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Composite of short coir fibres and natural rubber: effect of chemical modification, loading and orientation of fibre

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Coir fibre is considered to be a poor reinforcing fibre in rubber because of its low strength and lack of physical characteristics that are essential for a reinforcing fibre. Interfacial adhesion between coir and natural rubber (NR) was improved by treatment of the coir fibres with alkali (sodium hydroxide and sodium carbonate) and NR solution, and by the incorporation of HRH/RH bonding systems. Composites containing 10 mm long coir fibres were vulcanized at 150°C according to their respective cure times. Green strength measurements were carried out to measure the extent of fibre orientation. The effectiveness of green strength measurements in finding the degree of fibre orientation was also analysed. Tensile modulus, tensile strength and tear strength of the composites were analysed to investigate their performance. Anisotropic swelling studies were conducted to investigate the extent of fibre orientation and the fibre/rubber interfacial strength. Scanning electron microscopy was performed to study the fibre topology. © 1997 Elsevier Science Ltd. All rights reserved.

(Keywords: composites; coir fibres; natural rubber)

INTRODUCTION

Nowadays, fibre-reinforced rubber composites are of tremendous importance both in end-use applications and in the area of research and development. These composites exhibit the combined behaviour of the soft, elastic rubber matrix and the stiff, strong fibrous reinforcement. The use of short fibres makes it almost impossible to obtain the high level of reinforcement that can be attained with long fibres. Still, short-fibre composites are preferred in products such as V-belts[1] and hoses[2] because of their easy processability and high green strength, and the possibility of producing complex-shaped articles.

A considerable body of research has been carried out on glass, carbon, polyester and nylon as short-fibre reinforcement in rubber composites. A thorough investigation was done by Derringer[3] on the effects of the HRH (hexa–resorcinol–hydrated silica) bonding system, fibre orientation and fibre/matrix adhesion on carbon black-filled rubber composites containing different kinds of fibre such as rayon, glass and asbestos. Coran and Hamed[4] have reviewed the reinforcement of elastomers with short fibres. O'Connor[5] studied the processing and properties of natural rubber and nitrile rubber composites containing a variety of short fibres along with carbon black. Chakraborty *et al.*[6] studied the effect of silica and the concentrations of resorcinol and hexa in the HRH bonding system on jute-fibre-reinforced carboxylated nitrile rubber composites. The micromechanics of short-fibre-reinforced rubber composites was reviewed by Abrate[7]. Recently, in this laboratory, short sisal, pineapple, banana, oil palm and coir fibres have been used for reinforcing natural rubber[8–10], styrene–butadiene rubber[11,12], low-density polyethylene[13–16], polystyrene[17] and various thermosetting resins.

Coir is a versatile lignocellulosic fibre obtained from coconut trees (*Cocos nucifera*), which grow extensively in tropical countries. Because of its hard-wearing quality, durability and other advantages, it is used for making a wide variety of floor furnishing materials, yarn, rope, etc.[18]. But these traditional coir products consume only a small percentage of the potential total world production of coconut husk. Hence, apart from the conventional uses of coir as mentioned above, research and development efforts have been underway to find new use areas for coir, including utilization of coir as a reinforcement in polymer composites[19–21].

Coir is an inexpensive fibre among the various natural fibres available in world. Furthermore, it possesses the advantages of a lignocellulosic fibre. It is not brittle as glass fibres, is amenable to chemical modification, is non-toxic and poses no waste disposal problems. Unfortunately, the performance of coir as a reinforcement in polymer composites is unsatisfactory and not comparable even with other natural fibres. This inferior performance of coir is due to various factors such as its low cellulose content, high lignin and hemicellulose content, high microfibrillar angle, and large and variable diameter. The various physical properties of coir fibres are compared with those of other natural fibres in *Table 1*[22].

The efficiency of coir as a reinforcement in rubber composites can be improved by enhancing the interfacial adhesion between coir and rubber. This can be achieved

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Table 1 Comparison of cost and properties of coir fibres with other natural fibres

	Coir	Banana	Sisal	Pineapple	Jute
Diameter (μm)	100–460	80–250	50–200	20–80	—
Density (g cm^{-3})	1.15	1.35	1.45	1.44	1.45
Microfibrillar angle (°)	30–49	11	10–22	14–18	8.1
Cellulose/lignin content (%)	43/45	65/5	67/12	81/12	63/12
Elastic modulus (GPa)	4–6	8–20	9–12	43–81	20–22
Tenacity (MPa)	131–175	529–759	568–640	413–1627	533
Elongation (%)	15–40	1.0–3.5	3–7	0.8–1.6	1–1.2
Cost (relative to coir)	1	3	1.5	1.5	2

Table 2 Base formulation of mixes

Natural rubber (ISNR 5)	100
Zinc oxide (phr)	5
Stearic acid (phr)	1.5
TDQ[a] (phr)	1
Resorcinol (phr)	7.5
Hexa[b] (phr)	4.8
CBS[c] (phr)	0.6
Sulfur (phr)	2.5
Coir fibre (untreated) (phr)	30
Coir fibre (treated) (phr)	30

Description of mixes

Mix I—Untreated fibre + NR[d] solution + TDI[e] solution

Mix J—Sodium hydroxide-treated fibre + NR solution + TDI solution (J_{10}, J_{20}, J_{30}, J_{40} and J_{60} contain respectively 10, 20, 30, 40 and 60 phr coir fibre subjected to this treatment)

