INVESTIGATION OF MECHANICAL, THERMAL, AND MORPHOLOGICAL PROPERTIES OF EPDM COMPOUNDS CONTAINING RECLAIMED RUBBER

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ABSTRACT

The disposal of polymeric and hazardous materials is an important global issue. Thus, the mechanochemical reclaiming of waste EPDM rubber was carried out using disulfide oil (DSO), oily waste produced in gas refineries, as a reclaiming agent at a specific operation condition. Reclaimed rubber was characterized by crosslink density, sol-gel, and Mooney viscosity measurements. Following the study, blending of different portions of reclaimed rubber (20, 40, 60, and 80 wt%) with virgin EPDM rubber was carried out. Results showed that the addition of 60 wt% of reclaimed rubber to the virgin EPDM rubber had no adverse effect on the scorch and optimum curing time. Also, mechanical properties of revulcanized rubber improved when 40 wt% of virgin EPDM rubber was replaced with the reclaimed rubber. Scanning electron microscope and thermal gravimetric analyses of the samples revealed the effect of addition of reclaimed rubber on the rubber matrix and thermal stability of revulcanized samples, respectively. This study proposes a new route for solving the problem of disposal of the waste materials. [doi:10.5254/rct.18.81651]

INTRODUCTION

Ethylene–propylene–diene rubber (EPDM) is widely used in the automobile industry, adding a large volume of waste rubber to the environment each year. To save raw materials and resolve pollution problems, the recycling of the polymeric material appears so important. Rubber can be recycled in different ways, including rubber incineration and pyrolysis,[1–3] application of ground rubber powder in civil engineering,[4,5] grinding of the vulcanized materials and using as filler in rubber and plastics,[6–11], and so forth. However, devulcanization is the best way for reusing of waste rubber, which broadens the possibilities for utilization of the recycled material. Devulcanization is defined as a process of the scission of intermolecular bonds (C–S and/or S–S). Sometimes, a combination of crosslink scission and main-chain scission of the constitutive polymer chains, caused by heat, shear, and chemical reactions, has been defined as reclaiming.[12] Different methods of devulcanization have been reported, including physical reclaiming by means of a two-roll mill[13–15]; microwave[16,17]; ultrasonic,[18,19] catalyst,[20] mechanochemical,[21–24] thermomechanical,[25,26] and biotechnological[27,28] methods; devulcanization in supercritical carbon dioxide,[29,30] and the chemical reclaiming process.[31–33] The extrusion process for reclaiming of natural and synthetic rubber has been reported in many articles.[34–40] Organic disulfides or mercaptans are generally used in many industries as a reclaiming agent. These disulfides include diphenyl disulfide, dibenzyldisulfide, diamyl disulfide, bis (alkoxy aryl) disulfides, butyl mercaptan, phenol sulfides, and disulfides.[2] Jalilvand et al.[41] produced devulcanized EPDM using diphenyl disulfide as a devulcanizing agent in a laboratory intermeshing co-rotating twin-screw extruder. According to their results, shear stress is an important factor for increasing the percentage of devulcanization, and the temperature is the most important factor in determining sol fraction.[41] Jana et al.[42] used diallyl

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TABLE I
THERMOGRAVIMETRIC ANALYSIS TEST RESULTS FOR EPDM WASTE RUBBER

Property	Amount, wt%
Oil	5
EPDM	79
Carbon black and other fillers	16

disulfide and diphenyl disulfide as devulcanizing agents for mechanochemical devulcanization of NR. The most interesting observations is that the mechanical properties of revulcanized rubber obtained by devulcanizing in the presence of disulfide were better than those of the revulcanized rubber obtained by devulcanizing in absence of disulfide.[42,43] Shi and coworkers[44] compared the reclamation of ground tire rubber by diphenyl disulfide as a devulcanizing agent using four different methods. Finally, they recommended a reclaiming method with an oxygen-free medium, without high shear force, and at relatively low temperature.[44]

Disulfide oil (DSO) is an odorous and flammable compound produced as a by-product in gas refineries. DSO is generally rich in alkyl disulfides with small chain lengths, but the exact chemical composition can vary depending on types of sulfur contaminants in the treated feedstock. A constant challenge of gas refineries is the conservation and safe disposal of DSO because of its excessive sulfur. Even burning of the chemical due to high pollution is not a reasonable solution.[45] The authors' previous work showed that the DSO is an effective reclaiming agent.[46] It was confirmed that the blending of 40 wt% of reclaimed rubber (RR) produced by this method with the virgin EPDM not only had no adverse effect on the mechanical properties of the final product but also improved some mechanical properties, regardless of the carbon black in the waste rubber.

