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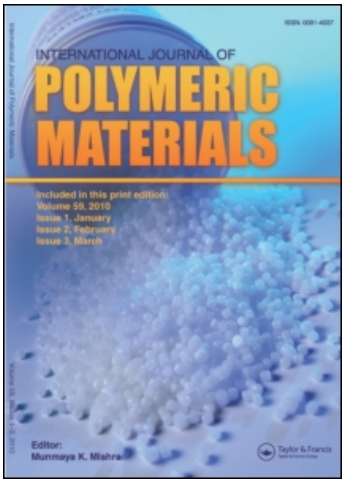

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Curing Characteristics and Mechanical Properties of Oil Palm Wood Flour Reinforced Epoxidized Natural Rubber Composites

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Curing Characteristics and Mechanical Properties of Oil Palm Wood Flour Reinforced Epoxidized Natural Rubber Composites

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The paper reports on the curing characteristics and mechanical properties of oil palm wood flour (OPWF) reinforced epoxidized natural rubber (ENR) composites. Three sizes of OPWF at different filler loadings were compounded with a two roll mill. The cure (t_{90}) and scorch times of all filler size decrease with increasing OPWF loading. Increasing OPWF loading in ENR compound resulted in reduction of tensile strength and elongation at break but increased tensile modulus, tear strength and hardness. The composites filled with smaller OPWF size showed higher tensile strength, tensile modulus and tear strength. Scanning electron microscope (SEM) micrographs showed that at lower filler loading, the fracture of composites occurred mainly due to the breakage of fibre with minimum pull-out of fibres from the matrix. However as the filler loading is increased, the fibre pull-out became very prominent due to the lack of adhesion between fibre and rubber matrix.

Keywords: Oil palm wood flour; curing characteristics; mechanical properties; epoxidized natural rubber; composite

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1. INTRODUCTION

Rubbers constitute a very important class of material because of their unique mechanical properties, such as elastic behaviour at very large deformation, energy absorbing capability, etc. Epoxidized natural rubber (ENR) is obtained from natural rubber by replacing some of the double bonds with epoxide groups [1]. ENR with 50 mole% epoxidation has been shown to be polar with properties similar to those

acrylonitrile butadiene rubber and butyl rubber. Numerous studies have been reported on ENR-filler interaction and reinforcement [1 – 3]. The most widely used fillers in rubber industries are carbon black and silica. The world energy crisis forces rubber producers to search for cheaper new filler materials which are viable to replace the conventional fillers either partially or totally.

In the present study, a relatively new type of wood-based filler is investigated. The filler used is derived from oil palm tree (*Elais guineensis*) component, called, oil palm frond (OPF). This material is the by product of palm oil industry. The effect of filler loading and size on the curing characteristics and mechanical properties of oil palm wood flour (OPWF) filled epoxidized natural rubber (ENR) composites will be reported. Scanning electron microscopy (SEM) studies are carried out to determine the failure mechanism of the composites.

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2. EXPERIMENTAL

2.1 Compounding Ingredients and Formulation

Table I shows the materials, their manufacturers, and loading used in this study. All materials were used as supplied. Oil palm frond (OPF) in fibrous form were ground into three sizes (270–500 μm), (180–270 μm) and (75–180 μm). These sizes were used in this study. Table II shows the chemical compositions of oil palm frond [4].

TABLE I Typical formulations of OPWF filled ENR 50 Compounds

Material	*Manufacturer*	*Formulation (phr)*
ENR 50	Guthrie (M)	100
Sulphur	Bayer (M) Ltd.	2.5
Zinc oxide	,,	5.0
Stearic acid	,,	2.0
CBS[a]	,,	0.5
Flectol H[b]	Monsanto (M) Ltd.	1.0
Fillers*	Sabutek Sdn. Bhd. (M)	0, 15, 30, 40, 50

[a] N-cyclohexyl-2-benzothiazole-2-sulphenamide.
[b] Poly-1,2-dihydro-2,2,4-trimethylquinoline.
* Three different sizes were used.