Mix K—Sodium hydroxide-treated fibre + liquid NR solution + TDI solution

Mix L—Untreated fibre + resorcinol + hexa

Mix M—Untreated fibre + resorcinol + hexa + 7.5 parts silica

Mix N—Sodium carbonate-treated fibre + resorcinol + hexa

Mix O—Sodium carbonate-treated fibre + resorcinol + hexa + 7.5 parts silica

[a] Polymerized 2,2,4-trimethyl-1,2-dihydroxyquinoline
[b] Hexamethylenetetramine
[c] *N*-Cyclohexyl-2-benzothiazyl sulfenamide
[d] Natural rubber
[e] Toluene diisocyanate

either by modifying the surface topology of coir by a suitable pretreatment or by selecting the proper components of the bonding system. It has been observed that sodium hydroxide treatment of coir fibre will enhance the bonding of coir with natural rubber matrix[23].

This paper deals with the effect of different chemical treatments of coir fibre on the mechanical properties of coir-fibre-reinforced natural rubber composites. The role of silica in the tricomponent 'hexa–resorcinol–hydrated silica' (HRH) dry bonding system in improving the fibre/rubber adhesion is also investigated. The extent of fibre orientation is determined from green strength and anisotropic swelling measurements. Scanning electron microscopy (SEM) studies were undertaken to characterize the fibre surface topology. Attempts have also been made to investigate the influence of fibre loading and orientation on the mechanical performance of the composites.

EXPERIMENTAL

Materials

Coir fibre was obtained from the local processing unit in Kollam, India. Coir contains cellulose (36–43%), lignin (41–45%), hemicellulose (0.15–0.25%) and pectins (3–4%) together with some water-soluble materials[24]. It has an average diameter of 100–400 μm and a specific gravity of 1.15. ISNR 5 (light colour) grade natural rubber used for the study was supplied by the Rubber Research Institute of India, Kottayam. All other ingredients used were of commercial grade.

Fibre preparation

Coir pith and other undesirable materials were separated from the coir fibre. It was then chopped to about 10 mm length. Sodium hydroxide and sodium carbonate treatments were carried out by soaking coir fibres in their 5% solutions for 48 h. Fibres were taken out, repeatedly washed with water and dried in air.

Both untreated and sodium hydroxide-treated fibres were given a pretreatment with a mixture of 1% natural rubber solution and 1% toluene diisocyanate (TDI) solution. Then these fibres were separated and oven dried. Similarly, sodium hydroxide-treated fibres were treated with a mixture of 6% depolymerized liquid natural rubber solution and 1% TDI solution.

Preparation of composites

The formulations of the mixes are given in *Table 2*. The composite materials were prepared in a two-roll laboratory mill (150 mm × 300 mm). The nip gap, mill/roll speed ratio and number of passes were kept the same for all mixes. The samples were milled for sufficient time to disperse the fibres in the matrix at a mill opening of 1.25 mm. The tricomponent dry bonding system consisting of resorcinol, hexamethylene tetramine and hydrated silica, was incorporated along with other ingredients. The coir fibres were incorporated at the end of the mixing process taking care to maintain the direction of compound flow, so that the majority of fibres are in the same direction.

Characterization

Green strength values were determined using dumb-bell shaped samples obtained from unvulcanized composites, at a stretching rate of 500% min^{-1}. Curing properties were measured in a Monsanto R-100 rheometer at a temperature of 150°C. The compounds were cured for their respective cure times. Tensile and tear specimens with longitudinal and transverse fibre orientations were punched out from the vulcanized composite.

Stress–strain measurements were carried out at a cross-head speed of 500 mm min^{-1} in an Instron tensile testing machine. Tensile and tear tests were conducted according to ASTM Standards D412-68 and D624-54, respectively. These tests were conducted both along and across the grain direction. The tensile moduli at 50%, 100% and 200% elongation were measured from the stress–strain graphs.

The anisotropic swelling experiments were carried out on rectangular specimens (2.5 cm × 1.0 cm × 0.3 cm) after an immersion period of 48 h in distilled toluene. Scanning electron microscopy studies were conducted in order to analyse the fracture behaviour of the composites.

RESULTS AND DISCUSSION

The primary role of a composite material in a product is to support the applied mechanical forces. A composite achieves this by load transfer between the matrix and the fibres induced by the shear deformation of the matrix around the fibres[25]. This shear deformation is produced because of the high Young's modulus of the fibre and the large difference between the mechanical properties of the composite's constituents.

The physical properties of short-fibre composites are intermediate between those of composites containing continuous fibres and particulate-filled composites. Besides component properties, the fibre orientation and the fibre/matrix interfacial adhesion are two critical parameters that affect the performance of any short-fibre-reinforced rubber composite. But the direct estimation of these parameters is difficult. This creates problems in establishing the actual morphology of these composites and so the prediction of the composite performance on the basis of interface morphology and the extent of fibre orientation becomes difficult.

Different methods have been used to measure the extent of fibre orientation. Digital image processing was used by Gadala-Maria and Parsi[26] to measure the distribution of fibre orientation. The estimation of fibre orientation of short-fibre-reinforced thermoplastic composites by image analysis has been reported by Fischer and Eyerer[27], but this method is not so dependable in the case of rubber composites because of the difficulty in achieving the required surface finish of the test specimens. Thus indirect methods such as enhancement in mechanical properties and anisotropic swelling studies must be relied upon in order to get information about these important parameters.

