

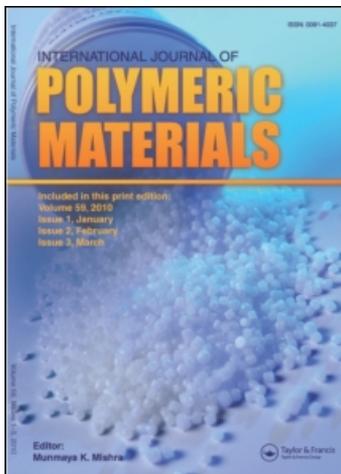
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### Green Composites from Natural Rubber and Oil Palm Fiber: Physical and Mechanical Properties

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## Green Composites from Natural Rubber and Oil Palm Fiber: Physical and Mechanical Properties

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*Natural rubber (NR) composites were prepared by incorporating short oil palm fibers of different lengths (viz., 2, 6, 10, and 14 mm) into natural rubber matrix in a mixing mill according to a base formulation. The curing characteristics of the mixes were studied and the samples were vulcanized at 150°C. The vulcanization parameters, processability characteristics, and tensile properties of these composites were analyzed. The effects of fiber length, orientation, loading, and fiber-matrix interaction on the mechanical properties of the green composites were studied. The reinforcement property of the alkali-treated fiber was compared with that of the untreated one. The extent of fiber orientation was studied from green strength measurements. From anisotropic swelling studies, the extent of fiber alignment and the strength of fiber-rubber interface adhesion were analyzed. Scanning electron microscopic (SEM) studies were carried out to analyze the fiber surface morphology, fiber pullout, and fiber-rubber interface.*

**Keywords:** composites, mechanical properties, oil palm fiber

### INTRODUCTION

Over the past decades, short fiber reinforced composites have been of great interest as they give rise to composites with improved mechanical properties compared to those containing non-fibrous fillers [1].

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The biodegradability of the natural fibers can contribute to a healthy ecosystem, while their low cost favors the economic interest of industry. The properties of the composites depend on fiber concentration, fiber dispersion, fiber-matrix adhesion, fiber orientation, and aspect ratio of fiber [2]. Lignocellulosic fibers like jute, sisal, coir, pineapple and banana have been used as reinforcement in thermoset matrices [3–4]. Wright and Mathias [5] succeeded in preparing light weight materials from balsa wood and thermoplastic polymers. Investigation has been carried out by Hedenberg and Gatenholm [6] in the recycling of plastic and cellulosic waste into composite.

Significant research efforts are currently being spent in developing a new class of fully biodegradable green composites by combining natural fibers with biodegradable resins [7–8]. Green composites can be prepared using a variety of natural and synthetic biodegradable resins. Researchers have developed jute fabric/Biopol composites [9]. In these cases, chemical treatment of fiber surfaces and alkali treatment of fibers were carried out to increase the fiber/resin interface bonding and improve the strength of the composite. Hermann and others have reported [10] the tensile properties of unidirectional laminates from ramie, hemp, and flax fibers in Sconacell A (starch modified resin by Buna Sow Leuna). Some of these composites were found to have properties comparable to E-glass/epoxy composites and were found to be suitable for a variety of structural applications.

There appeared in the literature several reports describing composites in which the polymeric matrix is reinforced by natural fibers [11–14]. In all cases an improvement in strength of the composites was noted, especially when the length of the reinforcing fibers fell in an optimal range. Green composites were also prepared from starch and modified starch blends. Takagi and co-workers [15] have reported composites based on modified starch resin and mao, hemp and bamboo fibers. Composites with excellent flexural properties could be produced.

The incorporation of short fibers in rubber compounds imparts increased strength and stiffness to the rubber matrices. It is also possible to achieve reduction of cost of the molding compound, improved processability and improved physical, chemical, and electrical properties in the final compound. Derringer [16] used short glass fibers for reinforcing rubbers. The composites showed a high modulus, mechanical strength, and low creep. Coran et al. [17] studied the behavior of cellulose fiber elastomer composites and found that the properties depend on the type of elastomer used, fiber concentration, fiber aspect ratio, and fiber orientation. The micromechanics of short fiber reinforced rubber composites was reviewed by Abrate [18] whereas Murthy and De [19] studied the effect of fillers on short jute-fiber

reinforced natural rubber composites. Setua and De [20] studied the effect of bonding agents in short silk fiber reinforced natural rubber composites. The role of silica in promoting adhesion in natural rubber composites had been studied by Creasy et al. [21]. Arumugam et al. [22] investigated the effect of different bonding agents on the physical properties of coir fiber reinforced natural rubber composites. Recently in this laboratory, short pineapple fiber, sisal fiber, coir fiber, and banana fibers have been successfully used for the reinforcement of natural rubber, low density polyethylene, and thermosets [23]. Joseph et al. [24] reported on the mechanical properties of sisal fiber reinforced epoxy, phenol formaldehyde, and thermoplastic composites. Stress relaxation in short sisal fiber reinforced rubber composites was studied by Varghese et al. [25]. Short sisal fiber reinforced styrene butadiene rubber composite was studied by Prasanth Kumar et al. [26]. Geethamma [27–28] reported the properties of short coir fiber reinforced natural rubber composites.

