

# Recycling of EPDM Waste. I. Effect of Ground EPDM Vulcanizate on Properties of EPDM Rubber

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**ABSTRACT:** Processing, cure characteristics, and mechanical properties of EPDM rubber containing ground EPDM vulcanizate of known composition were studied. Mooney viscosity increases and Mooney scorch time decreases by the addition of the ground vulcanizate. At higher loadings of the ground rubber, the maximum rheometric torque decreases. On addition of ground waste, stress-strain properties and tear resistance increase, whereas heat buildup marginally increases, resilience marginally decreases, low-strain modulus remains constant, and abrasion resistance decreases. The interplay between the filler effect of the ground EPDM and the crosslink density changes of the EPDM matrix is believed to be the reason for the variation in mechanical properties. It is believed that sulfur migration occurs from the raw EPDM matrix (R-EPDM) to the ground waste EPDM (W-EPDM) particle while accelerator migration occurs from W-EPDM to R-EPDM. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 82: 3293–3303, 2001

**Key words:** recycling; EPDM waste; ground rubber; accelerator migration

## INTRODUCTION

Discarded rubber products constitute the major part of accumulating nonbiodegradable wastes. Development of an effective recycling technology continues to be an interesting motive because earlier efforts to recycle rubber wastes, such as incineration, pyrolysis, and land fills, ended up with ecological and quality problems. Reutilization of powdered waste rubber seems to be a viable economic alternative that is also environmentally friendly.

Powdered waste rubber has been used as a filler in mixtures with virgin rubbers and thermoplastics<sup>1–13</sup> and also for modifying concrete and asphalt.<sup>14,15</sup> Effects of ground rubber on mixing be-

havior, curing characteristics, and mechanical properties of virgin rubbers like NR and SBR were previously reported.<sup>16–19</sup> Studies were also conducted on mixing of powdered specialty elastomers with general-purpose rubbers.<sup>20,21</sup> Processability and migration of curatives in waste rubber-filled compounds were studied by several workers.<sup>19,22,23</sup> Phadke and De<sup>24</sup> observed that cryogenically ground waste rubber can be vulcanized to an acceptable level by compounding with additional sulfur and accelerator, whereas incorporation of additional activators caused only marginal improvement in physical properties. It is believed that waste rubber vulcanizate contains unreacted zinc oxide and stearic acid, which take part in subsequent curing. To improve the adhesion between virgin rubber and powdered rubber vulcanizate, surface modification of the powder was done and studies were made with the surface-modified powders.<sup>7,25–27</sup>

EPDM rubber in the form of discarded automotive parts and roofing contributes much to envi-

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ronmental pollution. Given that EPDM products contain high oil and filler loadings, it is possible that EPDM powder may act as a low-modulus reinforcing filler. The present work aims to study the potential of ground waste EPDM, abbreviated as W-EPDM, as filler in virgin or raw EPDM (R-EPDM). EPDM vulcanizate after aging was taken as a substitute for waste EPDM (W-EPDM). This investigation reports the results of studies on effects of ground W-EPDM on the Mooney viscosity, cure characteristics, and mechanical properties of the virgin EPDM rubber.

## EXPERIMENTAL

### Materials

The EPDM rubber used for this study was Royalene 521 supplied by the Uniroyal Chemical Company (Naugatuck, CT).

The ground rubber was made from a typical sulfur-cured EPDM vulcanizate whose composition in phr (parts per hundred rubber, by weight) was as follows: EPDM, 100; GPF carbon black, 120; oil (Sunpar 2280), 70; brown factice, 10; paraffin wax, 10; polyethylene glycol, 1.5; 2,2,4-trimethyl-1,2-dihydroquinoline (Accinox TQ), 0.5; sulfur, 1.5; mercaptobenzothiazole (MBT), 1.2; tetramethylthiuram disulfide (TMTD), 1.2; and zinc salt of dimethyl dithiocarbamate (ZDC), 2.0. Rubber chemicals were procured from the local market.

### Preparation of Ground Waste EPDM

Specimens (thickness, 2 mm) were molded from the EPDM compound at 150°C as per the cure time obtained from Monsanto rheometer model MDR-2000 (Monsanto, St. Louis, MO) and then aged at 100°C for 48 h. The mechanical properties of the EPDM vulcanizate before and after aging were determined as per ASTM standards, the results of which are given in Table I. Mechanical grinding of thick specimens (thickness, 12 mm), cured for an additional 10 min and then aged, was done at room temperature using a mechanical grinder (Ralliwolf TG-6 bench grinder, Mumbai, India) with a silicon carbide abrasive wheel rotating at 2950 rpm.

### Determination of Powder Density

The specific gravity of the ground rubber was determined as per ASTM D1817-96 with acetone as the noninteracting immersion liquid.

**Table I Mechanical Properties of EPDM Vulcanizate Used for Making Ground Waste EPDM (W-EPDM)**

Property	Before Aging	After Aging
Tensile strength, MPa	7.04	7.01
Elongation at break, %	430	260
Modulus at 100% elongation, MPa	2.18	3.31
Tension set (at 100% elongation), %	4	4
Tear strength, kN/m	33.0	31.5
Hardness, Shore A	55	64
Abrasion loss, cm <sup>3</sup> /h	2.3	— <sup>a</sup>
Hysteresis loss (W <sub>1</sub> ), (J/m <sup>2</sup> ) × 10 <sup>-6</sup>	0.032	0.046

<sup>a</sup> Not determined.