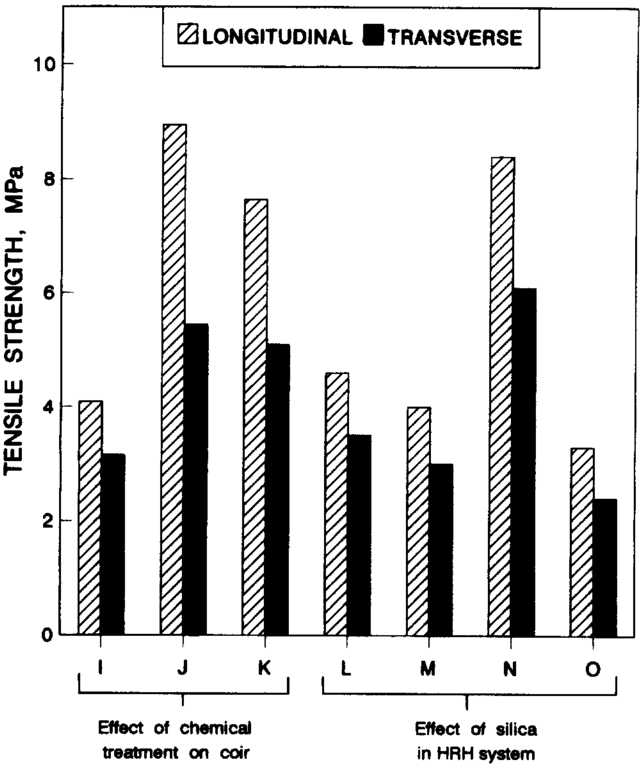

Figure 1 Tensile strength of the mixes

Effect of chemical treatment on interfacial adhesion

The interface is a region at least several molecular layers thick whose properties are intermediate between those of the fibre and matrix phases because of the peculiar restrictions on molecular motion in this zone. Matrix molecules may be anchored to the fibre surface by chemical reaction or adsorption, which determine the extent of interfacial adhesion. The interface may be composed of an additional constituent added to the composite as a bonding agent or as an interlayer between the two components of the composite. In the present work, the interfacial adhesion was improved by two means.

By the formation of an interlayer. The interlayer was produced by giving a coating of either natural rubber or depolymerized natural rubber to the surface of the coir fibres. *Figure 1* shows the influence of the nature of the interfacial region on the tensile strength of coir/rubber composites. Mix J contains sodium hydroxide-treated coir fibres which were subjected to a pretreatment with natural rubber and TDI solution. The tensile strength of mix J is about 120% higher than that of mix I, the latter containing raw coir fibres subjected to treatment with natural rubber and TDI solution. So it is clear that even though both raw and alkali-treated coir fibres were subjected to the same chemical treatment, the latter are better at reinforcing the natural rubber matrix. This indicates that a pretreatment of natural rubber and TDI solution to raw coir is not sufficient to produce good interfacial adhesion with the rubber matrix because of the waxy cuticle layer on the surface of the raw coir fibre. This is evident from the SEM photographs of the

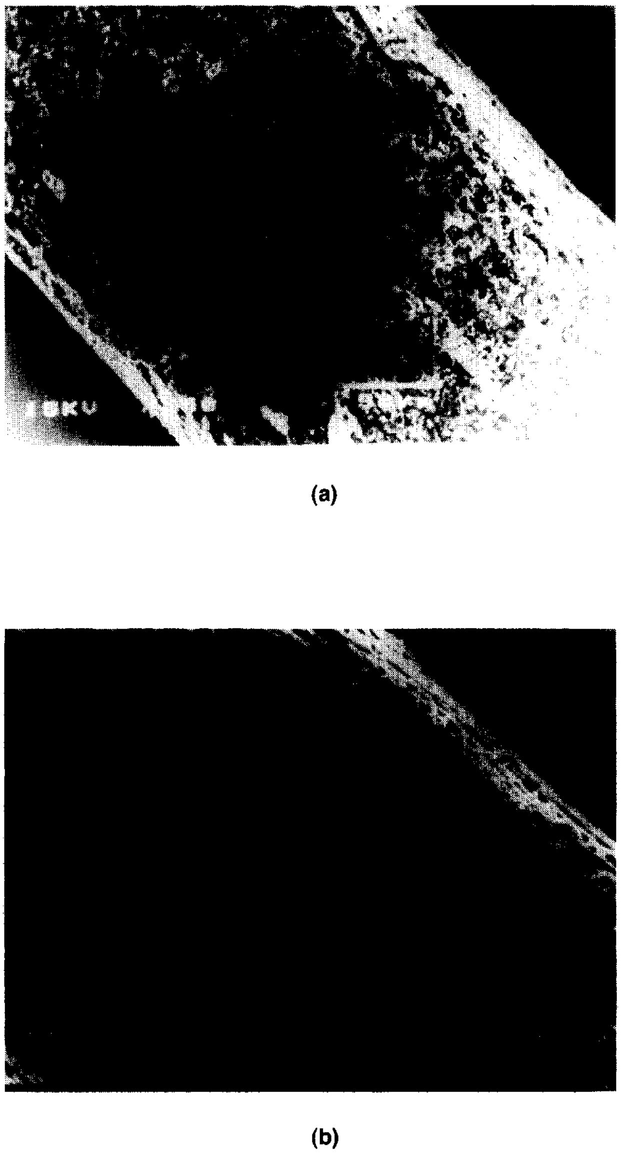

Figure 2 SEM micrographs of the surface of (a) an untreated coir fibre (globular protrusions on the surface) and (b) an alkali-treated coir fibre (voids are formed on the surface)

coir fibre surface. The surface of raw coir fibre contains globular protrusions identified as silicified stigmata (*Figure 2a*). It has been found that voids are produced on the surface of coir by alkali treatment as a result of the removal of these globular protrusions (*Figure 2b*). The loss of cuticle by the breakage of alkali-sensitive bonds makes the fibre surface rough and, as a result, the extent of interaction with natural rubber solution will be greater owing to the increased surface area of these fibres.

