Effect of Adhesive-Coated Glass Fiber in Natural Rubber (NR), Acrylonitrile Rubber (NBR), and Ethylene–Propylene–Diene Rubber (EPDM) Formulations. I. Effect of Adhesive-Coated Glass Fiber on the Curing and Tensile Properties of NR, NBR, and EPDM Formulations

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ABSTRACT: Adhesive-coated glass fibers (3 and 6 mm in length) were added at loadings of 10, 20, and 30 phr in natural rubber (NR), nitrile rubber (NBR), and ethylene–propylene–diene comonomer (EPDM) formulations in both plain and carbon black mixes. The compounds were mixed in a two-roll mill and were characterized for their cure properties, tensile, tear, and Mullin's effect. In NR mixes, all of the formulations showed reversion in cure behavior, suggesting that NR remained unaffected. In NBR and EPDM mixes, almost all of the mechanical properties of the fiber improved. The result was more significant in EPDM than in NBR. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 91: 1111–1123, 2004

Key words: elastomers; rubber; curing of polymers

INTRODUCTION

Rubber is a complex material in general. It exhibits a unique combination of physical properties, although at the same time, a virtually infinite number of vulcanized rubber compounds are possible, yielding a very wide range of properties.

Most practical rubber compounds are based on an elastomer, a curing system, and a reinforcing filler. The various reinforcing fillers in industrial practice include carbon black, silica, and clay. Recently, industries have been moving toward fiber reinforcing fillers.

Glass fibers have been commercially available for around 50 years. There are several areas where these specially treated glass fibers are being used in combination with rubber compounds to provide good modulus control. The difference in behaviors between rubber and glass is large, and by careful choice of application and design, the advantages of both materials can be obtained. The final properties of the composites can be tailored to match industrial needs.

In this context, we undertook a study was undertaken to evaluate the effect of adhesive-coated glass fiber (commercially available as RICS) in formulations based on natural rubber (NR), nitrile rubber (NBR), and ethylene–propylene–diene comonomer (EPDM). The kind of coating given to the glass fiber used in this study was suitable for synthetic rubbers in general. NR was studied along with NBR and EPDM as a reference material.

The glass fiber used is stated to be of E glass with length between 3–12 mm. Fibers 3 and 6 mm in length were used. The product literature (for RICS) stated that the adhesive formulation was resorcinol formaldehyde latex based on a blend of vinyl pyridine, polybutadiene lattices containing some process additives.

Breakage of glass fiber during rubber processing is known. The object of this work was to

1. Ascertain indirectly if breakage occurred with the help of physicomechanical properties. The treated fiber was used as available in the most commonly consumed rubbers, NR, NBR, and EPDM.
2. The influence of carbon black on the preference of the coated fiber to these rubbers was also studied.

For convenience, the findings are reported in two parts: In part I, the preparation, cure, hardness, and tensile properties are reported, and in part II, the specific properties, such as fatigue, abrasion, and the effect of aging, are discussed.

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TABLE I
Rubber and Glass Fiber Specifications

Rubber	Grade	Specification
NR	RSS4	—
NBR	Aparene N553 NS	Bound ACN 34%; ML (1 + 4) at 100°C = 47
EPDM	Herlene 502	ML (1 + 4) at 125°C = 57; E/P weight = 63/37; ENB weight % = 4; Specific gravity = 0.86
Glass fiber	RICS	Diameter = 10 μm; RFL coat = 17%; Glass fiber = 83%; Specific gravity = 2.01

EXPERIMENTAL[1,2]

In this study, we studied the level of rubber impregnated chopped strands of glass fiber (RICS).We varied the RICS loading by 0, 10, 20, and 30 in gum rubber compounds and in compounds to which we added carbon black filler. We mixed 8 batches for each rubber formulation with various levels of RICS. Hence, we mixed 24 batches, and they were identified as B1, B2, B3, and so on to B24.

The batches were grouped as follows: B1–B4, non-carbon-black-reinforced NR compounds; B5–B8, carbon-black-reinforced NR compounds; B9–B12, non-carbon-black-reinforced NBR compounds; B13–B16, carbon-black-reinforced NBR compounds; B17–B20, non-carbon-black-reinforced EPDM compounds; and B21–B24, carbon-black-reinforced EPDM compounds.

The following characteristics were tested to evaluate the test mixes: cure characteristics, tensile properties, tear strength, hardness, and Mullin's effect.

Specification of rubber and glass fiber

The type, grade, and specification for each rubber used and for the glass fiber are listed in Table I.

TABLE II
NR Formulations Used to Study the Effect of Adhesive-Coated Glass Fiber Both in Plain and Carbon Black Mixes (phr)

Ingredient	B1	B2	B3	B4	B5	B6	B7	B8
NR	100	100	100	100	100	100	100	100
RICS (3 mm)	0	10	20	30	0	10	20	30
Zinc oxide	10	10	10	10	10	10	10	10
Stearic acid	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Carbon black (GPF)	0	0	0	0	40	40	40	40
TDO	1	1	1	1	1	1	1	1
Sulfur	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
MBTS	1	1	1	1	1	1	1	1
Total	116.5	126.5	136.5	146.5	156.5	166.5	176.5	186.5
Specific gravity	0.997	1.033	1.067	1.106	1.127	1.157	1.178	1.21

TDQ = polymerized 2,2,4-trimethyl-1,2-dihydroquinoline; MBTS = bis(2-benzothiazolyl) disulfide.

TABLE III
NBR Formulations Used to Study the Effect of Adhesive-Coated Glass Fiber Both in Plain and Carbon Black Mixes (phr)

Ingredient	B9	B10	B11	B12	B13	B14	B15	B16
NBR	100	100	100	100	100	100	100	100
Sulfur	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
RICS (6 mm)	0	10	20	30	0	10	20	30
Zinc oxide	5	5	5	5	5	5	5	5
Stearic acid	1	1	1	1	1	1	1	1
CI resin	5	5	5	5	5	5	5	5
Carbon black (HAF)	0	0	0	0	45	45	45	45
TDQ	2	2	2	2	2	2	2	2
MBTS	1	1	1	1	1	1	1	1
TMTD	1	1	1	1	1	1	1	1
Total	115.5	125.5	135.5	145.5	145.5	155.5	165.5	175.5
Specific gravity	1.027	1.066	1.109	1.140	1.172	1.2	1.229	1.253

TDQ = polymerized 2,2,4-trimethyl-1,2-dihydroquinoline; MBTS = bis(2-benzothiazolyl)disulfide; TMTD = tetra methyl thiuram disulfide.