TABLE II Chemical Compositions of Oil Palm Frond (OPF) [4]

Lignin	18.3%
Halocellulose	80.5%
Pentosan	na
Hot Water Solubility	12.4%
Alcohol-Benzene Solubility	5.0%
1% NaOH Solubility	na
Alpha-Cellulose	na
Ash	2.5%

2.2 Sample Preparation

Mixing was carried out on a laboratory size (160 mm × 320 mm) two roll mixing mill (Model XK-160) in accordance to the method described by ASTM D 3184-80. The respective cure times at 140°C as measured by t_{90} were then determined using a Monsanto Rheometer, model MDR 2000. The scorch times, torque, elastic modulus etc. were also determined from the rheograph.

2.3 Measurement of Mechanical Properties

The various rubber compounds were compression moulded at 140°C according to their respective t_{90}, into sheets. All samples were tested in the mill direction in the measurement of mechanical properties. Dumb-bell and crescent test pieces according to ISO 37 and ISO 34 respectively were then cut out. Tensile and tear tests were carried out on Monsanto Tensometer, T10 according to BS 903: Part A2 and BS 903: Part A3, respectively at 500 mm/min cross-head speed. The test for hardness was carried out by using the Shore type A Durometer according to ASTM 2240. All tests were conducted at room temperature (25°C).

2.4 Scanning Electron Microscopy

Examination of the fracture surface was carried out using a scanning electron microscope (SEM) model Leica Cambridge S-360. The objective was to get some idea on the mode of fracture. The fracture ends of the tensile specimens were mounted on aluminium stubs and sputter coated with a thin layer of gold to avoid electrical charging during examination.

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2.5 Rubber-fibre Interactions

Lorenz and Parks equation [5] has been applied to study rubber-fibre interaction. According to this equation:

$$\frac{Q_f}{Q_g} = ae^{-z} + b \tag{1}$$

where Q is defined as grams of solvent per gram of hydrocarbon and is calculated by

$$Q = \frac{\text{Swollen weight} - \text{Dried weight}}{\text{Original weight} \times 100/\text{Formula weight}} \tag{2}$$

The subscrips f and g in equation (1) refer to filled and gum vulcanizates, respectively. z is the ratio by weight of filler to rubber hydrocarbon in the vulcanizate, while a and b are constants. The higher the Q_f/Q_g values, the lower will be the extent of interaction between the fibre and the matrix.

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3. RESULTS AND DISCUSSION

3.1 Curing Characteristics

The values of t_{90}, scorch time and maximum torque of OPWF-ENR composites with different particle sizes are shown in Tables III to V. The t_{90} and scorch time of all particle sizes decrease with increasing OPWF loading, i.e. it show enhancement in cure rate. However at any filler loading, OPWF with larger particles size show shorter t_{90} and scorch time. The cure enhancement can be associated to the filler related parameters such as surface area, surface reactivity, particle size, moisture loading and metal oxide loading. In general, a faster cure rate is obtained with fillers having a low surface area, high moisture loading and high metal oxide loading [6]. Butler and Freakley [7] found that cure rate is directly related to the humidity and water loading of the rubber compound. However, in the present study, the cure enhancement of larger particle size of OPWF is probably due to its lower surface area.

TABLE III Cure time (t_{90}), scorch time and maximum torque of OPWF reinforced ENR vulcanizates with particle sizes of 270–500 μm

%Filler (phr)	*0*	*15*	*30*	*40*	*50*
t_{90} (min)	13.59	10.54	10.37	10.21	10.21
Scorch time (min)	3.54	2.00	1.46	1.46	1.42
Maximum torque (dNm)	8.43	10.47	12.10	13.60	14.28

TABLE IV Cure time (t_{90}), scorch time and maximum torque of OPWF reinforced ENR vulcanizates with particle sizes of 180–270 μm

%Filler (phr)	*0*	*15*	*30*	*40*	*50*
t_{90} (min)	13.59	11.26	10.47	10.42	10.23
Scorch time (min)	3.54	2.39	2.28	2.23	2.17
Maximum torque (dNm)	8.43	11.15	12.88	13.61	15.27

TABLE V Cure time (t_{90}), scorch time and maximum torque of OPWF reinforced ENR vulcanizates with particle sizes of 75–180 μm

