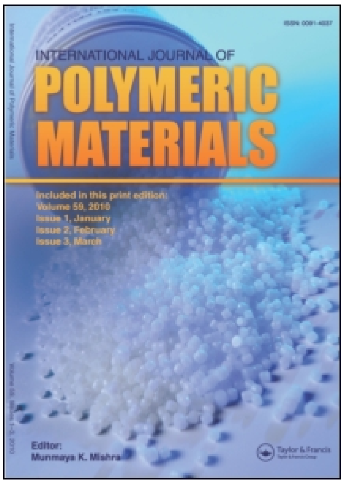

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

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To cite this Article Ismail, Hanafi , Jaffri, R. M. and Rozman, H. D.(2001) 'Oil Palm Wood Flour Filled Natural Rubber Composites: The Effects of Various Bonding Agents', International Journal of Polymeric Materials, 49: 3, 311 — 322
To link to this Article: DOI: 10.1080/00914030108039782
URL: http://dx.doi.org/10.1080/00914030108039782

Intern. J. Polymeric Mater., 2001, Vol. 49, pp. 311–322
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Oil Palm Wood Flour Filled Natural Rubber Composites: The Effects of Various Bonding Agents

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(Received 6 September 1999)

The effects of various bonding agents on curing characteristics and mechanical properties of oil palm wood flour (OPWF) filled natural rubber composites were examined. Compared to control compound the presence of various bonding agents increase the curing time, t_{90}, maximum torque (except phenol formaldehyde(PF) and resorsinol formaldehyde(RF)/Silica(Sil)), tensile strength, tensile modulus (except PF and RF/Sil), and hardness (except PF) but decrease the elongation at break and fatigue life of the composites. Swelling test results indicate that the presence of various bonding agents lead to stronger adhesion at the OPWF-rubber interface. Overall results indicate that RF/Sil/Hexa (Hexamethylene tetramine) is the most suitable bonding system for OPWF filled natural rubber composites.

Keywords: Oil palm wood flour; Natural rubber composites; Bonding agents; Curing characteristics; Bonding agents

1. INTRODUCTION

Short fibres are used in rubber compounding due to the considerable processing advantages, improvement in certain mechanical properties, and for economic consideration [1]. The main advantage of these fibres are their low cost, low density and resistance to breakage during processing [2]. In addition, these fibres offer an excellent opportunity to utilize an abundant source of such materials available from nature [3].

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Compounding of cellulosic fillers in rubbers increase the stiffness of the composites but tends to reduce strength. The poor strength properties results from a lack of adhesion between the hydrophobic rubber and hydrophilic filler. The incompatibility between the filler and rubber matrix can be overcome by modifying the filler-rubber interface [4–6].

In our previous work [7], we have reported the curing characteristics and mechanical properties of oil palm wood flour (OPWF) reinforced epoxidized natural rubber composites. The scorch time, t_2 and cure time, t_{90} decreased with increasing OPWF loading. The incorporation of OPWF also increases modulus and hardness but decrease tensile strength and elongation at break. In this paper, we report the effects of various bonding agents on the curing characteristics and mechanical properties of OPWF reinforced natural rubber composites. Lorenz and Parks equation [8] was used to study the rubber-filler interaction.

2. EXPERIMENTAL

2.1. Materials and Chemicals

The formulations of the composites are given in Table I. Composites were prepared using conventional vulcanization (CV) system. Natural rubber (SMR L) was obtained from Rubber Research Institute of Malaysia (RRIM). Oil palm empty fruit bunch (EFB) in fibrous form

TABLE I Formulation for oil palm wood flour reinforced natural rubber composites filled with different types of bonding system

Compound composition (phr)	*A**	*B*	*C*	*D*
SMR L	100	100	100	100
Zinc Oxide	0.7	0.7	0.7	0.7
Stearic acid	5	5	5	5
Flectol H	1	1	1	1
CBS	0.6	0.6	0.6	0.6
Sulphur	2.5	2.5	2.5	2.5
OPWF	15	15	15	15
Phenol formaldehyde (PF)	–	10	–	–
Resorsinol formaldehyde (RF)	–	–	5	5
Silica (Sil)	–	–	2	2
Hexamethylene tetramine (Hexa)	–	–	–	5

* Control composite (without bonding agents).

was obtained from Sabutek (M) Ltd., Teluk Intan, Perak, Malaysia. EFB fibres were ground into Oil Palm Wood Flour (OPWF) with the sizes of 180–270 μm. Other chemicals such as sulphur, zinc oxide, stearic acid, *n*-cyclohexyl benthiazyl sulphenamide (CBS), and silica were all purchased from Bayer (M) Ltd. Poly-1-2-dihydro-2,2,4-trimethylquinoline (Flectol H) was obtained from Monsanto Company (M). Bonding agents used in this study were phenol formaldehyde (Borden Chemical (M) Ltd.), hexamethylenetetramine (Fluka Chemical (M) Ltd.) and resorsinol formaldehyde (Lianco (M) Ltd.).