TABLE IV
EPDM Formulations Used to Study the Effect of Adhesive-Coated Glass Fiber Both in Plain and Carbon Black Mixes (phr)

Ingredient	B17	B18	B19	B20	B21	B22	B23	B24
NBR	100	100	100	100	100	100	100	100
RICS (3 and 6 mm)[a]	0	10	20	30	0	10	20	30
Zinc oxide	5	5	5	5	5	5	5	5
Stearic acid	2	2	2	2	2	2	2	2
CI resin	5	5	5	5	5	5	5	5
Carbon black (HAF)	0	0	0	0	45	45	45	45
TDQ	5	5	5	5	5	5	5	5
Sulfur	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
MBTS	1	1	1	1	1	1	1	1
TMTD	1	1	1	1	1	1	1	1
Total	119.5	129.5	139.5	149.5	164.5	174.5	184.5	194.5
Specific gravity	0.92	0.960	0.994	1.028	1.057	1.091	1.117	1.142

TDQ = polymerized 2,2,4-trimethyl-1,2-dihydroquinoline; MBTS = bis(2-benzothiazolyl)disulfide; TMTD = tetramethyl Thiuram Disulfide.

[a] 3-mm RICS was used in formulations B17–B20 and 6-mm RICS was used in formulations B21–B24.

Test equipment used

We used a shore A hardness durometer, a tensile testing machine, and an oscillating disc rheometer (Monsanto ODR2000).

Formulations

See Tables II–IV for the NR, NBR, and EPDM formulations used to study the effects of adhesive-coated glass fiber in both plain and carbon black mixes.

Mixing

The test mixes were prepared in lab-size open-mixing mill (150 × 325 mm) with the sequence listed in Table V.

The rubber and fiber were mixed thoroughly in a mixing mill for better dispersion.

TABLE V
Order of Mixing

Step	Batches B1–B8	Batches B9–B16	Batches B17–B24
1	NR	NBR	EPDM
2	RICS	Sulfur	RICS
3	Zinc oxide	RICS	Zinc oxide
4	Stearic acid	Zinc oxide	Stearic acid
5	Carbon black	Stearic acid	CI resin
6	TDQ	CI resin	Carbon black
7	Sulfur	Carbon black	TDQ
8	MBTS	TDQ	Sulfur
9		MBTS	MBTS
10		TMTD	TMTD

TDQ = polymerized 2,2,4-trimethyl-1,2-dihydroquinoline; MBTS = bis(2-benzothiazolyl)disulfide; TMTD = tetramethyl thiuram disulfide.

Preparation of test specimens

The specimens, tests, and vulcanization times and temperatures are listed in Table VI.

RESULTS AND DISCUSSION

Important parameters[3] in fiber reinforcement

In vulcanizates with short-fibers added, the properties are affected by (1) fiber loading, (2) retention of the aspect ratio, (3) fiber orientation, (4) adhesion, and (5) fiber dispersion.

The second variable was effectively studied by keeping the other parameters relatively under control. However, the aspect ratio of the fiber was expected to attain a comparable value in the vulcanizates, notwithstanding the extent of fiber breakage during (open-mill) mixing, provided the fiber became a part of the matrix, which was possible only with adhesion. The proprietary coating was thought to provide this. Random fiber distribution was also expected because in sheet-out from the mill, no directionality was attempted. As the fiber was adhesive-activated, the influence of the adhesion level also did not arise in all of the test formulations.

TABLE VI
Test Specimens

		Vulcanization	
Test	Specimen	Temperature (°C)	Time (min)
Tensile and tear	Slab	150	15
Hardness	Button	150	15

TABLE VII
Rheometer Cure Data of NR Mixes with Various Levels of Adhesive-Coate Glass Fiber

Batch	M_L (lb in.)	M_H (lb in.)	t_{s2} (min)	t_{90} (min)	t_{98} (min)	Cure rate $[100/(t_{90} - t_{s2})]$
B1	2.48	15.65	1.2	1.95	3.32	22.22
B2	1.75	18.55	0.9	1.56	2.8	22.25
B3	1.9	21.7	0.9	1.56	2.9	24.88
B4	2.45	22.05	1	1.67	2.65	27.32
B5	2.75	23.5	0.8	1.41	3.1	18.94
B6	2.65	26.5	0.74	1.62	3	18.32
B7	1.9	26	0.69	1.65	2.61	20.58
B8	2	28.58	0.67	1.5	2.65	20.58

Study of NR formulations

NR mixing did not pose any problem with fiber loading. Green NR mixes without carbon black were brick red in color with a fiber loading at 30 phr. When the green mix was stretched, it presented a leathery appearance.

Cure characteristics

The experimental data are given in Table VII. Fiber loading in nonblack NR mixes showed decrease in optimum cure time (t_{90}) and scorch time (t_{S2}). An increase in the cure rate and a reduction in reversion time was noticed with increased fiber loading, as shown in Table VII. In the NR black mix, the fiber increased the maximum torque and decreased the minimum torque and reversion time, as shown in Table VII. This trend was beneficial from the process point of view. At the same time, t_{S2} was constant for batches B5 and B6 (0- and 10-phr fiber, respectively) and decreased for B7 and B8. t_{90} increased for B6 and decreased for B8.

In general, NR plain mixes (B1 to B4) showed better scorch safety, faster cure, and lower viscosity (M_L) than black-added mixes (B5–B8). This trend is normally expected because carbon blacks do affect cure characteristics. A similar trend was observed in the presence of coated glass fibers. The processability of NR mixes was also not greatly impaired if one considered low M_L values as process indicators.

