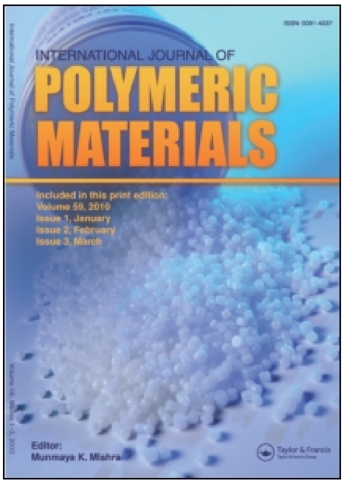

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PHYSICO-MECHANICAL PROPERTIES OF EPDM/NYLON-6 SHORT FIBER COMPOSITES

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PHYSICO-MECHANICAL PROPERTIES OF EPDM/NYLON–6 SHORT FIBER COMPOSITES

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The effect of the adhesion system components HRH (hydrated silica, resorcinol and hexamethylenetetramine) on the physico-mechanical properties of EPDM mixes was studied. The effect of different volume fractions of nylon–6 short fibers on these properties of EPDM vulcanizates was also studied before and after ageing. The results showed that the shorter the length of the fibers, the greater would be their efficiency in EPDM vulcanizates. The experimental results revealed that the best concentration of reinforcing fibers that leads to high stress at low strain was 25 phr. The presence of reinforcing fibers and the adhesion system greatly improve the ageing resistance of the rubber composites. This may be attributed to further formation of crosslinked resin, which balances the degradation that occurs during ageing.

Keywords: EPDM vulcanizates, physico-mechanical properties, nylon-6, short fibers, composites

INTRODUCTION

Rubber products generally undergo dynamic stress during service. Their resistance to dynamic load application is important. Most rubber products are composites, reinforced with textiles [1]. The short fibers to reinforce rubber articles, such as hoses and belts, have attracted the attention of several researchers because of their advantages in mechanical properties, good dispersion and good adhesion to rubber matrix [2–5]. The adhesion between many types of commercial fibers and most elastomers has been improved by the discovery of the tricomponent system consisting of HRH [6–9]. The adhesion between rubber and short fibers is based on the in-situ formation of

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resorcinol-containing resin in the rubber vulcanizates [10–12]. The mechanical properties of the composites, such as modulus, tensile strength at break and ultimate elongation, depend on fiber orientation, fiber aspect ratio, and adhesion between fiber and matrix [13–21].

In this paper results are reported of the investigation on the effect of the adhesion system on the physico-mechanical properties of EPDM rubber reinforced by nylon–6 short fiber composites before and after ageing, as well as the selection of the most suitable concentration of the fiber.

MATERIALS AND TECHNIQUES

Materials

Rubber: Ethylene propylene diene terpolymer (EPDM-6505, unsaturation 9%, approximate ethylene 55%).

Fiber: Nylon–6 short fiber, denier 6, chopped to about 113 mm produced in a local carpet factory.

Rubber ingredients: They were of grades customarily used in practice.

Solvents and chemicals: All solvents and chemical reagents were of pure grade.

Techniques

All rubber mixes were prepared on a two roll mill of diameter 470 mm and width 300 mm, with speed of slow roll at 24 rev./min., and gear ratio of 1:1:4. The role temperature was kept at about 50°C during mixing. Care was taken to ensure fiber orientation in the mill direction. The compounded rubber was left overnight before vulcanization. The rheometric characteristics of EPDM mixes were studied [22] using a Monsanto oscillating disc rheometer R-100. Vulcanization press was operated at 152 ± 1°C under pressure of about 4 MPa. The mechanical properties were determined according to standard methods [23] using a model 5586 Instron universal testing machine. Swelling was conducted using toluene [24]. Ageing of the samples was carried out at 90°C for different time periods in a good aerated electric oven [25].