2.2. Preparation of Rubber Composites and Determination of Cure Characteristics

Mixing was done on a conventional laboratory two roll mill size (160 × 320 mm) according to ASTM designation D3184-80. The total mixing time has been kept to a minimum to avoid sticking of the rubber compound to the mill rolls. The cure times at 150°C as indicated by the respective, t_{90}, were then determined using a Monsanto Rheometer, Model MDR 2000 (Moving die rheometer). Vulcanizates were conditioned for 24 hrs before testing. All properties were measured along the grain direction.

2.3. Measurement of Mechanical Properties

The various rubber compounds were compression moulded at 150°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 test pieces according to ISO 37 were then cut out. Tensile test was carried out on a Monsanto Tensometer, T10 according to BS 903: Part A2 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. Measurement of Fatigue Life

The various rubber compounds were compression moulded at 150°C according to their respective t_{90}, into rectangular sheets (22.9 × 7.6 × 0.15 cm) with beaded edges. Individual dumbbell samples were cut at right angles to the grain using a BS type E dumbbell cutter. Fatigue tests of the vulcanizates were then carried out on a Monsanto Fatigue To Failure Tester (FTFT). The samples were subjected to repeated cyclic strain at 100 rpm. The extension ratio used ranged from 1.6 to 2.4. Six specimens were used for each test. The numbers of cycles were recorded automatically. The fatigue life in kilocycles (kc) for each sample was computed as the J.I.S. average, which was obtained from the four highest values recorded using the formula:

$$\text{J.I.S. average} = 0.5A + 0.3B + 0.1(C + D)$$

where A is the highest value followed by B, C and D.

2.5. Rubber-fibre Interactions

Cured test pieces of dimension 30 × 5 × 2 mm were swollen in toluene (solvent) until equilibrium swelling, which normally took 72 h at 25°C was achieved. Lorenz and Parks equation [8] has been applied to study rubber-fibre interaction. According to this equation:

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

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 Eq. (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.

3. RESULTS AND DISCUSSION

Figure 1 shows that there is a significant difference in curing time between composites with bonding agents and composite without bonding agent (control composite). The presence of bonding agents

