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# EFFECT OF FIBRE LOADING ON INTERFACIAL ADHESION AND ENGINEERING PROPERTIES OF SISAL/SBR COMPOSITES

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**Abstract::** Styrene Butadiene Rubber Composites were prepared by incorporating short sisal fibres of different concentrations having a length of 6mm. into the SBR matrix on a mixing mill according to a base formulation. The curing behaviour of the mixes were studied. The mechanical properties were found to increase along and across the grain direction. But resilience was found to decrease on the addition of fibres. The adhesion between the fibre and rubber was enhanced by the incorporation of a two component dry bonding system consisting of resorcinol and hexomethylene tetramine. The failure behaviour in composites have been analysed by Scanning Electron Microscopy to understand the fibre-matrix interfacial interaction.

## 1. Introduction

Short fibres are used in rubber compounding due to the considerable processing advantages, improvement in certain mechanical properties and for economic considerations (1). The short fibre composites have been studied by several researchers in an attempt to produce reinforced products like V-belts (3), hoses (4), tire treads (5), and complex shaped articles (6). The various natural fibres such as jute, bagasse, asbestos, flax and cotton have been used to reinforce various types of rubber including NR, SBR and BR (7).

De and co-workers have studied the use of jute and silk fibres as reinforcing fillers for natural rubber (NR) and carboxylated nitrile rubbers (2,8). Recently, in our laboratory, sisal and coconut fibres have been used as reinforcing fillers for NR (9,10). Murthy et al. have studied the effect of mechanical properties on the jute fibre reinforced SBR compos-

ites. (11). However, no serious attempt has been made so far to evaluate the use of the sisal fibre as a reinforcing filler for SBR matrix. This paper reports the results of the studies on the effect of fibre concentration and bonding agent on the physical and mechanical properties of SBR composites.

## 2. Experimental

The raw sisal fibre was chopped to a length of 6mm. Sisal fibres contain cellulose 78%, hemicellulose and pectin 10%, lignin 8%, waxed 2% and ashes 1% (12). It has an average diameter of 100-300µ m. and a specific gravity of 1.45 (13). The fibres were washed with water to remove the undesirable materials and dried in an air oven at 70°C for 5 hrs. before mixing.

Styrene Butadiene Rubber (SBR-1502) used for the study was the technically specified form of rubber. All ingre-

Table 1. Formulation of mixes

Ingredients	Mixes						
	A	C	E	F	G	H	L
SBR - 1502	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Sulphur	2.2	2.2	2.2	2.2	2.2	2.2	2.2
Stearic acid	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Zinc oxide	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Resorcinol							5.0
Hexa <sup>a</sup>						2.5	
CBS <sup>b</sup>	1.0	1.0	1.0	1.0	1.0	1.0	1.0
TDQ <sup>c</sup>	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Sisal Fibre (Untreated)		35.0	5.0	10.0	15.0	20.0	35.0

a - Hexamethylene tetramine

b - N - cyclohexyl benzothiazyl sulphonamide

c - 2,2,4 - Trimethyl 1,2 - Dihydro quinoline polymerised

dients including hexa and resorcinol incorporated into the SBR matrix were of laboratory reagent grade. The recipe used in this work is shown in table 1.

Mixes were prepared by means of a two-roll laboratory mixing mill (150mm x 300mm). The curing characteristics were studied by a Monsanto Rheometer R-100. The samples were vulcanized at 150°C and measurements were carried out in a Universal Testing machine (ZWICK - 1474) at a crosshead speed of 50 cm/min.. The Tensile strength and tear strength were measured according to ASTM methods D412 - 51 T and D624 - 54, respectively. Figure 1 shows the longitudinal and transverse fibre orientation on tensile and tear samples having fractured surfaces.

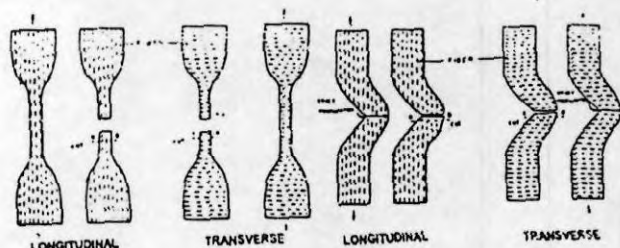


Fig 1. Tensile and Tear samples showing longitudinal and transverse orientation having fractured surfaces.

Extent of fibre breakage was analyzed to examine the fibre length distribution (Figure 2). Resilience was determined by Dunlop tripsometer, while hardness was measured by Shore - A Hardness tester.