RESULTS AND DISCUSSION

I. Effect of HRH System

The components of the adhesion system (HRH) were incorporated in EPDM mixes, the formulation of which are shown in Table 1. The effect of each component on the reheological characteristics and

TABLE 1 Rubber Formulations Containing the Components of HRH System

Formula No.	W1	W2	W3	W4	W5	W6
EPDM	100	100	100	100	100	100
ZnO	5	5	5	5	5	5
Stearic acid	1	1	1	1	1	1
SRF	40	40	40	40	40	40
Processing oil	3	3	3	3	3	3
Sulphur	1	1	1	1	1	1
MBT	0.8	0.8	0.8	0.8	0.8	0.8
TMTD	1.5	1.5	1.5	1.5	1.5	1.5
Resorcinol	—	—	5	5	5	—
Hydrated silica	—	5	5	5	5	—
Vulcacite H	—	—	—	3.2	3.2	—
Nylon fibers	—	—	—	—	20	20

*SRF: Semi reinforcing furnace black, MBT: Mercapto-benzo-thiazole, TMTD: Tetramethyle thiuram disulfide, Vulcacite H: Hexamethylene tetramine.

the physico-mechanical properties of the vulcanizates were determined and are listed in Table 2. From the data given in Table 2 it is clear that the addition of silica increased the optimum cure time (tc_{90}) and decreased the cure rate index (CRI), which is attributed to the

TABLE 2 Effect of HRH System on the Properties of EPDM Rubber Vulcanizates

Formula No.	W1	W2	W3	W4	W5	W6
Rheometric properties						
ML (dN m)	16	15		19	17	22
MH (dN m)	112	116		61	76	110
tc_{90} (min)	14	16.5	Uncured	22	21.5	14.5
ts_2 (min)	2.5	2.5		1.5	1.2	3
CRI $(min)^{-1}$	8.7	7.1		4.6	4.9	8.7
Physicochemical properties (longitudinal direction)						
T.S. (MPa)	17.2	18.8	—	14.6	5.9	10.2
E (%)	461.6	547.9	—	484.5	91.9	314.1
Y.M.(MPa)	3.9	4.5	—	7.7	21.4	38.9
Physicochemical properties (Transversal direction)						
T.S. (MPa)	15.9	18.3	—	12.1	4.8	7.5
E (%)	378.5	433.1	—	462.1	100.3	342.1
Y.M.(MPa)	3.0	4.7	—	5.9	14.6	13.2
Q (%)	150	151	—	183	150	263

Where: ML: minimum torque, MH: maximum torque, tc_{90}: Optimum cure time, ts_2: Scorch time, CRI: Cure rate index, T.S: Tensile strength, E: Elongation at break, Y.M.: Young's modulus, Q: Equilibrium swelling.

retarding effect of silica. On the other hand, the addition of silica slightly increases the tensile strength and elongation at break.

The rubber mixes that contained resorcinol with silica did not cure, indicating that resorcinol prevents the vulcanization reaction. The addition of the three components of HRH system led to a decrease in the scorch time and an increase in the cure time with respect to control sample (W1), which indicates that the vulcanization reaction starts faster but with slower rate. Also, it is clear that the HRH system slightly decreases the tensile strength, increases the elongation at break and slightly increases the equilibrium swelling in toluene.

The addition of 20 phr nylon−6 short fibers in presence of the adhesion system (W5) decreased the tensile strength and sharply decreased the elongation at break. On the other hand, it is clear that the effect of fibers without the adhesion system (W6) is lower than that in the presence of it, especially on the elongation at break. But nylon fibers led to a sharp decrease in both longitudinal (warp) and transversal (weft) tensile strength of the vulcanizates.

II. Effect of Fiber Concentration

Nylon−6 short fibers were incorporated in EPDM rubber mixes with different concentrations up to 35 phr as shown in Table 3. The rheometric characteristics of the mixes were determined and are given in Table 3 as well. From these data, it is clear that the addition of nylon−6 short fibers increased the maximum torque, while there is almost no effect on both the optimum cure time and the scorch time.