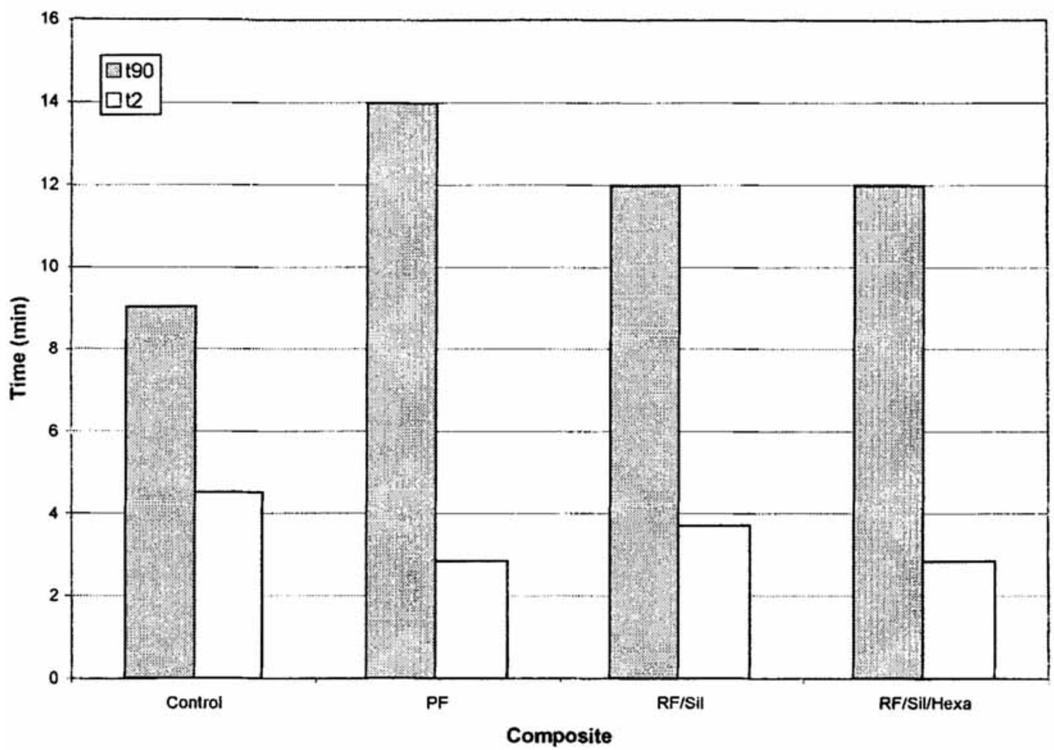

FIGURE 1 Comparison effect of various bonding agents and control composite on curing time (t_{90}) and scorch time (t_2).

in composites have prolonged the curing time. Chakraborty *et al.* [9] in their study on short jute fibre reinforced carboxylated nitrile rubber found that the slowing curing for composites with bonding agents was due to better bonding between fibre and rubbers. In our study, the similar observation can be seen in Figure 2 where the cure rate ($100/(t_{90} - t_2)$) of composites with bonding agents are lower than control composite. It can be seen also in Figure 1 that the addition of various bonding agents resulted in a small reduction of scorch time, t_2. Figure 3 shows the combination of RF/Sil/Hexa bonding system give the highest torque compared to control composite and composites with two other bonding systems. This indicates that RF/Sil/Hexa bonding system is the most suitable bonding system used in this study. The used of this bonding system produced strong bonding at the OPWF/rubber matrix interface and, consequently, the composites became stronger, harder and stiffer. Kumar *et al.* [10] reported that the adhesion between short sisal fibre and the styrene-butadiene rubber was enhanced by the addition of a dry bonding system consisting of resorcinol and hexamethylene tetramine.

The evidence of enhancement adhesion between OPWF and rubber matrix is shown in Figure 4. Figure 4 shows the effect of bonding

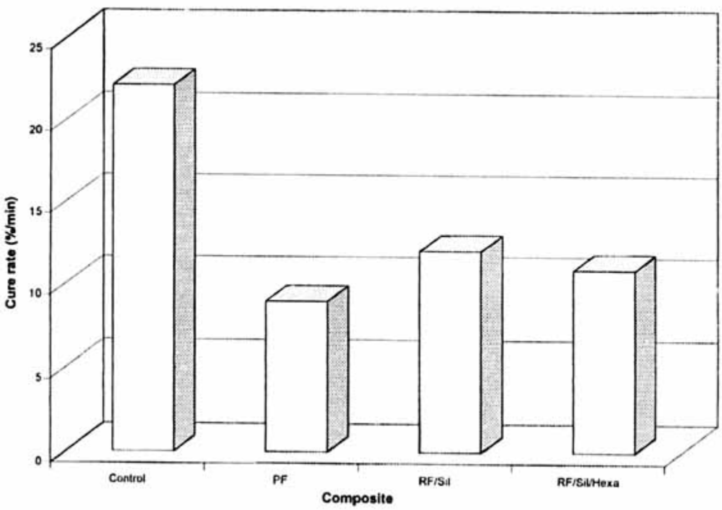

FIGURE 2 Comparison effect of various bonding agents and control composite on cure rate.

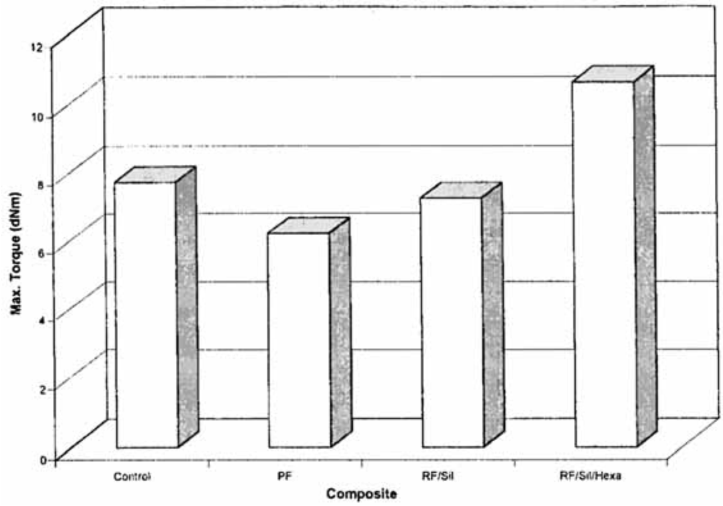

FIGURE 3 Effect of various bonding agents on maximum torque of OPWF filled natural rubber composites.