### Determination of Particle Size and Shape

The particle size and shape of the powdered rubber were determined by a Cam Scan Series 2 scanning electron microscope (Cambridge Scanning Co. Ltd., England) using gold-coated W-EPDM samples obtained as such from the grinder and after being dispersed ultrasonically over the mount. Typical aggregates of W-EPDM samples were also examined using an optical microscope Leitz Metallux-3 (Leitz Wetzlar, Germany), with a ×100 magnification.

### Mixing

The compositions selected for the study are given in Table II. Mixing was done on a two-roll mill (6 × 13 in.; Schwabenthan, Germany) as per the standard procedure.

### Determination of Mooney Viscosity and Scorch Time

Mooney viscosity of the compounds was determined using the Mooney viscometer (Negretti MK III; Negretti Automation Ltd., Buckinghamshire, UK) at 120°C as per ASTM D1646-96a. The time for a rise of 5 units from the minimum torque was taken as the scorch time at that temperature.

### Determination of Cure Characteristics

Cure characteristics of all the compositions were determined using Monsanto Moving Die Rheometer (MDR-2000) at 160°C.

**Table II Formulations Used**

Ingredient	Mix Number					
	W <sub>0</sub>	W <sub>20</sub>	W <sub>40</sub>	W <sub>50</sub>	W <sub>60</sub>	W <sub>100</sub>
R-EPDM <sup>a</sup>	100	100	100	100	100	100
W-EPDM <sup>b</sup>	0	20	40	50	60	100
ZnO	5.0	5.0	5.0	5.0	5.0	5.0
Stearic acid	1.0	1.0	1.0	1.0	1.0	1.0
TMTD <sup>c</sup>	1.0	1.0	1.0	1.0	1.0	1.0
MBT <sup>d</sup>	0.5	0.5	0.5	0.5	0.5	0.5
Sulfur	1.5	1.5	1.5	1.5	1.5	1.5

<sup>a</sup> Raw EPDM.

<sup>b</sup> Vulcanized EPDM powder that was aged (considered as waste EPDM rubber).

<sup>c</sup> Tetramethylthiuram disulfide.

<sup>d</sup> Mercaptobenzothiazole.

### Molding

Thin sheets of about 2-mm thickness were prepared from the compounds by compression molding at 160°C and 5 MPa pressure in a hydraulic press according to the optimum cure times obtained from MDR-2000.

### Determination of Mechanical Properties

Dumbbell-shaped and non-nicked-angle specimens were punched out, whereas abrasion testing samples and resilience samples were molded in standard molds. Tensile testing was done using Universal Testing Machine (Zwick Model 1445, Zwick GmbH & Co., Ulm, Germany) as per ASTM D412-98a at a crosshead speed of 500 mm/min. Tear strength was determined on the angle specimens (die C) using Zwick 1445 as per ASTM D624-98. Tension set at 100% elongation was determined on dumbbell-shaped samples as per ASTM D412-98a. Hardness was determined using a Shore A Durometer as per ASTM D2240-97. Hysteresis loss was determined on dumbbell-shaped samples by elongating up to a strain level of 100% using Zwick 1445 as per ASTM D412 (1980). Abrasion resistance was determined in a DuPont abrader (ASTM D394; DuPont, Wilmington, DE) and expressed as abrasion loss, which is the volume in cm<sup>3</sup> abraded from a test piece per hour. Heat buildup was determined using a Goodrich Flexometer as per ASTM D623-93. Rebound resilience of the vulcanizates was determined using Dunlop Tripsometer as per BS 903 Part 22 (1950).

### SEM Studies of Fractured Surfaces

SEM photomicrographs of tensile and tear fractured surfaces were taken using a JEOL JSM

5800 scanning electron microscope (JEOL, Peabody, MA). The samples were gold coated prior to examination under the microscope.

### Dynamic Mechanical Analysis

Dynamic moduli were determined using a Dynamic Mechanical Thermal Analyser (DMTA MK II; Polymer Laboratory, Loughborough, UK). The testing was done in bending mode at a frequency of 1 Hz from -80°C to 80°C at a heating rate of 2°C/min. The data were analyzed using a Compaq computer (version 1.2).

### Aging Studies

Tensile specimens were aged at 150°C for 72 h in an air-circulating aging oven and the tensile properties of the aged samples were determined as per ASTM D573 (1994).

### Swelling Studies

Samples of approximately 1 cm diameter and 0.25 cm thickness were punched out and allowed to swell in *n*-heptane. Swollen samples taken out after specific intervals were blotted with filter paper and weighed quickly. The apparent crosslink density was taken as 1/*Q*, where *Q* is defined as the grams of solvent per gram of rubber hydrocarbon.<sup>28</sup>

Swelling value (*Q*)

$$= \frac{\text{Swollen weight} - \text{dried weight}}{\text{Initial weight} \times \text{rubber hydrocarbon content/formula weight}} \quad (1)$$

Rubber hydrocarbon (RH) content includes RH both from R-EPDM and from W-EPDM present in the formulation.