The physico-mechanical properties were determined and presented in Figures 1−4. Figure 1 presents the dependence of both longitudinal and transversal tensile strength on the fiber concentration. It is clear

TABLE 3 Effect of Fiber Concentration on the Rheometric Characteristics of EPDM Formulations

Formula No.	W4	W7	W8	W9	W5	W10	W11	W12
Nylon 6–Fibers	—	5	10	15	20	25	30	35
Rheometric Characteristics								
ML (dN m)	19	20	17	18	17	24	25	22
MH (dN m)	61	64	84	72	76	62	76	70
tc_{90} (min)	25	25	20	16.5	21.5	17.5	22.5	19
ts_2 (min)	1.5	1.5	1.5	1.5	1.2	2	1.5	1.5
CRI $(min)^{-1}$	4.2	4.2	5.4	6.7	4.9	6.4	4.7	5.7

Base recipe (phr): EPDM 6505-100, ZnO-5; Stearic acid-1; SRF-40; proc. oil-3; resorcinol-5; hydrated silica-5; vulcacite H-3; sulphur-1, MBT-0.8, TMTD-1.5.

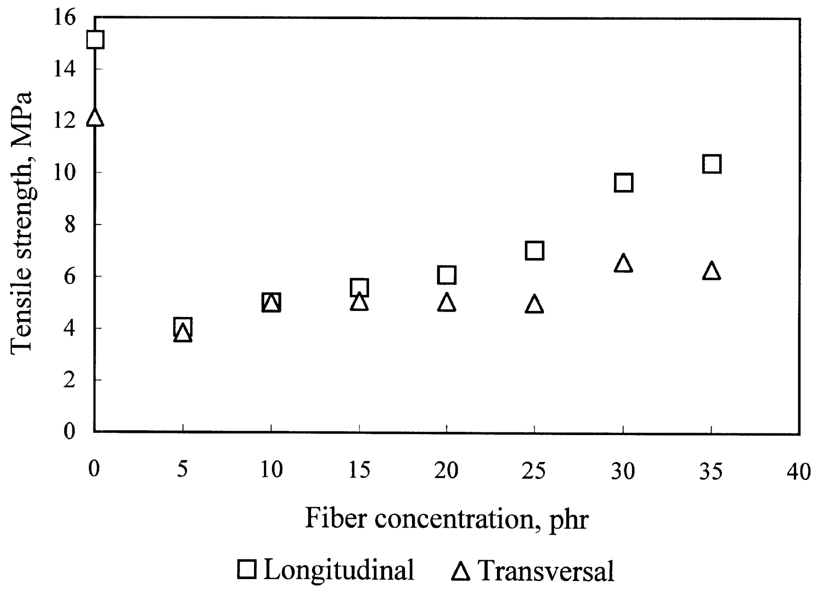

FIGURE 1 The dependence of both longitudinal and transversal tensile strength on the fiber concentration.

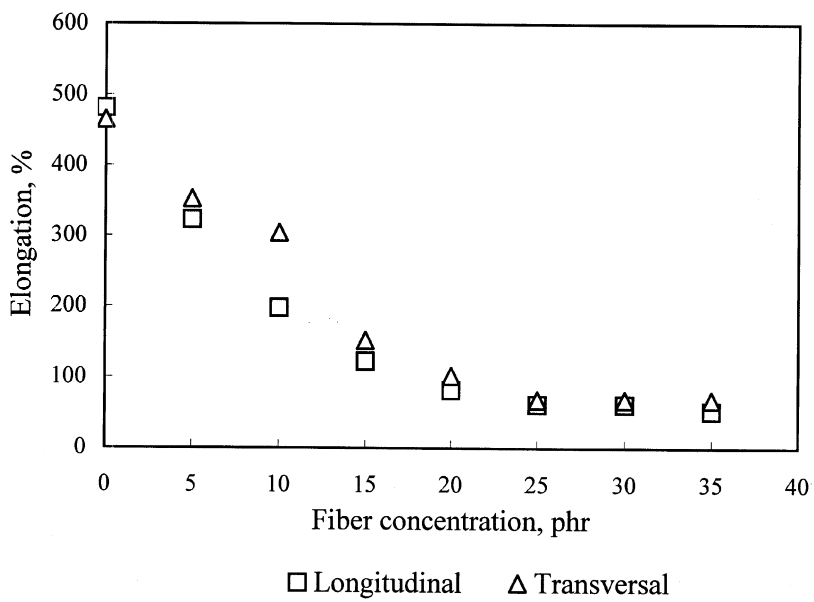

FIGURE 2 The dependence of both longitudinal and transversal elongation at break on the fiber concentration.