The main objective of this study is the use of DSO to reclaim EPDM waste rubber, blending of the RR with virgin EPDM rubber in different proportions other than our previous work, and investigation of mechanical properties, morphology, and thermal stability of the revulcanized samples.

EXPERIMENTAL

MATERIALS

Waste EPDM rubber obtained from Part Lastic Group Co. (Mashhad, Iran) had an average particle size of less than 1 mm and was characterized using thermogravimetric analysis (TGA-50, Shimadzu, Japan). The composition is given in Table I.[47,48] Virgin EPDM (KEP 270, Kumho Polychem, Korea), carbon black (N330, Pars Co., Saveh, Iran), process oil (oil 840, Behran Co., Tehran, Iran), ZnO (Pars Rangineh Co., Tehran, Iran), stearic acid (Uni Chemical, Malaysia), antioxidant (NA 4010), 2-2′-Dithiobis(benzothiazole) (MBTS), Tetramethylthiuram Disulfide (TMTD), zinc dibutyldithiocarbamate (Perkacit ZDBC) and elemental sulfur from Bayer Co. (Germany) were used as such. DSO was supplied by South Pars Oil and Gas Co. (Iran) and was used as a reclaiming agent. Toluene and acetone were supplied by Shazand Petrochemical Co. (Arak, Iran).

RECLAIMING WASTE EPDM RUBBER

Waste EPDM rubber was mixed with an appropriate amount of process oil (5 phr) and DSO (7 phr) and reclaimed using a laboratory intermeshing co-rotating twin-screw extruder (model TSE 20, Brabender Co., Germany) at the specific temperature (290 °C). The extruder had five heating/cooling zones and a screw diameter of 20 mm with a length/diameter ratio of 40. The feed rate and screw speed (120 rpm) were kept constant in our study. Finally, the reclaimed samples were rolled by using a laboratory two-roll mill.

CHARACTERIZATION

Swelling Test. — The swelling test method was used to determine the sol fraction, crosslink density (CLD), and the percentage of devulcanization of the reclaimed samples in accordance with ASTM D 6814-02 (2002). Based on this standard, a small amount of specimen (5 g) was placed in the soxhlet apparatus for 16 h to remove low-molecular-weight substances such as processing oil. After that, the sample was dried (at 80 °C), weighed (m1), and swollen by means of toluene (400 mL) for 72 h at room temperature. Toluene was replaced every 24 h to reach equilibrium swelling. Then, the specimens were taken out from the toluene, weighed (ms), and dried to a constant weight at 80 °C (m2).

The sol fraction, an indication of the rupture of the rubber network during the process,[40] was calculated according to Eq. 1:

$$\text{Sol fraction}\,(\%) = \frac{m_1 - m_2}{m_1} \times 100 \tag{1}$$

The CLD of the gel part of the samples was determined based on the Flory–Rehner equation (Eq. 2).[49] The interaction parameter (χ) for the toluene-EPDM system was determined using Eq. 3.[50]

$$V_e = \frac{-\left[\ln(1 - V_r) + V_r + \chi V_r^2\right]}{\left[V_l(V_r^{1/3} - V_r/2)\right]} \tag{2}$$

$$\chi = 0.429 + 0.218\,V_r \tag{3}$$

where V_e, V_r, and V_1 are the crosslink density, rubber volume fraction in the swollen sample, and molar volume of the solvent, respectively.