When alkali-treated coir is given a coating of depolymerized natural rubber and TDI solution (mix K) it exhibit certain anomalous characteristics. The stress–strain curves of mixes I, J and K in the longitudinal direction are shown in *Figure 3*. The tensile strength of mix K is lower than that of mix J. The moduli values at three different elongations, i.e. 50%, 100% and 200%, are highest for mix J (*Table 3*).

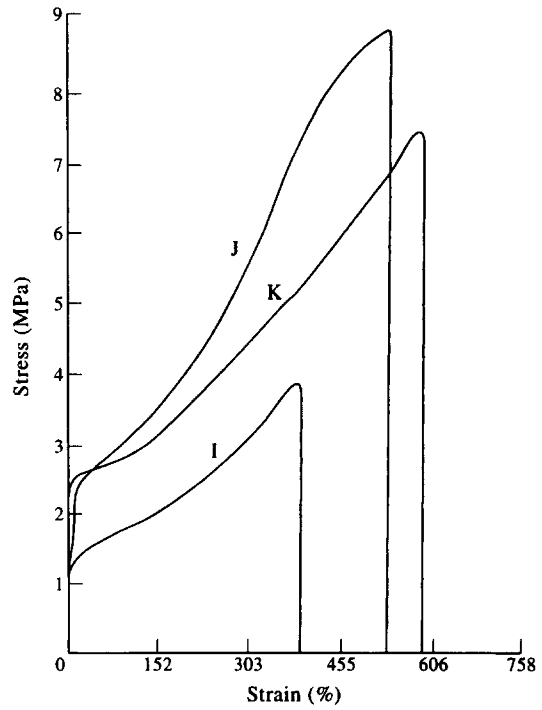

Figure 3 Stress–strain curves of mixes I, J and K (longitudinal)

Table 3 Tensile moduli and elongation at break of the mixes

Mix	Orientation[a]	Modulus at 50% elongation (MPa)	Modulus at 100% elongation (MPa)	Modulus at 200% elongation (MPa)	Elongation at break (%)
I	L	1.6	1.9	2.3	386
	T	0.9	1.2	1.5	404
J	L	2.7	3.1	4.2	537
	T	1.3	1.6	2.2	719
K	L	2.7	2.8	3.6	590
	T	1.1	1.4	1.8	618
L	L	2.4	2.5	2.7	650
	T	1.2	1.5	1.7	703
M	L	2.0	2.1	2.2	683
	T	1.3	1.4	1.5	790
N	L	2.9	3.0	3.2	550
	T	1.5	1.8	2.1	574
O	L	1.5	1.7	2.0	751
	T	0.8	1.2	1.3	800