Oil palm fiber is a lignocellulosic fiber obtained from the empty fruit and bunch fibrous mesocarp of oil palm tree (*Elaeis guineensis*), cultivated extensively in the tropics. West Africa, South East Asian countries like Malaysia and Indonesia, Latin American countries, and India are the major oil palm cultivating countries. Oil palm fibers are hard and tough and show similarity to coir fibers in cellular structure. The oil palm fibers, which are obtained as waste material after oil extraction, can be used as cordage and floor furnishing materials, fiber foams, and cushions in vehicles. Oil palm fiber is used as a reinforcement in clay, cement, and different polymers. Sreekala [29] studied the properties of oil palm fiber reinforced phenol formaldehyde composites. Ismail [30] studied the effect of different bonding agents on oil palm reinforced polymer composites.

This study deals with the optimization of the oil palm fiber in natural rubber composites. The vulcanization parameters, processability characteristics, and stress-strain properties were analyzed. The extent of fiber orientation in the composites was determined from green strength measurements. From anisotropic swelling studies, the extent of fiber alignment and the strength of the fiber–rubber interface adhesion were analyzed. Scanning electron microscope was carried out to investigate the fiber surface morphology, fiber pull out, and fiber–rubber interface.

## EXPERIMENTAL

Oil palm fiber was obtained from Oil Palm India Ltd., Kottayam, Kerala, India. The properties of oil palm empty fruit bunch fiber are presented in Table 1. The empty fruit bunch was subjected to wetting.

**TABLE 1** Characteristics of Oil Palm Empty Fruit Bunch Fiber

Property	Percentage
Chemical constituents (%)	
Cellulose	65
Hemi cellulose	—
Lignin	19
Wax	—
Ash	2
Physical properties	
Diameter ( $\mu\text{m}$ )	150–500
Density (g/cc)	0.7
Tensile strength (MPa)	248
Young's modulus (MPa)	6700
Microfibrillar angle ( $^{\circ}$ )	46
Elongation at break (%)	14

Fibers were then cleaned, washed, and dried in an air oven at  $80^{\circ}\text{C}$  for 5 h and kept in polythene bags to prevent moisture absorption. Then they were chopped to different lengths, for example, 2, 6, 10, and 14 mm. Natural rubber used for the study was ISNR-5 (light color) grade obtained from Rubber Research Institute of India, Kottayam. The actual values of the specification parameters for the natural rubber used in this study are given in Table 2. All other ingredients were of commercial grade.

### Preparation of Composites

The formulations of the mixes are given in Table 3. Natural rubber was masticated on the mill for 2 min followed by the addition of the ingredients. The chemical constituents and physical properties of the fiber are given in Table 1. The composites were prepared in a laboratory two roll mill ( $150 \times 300$  mm) at a nip gap of 1.3 mm. The

**TABLE 2** Properties of Natural Rubber

Physical properties	Percentage
Dirt content by mass	0.03
Volatile matter by mass	0.50
Nitrogen by mass	0.30
Ash by mass	0.40
Initial plasticity, $P_0$	38
Plasticity retention index, PRI	78

**TABLE 3** Formulation of the Mixes (Phr/weight)

Ingredients	Gum	A	B	C	D	E	F	G	P	Q	R	S	T
Natural rubber	100	100	100	100	100	100	100	100	100	100	100	100	100
Stearic acid	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Zinc oxide	5	5	5	5	5	5	5	5	5	5	5	5	5
TDQ <sup>a</sup>	1	1	1	1	1	1	1	1	1	1	1	1	1
CBS <sup>b</sup>	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
Sulphur	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Oil palm fiber (untreated)	—	35	35	35	35	35	35	35	5	10	20	35	50
Oil palm fiber (NaOH treated)	—	—	—	—	—	5%	10%	15%	—	—	—	—	—
						(1/2h)	(1/2h)	(1/2h)					
Fiber length (mm)	—	2	6	10	14	6	6	6	6	6	6	6	6

<sup>a</sup>2,2,4-Trimethyl-1,2-dihydroxy guinoline polymerized.<sup>b</sup>N-Cyclohexyl-2-benzothiazyl sulphenamamide.

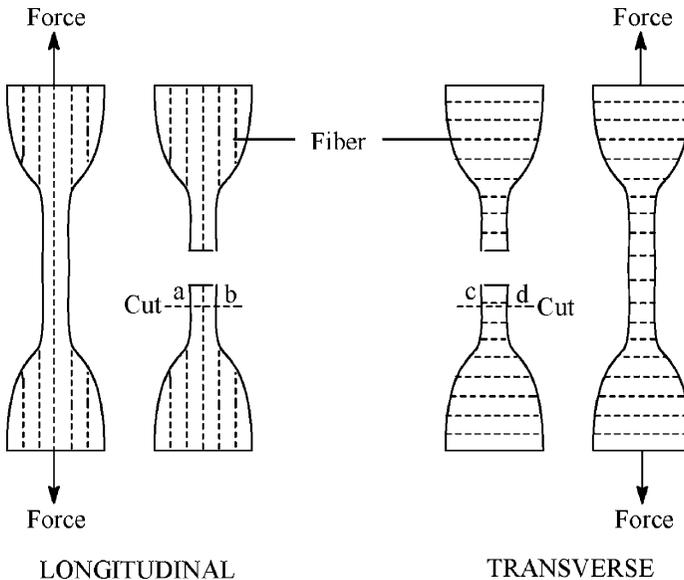
nip gap, mill roll speed ratio, mixing time, and the number of passes were kept the same in all mixes. Orientation of the fiber in the mill grain direction was achieved by repeated passing of the uncured compound through a tight nip gap.

## Property Measurement

Fiber breakage analysis was carried out by dissolving 1g of the uncured composite in toluene; fibers did separate out from the solution. Fiber distribution was analysed using a travelling microscope.