Tensile strength

Tensile strength decreased with increasing fiber concentration in nonblack NR compounds (B1–B4), as shown in the graph (Fig. 1). However, the initial modulus increased. However, with black reinforcement, NR compounds (B5–B8, Fig. 2) did not show a corresponding increase with the fiber loading. This may have been because carbon black particulate reinforcement is more dominant than fiber reinforcement. A further reduced tensile strength[4] may have been due to one or a combination of the following: (1) inhomogeneity, (2) a decrease in the crosslink density with increasing fiber loading, and (3) fiber loading interfering with the crystallization of NR

Elongation at break

Similar to tensile strength, elongation at break showed more significant decreases in nonblack mixes (Fig. 1)

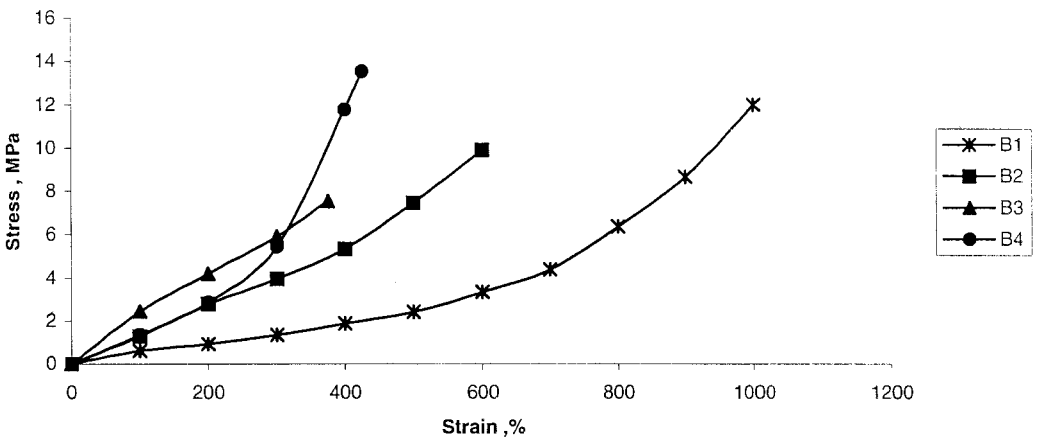

Figure 1 Stress–strain diagrams of NR mixes without carbon black with various concentrations of treated glass fiber.

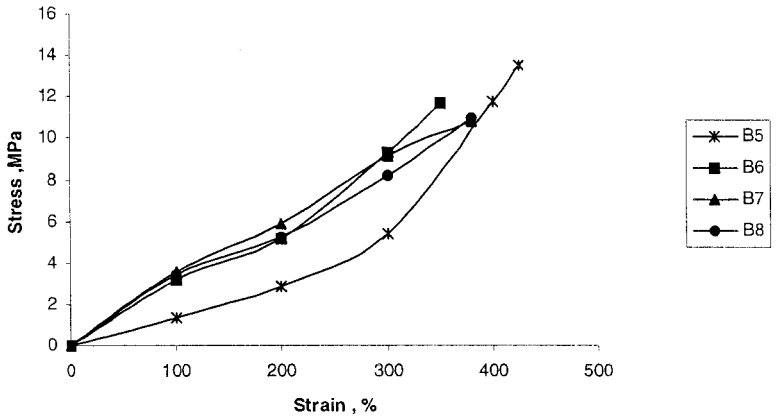

Figure 2 Stress–strain diagrams of NR mixes with carbon black with various concentrations of treated glass fiber.

than in black-reinforced mixes (Fig. 2). This may have been due to (1) a reduction of chain extensibility, (2) the fact that the fiber of low extensibility relative to the elastomeric matrix was controlling the overall deformation, and (3) the fact that fiber particles may have prevented the NR matrix from undergoing a true affine deformation.[5]

Modulus at 300% elongation (M300)

Modulus (M300) values did not exhibit a well-defined trend in plain mixes. However, better clarity was noticed with fiber loading in black formulations (B5–B8). Nonblack NR compounds (B1–B4, Fig. 1) showed an increase in modulus for 10- and 20-phr fiber loadings, but at a 30-phr loading, the modulus decreased. However, with carbon black reinforcements, NR had almost the same modulus. The benefit of fiber in the enhancement of the modulus was perhaps offset by the interference of strain-induced crystallization.

Tear strength

Tear strength increased with increasing in RICS loading in nonblack NR compounds (B1–B4, Fig. 3). This may have been due to fiber orientation being perpendicular to the direction of propagation of tear in the rubber matrix rather than being oriented parallel to it in general,[4] even though in this study, no specific fiber orientation was attempted. In carbon-black-reinforced NR, the tear strength increased for 10-phr fiber loading (B6) but decreased for 20- and 30-phr loadings of fiber (B7 and B8, Fig. 4). This may have been to the influence of carbon black in the fiber orientation.

Hardness

Hardness increased for the compounds with 10 phr of added fiber, as shown in Figure 5, and remained almost unchanged with 20 and 30 phr of added fiber in nonblack NR compounds. In black-filled compounds, hardness increased with increasing fiber addition, as shown in Figure 6. This observation suggests that (because hardness is related to shear modulus) fiber concentration had little influence on modulus as well.

Mullin's effect

Repeated stretching of the tensile specimen gave a favorable result for both nonblack and black-reinforced compounds (B1–B8, Figs. 7 and 8). There was no significant change in tensile strength. This may have been due to good fiber–rubber interaction, which implies that working on the vulcanizates may have

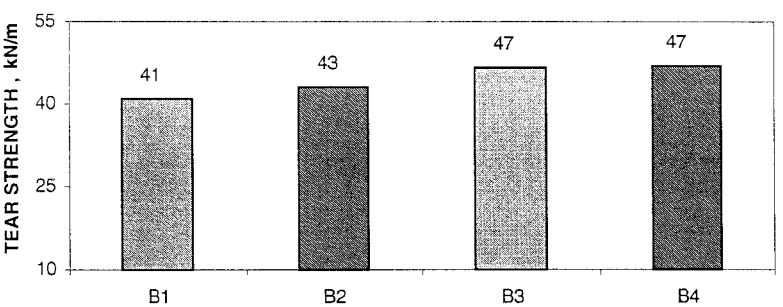

Figure 3 Tear strength of NR mixes without carbon black with various concentrations of adhesive-coated glass fiber.

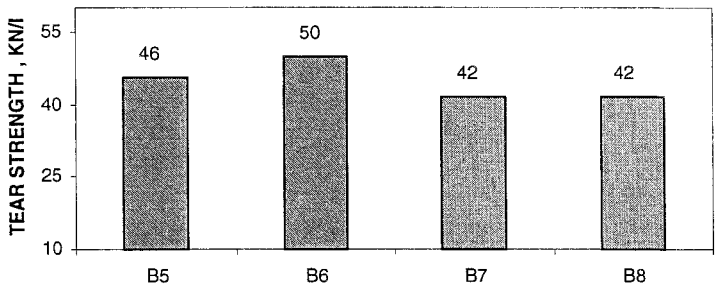

Figure 4 Tear strength of NR mixes with carbon black with various concentrations of adhesive-coated glass fiber.