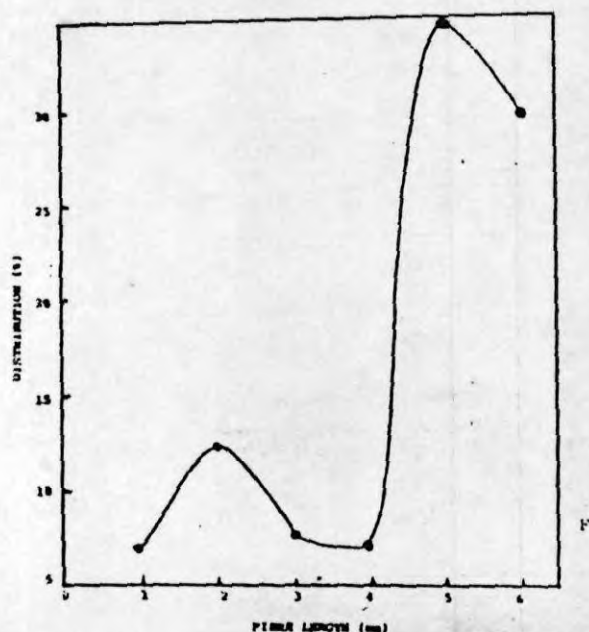


Fig 2. Effect of Fibre length on Distribution of fibres

The fracture surface were examined by using a JEOL scanning electron microscope to understand the fibre fracture and interfacial adhesion.

### 3. Results and Discussion

#### Curing Characteristics

The cure behaviour of various mixes are given in figure 3. The initial decrease in the torque to a minimum value is because of the softening of the rubber matrix. The rheographs show a drastic increase in the maximum torque from its minimum value. This is due to the crosslinking of the rubber matrix. The curves in the rheographs cross over each other because of differences in their cure times. The maximum torque value increases gradually with increase in fibre content due to the increase in the viscosity of the mixes.

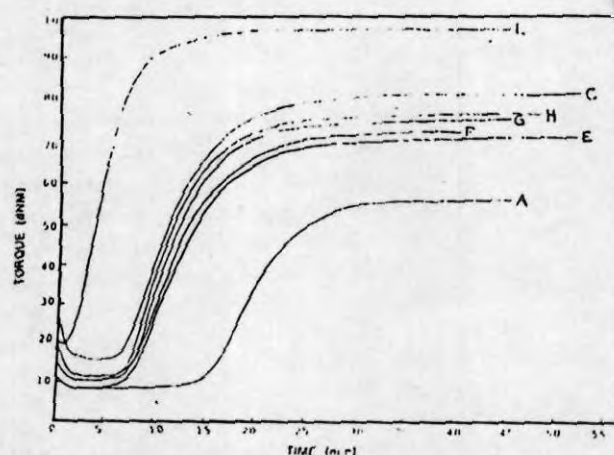


Fig 3. Rheographs of mixes

#### Mechanical properties

The effect of volume per cent of fibre on the tensile moduli of the composites is shown in figure 4. Modulus shows a rapid increase upto 14.5% volume loading and then shows a levelling off in the case of longitudinal orientation. But in the case of transverse orientation modulus at different elongations viz, 10, 20 and 30% increases gradually as the volume per cent of fibre reaches upto 17.7%. After 14.5% a binding is occurred between the surface of the sisal fibre and the SBR matrix. Hence, the tensile moduli is increased with the increase in fibre loading.

Tensile strength in the longitudinal orientation increased upto a loading of 17.7 volume per cent of fibre (Figure 5). Above that loading, the incorporation of fibres into SBR matrix is not possible due to the entanglement of fibres causing fibre breakage. In the transverse direction, the values of tensile strength show an initial drop and then gradually increases. This is due to the weakening of the rubber matrix by

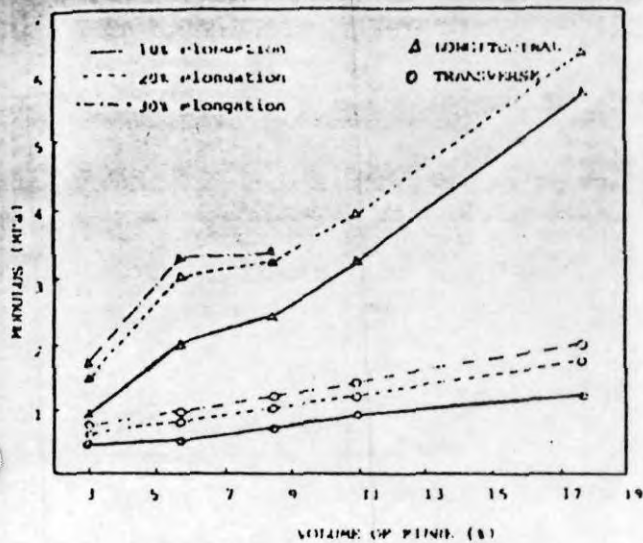


Fig 4. Effect of Volume of fibre and orientation on Modulus at 10%, 20% and 30% elongation.