**Table III Compositions Prepared for Studying the Effect of Curative Migration from W-EPDM to R-EPDM**

Ingredient	W <sub>50</sub>	W' <sub>50</sub>	T0	L0	TL0	WT0	WL0	WTL0
R-EPDM	100	100	100	100	100	100	100	100
W-EPDM	50	50	—	—	—	50	50	50
ZnO	5	—	—	5	—	—	5	—
Stearic acid	1	—	—	1	—	—	1	—
TMTD	1	—	1	—	—	1	—	—
MBT	0.5	—	0.5	—	—	0.5	—	—
Sulfur	1.5	—	1.5	1.5	1.5	1.5	1.5	1.5

### Rheometric Studies on Migration of Activators and Curatives Using Control Compounds

The effect of migration of additives was studied by preparing control compounds with and without additional activators, curatives, as well as with and without W-EPDM, as shown in Table III. The Mooney scorch time, curing characteristics, and physical properties were determined.

## RESULTS AND DISCUSSION

### Characterization of Ground EPDM

SEM and optical photomicrographs of the ambiently ground EPDM vulcanizate are shown in Figure 1. The ground rubber is porous, like aggregated chain structures [Fig. 1(a)], wherein the aggregates exist in clusters. The optical photomicrograph of a typical aggregate of W-EPDM is shown in Figure 1(b). Figure 1(c) is the SEM photomicrograph of a typical particle in an aggregate, which was broken by the ultrasonic dispersion technique. Particle size distribution is shown in Figure 2. Padovan et al.<sup>29</sup> previously studied the topology of wear particles and proposed a mechanism of aggregate formation during wear. Specific gravity of the powder was found to be 1.06.

### Mooney Viscosity and Scorch Time

It was observed that the Mooney viscosity increases with the addition of W-EPDM and the curve obtained by plotting the relative Mooney viscosity against the volume fraction of W-EPDM (Fig. 3) can be expressed by the following equation:

$$M_f = M_g(1 + 0.34c + 3.16c^2) \quad (2)$$

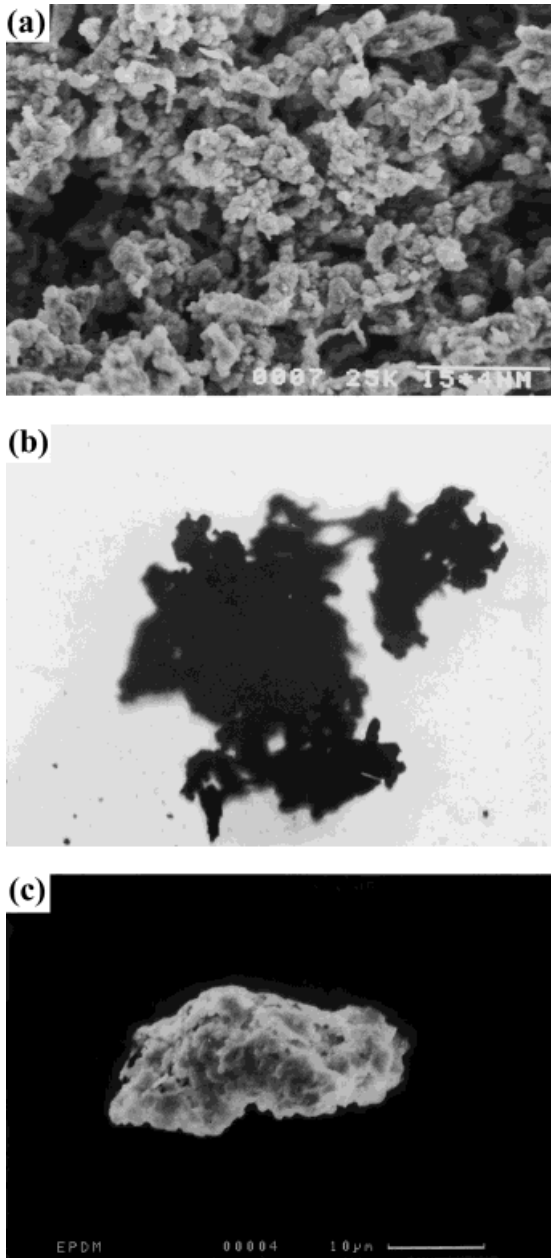
where  $M_f$  is the Mooney viscosity of compounds containing W-EPDM,  $M_g$  is the Mooney viscosity of the composition without W-EPDM, and  $c$  is the volume fraction of W-EPDM in the compositions.

It is believed that the highly aggregated and convoluted structure of W-EPDM powder contains void space into which the matrix rubber is trapped, thereby increasing the effective volume fraction of W-EPDM and hence the higher Mooney viscosity of W-EPDM-filled R-EPDM matrix. Ghosh et al.<sup>30</sup> also made similar observations while studying the effect of ground silicone vulcanizate on properties of silicone rubber compound. Gibala et al.<sup>23</sup> reported that the spongy structure of ambiently ground rubber is responsible for the higher Mooney viscosity of SBR compound than that containing cryogenically ground rubber powder.