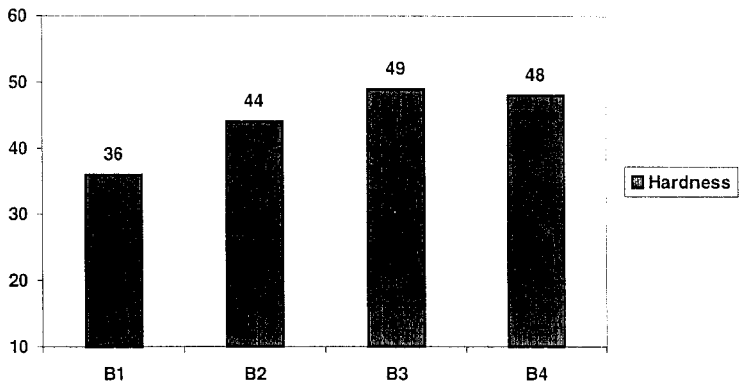

Figure 5 Hardness of NR mixes without carbon black with various concentrations of adhesive-coated glass fiber.

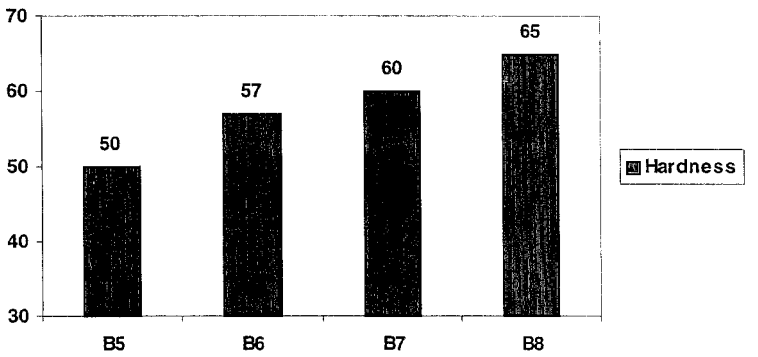

Figure 6 Hardness of NR mixes with carbon black with various concentrations of adhesive-coated glass fiber.

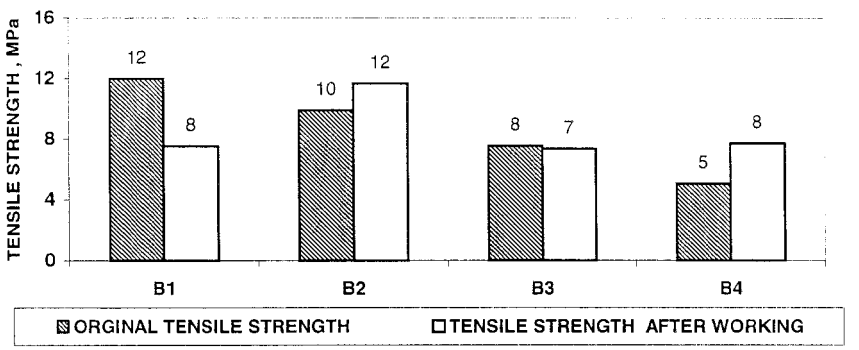

Figure 7 Effect of adhesive-coated glass fiber concentration on NR mixes without carbon black after repeated stretching.

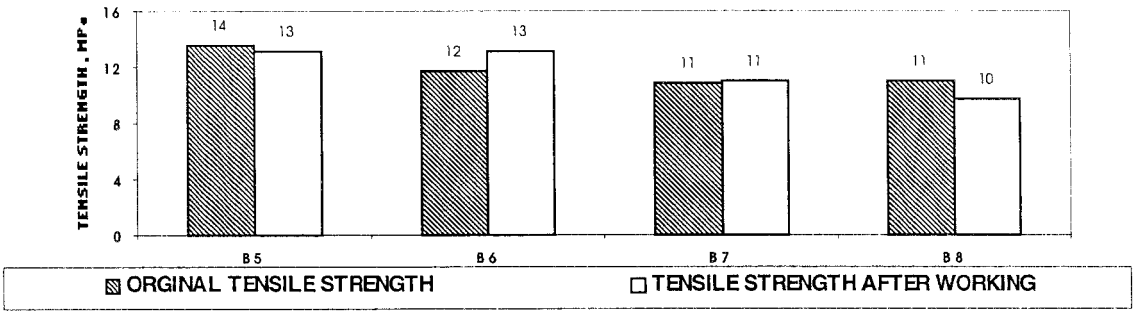

Figure 8 Effect of adhesive-coated glass fiber concentration on NR black mixes after repeated stretching.

also eliminated weak sites and inhomogeneities. For carbon-black-reinforced fiber-added NR compounds, initially, B5 showed softening (Fig. 8). This softening occurred when the number and strength of filler–rubber bonding sites were large.

Study of NBR formulations

NBR mixing did not cause any difficulty during the fiber addition, although the incorporation took a little longer than NR. Green mixes of NBR without black were brick red in color with RICS loading (B12).

Cure characteristics

RICS loading in nonblack NBR mixes showed an increase in minimum torque (M_L) and maximum torque (M_H). The cure rate increased up to 20 phr of fiber in B9–B11 and decreased in B12 (30 phr of RICS), as shown in Table VIII. t_{S2} also decreased. The interference of carbon black in the cure mechanism was stronger in NBR than in NR in the presence of treated glass fiber.