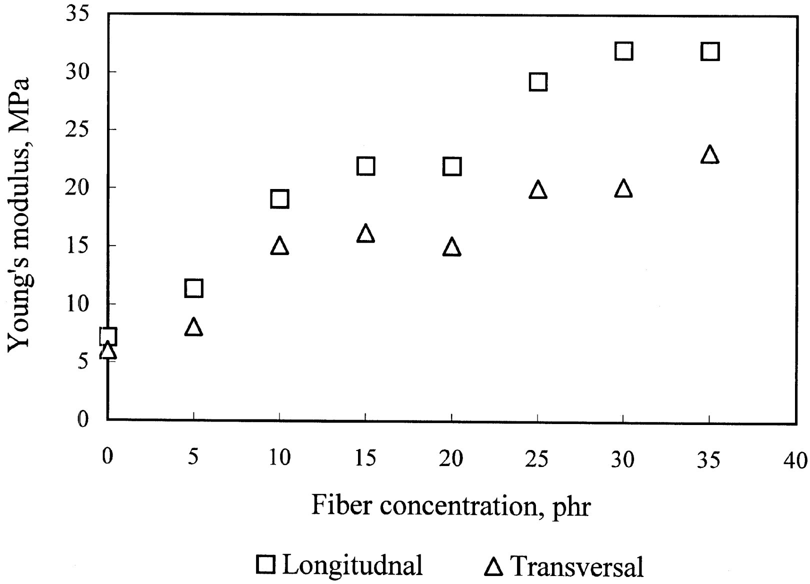

FIGURE 3 The dependence of both longitudinal and transversal Young's modulus on the fiber concentration.

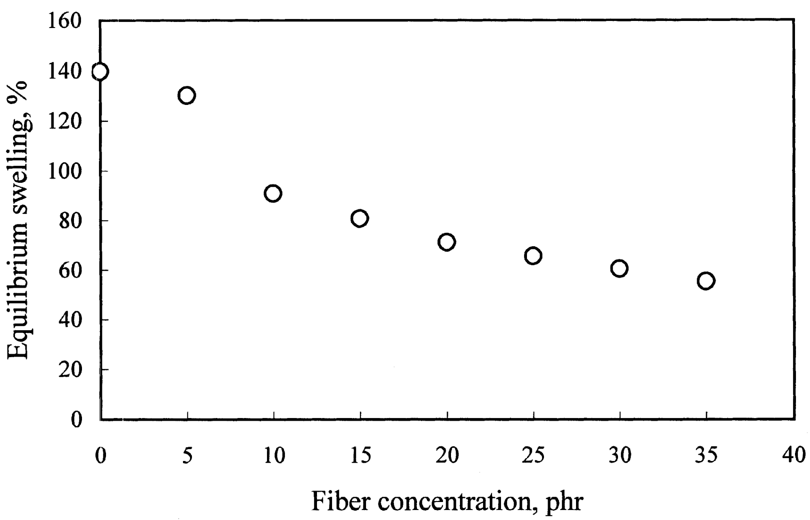

FIGURE 4 The dependence of equilibrium swelling in toluene on the fiber concentration.

that there is a sharp decrease in tensile strength in both directions with the initial loading of fiber followed by an increase in the tensile strength with the increase in fiber concentration up to 30 phr. Also, it is noticed that the increase of tensile strength in the longitudinal direction is higher than that in transversal one. Figure 2 shows the dependence of the elongation at break versus the fiber concentration. It is clear from the figure that addition of fiber leads to a sharp decrease in the elongation at break in both directions up to 25 phr, then the elongation remains constant with further increase in fiber concentration. On the other hand, it is noticed that the longitudinal elongation is lower than the transversal one for all fiber concentrations, which indicates that the fibers were well oriented in the rubber matrix in the mill direction.

The relation between Young'modulus in both directions and the fiber concentration is shown in Figure 3. From the data it is clear that Young's modulus increases with increasing fiber concentration up to 30 phr. Also it is noticed that the increase in Young's modulus in the longitudinal direction is higher than that in transversal one.