The scorch time of the compositions decreases with increase in W-EPDM content (Table IV). This may be ascribed to the migration of accelerators from W-EPDM to R-EPDM. Phadke et al.<sup>24</sup> also observed a decrease in Mooney scorch time on incorporation of cryoground waste rubber into natural rubber compound. Gibala and Hamed<sup>22</sup> also observed decreased scorch time in SBR compounds on incorporation of powdered rubber vulcanizate, which was ascribed to the migration of accelerator from the ground vulcanizate to the matrix. The migration of curatives is discussed below.

Results of Monsanto rheometric studies are summarized in Table IV. The minimum torque continuously increases with increases in W-EPDM content. W-EPDM acts as a filler and the effect is similar to that observed in the case of Mooney viscosity measurements. The maximum torque increases up to 50 phr W-EPDM, beyond which it drops. The initial rise of torque may be



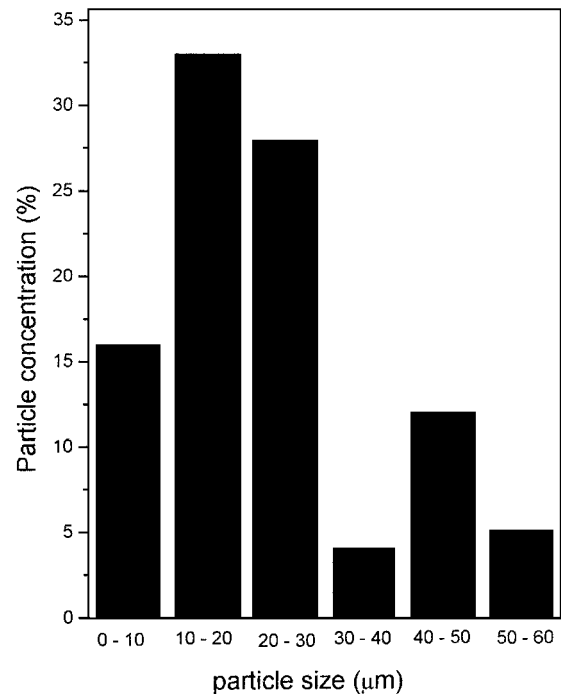


**Figure 1** (a) SEM photomicrographs of W-EPDM at low magnification showing clusters of aggregates. (b) Optical micrograph ( $\times 100$ ) of a typical aggregate. (c) SEM photomicrograph of a typical particle in an aggregate.

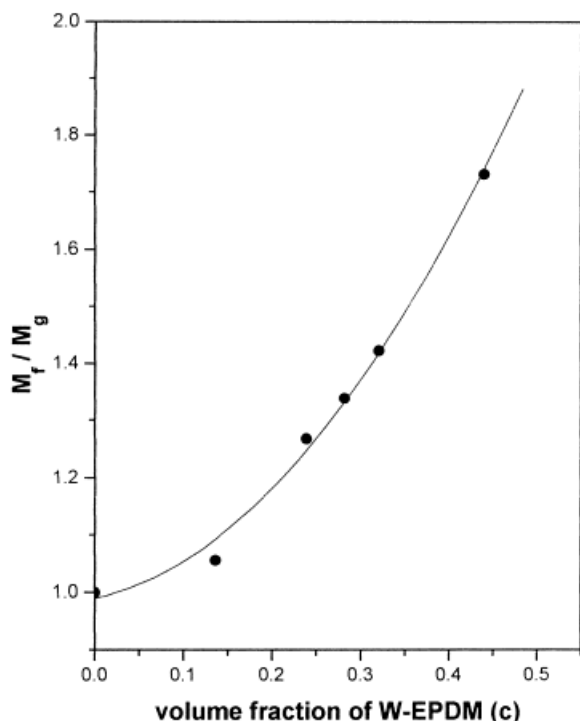
the result of greater interaction between the filler (W-EPDM) and the R-EPDM, whereas at higher concentrations the filler effect is overcome by the migration of sulfur from R-EPDM into the dispersed W-EPDM phase, thereby lowering the crosslinking efficiency of the matrix (R-EPDM). Phadke et al.<sup>19</sup> first reported the migration of

sulfur from the rubber-matrix phase (natural rubber compound) to the cryogenically ground waste rubber during curing, causing a drop in maximum rheometric torque. Cryogenically ground waste rubber was reported to contain a negligible amount of free sulfur. The rate of sulfur diffusion is proportional to the concentration gradient of free sulfur between the two phases.<sup>31</sup> Morita<sup>32</sup> described that diffusion of MBT into the matrix would reduce the scorch time and the migration of amine, a decomposition product of sulfenamides, may accelerate curing. Curative diffusion in blends of dissimilar elastomers was previously studied by Zapp.<sup>33</sup> Layer<sup>34</sup> studied recuring of vulcanizates by reheating cured networks swollen with sulfur or accelerator. It was suggested that accelerator fragments are not irreversibly bound to the network and might be available for subsequent crosslinking at a later stage.<sup>35</sup> Hamed et al.<sup>22</sup> also reported migration of sulfur from virgin rubber compound to waste rubber phase.