Tensile strength

The tensile strength of plain NBR mixes with various fiber levels did not exhibit a smooth trend (Fig. 9). However, a prominent increase in yield strength was noticed with increasing fiber concentration. The mechanism suggested for such increase in yielding was that of lack of compatibility.[4] However, at the same time for vulcanizates of B10 and B11 (10- and 20-phr fiber loadings), the tensile strength increased. This may

TABLE VIII
Rheometer Data for Various Fiber Concentrations in the NBR Mixes

Batch	M_L (lb in.)	M_H (lb in.)	t_{s2} (min)	t_{90} (min)	Cure rate $100/(t_{90} - t_{s2})$
B9	2.05	16.95	1.13	1.6	35.46
B10	2.35	19.25	1.07	1.5	38.76
B11	2.63	21.1	1.05	1.47	39.68
B12	2.95	22.9	1.05	1.5	37.04
B13	5.02	28.45	0.85	1.28	41.67
B14	7.55	30.5	0.79	1.2	40.65
B15	5.75	31	0.83	1.25	39.68
B16	6.05	32.25	0.85	1.325	35.09

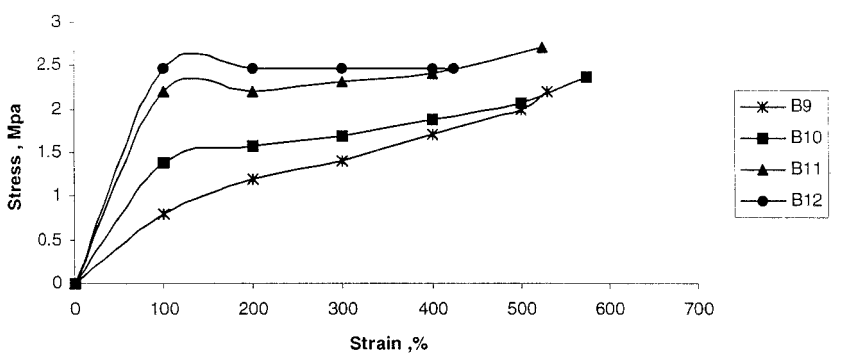

Figure 9 Stress–strain diagrams of NBR mixes without carbon black with various concentrations of adhesive-coated glass fiber.

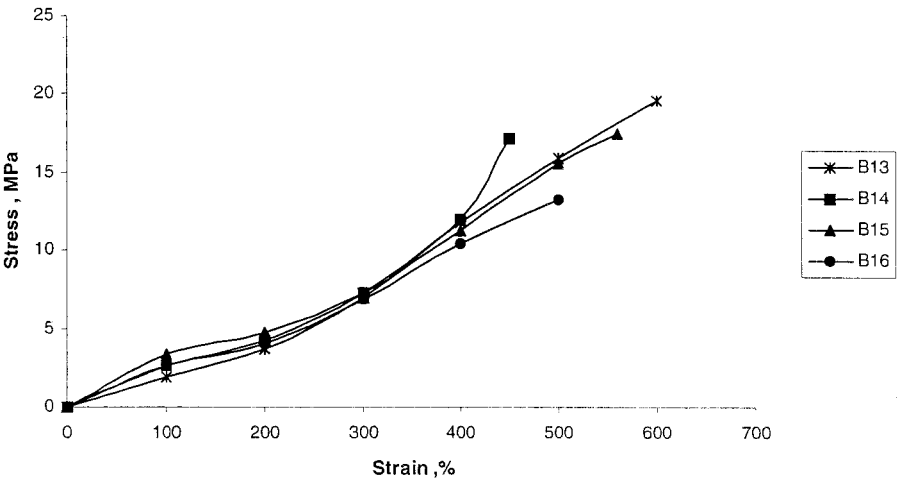

Figure 10 Stress–strain diagrams of NBR mixes with carbon black with various concentrations of adhesive-coated glass fiber.

have been due to the presence of just the required quantity of fiber, which enhanced the strength in non-crystallizing rubber compared to a gum compound. It may also have been due to good bonding between RICS and the NBR matrix. In carbon-black-reinforced compounds at higher fiber concentrations, the tensile strength decreased (Fig. 10). Further, the yielding phenomenon noticed in plain mixes was absent here. Compared with B16 (30 phr of fiber), B14 and B15 (10 and 20 phr of fiber) had good tensile strengths (Fig. 10). A lack of bonding between the fiber and NBR matrix at a higher dosage in the presence of carbon black may have been the reason for this.

Modulus

Increasing fiber concentration in B9–B12 (nonblack compounds) resulted in increased moduli. Carbon-black-reinforced compounds (B14–B16) showed a slight increase in moduli with increasing fiber loading. However, the increase for B14–B16 was lower compared to the nonblack compounds (B10–B12). This may have been due to carbon black interference with RICS and the NBR matrix. B16 (30 phr of fiber) showed a decrease in modulus compared to B14 and B15 (10 and 20 phr of fiber). This may have been due to the fiber concentration exceeding the optimum level.

Elongation at break

An increase in fiber addition resulted in a decrease in the elongation at break of B9–B12 (without carbon black compounds, Fig. 9). This may have been due to strong fiber–NBR matrix interaction and immobilization of the rubber chains. For B13–B16, the higher elongation (in Fig. 10) may have been due to the interference of carbon black with the fiber.

Tear strength

Tear strength increased with increasing fiber addition in nonblack compounds (B9–B12, Fig. 11). The effect was more pronounced with carbon black addition (Fig. 12). There was a substantial increase in tear strength. This may have been due to the limitation of the amorphous rubber matrix. The trend in fiber with

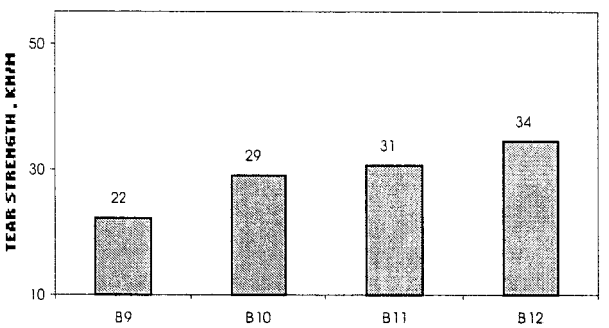

Figure 11 Tear strength of NBR mixes without carbon black with various concentrations of adhesive-coated glass fiber.

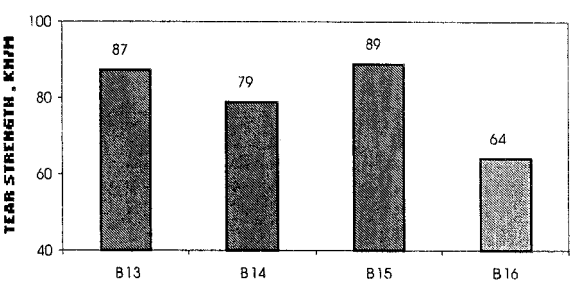

Figure 12 Tear strength of NBR mixes with carbon black with various concentrations of adhesive-coated glass fiber.