[a] L = longitudinal; T = transverse

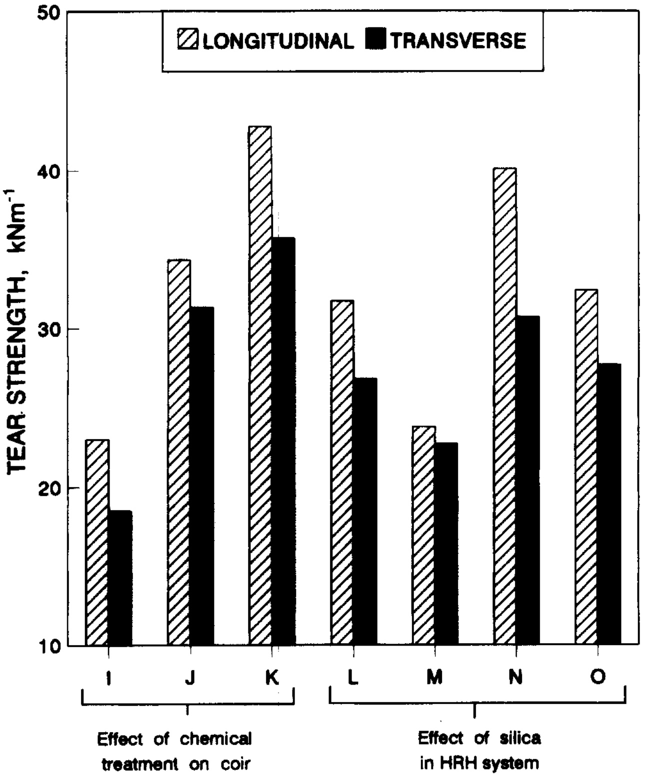

Figure 4 Tear strength of the mixes

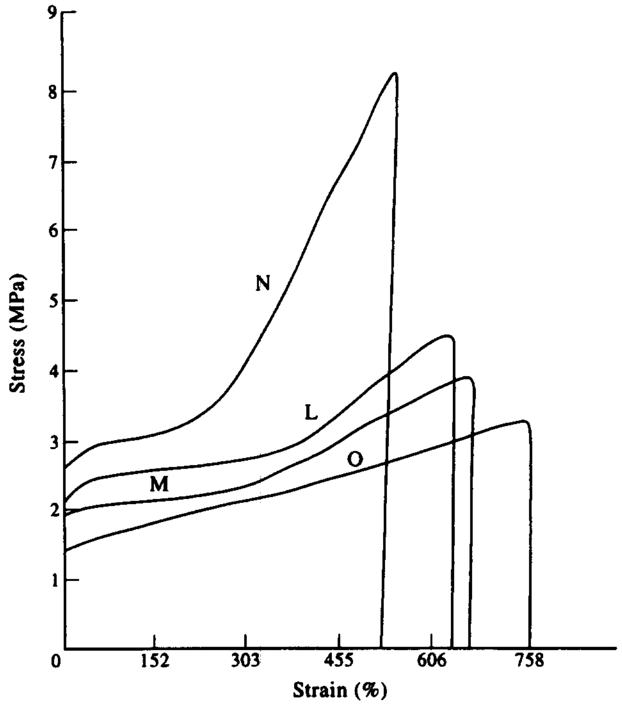

Figure 5 Stress–strain curves of mixes L, M, N and O

This indicates the improved stiffness of mix J. Moreover, the elongation at break values of mix J are smaller than those of mix K. Nevertheless, the tear strength of mix K is higher than that of mix J by about 25% (*Figure 4*). Thus it is clear that the performance of mix J is better than that of mix K in certain respects only. Intensive studies are going on in this laboratory to investigate the peculiar behaviour of liquid natural rubber as a size on coir fibre.

By the incorporation of HRH bonding system. Chakraborty *et al.*[6] found that silica is not necessary in the bonding system for jute-fibre-reinforced carboxylated nitrile rubber composites. Murty and De[28] observed that 5 phr silica is essential for enhancing fibre/rubber adhesion in short-jute-fibre/natural-rubber composites. But Varghese *et al.*[29] reported that in the natural rubber matrix, silica is not needed in the bonding system when sisal fibre is used as a reinforcement. Hence it is clear that nature of both the rubber matrix and the reinforcing fibre determines whether silica is needed or not as one of the components.. Analysis of stress–strain curves (*Figure 5*) shows that silica is not needed in the bonding system for coir-fibre-reinforced natural rubber composites. Raw coir fibres are used as reinforcement in mixes L and M, but mix L exhibits higher tensile strength than mix M. This can be explained by the presence of the unnecessary component of silica in the bonding system in mix M. Similarly, the tensile strength of mix N is higher than that of mix O even though both of these mixes contain sodium carbonate-treated coir fibre. This is also due to the presence of silica in mix O.

Likewise, the tensile moduli of mixes L and N for all the three different percentage elongations are greater than those of mixes M and O, in both longitudinal and transverse directions (*Table 3*). The elongation at break values of mixes L and N are lower than those of mixes M and O. This confirms the better fibre/rubber adhesion in the former mixes. *Figure 4* shows the tear strength of mixes L, M, N and O. These mixes exhibit the same trend as in tensile strength and moduli. Thus from the mechanical properties of the composites it can be observed that silica is not essential in the bonding system in order to achieve good interfacial adhesion.

Effect of fibre orientation on mechanical properties

The fibre-reinforced composites exhibit anisotropy in mechanical properties because of fibre orientation along the flow direction during processing. The performance of any short-fibre-reinforced polymer composite is dependent upon the angle between the directions of fibre orientation and the applied force or, in other words, the extent of fibre orientation. The efficiency of stress transfer is higher if fibres are aligned parallel to the direction of application of force. Thus processing techniques aim to orient the fibres in the preferred direction to meet the anticipated loads on fabricated products. Moghe[30] studied the effects of milling parameters such as the number of passes, nip gap, roll/speed ratio, etc. on fibre orientation in rubber composites. He concluded that only the nip gap has a significant effect on the fibre orientation and that the majority of fibres are oriented in the first pass itself.

Table 3 shows that even if the moduli values in the longitudinal direction are higher than those in the transverse direction, the stiffness across the fibre direction is somewhat similar to that of the rubber matrix. The moduli of mix J is higher than those of mixes I and K for the three different elongations. This is because of the higher fibre/matrix adhesion that exists in the composites containing alkali-treated coir fibres coated with natural rubber solution. This increase in modulus is similar to that achieved by the usual chemical crosslinking reaction. But the use of short fibres leads to anisotropic behaviour of the modulus, which is rather unlikely in the normal crosslinking reaction.

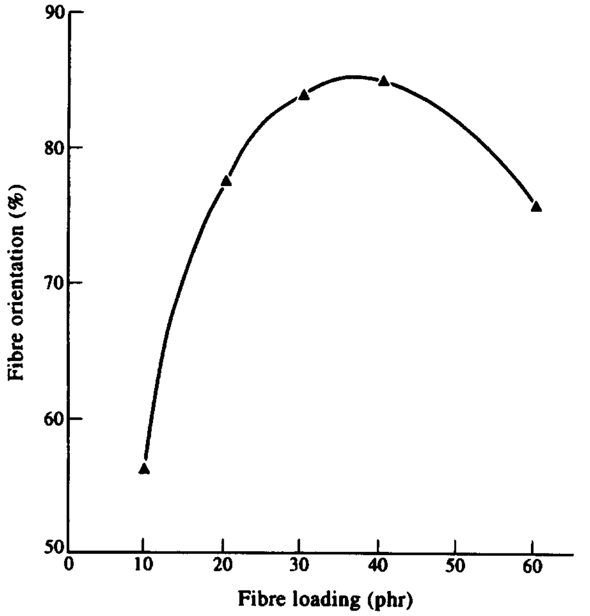

Figure 6 Variation of the percentage fibre orientation with fibre loading

Extent of fibre orientation from green strength measurements

A very important advantage of short-fibre-reinforced rubber composite is their enhanced green strength which facilitates shape retention prior to complete cure. This in turn leads to early demoulding, which can reduce the moulding cycle time[31]. The green strength of short-fibre-reinforced composites depends upon the degree of fibre orientation and so the latter can be obtained from the expression[32]

$$\% \text{ Fibre orientation} = \frac{S_L/S_{G,L}}{S_L/S_{G,L} + S_T/S_{G,T}} \quad (1)$$

where S represents green strength and subscripts G, L and T represent gum, longitudinal and transverse, respectively.