From Table IV it can be seen that the difference between the maximum and minimum rheometric torques ( $M_H - M_L = \Delta\text{torque}$ ) increases with W-EPDM loading up to 50 phr and then decreases. At loadings up to 50 phr, the  $\Delta\text{torque}$  change is similar to that observed in the case of reinforcing fillers. As shown in Table V, the apparent degree of crosslinking increases up to 40



**Figure 2** Particle size distribution of W-EPDM.



**Figure 3** Plot of relative Mooney viscosity against W-EPDM concentration at 120°C.

phr of W-EPDM and then gradually decreases at higher W-EPDM loadings. Results of solvent swelling studies are in conformity with the results of Monsanto rheometric studies. Below 50 phr of W-EPDM, the filler effect dominates over the effect of crosslinking and beyond 50 phr loading of W-EPDM, sulfur migration overshadows the filler effect and  $\Delta$ torque decreases.

### Mechanical Properties

Mechanical properties are summarized in Table V and the stress-strain properties are displayed in Figure 4. As the W-EPDM content increases, the

tensile strength, elongation at break, tear strength, and hysteresis increase, whereas the hardness and low-strain modulus remain almost constant. Whereas the increase in stress-strain properties, tear strength, and hysteresis can be ascribed to the filler effect, constancy in hardness and low-strain modulus are attributed to the interplay of opposing factors, that is, the filler effect being countered by the lowering of crosslink density. The abrasion resistance, however, is controlled more by the crosslinking factor and accordingly abrasion loss increases with increase in W-EPDM loading. Higher loading of plasticizer with increasing W-EPDM content may also contribute to the increasing abrasion loss. Dependence of mechanical properties on the interplay between fillers and network structure was previously reported.<sup>36</sup> Increase of carbon black content with increasing W-EPDM in the compound results in a decrease of rebound resilience and an increase of heat buildup resulting from the filler effect. SEM photomicrographs of tensile fracture surfaces are shown in Figure 5. It is evident that, in the absence of waste rubber powder, gum EPDM shows a large number of tear lines and the strength is poor.<sup>36</sup> At 20 phr loading of waste rubber the fracture mode does not change, but at 50 and 100 phr loadings the fractographs are similar to that of carbon black-filled rubbers.<sup>37</sup>

The observations on stress-strain behavior made here are in contrast to the results obtained by other investigators. Gibala<sup>17</sup> reported that ground rubber acted as a stress-raising flaw in tensile testing, while increasing the tear resistance. While studying the effect of incorporation of cryoground rubber powder in unfilled (gum) and carbon black-filled natural rubber compounds, it was observed by Phadke et al.<sup>38</sup> that the addition of cryoground rubber powder had a detrimental effect on most of the vulcanizate

**Table IV** Effect of W-EPDM on Curing Characteristics of EPDM Compound<sup>a</sup>

Curing Characteristic	Mix Number					
	W <sub>0</sub>	W <sub>20</sub>	W <sub>40</sub>	W <sub>50</sub>	W <sub>60</sub>	W <sub>100</sub>
Minimum torque ( $M_L$ ), dN · m	0.35	0.52	0.61	0.70	0.89	1.07
Maximum torque ( $M_H$ ), dN · m	9.39	10.25	10.38	10.51	10.16	9.73
$\Delta$ torque ( $M_H - M_L$ ), dN · m	9.04	9.73	9.77	9.81	9.27	8.66
Optimum cure time ( $t_{90}$ ), min	7.0	6.5	6.0	7.0	6.6	7.3
Mooney scorch time, min	33.0	18.0	14.0	13.0	13.0	10.0

<sup>a</sup> Rheometric torques at 160°C and Mooney scorch time at 120°C.

**Table V** Effects of W-EPDM on Mechanical Properties of EPDM Rubber Vulcanizate

Property	Mix Number <sup>a</sup>					
	W <sub>0</sub>	W <sub>20</sub>	W <sub>40</sub>	W <sub>50</sub>	W <sub>60</sub>	W <sub>100</sub>
Tensile strength, MPa	1.50 (1.27)	2.41 (2.10)	3.37 (3.40)	3.60 (3.37)	4.18 (4.17)	5.50 (5.43)
Elongation at break, %	157 (107)	231 (146)	295 (156)	305 (179)	337 (214)	415 (223)
Modulus at 100% elongation, MPa	1.16 (1.27)	1.23 (1.46)	1.26 (1.71)	1.31 (1.81)	1.32 (1.83)	1.33 (2.12)
Tear strength, kN/m	8.9	13.0	14.3	16.5	16.1	19.4
Tension set at 100% elongation, %	— <sup>b</sup>	2	2	2	2	2
Abrasion loss, cm <sup>3</sup> /h	0.35	0.46	0.42	0.57	0.50	0.78
Heat build up ( $\Delta T$ ) at 50°C, °C	1	4	4	4	5	6
Rebound resilience, %	75	74	71	70	69	67
Hysteresis loss (W <sub>1</sub> ), <sup>c</sup> (J/m <sup>2</sup> ) × 10 <sup>-6</sup>	0.003	0.004	0.006	0.006	0.006	0.008
Hardness, Shore A	45	45	46	46	46	45
1/Q <sup>d</sup>	0.437 (0.573)	0.448 (0.583)	0.449 (0.546)	0.436 (0.532)	0.425 (0.526)	0.417 (0.551)

<sup>a</sup> Values in parentheses are for aged samples.