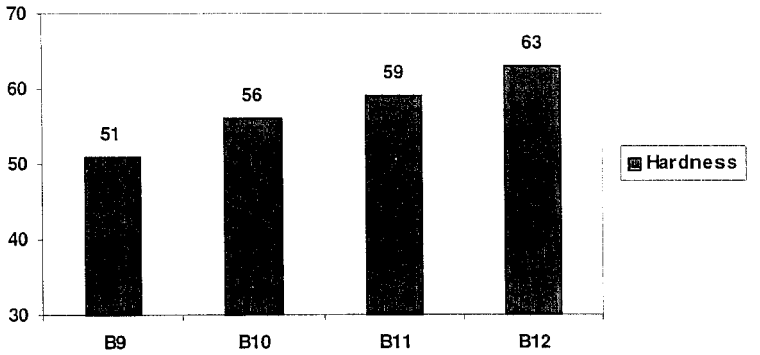

Figure 13 Hardness (shore A) of NBR mixes without carbon black with various concentrations of adhesive-coated glass fiber.

carbon black addition had a definite positive effect on tear strength (B12 and B16, Figs. 11 and 12).

Hardness

Hardness increased with increasing fiber addition in nonblack NBR compounds, as shown in Figure 13. Black-filled NBR compounds did not show a pronounced increase in hardness with increasing RICS loading, as shown in Figure 14. Hardness is a measure of low strain modulus. The increase in this modulus was minimal with RICS in black compounds compared with nonblack mixes. A similar observation was made for tensile modulus as well, as mentioned previously.

Mullin's effect

The test for Mullin's effect showed a positive result for bonding between the fiber and the rubber matrix. Both black-filled compounds (B13–B16, Fig. 15) and nonblack-filled compounds (B9–B12, Fig. 16) showed a similar trend. Similar to NR, the initial working on the

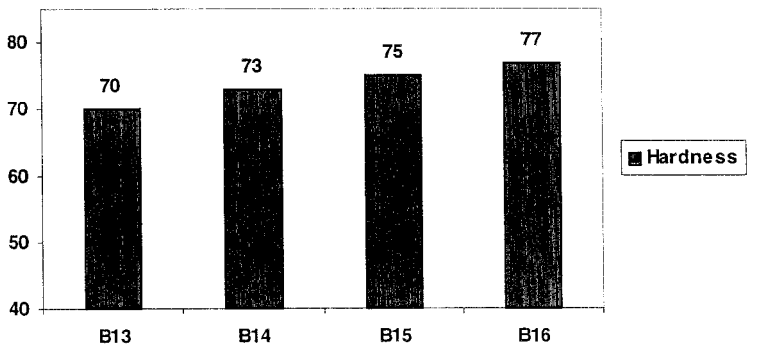

Figure 14 Hardness (shore A) of NBR mixes with carbon black with various concentrations of adhesive-coated glass fiber.

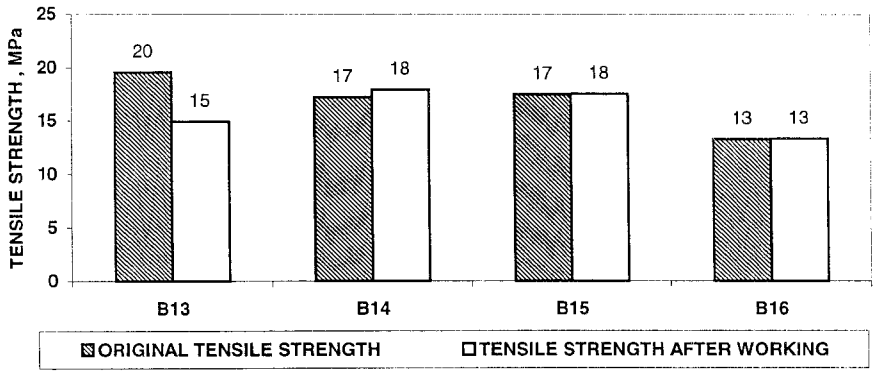

Figure 15 Tensile strength of NBR mixes with carbon black with various concentrations of adhesive-coated glass fiber after repeated stretching.

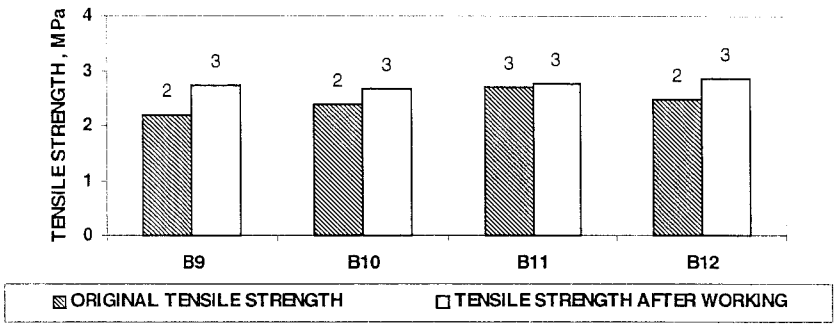

Figure 16 Tensile strength of NBR mixes without carbon black with various concentrations of adhesive-coated glass fiber after repeated stretching.

vulcanizates improved the rubber–fiber interaction by eliminating flaws and defects.

Study of EPDM formulations

The incorporation of fiber in EPDM took longer than in NR and NBR. The nip gap was reduced to 0.5 mm for the EPDM mixing during the incorporation of RICS. The mixes appeared leathery.

Cure characteristics

Unlike in NR and NBR, there was a gradual increase in compound viscosity (M_L) and cure torque (M_H) with RICS loading. A decrease in t_{S2} and t_{90} and an increase in the cure rate were other features of EPDM seen with increasing fiber concentration (Table IX). In black EPDM mixes, an increase in the M_L of B22 and a decrease in B24 were noticed. A decrease in t_{S2} and an increase in the cure rate were noticed with increasing RICS loading. Such trends showed that the compounds became more cure-active in the presence of coated fiber. The level of cure activity was, of course, controlled by the carbon black. Unlike NR or NBR, the increase in cure rate with fiber loading was three times and two times, respectively, in nonblack compounds (B17–B20) and in black compounds (B21–B24).

It appeared that the process benefits were greater if the rubber matrix chosen was weaker (i.e., EPDM compared to NR). This proposition needed to be verified in terms of mechanical properties such as tensile properties and tear strength.