Green strength values were determined using dumb-bell-shaped samples obtained from un-vulcanized composites on a Zwick Universal testing machine Model 1474 at a stretching rate of 500%/min in accordance with ASTM D624-81. Curing properties were measured in a Monsanto R-100 rheometer at a temperature of 150°C.

Stress-strain measurements were carried out at a crosshead speed of 500 mm/min. Tensile strength and tear strength was measured according to ASTM methods D 412-68 and D624-54, respectively. The tensile tests were done with dumb-bell samples cut at different angles with respect to the orientation of fibers. Figure 1 shows schematic representation of the longitudinal transverse fiber orientation.



**FIGURE 1** Schematic representation of the tensile samples with fiber orientation and corresponding fractured surfaces.

Anisotropic swelling studies were carried out with rectangular samples cut at different angles with respect to orientation of the fiber from the tensile sheets and swollen in toluene at room temperature for 3 days. The length, breadth, and thickness of the samples were measured before and after swelling.

Scanning electron microscopy (SEM) studies were conducted with a Jeol JSM 5800 to analyze the fracture behavior of the composites. The fracture ends of the tensile and tear specimens were mounted on aluminium stubs and gold-coated to avoid electrical charging during examination.

## RESULTS AND DISCUSSION

### Fiber Breakage Analysis

The extent of fiber breakage during mixing was estimated by immersing 1 g of the unvulcanized mix containing oil palm fiber in toluene to dissolve the rubber component and thus separate the fibers intact. One hundred counts were made to determine the fiber length distributions. The fibers were washed with toluene to ensure complete removal of rubber from the surfaces. The washed fibers were then collected and examined under a travelling microscope. The control of the fiber length and aspect ratio of the fibers in a rubber matrix is difficult because of fiber breakage during processing. The severity of fiber breakage depends mainly on the type of fiber, the initial aspect ratio, and the magnitude of stress and strain experienced by the fibers during processing. The breakage of fibers due to high shear forces caused during mixing can be indicated by a fiber length distribution curve [31]. The distribution of fiber lengths can be represented in terms of moments of the distribution. The number and weight-average fiber lengths can be defined as:

$$L_n = \frac{\sum N_i L_i}{\sum N_i} \quad (1)$$

$$L_w = \frac{\sum N_i L_i^2}{\sum N_i L_i} \quad (2)$$

where  $L_n$  is the number-average fiber length,  $L_w$  the weight-average fiber length, and  $N_i$  the number of fibers having length  $L_i$ .

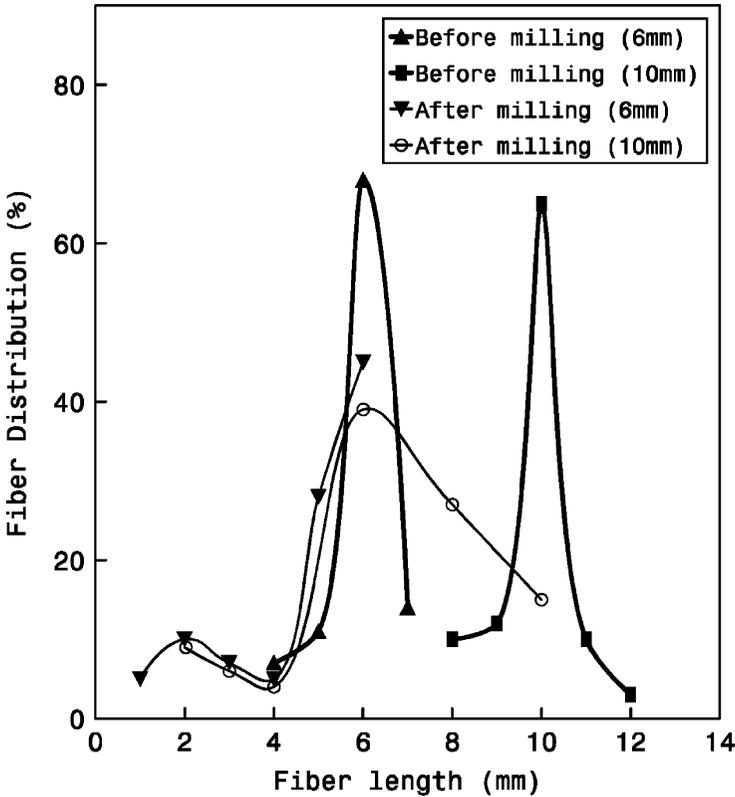
The value of  $L_w/L_n$ , the polydispersity index, can be taken as a measure of fiber length distribution. The values of  $L_n$ ,  $L_w$ , and  $L_w/L_n$  were calculated based on 100 fibers for the chopped oil palm fibers extracted from the mix. The fiber length distribution indices of the untreated oil palm fibers before and after mixing are given in Table 4. The value of

**TABLE 4** Fiber Length Distribution Index

Oil palm fiber (6 mm)	$L_n$ (mm)	$L_w$ (mm)	$L_w/L_n$
Before mixing	5.92	5.99	1.012
After mixing	4.68	4.75	1.014

$L_w/L_n$  remains about the same before and after processing indicating that no considerable fiber breakage occurred during mixing.

The fiber length distribution curves of the composites A and B, which contains untreated oil palm fibers of initial length 6 mm and 10 mm, is given in Figure 2. After mixing the majority of fibers were distributed between 5.7 and 6 mm. The breakage of oil palm fibers



**FIGURE 2** Fiber length distribution curve of the composites containing oil palm fiber before and after milling.

during mixing was low because oil palm fiber is a lignocellulosic fiber, it undergoes bending and curling rather than breaking during milling. The average diameter of the oil palm fiber (0.15 mm) remained the same after mixing.