%Filler (phr)	*0*	*15*	*30*	*40*	*50*
t_{90} (min)	13.59	12.20	12.11	11.59	11.56
Scorch time (min)	3.54	2.45	2.34	2.28	2.23
Maximum torque (dNm)	8.43	11.55	13.04	14.72	16.12

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Tables III to V also show that the addition of OPWF increases the torque value of the NER compound. The marked increment in the maximum torque with increasing filler loadings indicates that the presence of fillers in the rubber matrix has reduced the mobility of the macromolecular chains of the rubber. These impart more restriction to the deformation and consequently increase the OPWF-ENR composites stiffness [8]. The maximum torque of all the vulcanizates shows tht OPWF with smallest particle size has the highest torque. It seems from the results that the trend is affected by the particle size of the fillers. According to Ishak and Bakar [9] the smaller the particle size, hence the larger surface area, the greater the interaction between the filler and rubber matrix. Thus a higher restriction to molecular motion of the macromolecules is expected. In other words, the addition of fillers of a smaller size tends to impose extra resistance to flow.

3.2 Mechanical Properties

The effect of filler loading and particle size on tensile strength of OPWF-ENR composites is shown in Figure 1. It can be seen that the tensile strength of the composites decreases steadily with filler loading. This rather poor strength properties may be attributed to the geometry of the OPWF fillers. For irregularly shaped fillers, the strength of the composites decreases due to the inability of the filler to support stress transferred from the polymer matrix. The presence of such fillers is evident from our SEM study as shown in Figure 2. It can be seen that OPWF actual shape is between the extremes of spheres and long fibres, i.e. they exist as a mixture of irregular particles and short fibres. The results obtained are in agreement with the commonly observed data in other particulate filler systems. The smaller particle size filler exhibits higher tensile strength especially at loading below 30 phr. The better filler dispersion and filler-matrix interaction may be the two main fac-

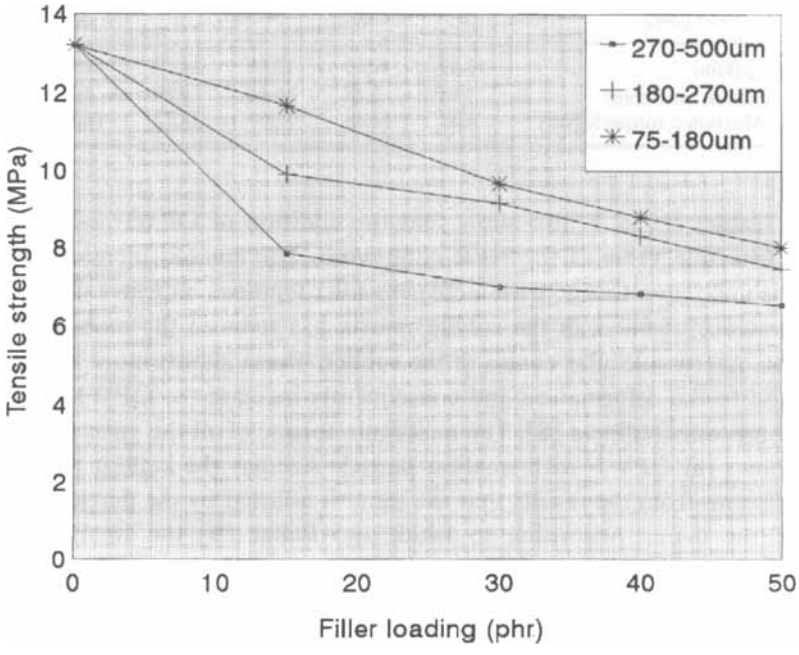

FIGURE 1 Relationship between tensile strength, filler loading and particle size of OPWF reinforced ENR composites.

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FIGURE 2 SEM micrograph of OPWF reinforced ENR composites after tensile fracture at 50 phr of filler (particle sizes: 180 – 270 µm) and magnification 50 *x*.

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tors responsible for the observed trend. Leidner *et al.* [10] and Maiti *et al.* [11] also reported the similar observations.

Modulus is one of the basic properties of the composites studied. Figure 3shows the modulus at 300% elongation (M300) of the OPWF-ENR composites increases steadily with increasing filler loading. This observation highlights the fact that the incorporation of fillers into the rubber matrix can improve the stiffness of the composites. As expected, the smaller-sized fillers gave higher M300 values that larger filler size [12 – 13]. Murty *et al.* [14] found that modulus value increases when the fibre concentration is increased for natural rubber-jute, SBR-jute, SBR-glass and natural rubber-glass composites.