Tensile strength

In EPDM, an increase in tensile strength was noted in non-black-filled compounds with fiber, as shown in Figure 17. Compounds with 10- and 20-phr loaded RICS (B18 and B19) almost had the same tensile strength, and 30-phr loaded RICS (B20) had slightly lower tensile strengths compared to B18 and B19. This may have been because a higher RICS loading reinforced the matrix, facilitating stress transfer, which overweighed the dilution effect.[3] In black-added mixes, carbon black may have increased the tensile strength, but the trend was not as smooth, as shown in Figure 17. B22 showed a higher tensile strength compared to B23 and B24 (Fig. 18). This decrease in tensile strength may have been due to the introduction of flaws at the fiber ends where high stress concentration occurred, causing bonds between the fiber and EPDM to break.[5] The increase in tensile strength with RICS loading may have been due to the (1) adhesive agent being favorable to EPDM, (2) a better dispersion of fiber in EPDM matrix, and (3) better compatibility.

TABLE IX
Rheometer Cure Data of EPDM Mixes with Various Concentrations of Adhesive-Coated Glass Fiber

Batch	M_L (lb in.)	M_H (lb in.)	t_{s2} (min)	t_{90} (min)	Cure Rate $100/(T_{90} - t_{s2})$
B17	3.75	18.95	1.68	4.05	7.03
B18	4.39	16.63	1.4	2.55	14.49
B19	5	17.4	1.08	1.95	19.16
B20	5.45	16.65	1.01	1.77	21.93
B21	8.3	35.4	0.89	2.32	11.66
B22	9.45	32.45	0.81	1.81	16.67
B23	10	30.58	0.66	1.55	18.73
B24	9.6	28.4	0.76	1.5	22.52

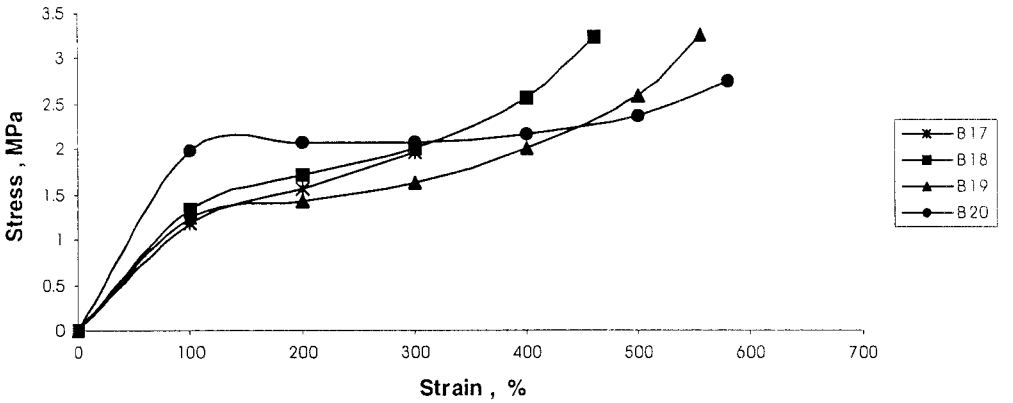

Figure 17 Tensile stress–strain diagrams of EPDM mixes without carbon black with various concentrations of adhesive-coated glass fiber.

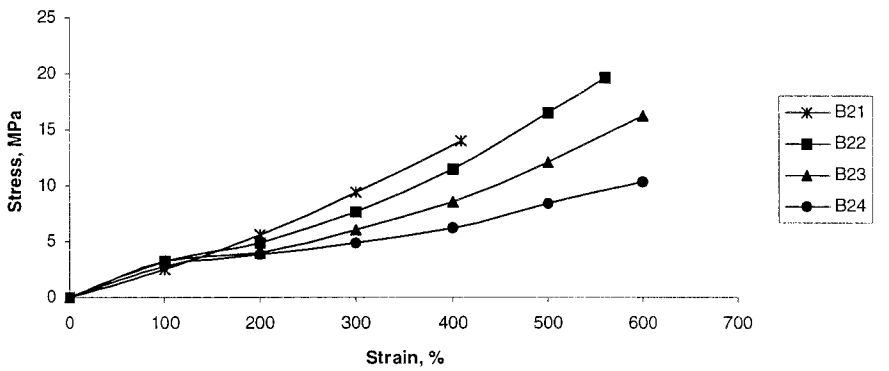

Figure 18 Tensile stress–strain diagrams of EPDM mixes with carbon black with various concentrations of adhesive-coated glass fiber.

Elongation at break

An increase in elongation at break was noticed in the nonblack compounds with increasing fiber loading (B17–B20). For black-filled compounds also, an increase in the fiber loading increased the elongation at break (Fig. 18). However, the variation was not very significant. An increase in elongation at break may have been due to unbonded RICS pulling away from rather than reinforcing the EPDM matrix[5] or to the fiber becoming an integral part of the matrix.

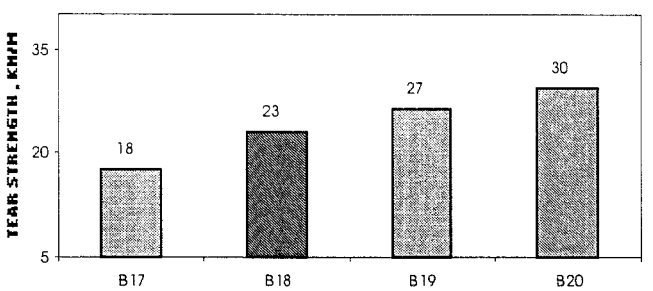

Figure 19 Tear strength of EPDM mixes without carbon black with various concentrations of adhesive-coated glass fiber.

Modulus

An increase in fiber loading in nonblack compounds resulted in an increase in the modulus, but a smooth trend was not observed (Fig. 17). B18 (10 phr of fiber) showed a higher modulus compared to other compounds (Fig. 17). In carbon-black-filled compounds, the fiber increased the moduli. B22 (10-phr fiber loading) showed a very high modulus (Fig. 18) among the fiber-loaded mixes. Increases in moduli may have occurred because the fiber at 10 phr was just the optimum above the concentration at which structural defects became overriding.

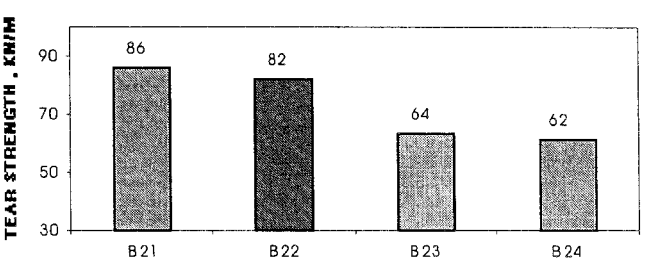

Figure 20 Tear strength of EPDM mixes with carbon black with various concentrations of adhesive-coated glass fiber.