Synthetic fibers like glass and carbon undergo severe breakage during mixing. Studies on glass and carbon fibers by O'Connor [32] showed that they are poor in improving mechanical properties compared to cellulosic fibers.

## Cure Characteristics

The processability of the compounds was studied from the rheographs. The minimum torque in the rheograph gives an indication of the filler content in the rubber, whereas the maximum torque in the rheograph is a measure of the crosslink density and stiffness in the rubber. In general, for all the mixes (Figure 3), the torque initially decreased, then increased, and finally levelled off. The initial decrease in torque to a minimum value was due to the softening of the rubber matrix, whereas the increase in torque was due to the crosslinking of the rubber. The levelling off is an indication of the completion of curing. Generally, the presence of fibers increased the maximum torque.

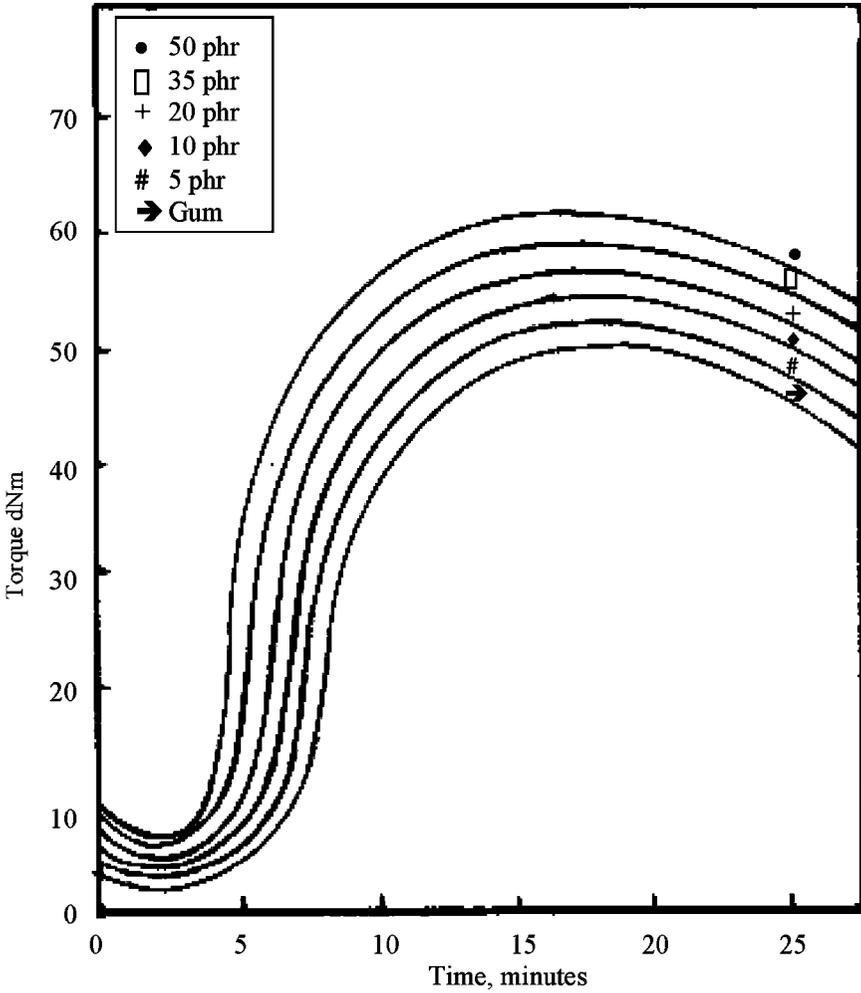
## Effect of Fiber Loading

Figure 3 shows the rheographs of gum 5, 10, 20, 35, and 50 phr mixes. The presence of fibers generated an increase in the viscosity of the mixes. The increment in torque values with increasing fiber loading indicated that as more and more fiber got into the rubber matrix, the mobility of the macromolecular chains of the rubber decreased, which resulted in more rigid vulcanizates. Table 5 shows the maximum torque, minimum torque, and cure time values for various mixes. The cure time was independent of the fiber loading.

## Mechanical Properties

### Effect of Fiber Length

The properties of short fiber reinforced elastomer composites depend on the degree to which an applied load is transmitted to the fibers. The extent of load transmittance is a function of the fiber length and the magnitude of the fiber–matrix interaction. At a critical fiber length ( $l_c$ ), the load transmittance from the matrix to the fiber is at a maximum. If  $l_c$  was greater than the length of the fiber, the stressed fiber debonds from the matrix and the composite fails at a low load.



**FIGURE 3** Rheographs of mixes gum, 5, 10, 20, 35, and 50 phr mixes.

**TABLE 5** Vulcanization Characteristics of Mixes

Mixes	$T_{\max}$ (dNm)	$T_{\min}$ (dNm)	$\Delta T$ (dNm)	$t_{90}$ (min)
Gum	56	7	49	14.9
P	57	5	52	10.6
Q	58	4	54	10.7
R	59	4	55	10.8
S	63	6	57	11.0
T	64	6	58	10.9

**TABLE 6** Effect of Fiber Length on the Mechanical Properties of the Mixes

Properties	Orientation	Gum	A (2 mm)	B (6 mm)	C (10 mm)	D (14 mm)
Modulus at 100% strain (MPa)	L	0.38	2.83	2.93	2.73	2.62
	T	0.37	2.79	2.85	2.68	2.53
Tensile strength (MPa)	L	21.36	9.10	9.50	8.51	8.42
	T	21.10	8.00	8.70	7.93	7.67
Tear strength (kN/m)	L	38.13	27.31	28.43	25.14	24.00
	T	35.06	25.93	26.55	21.32	20.00
Elongation at break (%)	L	1196	684	693	678	669
	T	1156	614	621	609	585

Table 6 shows the effect of fiber length on the tensile strength, elongation at break, and tensile modulus at 100% elongation. All of these properties were at a maximum when the length of the oil palm fiber was 6 mm. At higher fiber lengths, a decrease in the properties were found. This was due to the fiber entanglements prevalent at longer fiber length. From the overall mechanical property studies it was found that 6 mm was the optimum fiber length for oil palm fiber reinforced in NR matrix.