Because of the presence of filler in the waste EPDM, V_r is modified according to Eq. 4.[51]

$$V_r = \frac{1}{1 + \frac{d_r}{d_s}\left(\frac{1-f_{sol}}{1-f_{sol}-f_{fill}}\right)\left(\frac{m_s}{m_2} - 1\right)} \tag{4}$$

where d_r and d_s are the densities of the rubber and solvent, respectively. f_{sol} is the sol fraction of the initial sample, f_{fill} is the initial weight fraction of the filler in the sample, w_s is the weight of the swollen gel. and w_0 is the weight of the dried sample. The density of the rubber was determined using the digital densitometer Precisa xr-405.

The degree of devulcanization was calculated according to Eq. 5:

$$\text{Devulcanization}\,(\%) = \frac{V_{e2} - V_{e1}}{V_{e1}} \times 100 \tag{5}$$

where V_{e1} and V_{e2} are the crosslink density before and after the devulcanization, respectively.

Horikx[52] developed a theory for further understanding the reaction taking place during the reclamation process. According to this theory, two limited cases were tracked: (1) when main chain scission occurs, a relative decrease in crosslink density is calculated using Eq. 6:

$$1 - \frac{V_f}{V_i} = 1 - \frac{(1 - S_2^{1/2})^2}{(1 - S_1^{1/2})^2} \tag{6}$$

where V_i and V_f are the crosslink density before and after the devulcanization. S_1 and S_2 are the sol

TABLE II
REVULCANIZATION FORMULATIONS

Ingredient	A[a]	B	C	D	E
EPDM	100	80	60	40	20
RR	—	20	40	60	80
N330	40	40	40	40	40
Oil 840	5	5	5	5	5
NA 4010	1	1	1	1	1
ZnO	4	4	4	4	4
Stearic acid	1.5	1.5	1.5	1.5	1.5
MBTS	0.8	0.8	0.8	0.8	0.8
TMTD	0.3	0.3	0.3	0.3	0.3
ZDBC	0.3	0.3	0.3	0.3	0.3
S	1.8	1.8	1.8	1.8	1.8

[a] Reference compound.

fractions before and after the devulcanization, respectively. (2) When crosslink scission occurs, the relative decrease in crosslink density is calculated using Eq. 7:

$$1-\frac{V_f}{V_i}=1-\frac{\gamma_f(1-S_2^{1/2})^2}{\gamma_i(1-S_1^{1/2})^2} \tag{7}$$

where the parameters $\gamma_f$and γ_i are the crosslink index and refer the average number of crosslinks per chain in the residual gel before and after the devulcanization, respectively. Horikx's theory is restricted in two mentioned cases and cannot explain the situations in which there is a combination of both main chain and crosslink scission.[53]

Mooney Viscosity. — Mooney viscosity of the RR was determined using a Mooney MV2000 apparatus from Alpha Technologies according to ISO 289 (2005; ML 121 °C [1 + 4] min).

Curing Behavior and Compounding. — The RR was blended with the virgin EPDM rubber at different rations (20, 40, 60, and 80 wt%), and then the blends were compounded with the ingredients according to Table II on the laboratory mill for about 15 min to obtain homogeneous EPDM compounds.

The cure characteristics of the RR were measured at 160 °C for up to 20 min (ISO 3417, 2008) by means of an oscillating disk rheometer curemeter (ODR Gotech testing machines, model GT-M2000-F, Taiwan).

The cure rate index (CRI) was calculated via Eq. 8:

$$CRI=\frac{100}{t_{90}-t_{s2}} \tag{8}$$

The resulting compounds were then cured at 160 °C for the optimum curing time of t_{90} by compression molding at a pressure of 20 MPa.

Mechanical Properties. — The tensile strength, elongation at break, and modulus at different elongations of the samples were measured using a tensile testing machine (model 6025, Instron, High Wycombe, UK) according to ASTM D412-98a (2002) at room temperature. For each sample, the average of three replicates measurements is reported.

TABLE III
CHARACTERIZATION OF THE RECLAIMED RUBBER

Sample No.	Density, g/cm^3	CLD × 10^{-3}, mol/cm^3	Devulcanization, %	Sol fraction, %	Mooney viscosity
Reference[a]	1.055	0.2999	0	2.0	—
RR	1.160	0.0814	73	20.0	37

[a] EPDM waste rubber.

The Shore A hardness was determined based on ASTM D 2240-02 (2002) with a Zwick durometer hardness tester. The value of the hardness is the average of three different measured points on the same sample.