Figure 4 represents the relation of the equilibrium swelling in toluene versus fiber concentration. It is clear that the equilibrium swelling sharply decreases with the increase of the fiber concentration up to 20 phr, then a slight decrease occurs with further increase of fiber concentration.

Based on this discussion of results it can be concluded that the adhesion of nylon–6 short fibers to rubber mixes leads to rubber vulcanizates with high stress at low strain.

III. Effect of Ageing on the Fiber Loaded Vulcanizates

Vulcanizates containing different concentrations of nylon–6 short fibers were subjected to thermal oxidative ageing at 90°C for different time periods. The physico-mechanical properties of the aged samples were determined and the retained values were calculated and are presented in Tables 4–6.

From Table 4 it is clear that the tensile strength decreased with the ageing time for sample W4, which is without fiber. While the tensile strength of the other samples containing fibers with different concentrations remained approximately constant or slightly increased. This can be attributed to further formation of resorcinol-containing resin with ageing time, which enhances the adhesion between the rubber and the fiber during ageing.

The same observation can be obtained from the elongation and swelling data shown in Tables 5 and 6.

TABLE 4 Retained Tensile Strength of Rubber Vulcanizates

Formula No.		W4	W7	W8	W9	W5	W10	W11	W12
Retained T.S% (longitudinal direction)									
Time (days)	0	100	100	100	100	100	100	100	100
	2	98.4	105.7	101.6	98.9	112.3	98.7	96.5	106.0
	4	89.0	104.9	97.1	104.9	112.4	105.7	101.3	95.2
	6	75.8	101.3	98.9	110.7	105.2	94.3	99.1	102.1
	7	73.9	102.5	98.2	118.8	106.4	97.0	103.9	104.8
Retained T.S% (transversal direction)									
Time (days)	0	100	100	100	100	100	100	100	100
	2	99.7	100	108.8	104.7	104.0	89.1	102.8	97.9
	4	98.3	106.6	110.5	105.4	105.7	91.9	103.4	87.6
	6	91.6	106.1	111.6	106.6	105.5	91.0	104.1	86.6
	7	91.0	108.5	112.0	106.2	107.2	93.6	105.3	81.0

TABLE 5 Retained Elongation at Break of Rubber Vulcanizates

Formula No.		W4	W7	W8	W9	W5	W10	W11	W12
Retained E% (longitudinal direction)									
Time (days)	0	100	100	100	100	100	100	100	100
	2	90.1	91.2	83.6	92.8	84.6	90.6	90.4	93.0
	4	89.1	88.2	73.8	87.5	85.1	98.2	112	93.1
	6	85.6	87.8	71.4	80.7	84.3	98.1	101	95
	7	82.0	87.6	69.7	73.8	83.7	101	101	98.7
Retained E% (transversal direction)									
Time (days)	0	100	100	100	100	100	100	100	100
	2	88.3	96.8	90.0	91.0	106	112	98.0	105
	4	86.8	95.4	70.1	88.0	105	112	98.4	105
	6	86.7	92.3	61.1	81.2	102	111	98.8	106
	7	85.0	91.3	60.1	80.9	101	106	98.4	106

TABLE 6 Retained Equilibrium Swelling in Toluene of EPDM Rubber Vulcanizates

Formula No.		W4	W7	W8	W9	W5	W10	W11	W12
Time (days)	0	100	100	100	100	100	100	100	100
	2	98.7	95.3	95.6	91.8	89.7	97.8	93.2	96.4
	4	89.9	91.5	88.0	88.9	85.3	94.3	92.4	96.0
	6	85.5	82.9	72.6	79.0	82.4	89.2	90.5	95.8
	7	78.8	70.0	70.1	77.6	81.9	88.2	84.7	92.2

CONCLUSION

The following conclusions can be drawn from the experimental data presented in the paper:

1. The presence of nylon−6 short fibers increases the efficiency of the reinforcement of EPDM rubber vulcanizates.
2. The best volume fraction of reinforcing fiber that leads to high stress at low strain was found to be 25 phr.
3. The presence of reinforcing nylon−6 fibers and the adhesion system greatly improves the ageing resistance of the rubber vulcanizates. This may be attributed to further formation of crosslinked resin, which balances the degradation that occurs during ageing.

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