### Effect of Fiber Loading

The effect of fiber loading on the tensile properties of oil palm fiber reinforced NR composites are presented in Table 7. Generally, the tensile strength and tear strength decreases with increase of fiber loading. The behavior of the composites containing 5, 10, 20, 35, and 50 phr untreated oil palm fiber was studied. The modulus increased for natural fiber reinforced rubber composites in the longitudinal direction. In the transverse direction also the same trend has prevailed.

When fiber reinforced rubber composites are subjected to load, the fibers act as carriers of the load, and stress is transferred from

**TABLE 7** Effect of Fiber Loading on the Mechanical Properties of the Mixes

Properties	Orientation	P	Q	R	S	T
Modulus at 200% strain (MPa)	L	0.82	1.27	1.55	1.73	1.91
	T	0.73	0.94	1.2	1.62	1.7
Tensile strength (MPa)	L	19.2	18.1	12.2	9.59	7.28
	T	17.5	16.3	10.4	9.14	5.97
Elongation at break (%)	L	1082	1045	840	693	496
	T	940	861	719	621	386

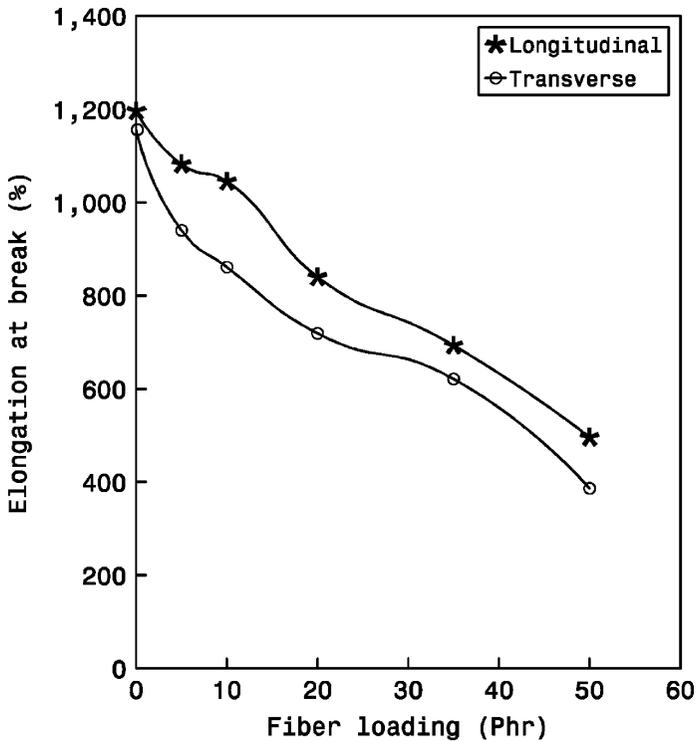
the matrix along the fibers, which leads to effective and uniform stress distribution, which results in good mechanical properties for the composites. The uniform distribution of stress is dependent on two factors: (1) the population and (2) the orientation of the fibers. At low levels of fiber loading, the orientation of the fibers is poor, and the fibers are not capable of transferring load to one another, and stress gets accumulated at certain points in the composite, which leads to a low tensile strength. At high levels of fiber loading, the increased population of fibers leads to agglomeration, and stress transfer gets blocked. At intermediate levels of loading (35 phr), the population of the fibers is just right for maximum orientation, and the fibers actively participate in stress transfer.

The longitudinal orientation of fibers resulted in a higher tensile strength than transversely oriented, as shown in Table 7. When the fibers were longitudinally oriented, the fibers were aligned in the direction of strain and the fibers transferred stress uniformly. When transversely oriented, the fibers were aligned perpendicular to the direction of load and they could not take part in stress transfer. As fiber loading increased the tear strength also decreased as the increased the strain in the matrix between closely packed fibers increased the tearing and reduced the tear strength.

Figure 4 shows the variation of elongation at break (%) with fiber loading. Elongation at break values showed a reduction with increasing fiber loading: the stiffness and brittleness of the composites increased gradually with an associated decrease in the elongation at break. This reduced the composites resilience and led to lower resistance to break. The stress strain curves of the mixes gum P, Q, R, S, and T are shown in Figure 5. From the figure it can be observed that the tensile strength decreased with increased fiber loading. Natural rubber inherently possesses high strength due to strain induced crystallisation. When fibers are incorporated into natural rubber, the regular arrangement of rubber molecules is disrupted and hence the crystallization ability will decrease. This may be the reason for the lower tensile properties of fiber reinforced natural rubber than the gum compounds.

The variation of modulus at 200% elongation with fiber loading is given in Figure 6. The value showed a steady increase with fiber loading. This is because at higher fiber loadings the composites are able to withstand more loads.