The tear strength of the rubber vulcanizate was measured using a Hiwa Materials Testing-200 (Tehran, Iran) in accordance with ASTM 624 (2001).

The cylindrical samples (12.5 mm thick and 29.0 mm in diameter) were used to determine the compression set of the revulcanized samples (cured at 180 °C for 13 min). The test was performed at 70 °C for 24 h using a compression set testing apparatus (Taha Ghaleb Toos Co., Mashhad, Iran) with accordance to ASTM D395-03 (2003). Finally, the compression set was calculated using the following expression (Eq. 9):

$$\text{Compression set} = \frac{\text{Initial thickness} - \text{final thickness}}{\text{Initial thickness}} \times 100 \tag{9}$$

The resilience was measured using cylindrical samples, 12 mm thick and 50 mm in diameter. The samples were placed in a Dunlop Tripsometer (Zwick, Ulm, Germany) according to the test method described by ASTM D2632–01 (2008) at room temperature. This was repeated at three different positions on the same sample, and the median of the three readings was reported.

Morphology of RR/EPDM Blends. — The physical properties of the polymer blends are always affected by the resulting morphology of the blends.[54,55] The scanning electron micrographs (SEMs) of the cryogenically fractured surfaces of compression-molded specimens of different RR/EPDM were observed by a LEO 1450 VP SEM (Zeiss, Oberkochen, Germany).

Thermal Stability. — The measurements of the sample weight loss were carried out on a TGA-50 thermogravimeter (Shimadzu, Kyoto, Japan). All of the samples used in the thermogravimetric measurements were similar in weight (4.5 ± 0.1 mg) and heated from room temperature to 800 °C at a heating rate of 10 °C/ min.

RESULTS AND DISCUSSION

CHARACTERIZATION OF RR (SWELLING TEST AND MOONEY VISCOSITY)

The swelling test results and the viscosity of the RR are shown in Table III.

The results show that the DSO is an effective devulcanizing agent in mechanochemical reclaiming of EPDM waste rubber (73% decrease in crosslink density).

The Mooney viscosity value was obtained at 37 MU, which is a measure of flowability and processability of elastomeric compounds. EPDM RR with Mooney viscosity between 45 and 70 MU is suitable to be blended with EPDM virgin rubber, as mentioned in the literature.[56]

The sol fractions of the devulcanized rubber versus decrease in crosslink density are shown in Figure 1 as predicted by Horikx's theory. The obtained experimental point is located between the two curves (solid and dash lines); that is, the reaction of the devulcanization is proceeded via both crosslink scission and main chain scission.

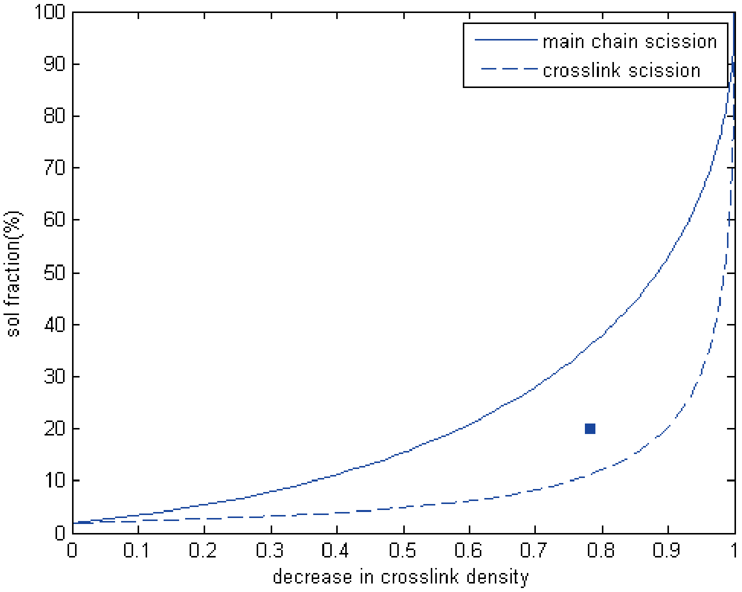


FIG. 1. — Sol fraction of reclaimed rubber versus relative decrease in crosslink density. These two curves are plotted according to Eqs. 6 and 7, in which is S_1 was determined with the value of 2% by swelling test and γ_i was calculated according to the equation $S_i = \frac{1}{(1+\gamma_i-\gamma_i S_i)^2}$.[58]

CURE CHARACTERISTICS

The cure characteristics are given in Table IV.