With increase in OPWF loading, the stiffness and brittleness of the composite increased gradually with an associated decrease in the elongation at break (see Fig. 4). It is interesting to note tht the EB of the smaller filler size composites is higher than the bigger one up to a filler loading of 30 phr. At 50 phr, the EB of all composites converge to more or less similar values, irrespective of filler sizes. Below 30 phr, the better filler dispersion of the smaller OPWF fillers in the ENR 50 matrix reduces the tendency of filler–filler interactions from taking

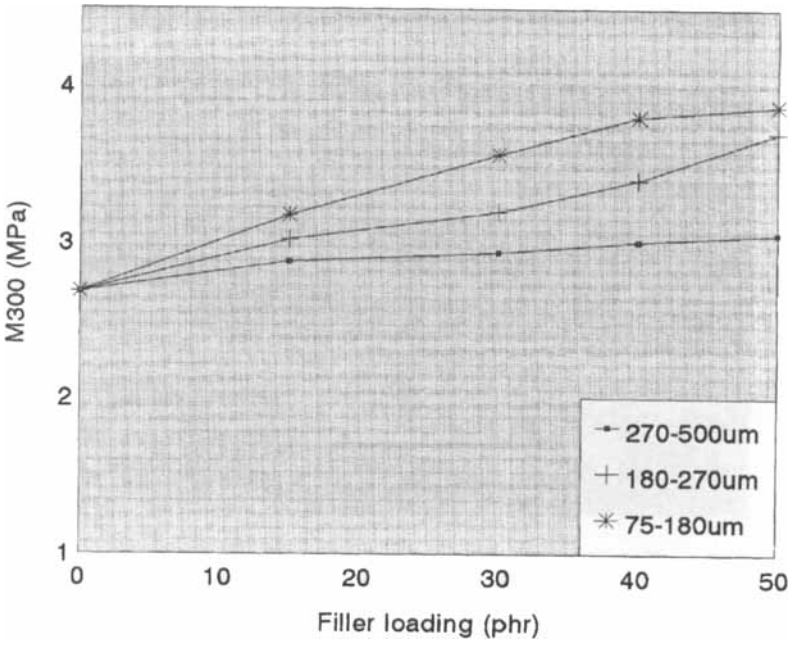

FIGURE 3 Relationship between M300, filler loading and particle size of OPWF reinforced ENR composites.

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place. Thus the samples can be elongated to a much higher value. However, at the highest filler loading i.e. 50 phr, the degree of filler–filler interactions became more prominent even in the case of the smallest filler size composites. Consequently, dramatic reduction in EB was observed. The increment in filler loading will eventually resulted in the reduction of the deformability of a rigid interface between the filler and the rubber matrix [15].

The effect of filler loading and particle size on tear strength of the composites is shown in Figure 5. It can be seen that the tear strength increases with increasing filler loading. The smallest filler i.e. particle-size of 75–180 μm, has a slightly higher tear strength than the bigger filler. The increment in tear strength may be due to the obstruction caused by the short fibre to the tear paths, thus making crack propagation more difficult [16]. Akhtar *et al.* [17] also found that the tear strength increased with increasing fibre loading in their work with silk fibre reinforced polyethylene-rubber blends.