The effect of fibre loading on percentage orientation of chemically treated fibre with natural rubber and TDI solution is shown in *Figure 6*. The percentage orientation is the lowest when the fibre loading is small, i.e. 10 phr. At low levels of fibre loading, the fibres can assume a multitude of alignment directions and so the freedom of movement is more. Thus the chaoticity of their distribution is increased. The percentage orientation increases as the loading of fibre increases. A sharp increase of about 38% is observed for the compound containing 20 phr fibre compared with 10 phr fibre loading. But the increase is only about 8% for the composite containing 30 phr fibre over that for 20 phr fibre loading. After 30 phr fibre loading, this increase is gradual and it even decreases after 40 phr fibre loading. It is clear that, at 60 phr fibre loading, the fibres cannot orient themselves in the unidirectional fashion that has occurred in composites containing 20, 30 or 40 phr fibre loading because of entanglement as a result of the overpopulation of fibres.

Drawbacks of green strength measurements

Fibres orient during processing and consequent fabrication depending upon the nature of the flow, i.e. convergent, divergent, elongational or shear. If flow is of the convergent type, the fibres align themselves in the direction of flow (parallel) and divergent flow leads to the alignment of fibres away from the direction of flow (transverse). In elongational flow, the fibre orientation take places mainly in the direction of the applied force. In shear flow, fibre orientation can be from random to unidirectional depending on shear rate.

The fibre orientation has a pronounced effect on the mode of composite fracture and thereby influences the mechanical properties[33]. The composite fracture occurs by different modes, for example, fibre breakage, shear failure or matrix failure, if the angle, θ, between the fibre orientation and the direction of application of force is in the range 0–10°, 10–60° and 60–90°, respectively. The maximum strength of a long-fibre-reinforced composite can be achieved if the angle between the principal fibre orientation and the direction of force application is 0°. Theoretically, the composite strength should be almost similar to the fibre strength when the angle is 0° under perfect conditions of fibre/matrix adhesion and fibre orientation. But this is not true in practice especially for short-fibre-reinforced composites, because the fibres cannot be stressed to their maximum because of the ineffective stress transfer near the fibre ends. The strength decreases as the angle θ increases from 0°. When the angle becomes 90°, fracture of the composite takes place mainly through the matrix. Here, the fibres do not contribute much to the strength of the composite. Thus the composite shows minimum strength when the angle is 90°.

The achievement of 100% orientation, i.e. $\theta = 0$, in short-fibre/rubber composites is quite impractical if the standard rubber processing and fabrication techniques are used. This was confirmed by the following observation. In the present study it was seen that the fibre orientation in the unvulcanized rubber composite that had been achieved by mill mixing was not maintained as such in the vulcanized sample. This is because shear flow occurs during compression moulding. So the complete effect of the extent of fibre orientation as calculated by green strength measurements may not be reflected in the properties of vulcanized rubber composites. Therefore, because of the uncontrolled distribution of fibre orientation, a clear demarcation between the modes of failure mentioned in the above paragraph is not easy and an overlap occurs between them.

Directional swelling studies

Information about anisotropy caused by the orientation of fibres and matrix/fibre adhesion can be obtained from equilibrium swelling studies. This would provide an idea about the extent of fibre orientation also. Recently, equilibrium swelling studies have been carried out for natural rubber composites containing sisal fibre by Varghese *et al.*[34]. Estimation of the extent of fibre orientation and the fibre/matrix interfacial adhesion in coir-fibre-reinforced natural rubber composites by anisotropic swelling studies have been reported by Geethamma *et al.*[23]. The swelling ratio, a_θ, in a direction forming an angle θ with the fibre orientation given by Coran *et al.*[35] is as follows

$$a_\theta^2 = \left(a_T^2 - a_L^2\right) \sin^2 \theta + a_L^2 \quad (2)$$

where a_L and a_T are the dimensional swelling ratios in the longitudinal and transverse directions. *Figure 7* presents the dimensional swelling variation of mixes gum, I, J and K with angle θ in accordance with the above equation, where various values of θ were assumed. For all four mixes, the swelling increases with θ and is found to be maximum when θ becomes 90°. This confirms that the preferential fibre orientation is in the longitudinal direction. The line corresponds to gum compound which does not