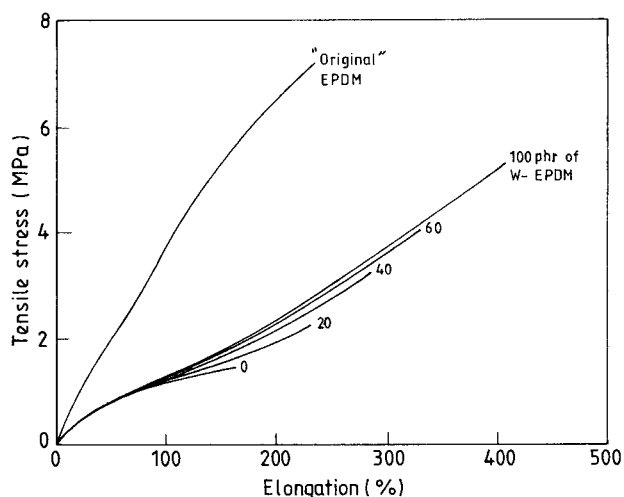
<sup>b</sup> Could not be determined; sample broke.

<sup>c</sup> First cycle.

<sup>d</sup> Determined by solvent swelling studies .

properties such as tensile strength, heat buildup, and abrasion. Tear strength, however, was not adversely affected.

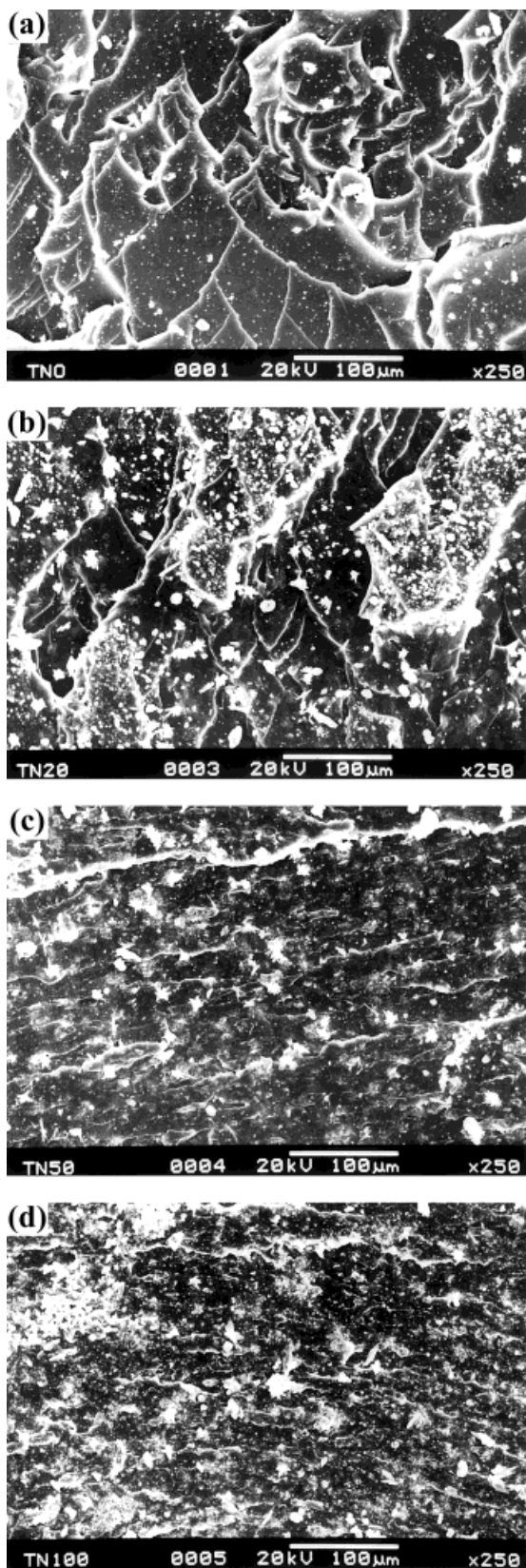
Burgoyne et al.<sup>8</sup> reported a 15% reduction in tensile strength for a compound containing 10% of