SEM studies have been used extensively to study fiber-matrix interaction. Figures 7(a) and (b) shows the scanning electron micrographs of fractured tensile specimens of the composites containing 35 phr fiber. SEM observations indicated that there was a certain difference in the

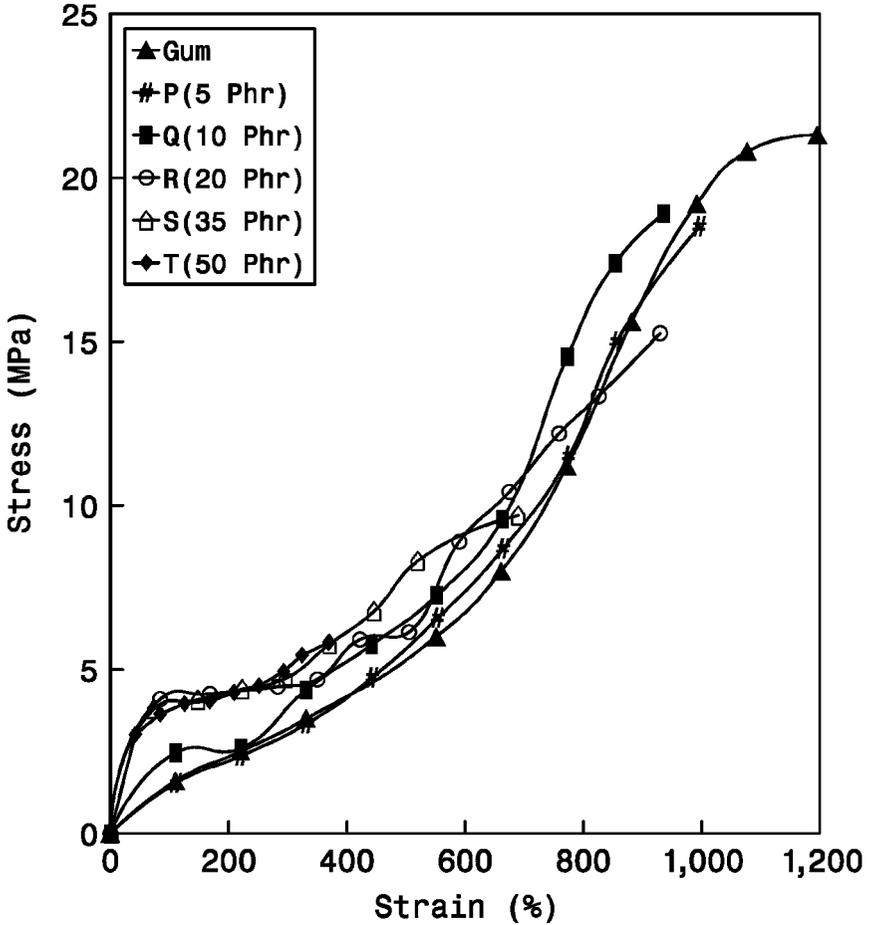


**FIGURE 4** Effect of fiber loading on the elongation at break values of oil palm fiber reinforced Natural Rubber composites (Longitudinal and Transverse).

fiber–matrix interaction depending on the loading of the composites. As shown in Figures 7(a) and (b), holes are seen as a result of the pullout of fibers from the rubber matrix. This pullout of fibers is evidence for poor fiber–matrix adhesion in the case of untreated oil palm fibers.

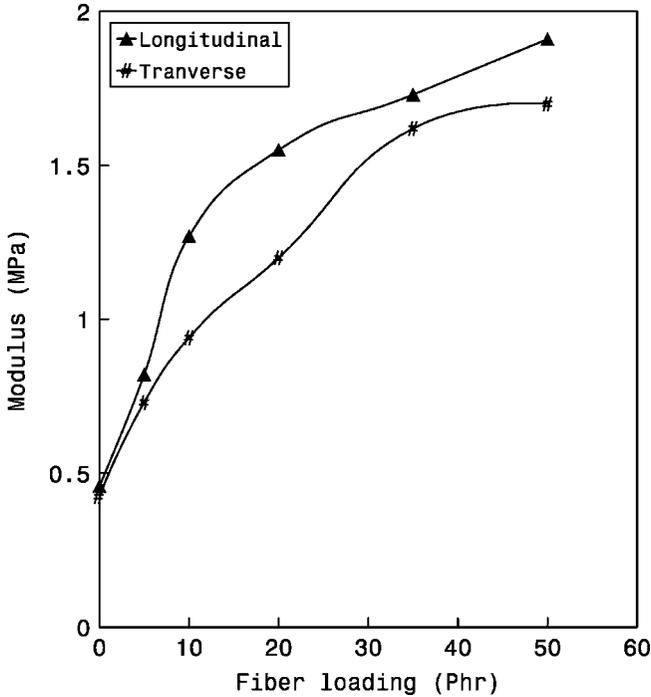
### Effect of Alkali Treatment

Good interfacial strength between fiber and rubber is an essential factor to achieve better reinforcement. The interfacial strength depends on surface topology of the fiber. In order to improve adhesion between oil palm fiber and natural rubber, fibers were treated with sodium hydroxide solutions of 5%, 10%, and 15% concentration for half an hour (E, F, G). The fiber changed color from pale yellow to brown. Fibers became thinner on alkali treatment. This may be because of dissolution and leaching out of fatty acids and/or phenolic compounds from the surface.