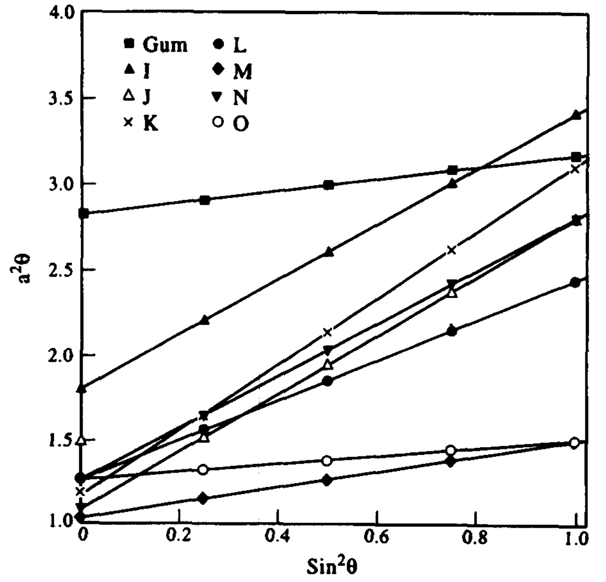

Figure 7 Swelling variation as a function of the angle of measurement, θ

Table 4 Anisotropic swelling values

Mix	a_L	a_T	a_Z
Gum	1.68	1.778	1.4
I	1.34	1.84	1.90
J	1.04	1.60	1.67
K	1.08	1.76	1.84
L	1.12	1.56	1.33
M	1.02	1.22	1.67
N	1.12	1.67	1.60
O	1.12	1.22	2.0

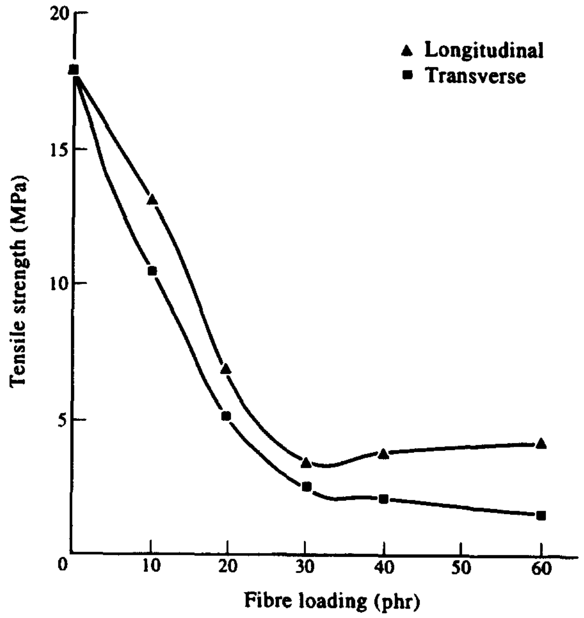

Figure 8 Effect of fibre loading on tensile strength

contain fibres, and is positioned above those for mixes I, J and K. This shows that short fibres restrict the transport of solvent into the composite.

Information about the extent of fibre orientation can also be obtained from these graphs. Noguchi *et al.*[36] showed that the mix corresponding to a line with higher slope possesses a greater degree of fibre alignment. In *Figure 7*, composite K possesses the highest slope value, 1.928, which indicates that the extent of fibre alignment is higher in mix K than in mixes I and J, whose slopes are 1.592 and 1.708, respectively.

Table 4 contains the swelling ratios of these mixes. For all the mixes the swelling ratio, a_L, in the longitudinal direction is lower than a_T and a_Z, which are the swelling ratios in the transverse and thickness directions, respectively. For mixes J and K, the a_L values are close to one which confirms the existence of a strong interface. The a_T values (the transverse swelling ratios) of mixes I, J and K show that, perpendicular to the preferred orientation, the swelling is independent of the nature of fibres and the value is roughly equal to that of the gum rubber. The fibre distribution is higher in the direction perpendicular to the application of pressure during moulding, i.e. swelling is reduced in the transverse direction because of the higher amount of fibres present. Hence in most cases the a_T values are less than a_Z values. Thus it is obvious that in coir-fibre-reinforced rubber composites swelling occurs in the thickness direction.

It is interesting to note that the a_L values of composites L and N are lower than those of M and O, respectively. The latter mixes differ from the former by the presence of silica. This indicates that the interface is stronger in the absence of silica. This again establishes the better performance of composites that do not contain silica. Also, the higher slopes of L and N compared with those of M and O, respectively, show the better fibre orientation in the former mixes (*Figure 7*).

The effect of fibre loading

The effect of fibre loading on tensile strength in short-fibre-reinforced rubber composites has widely been studied. Generally, the tensile strength initially drops up to a certain amount of fibre and then increases. This minimum volume of fibre is known as the critical volume above which the fibre reinforces the matrix. The critical volume varies with the nature of both fibre and matrix, fibre aspect ratio, fibre/matrix interfacial adhesion, etc. Setua and De[37] found that the critical loading is 40 and 7.5 phr in the case of jute and silk fibres, respectively.

In the present study, the behaviour of composites containing 10, 20, 30, 40 and 60 phr alkali-treated coir fibres which were subjected to pretreatment with the natural rubber and TDI solution as given in *Table 2* was analysed. From *Figure 8* it can be seen that the tensile strength in the longitudinal direction decreases sharply with increase in fibre loading up to 30 phr and then shows a slight increase for composites containing 40 and 60 phr fibre loading. In the transverse direction also, tensile strength drops with increase in fibre loading up to 30 phr after which the decrease is small. When the amount of fibres is not enough to restrain the matrix, large stresses will be developed at low strains and the distribution of these stresses will not be uniform. But after 30 phr fibre loading the fibres are sufficient to restrain the matrix, the stress distribution will be uniform and therefore the fibres start reinforcing the matrix. A similar trend was reported by Murty and De[38]. They compared the effect of carbon black on the mechanical properties of short-glass-fibre/natural rubber composites and found that tensile and tear strengths were decreased with fibre loading up to 40 vol%.