agents in reducing the Q_f/Q_g value of the composites. The lower value of Q_f/Q_g indicates the better rubber-OPWF interaction [8]. This means that the bonding agents increased the OPWF-rubber interaction. Various researchers [11–14] reported that the presence of tricomponent dry bonding system (*e.g.*, resorcinol-hexamethylene-tetramine-silica) is essential for the promotion of adhesion between the fibre and rubber matrix.

Figure 5 shows the composites with various bonding agents have higher tensile strength than the control composite. The presence of bonding agents lead to stronger adhesion at the OPWF/rubber matrix interface. The stress transfer become more efficient and consequently, enhances the tensile strength. Miwa *et al.* [15] reported that the strong adhesion between filler and rubber matrix resulted in higher shear strength at the interface and consequently enhanced the tensile strength.

Results for tensile modulus *i.e.*, modulus at 100% elongation (M100) and modulus at 300% elongation (M300) in Figure 6 again show that composite with RF/Sil/Hexa bonding system exhibit the highest value. This indicates that RF/Sil/Hexa is the most effective bonding system for OPWF filled natural rubber composites.

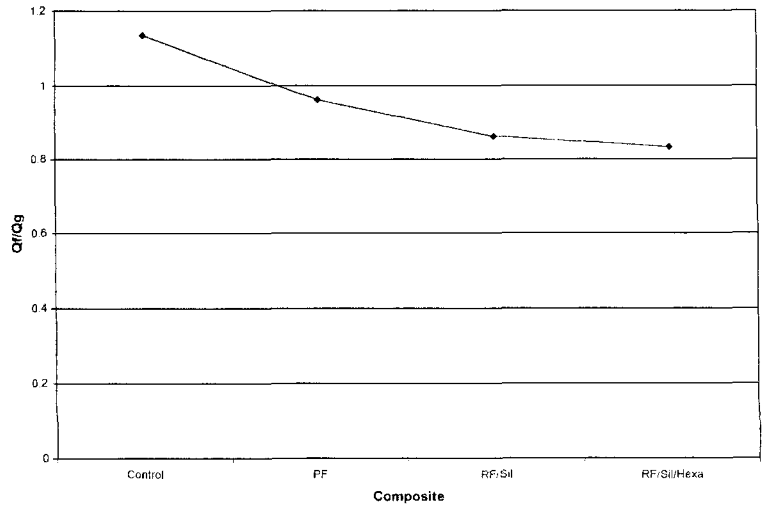

FIGURE 4 Effect of various bonding agents on Q_f/Q_g of OPWF filled natural rubber composites.

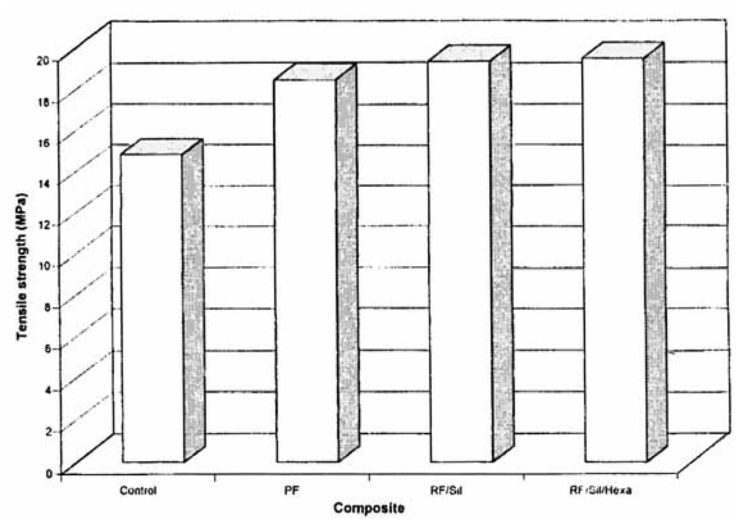

FIGURE 5 Comparison effect of various bonding agents and control composite on tensile strength.