The scorch time and induction period for compounds B, C, and D do not change with increasing RR content up to 60 wt%, indicating that RR participates in the crosslinking reactions as readily as EPDM does. However, the increasing RR content from 60 wt% to 80 wt% resulted in a decrease of the scorch time. This may be due to the presence of the active crosslinks in the RR.

The optimum curing times for the compounds containing RR are lower than that of the reference compound. This observation confirms that the increasing RR content accelerates the rate of the cure, which is in agreement with the CRI values.

The T_{min} value is a measure of the processability of rubber samples and an indication of the stock viscosity increase with increasing RR content (up to 80 wt%, which was studied).

TABLE IV
CURING PROPERTIES OF THE RUBBER COMPOUNDS

Compound No.	Scorch time (ts_2), m:s	Cure time (t_{90}), m:s	T_{min}, dN m	T_{max}, dN m	$\Delta torque$, dN m	CRI, s^{-1}
A	1:09	7:33	11.653	49.225	37.572	0.260
B	1:07	6:15	13.336	46.035	32.699	0.325
C	1:08	5:43	15.642	41.551	25.909	0.364
D	1:00	5:42	21.045	46.611	25.566	0.355
E	0:53	6:19	26.733	48.827	22.094	0.307

REVULCANIZATE PROPERTIES OF RR/VIRGIN EPDM BLENDS

The mechanical properties of the revulcanized rubber with different amounts of RR are shown in Table V.

It can be seen from Table V that the progressive increase in RR content in the blends had a different effect on various mechanical properties. With an increasing RR content up to 40 wt%, the tensile strength increased (14% increase), which lowered on further increase of RR content in the blend. No obvious trend was seen in the case of elongation at break.

The modulus of the revulcanized samples is higher than that of the original sample. The revulcanized rubber contains a certain amount of main chain degradation, which restricted the molecular chain mobility under tension force and hence increased its modulus.[43] This suggests that the presence of a small amount of devulcanized rubber in the blend was beneficial to the modulus of the final rubber vulcanizate.

The hardness of all of the revulcanized samples is higher than the original sample, possibly because of the presence of filler in the RR.

The tear strength of the revulcanized samples containing 20 and 40 wt% of RR is almost near the reference. However, by increasing the RR content up to 80 wt%, the tear strength declined up to 50%.

The addition of the RR content caused an increase in compression set and decrease in resilience gradually.

MORPHOLOGY STUDY

Figure 2a–e shows the micrographs of the cryogenically fractured surface of the compression-molded specimens (A, B, C, D, and E), all at a magnification of 5000×. The full dispersion of the filler particles in the rubber helps to maximize the reinforcing effect of the filler on the mechanical properties of the cured rubbers.[57] Based on Figure 2a for the reference compound, the sample has a rough and homogenous surface with no hole, suggesting that the strong interaction between the sol part and loosely cosslinked gel part occurred during the revulcanization, resulting in high tensile strength and elongation at break. However, by increasing the RR content, the number of crack paths in different directions increases, the surface will be smoother, and some holes appear. Thus, in these samples, detachment of unrevulcanized gel particles from the EPDM matrix resulted in lower crosslink density (in accordance to $\Delta torque$ data) and deterioration of tensile properties (Table V).