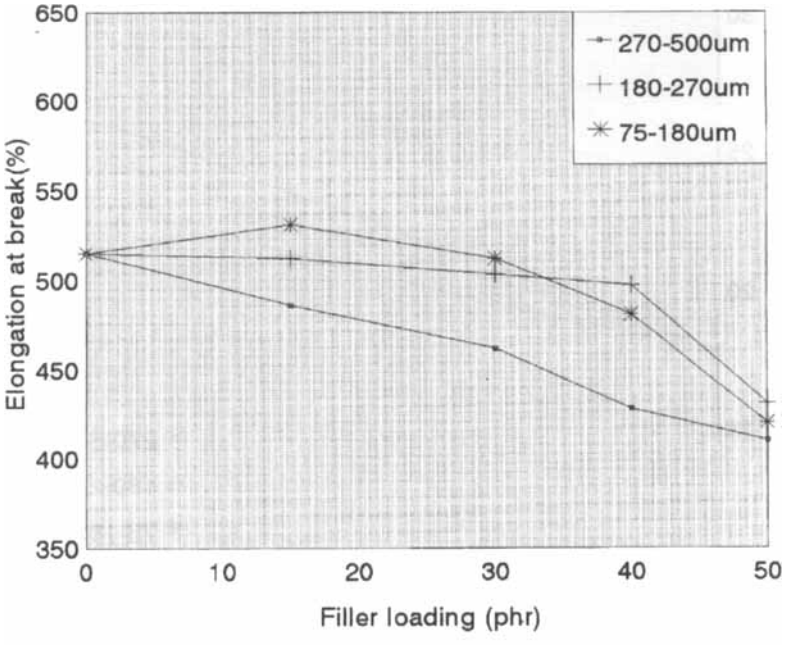

FIGURE 4 The effect of filler loading and particle size on elongation at break of OPWF reinforced ENR composites.

Figure 6 shows that the incorporation of OPWF into the ENR matrix increases the hardness of the composite. As the filler loading increases the composites became stiffer and harder. Thus an increase in the hardness with increasing filler loading was obtained. Chakraborty *et al.* [18] have obtained similar results. As expected, at any fibre loading, there was no significant effect of filler size on hardness.

3.3 Scanning Electron Microscopy (SEM)

Figure 7 shows the SEM micrographs for OPWF-ENR composites at three different loadings and 180–270 μm particle size. The comparison is made at a magnification of 150 *x*. It can be seen that there are very low pull-out of fibres on the fracture surface of lower filler loading (Fig. 7a). This indirectly implies that the presence of more rubber matrix at low filler loading has resulted in a better adhesion between the fillers and the rubber matrix. It is known that for short fibre reinforced composites

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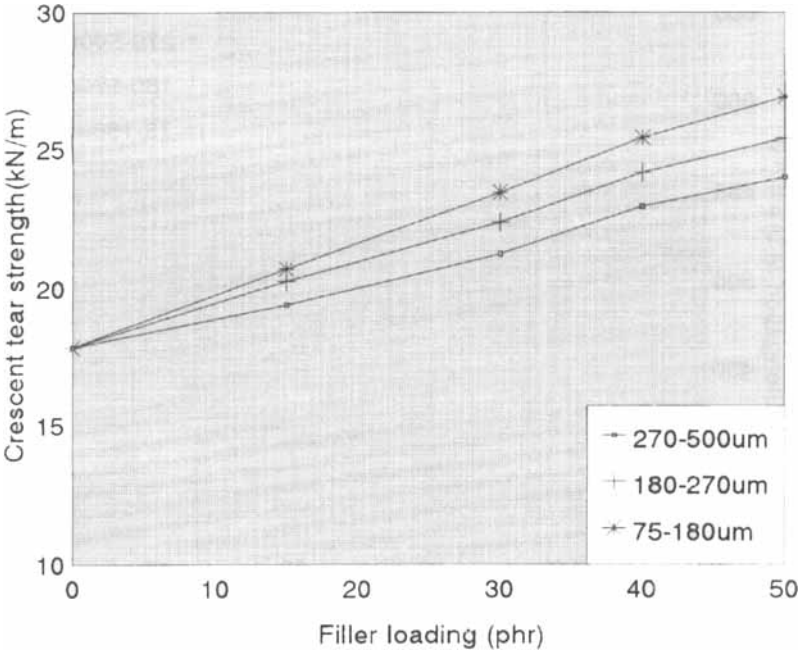

FIGURE 5 The effect of filler loading and particle size on Crescent ear strength of OPWF reinforced ENR composites.

the failure mechanisms is dominated by fibre de-bonding and pull-out processes [8]. This phenomenon is in fact evident in the present OPWF-ENR composites, especially at high filler loading whereby the filler particles were no longer adequately separated or wetted by rubber phase. Hence the fibre pull-out is very prominent due to the lack of adhesion between fibre and rubber matrix (see Figs. 7b and 7c). This mode of failure explains why the tensile strength of the composites decreases steadily with filler loading. The SEM micrograph shown in Figure 2 at a magnification of 50 x (50 phr) provides a further direct evidence for the presence of irregular shaped OPWF fillers which give rise to the poor strength properties of the composites.