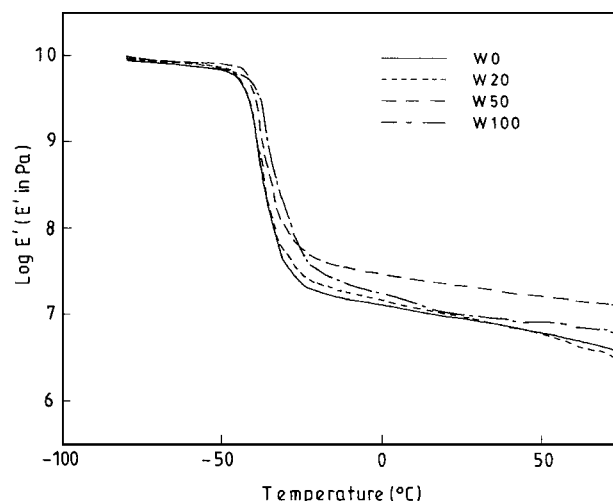
**Figure 4** Effect of W-EPDM loading on stress-strain behavior of EPDM vulcanizate. Stress-strain curve of “original” EPDM vulcanizate also shown.

ground vulcanizates (600–425  $\mu\text{m}$ ). Most of the earlier workers used ground rubber of large particle size. In the present investigation, particle size was smaller and it is believed that the highly aggregated structure consisting of fine particles breaks down to smaller units on milling, to produce uniformly distributed ground rubber particles in the matrix. Thus the chances of occurrence of stress-raising flaws in tensile test samples become diminished.

As seen in Figure 4, as the W-EPDM loading increases, the tensile stress continuously increases. The comparison between original carbon black-filled EPDM vulcanizate (before grinding) and the virgin EPDM vulcanizate containing W-EPDM shows that the incorporation of W-EPDM into virgin EPDM shows a reinforcing effect, although the strength is not as high as the “original” carbon black-filled vulcanizate. It may be noted that the original vulcanizate consists of 120 phr of carbon black, whereas in the case of W<sub>100</sub> (EPDM compound with 100 phr W-EPDM) the carbon black content is only 37 phr and it is proportionately less at lower loadings of W-EPDM. Accordingly, the tensile strength of W-



**Figure 5** SEM photomicrographs of tensile fracture surfaces: (a)  $W_0$ ; (b)  $W_{20}$ ; (c)  $W_{50}$ ; (d)  $W_{100}$ .



**Figure 6** Effect of W-EPDM loading on variation of storage modulus with temperature.

EPDM-filled EPDM is low when compared to that of the “original” vulcanizate before grinding.

#### Dynamic Mechanical Properties

The plots of storage modulus versus temperature are given in Figure 6, the results of which are summarized in Table VI. Storage modulus increases with the incorporation of W-EPDM up to a loading of 50 phr, beyond which (i.e., at 100 phr) it decreases. As discussed earlier, the initial increase in storage modulus is ascribed to the filler-reinforcement and the subsequent drop at 100 phr is ascribed to the lowering of crosslink density overshadowing the filler effect. Marginal increase in glass-to-rubber transition temperature ( $T_g$ ), obtained from  $\tan \delta$  versus temperature plots (not shown in the figure) at all loadings, is ascribed to the filler effect.

#### Aging Resistance

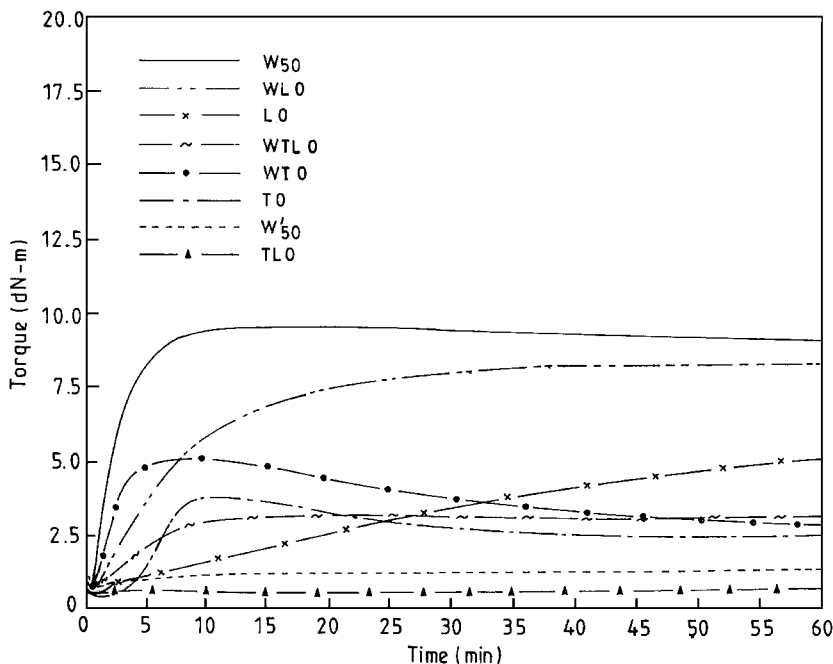
The aging resistance of the EPDM vulcanizate improves by the addition of W-EPDM. Results are

**Table VI** Results of Dynamic Mechanical Analyses

Property	Mix Number			
	$W_0$	$W_{20}$	$W_{50}$	$W_{100}$
$E'$ at 50°C, MPa	6.38	6.14	16.56	8.69
$T_g^a$ , °C	-35	-35	-34	-32

<sup>a</sup> Determined from  $\tan \delta$  versus temperature plots (not shown in the figure).