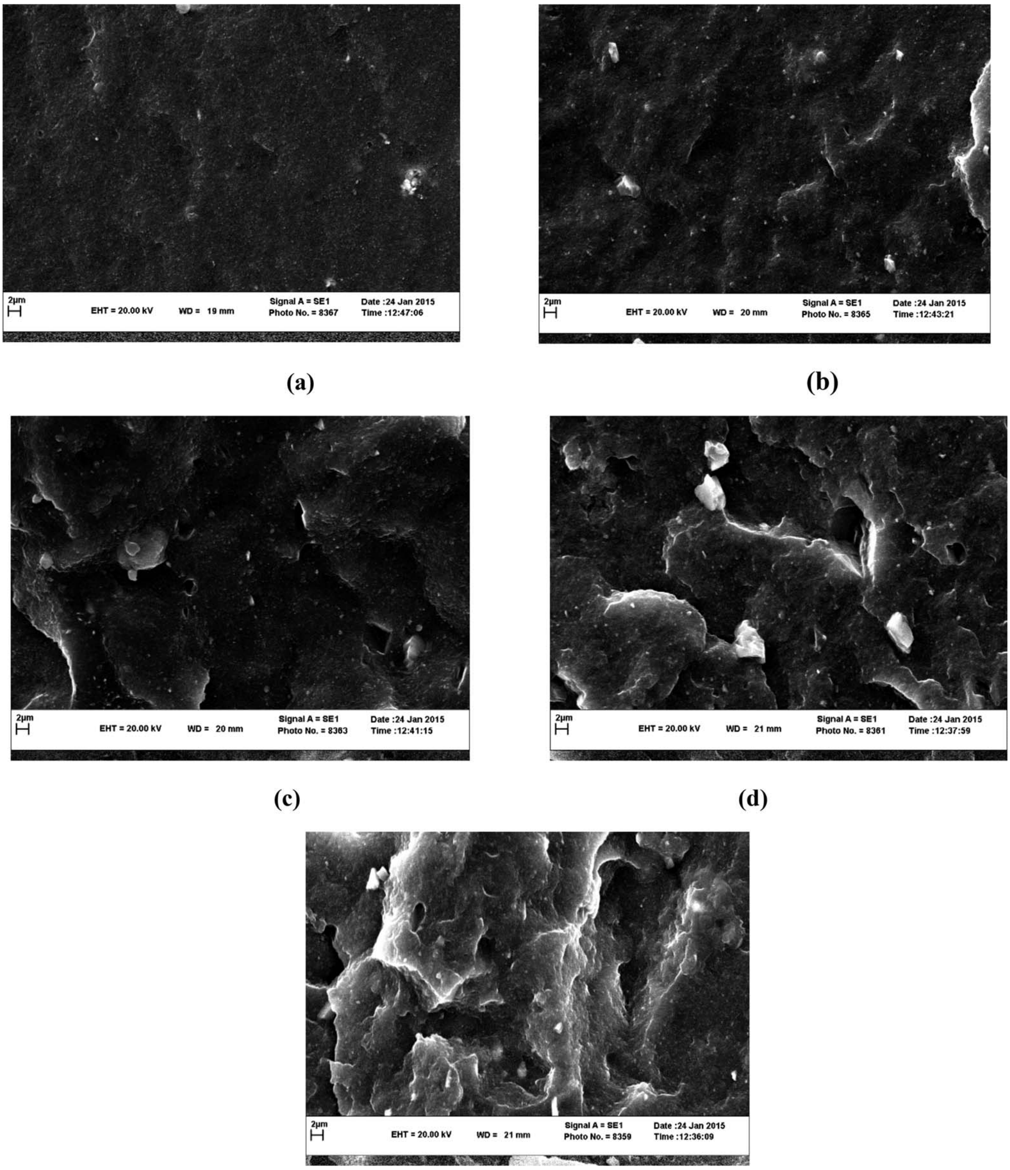
It is noted that the increased value of some mechanical properties compared with the reference is probably due to the dominant reinforcing effect of carbon black in these blends (B and C). This idea was proved in our previous study.[46] However, further increasing the RR may dominate the reclaimed phase over the virgin rubber (D and E).

THERMAL ANALYSIS

The TGA curves of the reference and revulcanized samples are shown in Figure 3. The TGA curves for all vulcanizates show three-step degradation. For compounds A and B, more similar steps were observed. The onset degradation temperature of the samples lowered with an increase in the RR content in the blend. The temperature of the other steps also showed a similar trend. Based on the values of $\Delta torque$ (Table IV), the degree of crosslinking decreases with increasing RR content, resulting in a decrease in the rigidity of the blends and consequent decrease in thermal stability.