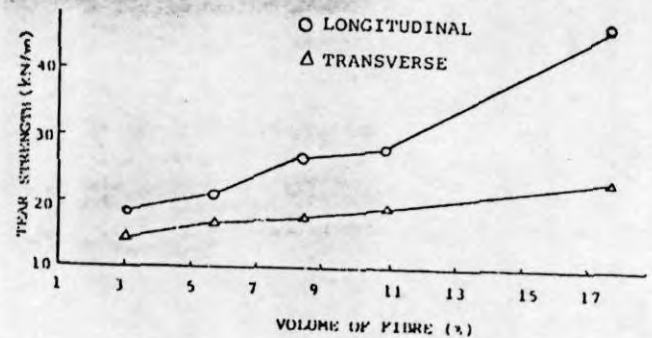


Fig 6. Effect of Volume of fibre and orientation on Tear strength.

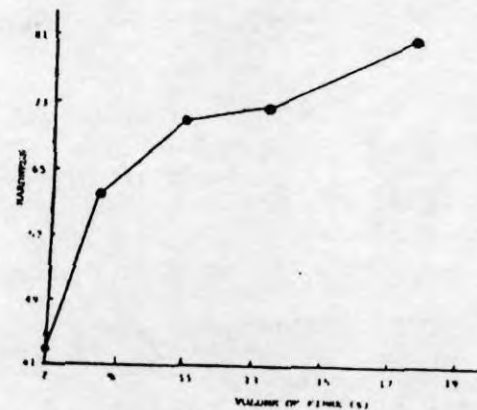


Fig 7. Effect of Volume of fibre on Hardness

the presence of transversely oriented fibers. In the case transverse or longitudinal orientation, at low fibre loading the matrix is not restrained by enough fibres and highly localised strains occur in the matrix at low stresses, causing the bond between the fibre and rubber to break leaving the matrix diluted by non reinforcing debonded fibres. As the fibre concentration increases in mixes from A to L, the stress is more evenly distributed in the composite.

The effect of fibre concentration on the tear strength of the composites in both orientations were investigated (Figure 6). The increase in tear strength on the addition of fibres is due

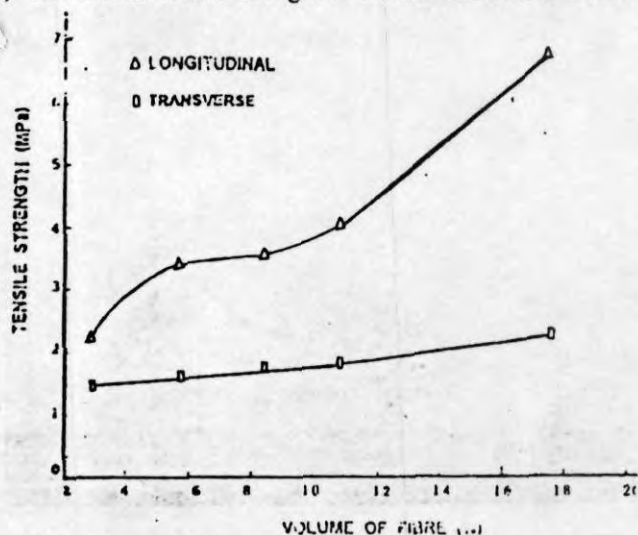


Fig 5. Effect of Volume of fibre and orientation on Tensile strength.

to the obstruction of tear paths caused by the short fibres. At 17.7 volume per cent of the fibre, the tear strength in both orientations registers a maximum value.