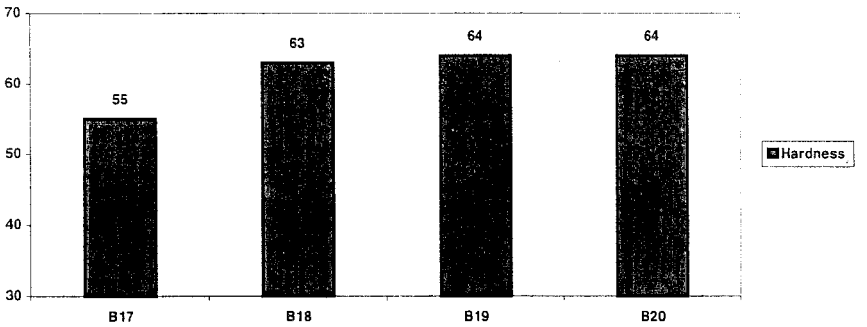

Figure 21 Hardness (shore A) of EPDM mixes without carbon black with various concentrations of adhesive-coated glass fiber.

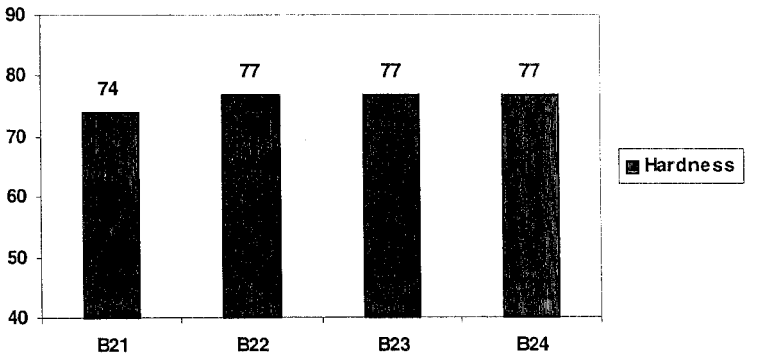

Figure 22 Hardness (shore A) of EPDM mixes with carbon black with various concentrations of adhesive-coated glass fiber.

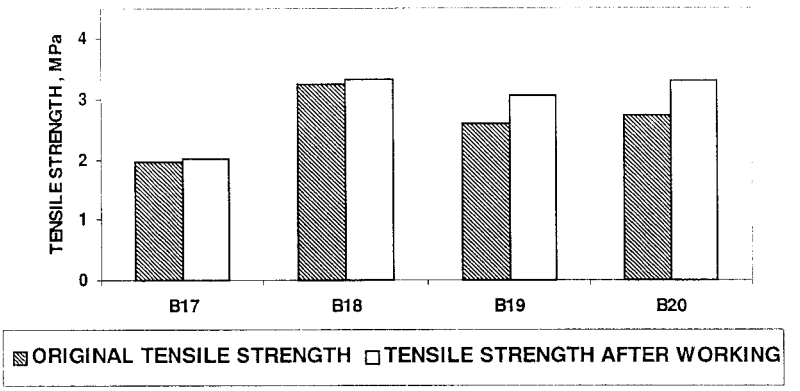

Figure 23 Effect of repeated stretching on EPDM mixes without carbon black with various concentrations of adhesive-coated glass fiber.

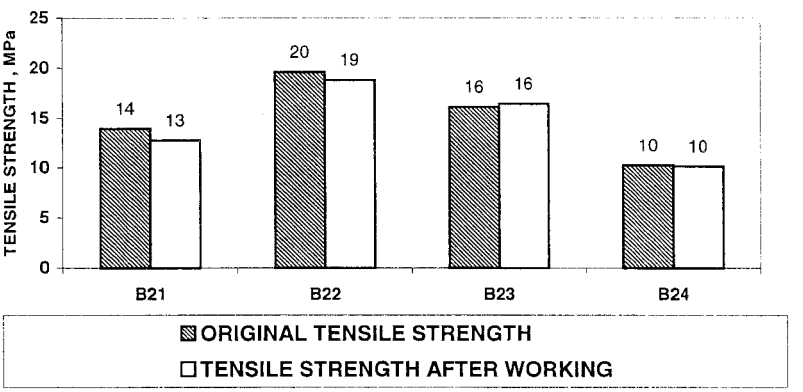

Figure 24 Effect of repeated stretching on EPDM mixes with carbon black with various concentrations of adhesive-coated glass fiber.

Tear strength

In nonblack compounds, fiber loading resulted in an increase in tear strength (Fig. 19). This may be due to the fiber getting oriented perpendicular to the direction of propagation of tear in the rubber.[3] In black-filled compounds, a decrease in tear strength was noticed with increasing fiber loading (Fig. 20). Unlike NR and NBR in EPDM, fiber alone increased the tear strength (Fig. 19), whereas when carbon black was present, a reversal was noticed (Fig. 20). These results indicate that fiber may have affected the carbon black–EPDM interaction as for as the tearing mechanism was concerned.

Hardness

EPDM compounds did not show a significant change in hardness with increasing fiber addition for nonblack and black compounds, as shown in Figures 21 and 22. The higher hardness in black compounds truly reflected the effect of carbon black, even in the presence of higher fiber loadings, for example, 30 phr (B24, Fig. 22). The initial increase by 3 units with 10 phr of fiber (B21 and B22) was similar to the observation made with M300. Regardless of the fiber addition hardness remained around 75 shores A in carbon-black-reinforced vulcanizates, a trend similar to that seen in NBR. However, in NR, the situation was different.

Mullin's effect

Mullin's effect showed better result for all of the vulcanizates (B17–B24, Figs. 23 and 24). Higher retention properties may have been due to better compatibility of the fiber–EPDM matrix, which was similar to NR and NBR.

CONCLUSIONS

The treated glass fiber had a bearing on the cure and technical properties of NR, NBR, and EPDM. The influence was more pronounced in NBR and EPDM. EPDM showed particularly higher cure rates. The retention of tensile strength after repeated stretching was also better. However, with tensile properties and tear strength, an irregular trend with fiber loading was noticed. Further hardness showed a trend that was dependent on the base polymer. The initial working on the vulcanizates did not impair the tensile strength.

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