Dzyura[39] reported that the minimum amount of fibres needed to restrain the matrix is smaller if the matrix strength

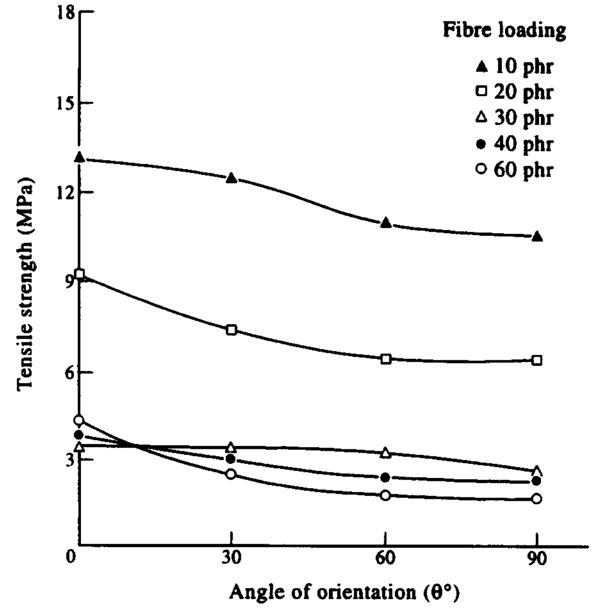

Figure 9 Variation of tensile strength with fibre loading and angle of orientation

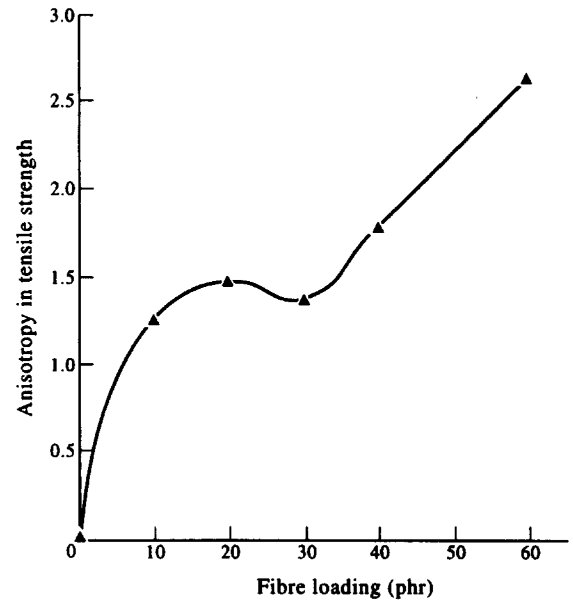

Figure 10 Influence of fibre loading on anisotropy in tensile strength

is higher. In the present study natural rubber has been used as matrix material which is a very strong rubber because of its strain-induced crystallization behaviour. However, in the present study, a critical fibre loading was not obtained even at 30 phr fibre level. A slight increase of tensile strength was observed only after 40 phr fibre loading. This may be due to the poor fibre/rubber adhesion. Hence experiments are underway in this laboratory to innovate a more fruitful pretreatment method for coir fibre.

The variation of composite tensile strength with the angle of orientation at different levels of fibre loading is given in *Figure 9*. Tensile strength decreases with increase in angle θ for all fibre loadings except at 30 phr loading. It has already been seen (*Figure 8*) that with up to 30 phr fibre loading the tensile strength decreased sharply and then remained almost constant up to 40 phr, i.e. the restraining of the matrix by fibres starts at a loading of about 30 phr. Hence the anisotropy of composites containing 30 phr fibre is comparatively low. This can be seen in *Figure 10*. It is obvious that the anisotropy in tensile strength is not noticeable at low fibre loadings. In contrast to the trend observed in extent of fibre orientation because of the anisotropy in green strength values (*Figure 6*), the anisotropy in tensile strength is improved for a composite containing 60 phr coir fibre. This is because shear flow occurs during moulding as explained earlier.

CONCLUSION

Different chemical treatments were tried on coir fibre in order to improve its efficiency as a reinforcement in natural rubber composites. It was found that composites containing alkali-treated coir fibres that had been subjected to a pretreatment with depolymerized liquid natural rubber solution exhibit improved tear strength and fibre orientation even though the tensile properties are marginally lower than those of composites containing coir fibres treated with NR and TDI solutions.

Tensile and tear testing have been performed to evaluate the role of silica in the tricomponent HRH dry bonding system. It was observed that silica is not an essential component in producing good coir/rubber interfacial adhesion.

Directional swelling studies were carried out to analyse the extent of fibre orientation in mixes I, J and K. It was observed that the extent of fibre orientation is the highest in mix K. The smaller a_L values of mixes L and N indicate their lower solvent intake and hence higher interfacial adhesion than those of mixes M and O, respectively.

The extent of fibre orientation in the composite was calculated from green strength measurements. It was observed that the extent of fibre orientation is highest in the natural rubber composite containing 40 phr coir fibre. However, the effect of high fibre orientation was not reflected in the tensile strength of this composite. Similarly, no improvement in the anisotropy in tensile strength was observed for composites containing 60 phr fibre as expected from the anisotropy in green strength measurements.

The variation of tensile strength with fibre loading was studied. The tensile strength decreased sharply up to 30 phr and showed only a slight increase even at a high fibre loading of 60 phr. This behaviour is explained on the basis of the shear flow that occurs during compression moulding and the poor interfacial adhesion.

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