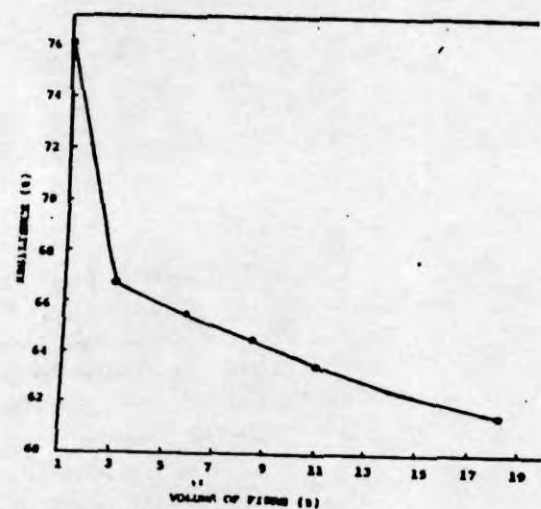


Fig 8. Effect of Volume of fibre on Resilience



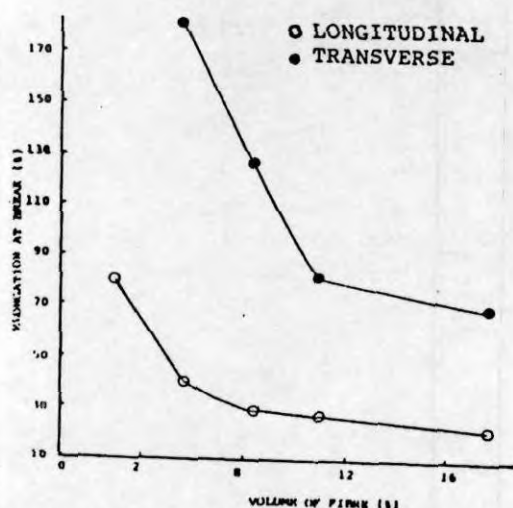


Fig 9. Effect of Volume of fibre and orientation on Elongation at break.

Figure 7 shows an increase in hardness and figure 8 shows a decrease in resilience due to the increase in fibre concentrations in the mixes. However, there was a corresponding steady decrease in the elongation of the fibre loaded samples, with increased loading in both orientations (Figure 9). The elongation at break in the transverse orientation also



Fig 10. SEM Photograph showing deep cracks developed on the sample.

registers the same trend but with consistently higher values. With increasing fibre loading, the hardness values of the composites increased gradually, with an associated decrease in the elongation at break. Due to the increased hardness, the composites become brittle, which results in the development of deep cracks over the fracture surface (Figure 10.).

#### Effect of bonding agent

The increase in maximum torque is more prominent in the presence of bonding agent. The hexa and resorcinol forms



Fig 11. SEM Photograph showing pull out of fibres

a resin which creates strong bonds between sisal fibre and SBR matrix. The optimum cure time is also found to decrease drastically with the addition of fibre and bonding agent (Table 1 and Figure 3).

Fibre filler composites containing 2.9% to 17.7% volume of fibre exhibit a marked change in the fracture topography (14). The fracture of composites was occurred in two modes: (i) breakage of fibre leading to failure and (ii) pull-out of several fibres from the matrix. In the case of longitudinally oriented fibres, the fibres are oriented perpendicular to the fracture direction. Hence, the breakage and pulling out of the fibres takes place, whereas for transversely oriented fibres the crack progresses in the direction of fibre alignment, experiencing, therefore, a lower resistance by the fibres.



Fig 12a. SEM Photograph of mix L showing better adhesion between fibre and matrix on longitudinal orientation

Table 2. Properties of mixes.

Mixes	Orien- tation	Tensile strength (MPa)	Elonga- tion at break (%)	Tear strength (kN/m)
A	L	2.36	373	13.89
	T	2.07	288	9.94
C	L	6.70	23	46.50
	T	2.24	69	23.30
L	L	8.10	15	56.25
	T	2.30	65	36.67

L- Longitudinal; T- Transverse.

The SEM studies revealed the adhesion between the fibre and matrix. SEM photographs show very short segment of fibres and holes left after the fibres are pulled out from the matrix (Figure, 11), while the other figures show better adhesion between the fibres and matrix due to the addition of bonding agent (Figures 12a & 12b).

Thus it is inferred that mix (L) showed higher mechanical properties compared to mix (C) (Table 2).

#### 4. Conclusions

1. The addition of sisal fibres to SBR offers good reinforcement and causes improvement in mechanical and physical properties due to polymer-filler interaction which gets further enhanced by the presence of bonding agent.

2. Mechanical anisotropy is observed at 17.7 volume per cent of fibre.

3. The bonding between sisal fibre and SBR matrix is poor and can be enhanced by the use of hexa-resorcinol bonding system.

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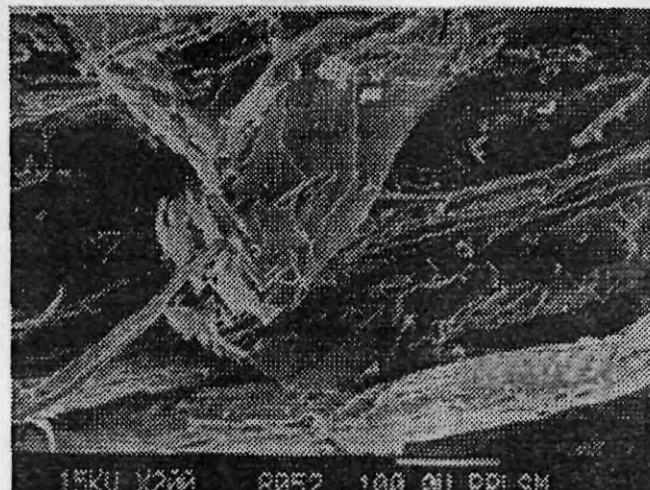


Fig 12b. SEM Photograph of mix L showing better adhesion between fibre and matrix on transverse orientation

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