**FIGURE 5** Stress strain curves of gum, P, Q, R, S, and T mixes (Longitudinal).

The tensile moduli of the composites containing 35 phr oil palm fiber in varying concentrations of NaOH are shown in Table 8. It can be seen that tensile moduli, tensile strength, and tear strength of longitudinally oriented composites at 200% elongation are maximum for 5% concentration. However, as NaOH concentration increases there is a decrease in properties. This may be due to an excessive removal of binding materials such as lignin, hemicellulose, and so on from the surface as the concentration of NaOH increases. This decreases the fibrous properties and as a consequence, the properties are decreased. This trend is true in both transverse and longitudinal directions.



**FIGURE 6** Variation of modulus with fiber loading.

Figure 8 shows the SEM of the fractured end of the tensile specimens of the mix E. In the figure, broken fiber ends can be seen instead of pullout phenomenon. The cracks on the fiber ends support the fact that the fibers have undergone more breakage rather than pullout, which affirms a better interfacial strength.

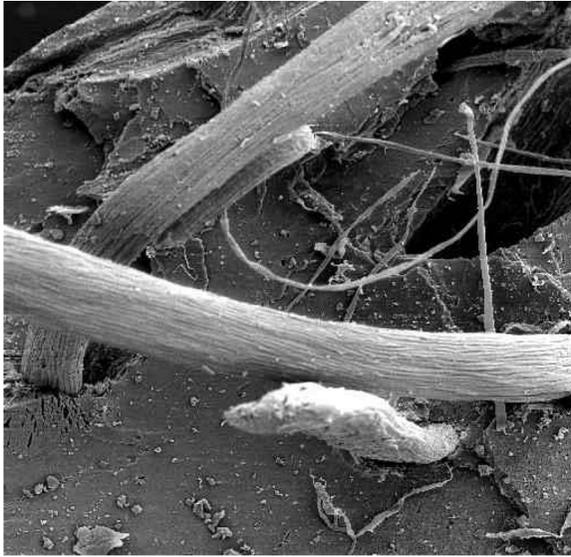
### Fiber Orientation

From the green strength values of various mixes, the extent of fiber orientation can be calculated using the equation [33]:

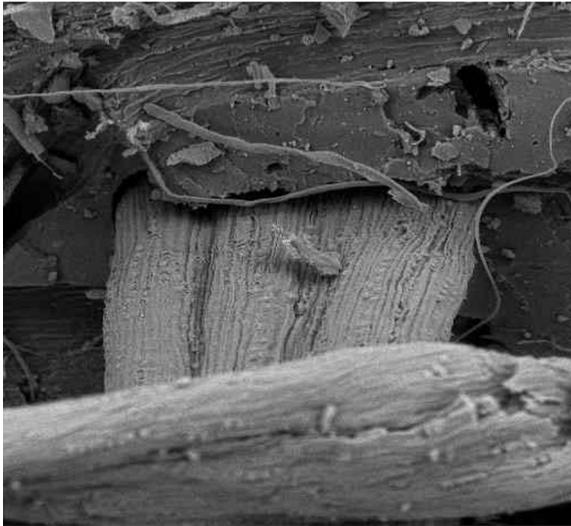
$$\% \text{ fiber orientation} = \frac{S_L/S_{G,L}}{S_L/S_{G,L} + S_T/S_{G,T}} \quad (3)$$

where S denotes the green strength of the composite and subscripts L, T, and G denote longitudinal, transverse, and gum compounds, respectively.

The effect of fiber loading on percentage orientation of the composites P, Q, R, S, T is given in Table 9. The percentage orientation



(a)



(b)

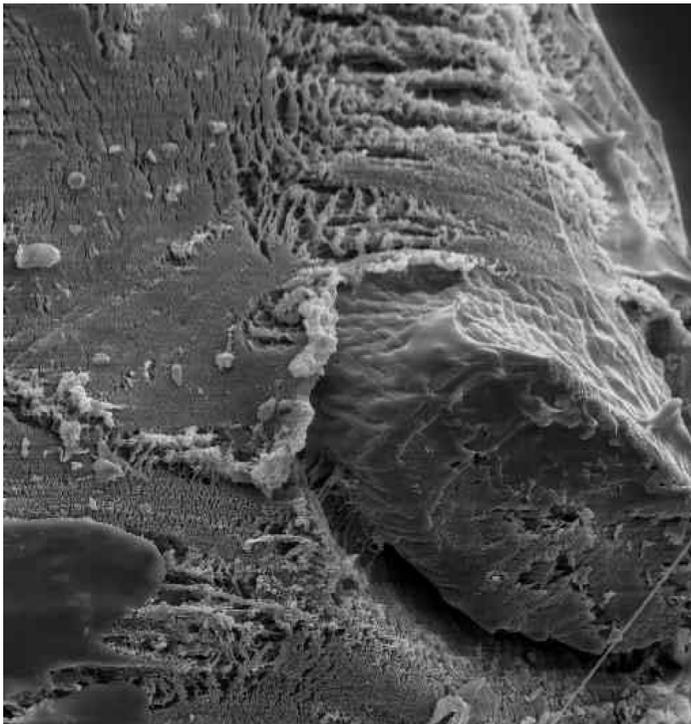
**FIGURE 7** Scanning Electron Micrographs of (a) tensile fracture surface showing fiber pull out and (b) poor fiber-matrix adhesion.

was the lowest when the fiber loading was small, that is, 5 phr. At low fiber loading, the fibers could randomly move around, which led to increased randomness and decreased levels of orientation. As the fiber

**TABLE 8** Effect of Soaking of Oil Palm Fibers in NaOH on Processability and Mechanical Properties

Properties	Orientation	Gum	E	F	G
Modulus at 200% strain (MPa)	L	0.38	1.92	1.85	1.80
	T	0.37	1.78	1.68	1.64
Tensile strength (MPa)	L	21.36	9.95	9.61	8.86
	T	21.10	9.52	9.31	8.32
Tear strength (kN/m)	L	38.13	30.42	28.49	25.50
	T	35.06	28.13	26.13	22.32
Elongation at break (%)	L	1196	730	690	670
	T	1156	710	655	610

loading is increased, the percentage orientation increased with a maximum value for the composites containing 35 phr fibers. When the fiber content is above 35 phr the percentage orientation decreased,

**FIGURE 8** Scanning Electron Micrographs of tensile fracture surface of mix E shows no fiber pull out, which indicates better fiber–rubber adhesion.

indicating that the fibers could not orient themselves because of entanglement caused by the increased population of fibers.