Figure 7 shows the composites with bonding agents have lower E_b than control composites. As previously discussed, the bonding agents improved the OPWF-rubber interaction. This better OPWF-rubber interaction was due to the specific bond formed between the OPWF

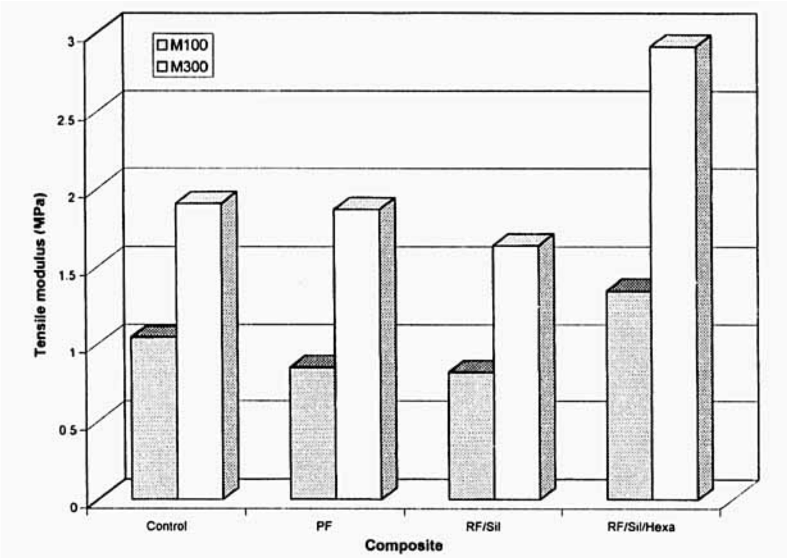

FIGURE 6 Comparison effect of various bonding agents and control composite on tensile modulus.

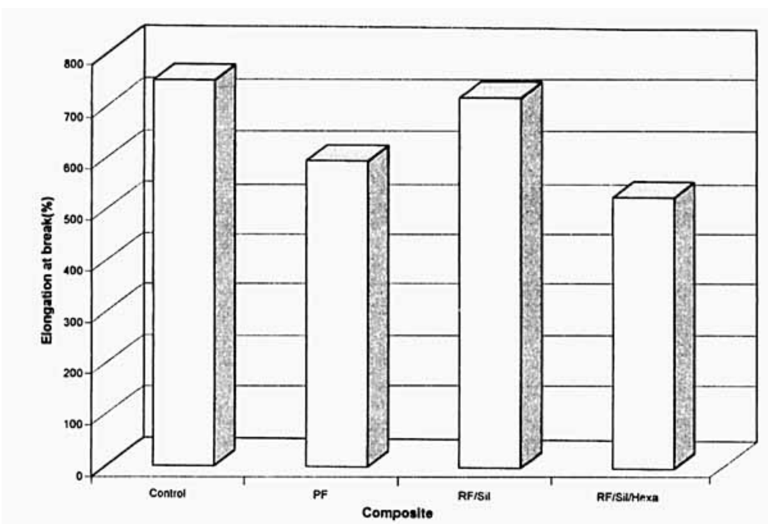

FIGURE 7 Effect of various bonding agents on E_b of OPWF filled natural rubber composites.

and rubber matrix which resulted in higher crosslink density of the vulcanizates. The increment in crosslink density can be seen from the plot of maximum torque – minimum torque (Fig. 8), fatigue life (Fig. 9)

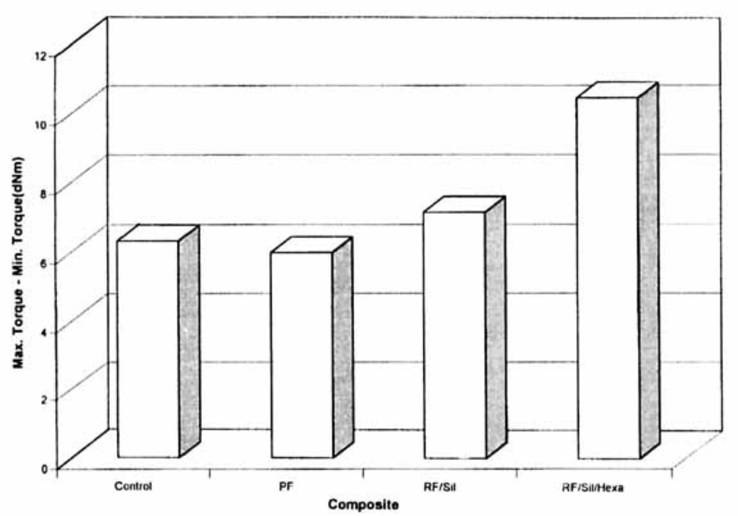

FIGURE 8 Effect of various bonding agents on maximum torque – minimum torque of OPWF filled natural rubber composites.