TABLE V
MECHANICAL PROPERTIES OF THE RUBBER COMPOUNDS

Compound No.	Tensile strength, MPa	Elongation at break, %	Hardness, Shore A	Modulus at 100%, MPa	Modulus at 200%, MPa	Tear strength, N/mm	Compression set, %	Resilience, %
A	14.3	268	68	3.2	9.1	38.1	13	60
B	14.6	228	68	4.6	12.3	38.8	31	55
C	16.3	244	72	5.8	13.4	40.3	46	44
D	11.3	259	79	4.4	9.2	27.9	50	33
E	7.6	163	85	5.4	—	18.1	53	23



(a) **(b)**

(c) **(d)**

FIG. 2. — SEM images of the cryogenically fractured surface of the compression-molded specimens. (a) Reference compound. (b) RR/EPDM (20/80). (c) RR/EPDM (40/60). (d) RR/EPDM (60/40) and RR/EPDM (80/20).

CONCLUSIONS

The mechanochemical devulcanization of waste EPDM rubber was carried out using a twin-screw extruder with the aid of DSO, an oily waste from gas refineries. The virgin EPDM rubber was blended with different amounts of RR (20, 40, 60, and 80 wt%). Characterization of RR, 73% decrease in CLD, sol fraction of 20%, and the value of 37 MU for Mooney viscosity showed the successful devulcanization of waste EPDM rubber by this method. The curing behavior of the

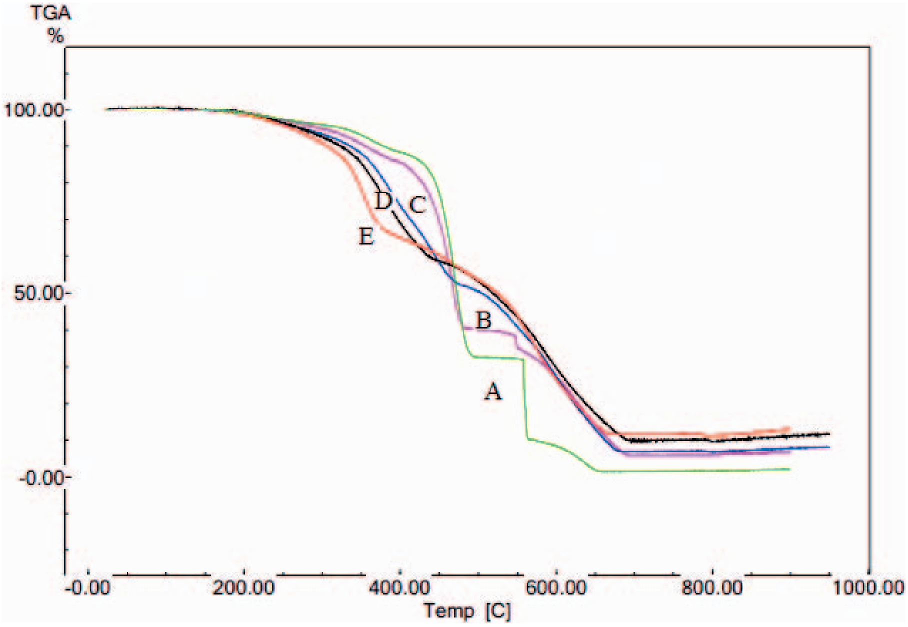


FIG. 3. — TGA curves for vulcanizate rubber.

rubber compounds showed that the addition of 60 wt% of RR had no adverse effect on the scorch and optimum curing time; likewise, the CLD (indicated by Δ*torque*) was decreased and the rate of cure was increased by the increase of RR content. Replacing 40 wt% of the virgin EPDM with RR had no adverse effect on mechanical properties. The increased value of some mechanical properties compared with the reference is probably due to the dominant reinforcing effect of carbon black in these blends (B and C); however, further increasing of RR may dominate the reclaimed phase over the virgin rubber (D and E). SEM images showed that increasing the RR content decreases the compatibility between the RR and virgin EPDM, resulting in degradation of mechanical properties. TGA studies showed that increasing the RR content decreases the onset degradation temperature, indicating a drop in thermal stability.

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