**Figure 7** Rheographs of control compounds for studying the accelerator migration from W-EPDM to R-EPDM using MDR-2000 at 160°C.

shown in Table V. It is apparent that a change in tensile strength on aging is minimal, whereas modulus increases and elongation at break decreases, which results from an increase in the apparent degree of crosslinking on aging, as shown under results of solvent swelling in Table V.

### Swelling Studies

The apparent crosslink density values ( $1/Q$ ) of the unaged and aged samples are given in Table V. The apparent crosslink density increases up to a loading of 40 phr of W-EPDM, beyond which the sulfur migration from R-EPDM to W-EPDM results in a gradual decrease of crosslink density. Results of solvent swelling studies are similar to the changes in torque in Monsanto rheographs. Aging causes an increase in apparent crosslink density in all formulations.

### Studies on Migration of Additives

To study the possible migration of additives from W-EPDM phase to R-EPDM matrix, Monsanto rheometric studies of different compositions (Table III) were undertaken.

Rheographs are shown in Figure 7 and curing characteristics are summarized in Table VII.  $W'_{50}$

corresponds to the composition  $W_{50}$  but without additional activators and curatives. It is apparent that in the absence of additional sulfur, W-EPDM alone cannot cure virgin (or raw) EPDM compound (R-EPDM). T0, L0, and TL0 are compositions without W-EPDM, whereas WT0, WL0, and WTL0 are the corresponding compositions containing W-EPDM. T0 and WT0 contain additional curatives (S, TMTD, and MBT), but no additional activators (ZnO and stearic acid). It is apparent that unutilized activators along with accelerator from W-EPDM take part in the crosslinking of R-EPDM, and scorch time drops from 37 min to 19 min on addition of W-EPDM along with a drop in optimum cure time and marginal improvement in physical properties. L0 and WL0 contain added activators and sulfur but no accelerator. It is evident that addition of W-EPDM causes a drastic increase in maximum rheometric torque and physical properties, along with a sharp drop in scorch time. The results are indicative of substantial migration of accelerators (TMTD and MBT) from W-EPDM to R-EPDM. TL0 and WTL0 contain only added sulfur but no accelerators and no activators. It is evident that additional activators are required for the completion of crosslinking reaction. It is concluded that accelerators migrate from the waste rubber to the matrix, but sulfur

**Table VII Curing Characteristics of the Compounds and Physical Properties of the Corresponding Vulcanizates<sup>a</sup>**

Property	W <sub>50</sub>	W' <sub>50</sub>	T0	L0	TL0	WT0	WL0	WTL0
Maximum torque ( $M_H$ ), dN · m	10.51	1.20	3.91	5.23	0.90	5.17	8.63	3.37
Minimum torque ( $M_L$ ), dN · m	0.70	0.72	0.41	0.47	0.47	0.71	0.73	0.81
Δtorque ( $M_H - M_L$ ), dN · m	9.81	0.48	3.50	4.76	0.43	4.46	7.90	2.56
Mooney scorch time ( $t_5$ ) at 120°C, min	13.0	— <sup>b</sup>	37.0	>60.0	— <sup>b</sup>	19.0	18.0	23.0
Optimum curing time at 160°C, min	7.0	—	7.3	49.5	—	4.4	23.5	12.3
Modulus at 100% elongation, MPa	1.31	—	—	0.82	—	0.84	1.12	0.71
Elongation at break, %	305	—	>1500 <sup>c</sup>	479	—	911	372	720
Tensile strength, MPa	3.60	—	>2.40	1.72	—	5.24	3.37	1.97
Tear strength, kN/m	16.5	—	10.7	11.4	—	17.5	17.6	11.1
1/Q <sup>d</sup>	0.377	—	0.126	0.255	—	0.229	0.342	0.124

<sup>a</sup> Refer to Table III.<sup>b</sup> Did not cure.<sup>c</sup> Did not break at 1500%.<sup>d</sup> Apparent crosslink density determined by solvent swelling.<sup>28</sup>

and activators are to be incorporated from outside for the completion of the curing reaction, in the compound containing both waste EPDM (W-EPDM) and raw EPDM (R-EPDM). Hamed et al.<sup>22</sup> also observed migration of accelerator from the waste rubber phase to fresh rubber compound.

## CONCLUSIONS

1. Ambiently ground EPDM vulcanizate (W-EPDM) acts as a filler in raw EPDM (R-EPDM) and its addition results in an increase of Mooney viscosity, but a decrease of scorch time, presumably as a result of the accelerator migration from W-EPDM to the matrix (R-EPDM).
2. At higher loadings of vulcanizate powder, the maximum rheometric torque decreases, which can be attributed to the sulfur migration from the matrix (R-EPDM) to W-EPDM, causing a decrease in apparent crosslink density of the matrix.
3. The improvement in tensile strength, tear strength, and elongation at break indicates the reinforcing nature of the carbon black-containing W-EPDM powder. In the case of low-strain modulus, the filler effect is coun-

terbalanced by the crosslinking effect. The marginal increase in heat buildup and drop in resilience are attributed to the filler effect.

4. Studies on processing and curing behavior and measurements of physical properties of control formulations of varying curative concentrations reveal that accelerators migrate from the W-EPDM to the matrix (R-EPDM).

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