3.4 Rubber-fibre Interactions

The relationship between Q_f/Q_g and filler loading in the OPWF-ENR composites is shown in Table VI. It can be seen that irrespective particle

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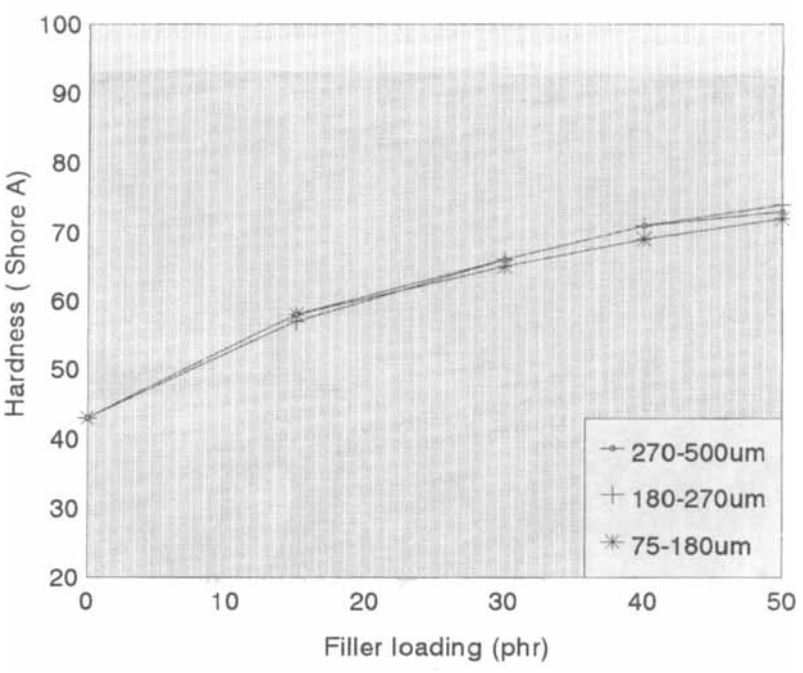
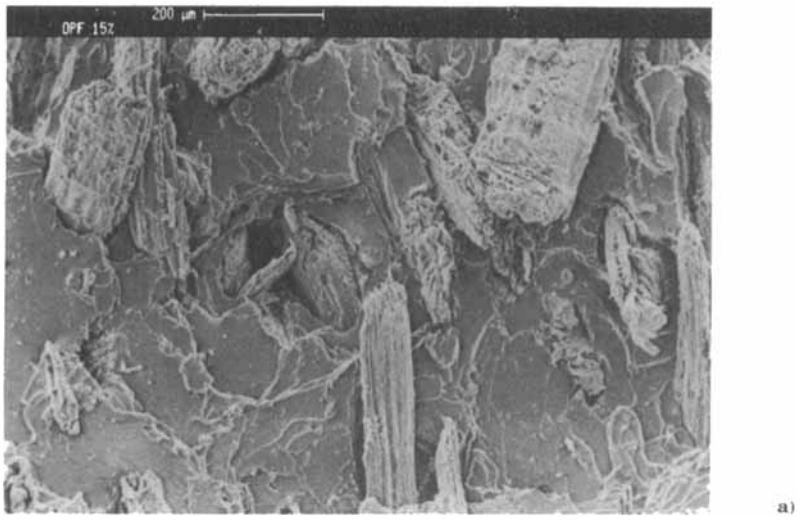


FIGURE 6 Relationship between hardness, filler loading and particle size of OPWF reinforced ENR composites.



a)

FIGURE 7 EM micrograph of OPWF reinforced ENR composites after tensile fracture at various filler loading (particle size: 180–270 µm). (a) 15 phr (mag. 150 *x*) (b) phr (mag. 150 *x*) (c) phr (mag. 150 *x*).