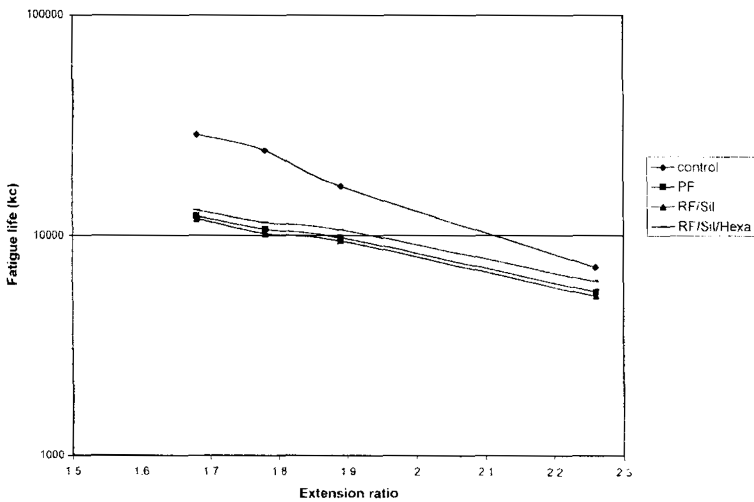

FIGURE 9 Comparison effect of various bonding agents and control composite on fatigue life.

and hardness (Fig. 10). The maximum torque–minimum torque (except PF and RF/Sil systems) and hardness (except PF system) of the composites with bonding agents are higher than control

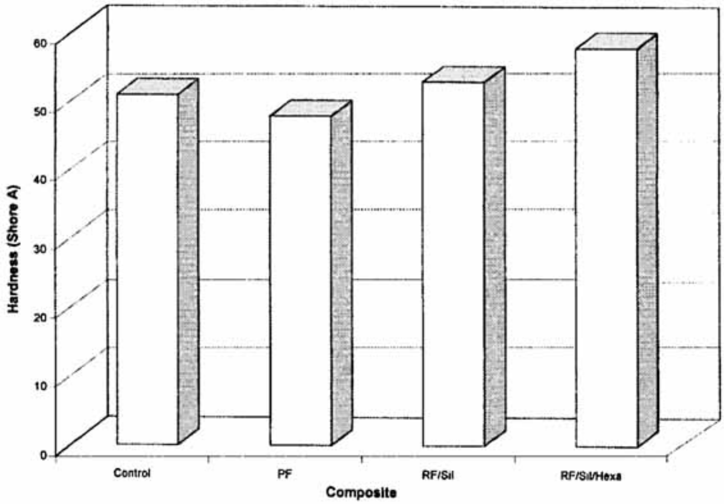

FIGURE 10 Comparison effect of various bonding agents and control composite on hardness.

composites. However for fatigue life, the reverse result is obtained where the control composite exhibit higher fatigue life than the composites with various bonding agents. The labile nature of the crosslink in the control composite (lower crosslink density) giving rise to a more 'relaxed' network and thereby allowing the chains to extend further without breakage at earlier stages compared to composites with different bonding systems.

4. CONCLUSION

1. The incorporation of bonding agents affects the curing characteristics of the composites. The curing time, t_{90}, maximum torque (except PF and RF/Sil systems), maximum torque – minimum torque (except PF system) increase with the addition of bonding agents. However the scorch time and cure rate show decreasing trend.
2. The incorporation of bonding agents also enhances the mechanical properties *viz.* tensile strength, tensile modulus (except PF and RF/Sil systems), and hardness (except PF system) but reduce the elongation at break and fatigue life of the composites.

3. The filler-rubber interaction is improved with the addition of various bonding agents.
4. Overall results indicate that RF/Sil/Hexa is the most effective bonding system for the OPWF filled natural rubber composites.

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