### Anisotropic Swelling Studies

Swelling is a uniform restrictive force induced on the vulcanizate samples. Because of the anisotropic nature of fiber reinforced rubber composites, swelling is restricted in the direction of fiber alignment. As a result swelling becomes anisotropic. The swelling behavior of fiber reinforced elastomeric composites has been studied by Das [34]. Recently, swelling behavior of natural rubber composites have been studied by Maya et al. [35] in various aromatic solvents. The swelling studies provide information on the strength of an interface, degree of dispersion of fibers and their alignment in the elastomeric matrix. Estimation of the extent of fiber orientation and the fiber–matrix interfacial adhesion in coir fiber reinforced natural rubber composites have been investigated by Geethamma et al. [28]. The swelling ratio  $a_\theta$

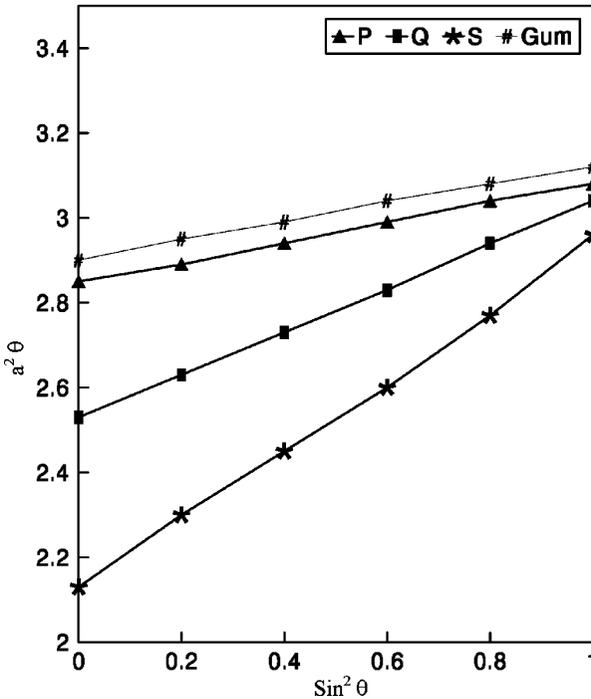


FIGURE 9 Swelling variation as a function of  $\theta$ .

**TABLE 9** Green Strength Values and Orientation

Mix (Phr)	S <sub>L</sub> (MPa)	S <sub>T</sub> (MPa)	Orientation (%)
Gum	0.248	0.236	—
P	0.59	0.56	50.2
Q	0.71	0.57	54.6
R	0.85	0.49	62.4
S	0.92	0.43	67.1
T	0.78	0.53	59.8

in a direction forming an angle  $\theta$  with the fiber orientation given by Coran et al. [36] is as follows:

$$a_{\theta}^2 = (a_T^2 - a_L^2)\sin^2\theta + a_L^2 \quad (4)$$

where  $a_L$  and  $a_T$  are the dimensional swelling ratios in the longitudinal and transverse directions. Figure 9 gives the dimensional swelling variation of mixes gum, P, Q, and S with angle  $\theta$  based on Eq. (4) where various values of  $\theta$  were assumed. For all mixes the swelling increases with  $\theta$  and is found to be maximum when  $\theta$  becomes  $90^\circ$ . This confirms that preferential fiber orientation is in the longitudinal direction. The line corresponding to the gum compounds, which does not contain fibers is positioned above those for mixes P, Q, and S. This showed that short fibers restricted the transport of solvent into the composite.

The extent of fiber alignment can be understood from the slope values given in Table 10. Noguchi et al. [37] showed that the mix corresponding to a line with a higher slope possess a greater degree of fiber alignment. Mix S showed the maximum slope value among the various mixes. This showed that the composite containing 35 phr fiber had the highest degree of fiber alignment.

**TABLE 10** Slope Values from the Anisotropic Swelling Studies

Mix	Slope values
Gum	0.1818
P	0.1912
Q	0.4445
S	0.5715

## CONCLUSION

The mechanical properties of green composites from natural rubber and oil palm fiber were investigated as a function of fiber length, orientation, and loading. Fiber breakage analysis revealed that the extent of breakage of fibers during milling was low. The mechanical properties of the composites in the longitudinal direction were superior to those in the transverse direction. Fiber length of 6 mm was found to be optimal to achieve good reinforcement in natural rubber composites. The addition of oil palm fiber led to a decrease in the tensile strength and tear strength but an increase in the modulus. Cure time was found to be independent of fiber loading. The fiber orientation and properties was higher for NaOH treated (5%) oil palm fibers than untreated ones. Anisotropic swelling studies indicated that the presence of short fibers restricted the entry of solvent into the composites. SEM studies revealed better adhesion between alkali treated oil palm fibers and natural rubber. Further studies on green composites from natural rubber, treated oil palm fibers, and cellulose (microfibrils) are in progress.

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