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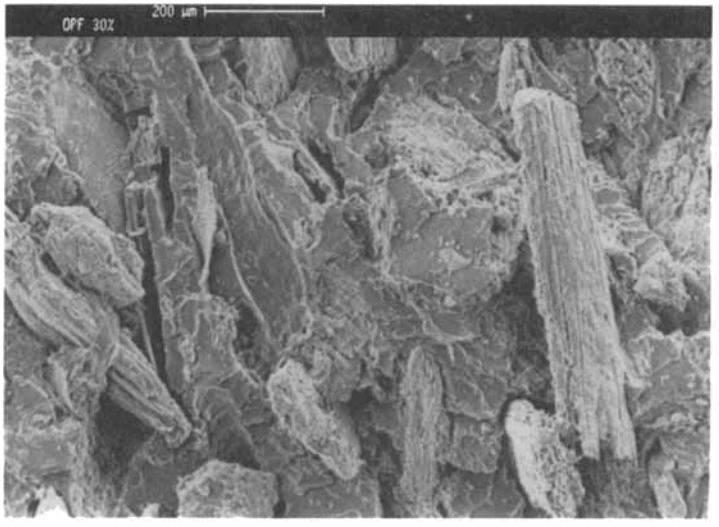
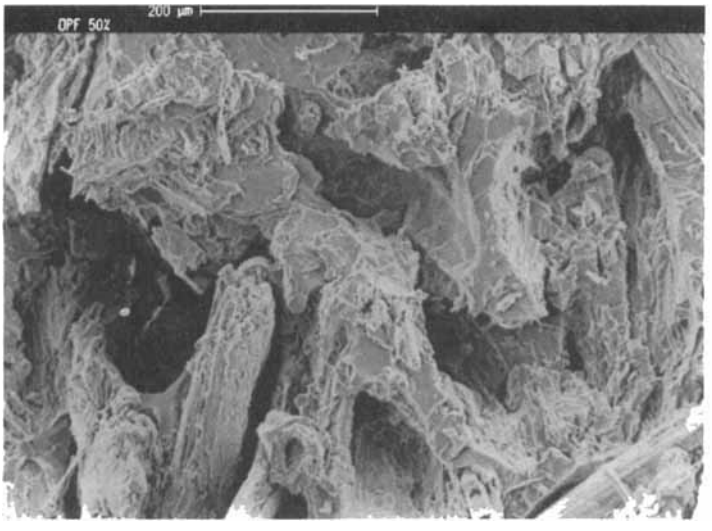


b)

FIGURE 7 (Continued).



c)

FIGURE 7 (Continued).

size of fillers, Q_f/Q_g values show increasing trend with filler loading. The higher Q_f/Q_g value confirm that the increasing filler loading has weaken the rubber-fibre interactions since the higher the Q_f/Q_g values,

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TABLE VI Relationship between Q_f/Q_g values, filler loadings and particle size of OPWF reinforced ENR composites

Particle size	*270–500 μm*	*180–270 μm*	*75–180 μm*
Loading (phr)		Q_f/Q_g	
0	1.000	1.000	1.000
15	1.072	1.036	1.013
30	1.111	1.051	1.035
40	1.123	1.072	1.062
50	1.156	1.085	1.071

the lower will be the extent of interaction between the fibre and the matrix. This result supports the proposed mode of failure as discussed in SEM study. However at any fibre loading, smallest particle size of filler shows the lowest Q_f/Q_g values which mean better rubber-fibre interaction. This result thus compliments our earlier observation that OPWF-ENR composites with the smallest particle size show the best overall mechanical properties, i.e. tensile strength, tensile modulus and tear strength than larger particle size.

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CONCLUSIONS

The incorporation of OPWF into the ENR compounds greatly influenced the processing characteristics of the composites. The cure (t_{90}) and socrch times of OPWF-ENR composites decreased with increasing filler loading. At any filler loading, OPWF with larger particles size show shorter cure and scorch times. The maximum torque increases with increasing fibre loading and OPWF with smallest particles size shows the highest torque values. The incorporation of OPWF also increases tear strength, modulus and hardness but decrease tensile strength and elongation at break. Again smaller particle size of OPWF gives better mechanical properties. Swelling test result shows that the increasing OPWF loading has weaken the matrix-fibre interphase and supports the proposed mode of failure as indicated by SEM study.

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