre increase the rate of stress relaxation over the corresponding unfilled vulcanizates; composites containing bonding agent exhibit slower relaxation than those without a bonding system, the effect of fibre orientation on the relaxation behavior appears to be marginal; pre-strain decreases the stress relaxation rate considerably, particularly for composites without bonding agent. Varghese et al. (1994) have studied the stress relaxation behavior of acetylated short sisal fibre reinforced natural rubber composites with special reference to the effects of strain level, fibre loading, bonding agent and temperature. They reported the existence of a single relaxation pattern in the unfilled stock and a two-stage relaxation mechanism for the acetylated fibre filled natural rubber composites. It was also observed that, for the composites in the absence of bonding agent, the rate of relaxation increased with strain level, but in the presence of bonding agent, the relaxation rate is almost independent of strain level, because of the strong fibre-matrix interface.

## CONCLUSIONS

The use of sisal fibre as reinforcing agent in polymer based composites were reviewed from viewpoints of status and future expectations of natural fibres in general, structure and properties of sisal fibre, fibre surface modifications, and physical and mechanical properties of sisal fibre based polymer composites. Sisal fibres have good potential as reinforcements in polymer (thermoplastics, thermosets and rubbers) composites. Due to the low density and high specific properties of sisal fibres, composites based on these fibres may have very good implications in the automotive and transportation industry. More over, reduced equipment abrasion and subsequent reduction of re-tooling costs will make these composites more attractive. The use of sisal fibres as a source of raw material in plastic industry not only provides a renewable resource, but could also generate a non-food source of economic development for farming and rural areas. Since Brazil is the one of the largest

sisal fibre producing countries in the world, sisal fibre reinforced polymer composites and the subsequent applications would be very attractive from the economic point of view. From the above descriptions, it became quite evident that newer composites using abundantly available sisal fibres are on the horizon, this brings new trends in composite materials. It is worth mentioning that these composites can be used as a substitute for wood. However, suitable cost-effective design and fabrication techniques for manufacture should be developed. Sisal fibre polymer composites with and without hybridization should be developed and characterized so as to arrive at a series of composites which may find use in several areas such as marine, structural, consumer articles and industrial applications. Thus it can be concluded that with systematic and persistent research there will be a good scope and better future for sisal fibre – polymer composites in the coming years.

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