



Applicability of Thermomechanically Devulcanized EPDM in Peroxide-Crosslinked EPDM Composites: Effect of Recycled EPDM

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Abstract: Recycling of ethylene propylene diene monomer (EPDM) rubber has attracted increasing attention as an effective strategy to reduce rubber waste while maintaining material performance. In this study, waste EPDM was devulcanized through a thermomechanical process and incorporated into peroxide-crosslinked EPDM (PO-EPDM) composites to evaluate its applicability as a recycled rubber component. The recycled EPDM (rEPDM) content was varied from 5 to 25 phr, and its effects on devulcanization behavior, revulcanization characteristics, mechanical properties, compression set, and morphology were systematically investigated. Horikx analysis confirmed that the optimized thermomechanical process preferentially promoted sulfur crosslink scission while minimizing polymer main-chain degradation. As the rEPDM content increased, the minimum and maximum cure torques gradually decreased, indicating improved processability but reduced crosslink density after revulcanization. Tensile strength, tear strength, and compression set remained comparable to those of the virgin compound when the rEPDM content was 15 phr or lower. However, at 20 phr and above, significant deterioration in mechanical performance and elastic recovery was observed due to reduced interfacial adhesion, particle agglomeration, and heterogeneous crosslink structures, as confirmed by scanning electron microscopy. These results demonstrate that devulcanized rEPDM can be effectively incorporated into PO-EPDM without significant performance loss up to 15 phr, providing a practical guideline for manufacturing high-performance recycled EPDM materials with improved sustainability.

Keywords: Thermomechanical devulcanization, Recycled EPDM, Peroxide crosslinking, Revulcanization kinetics, Selective crosslink scission

Introduction

Ethylene propylene diene monomer (EPDM) rubber is a synthetic rubber prepared by incorporating a small amount of diene monomer into a copolymer of ethylene and propylene, enabling sulfur vulcanization through the diene component. The main chain of EPDM consists predominantly of saturated hydrocarbon structures, which confer excellent chemical stability.¹⁻³ In addition, EPDM exhibits superior resistance to ultraviolet radiation and relatively high thermal aging resistance. Owing to these structural characteristics, EPDM is widely used in applications that require long-term reliability under outdoor exposure conditions.^{4,5} Because

EPDM is extensively applied in various industries, including automotive parts, construction waterproofing materials, and electrical insulation components, the volume of EPDM waste has continuously increased.^{6,7} Traditionally, end-of-life EPDM has been disposed of through landfilling or incineration; however, efficient recycling technologies are being actively investigated.^{8,9} Rather than utilizing recycled EPDM simply as an inert filler, a revulcanization strategy that selectively breaks crosslinks and re-integrates the material into the matrix network is required. Devulcanization selectively cleaves crosslink structures in waste rubber and restores reprocessability, but polymer-chain degradation may occur concurrently and reduce mechanical performance. Exposure to air at high temperatures can accelerate oxidative degradation, and excessive thermal input can induce random polymer-chain

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scission through radical reactions.¹⁰⁻¹³ Mechanical processing combined with shear can promote selective cleavage of sulfur crosslinks. Piriti et al. investigated recycling of vulcanized EPDM using both continuous and batch processes and reported a substantial reduction in crosslink density after devulcanization.¹⁴ Esser et al. examined blending liquid polymer (LP) with devulcanized EPDM and found that LP improved processability in the unvulcanized state.¹⁵ Dalina et al. applied microwave irradiation to EPDM waste for devulcanization.¹⁶ These studies indicate that restoring the properties of virgin EPDM remains challenging. Several studies have shown that peroxide-based crosslinking can promote formation of a new crosslinked network in EPDM through radical reactions.¹⁷⁻²² Because the reactivity and dispersion state of recycled EPDM can vary, it is necessary to systematically examine the effects of recycled content on curing behavior, mechanical performance, compression set, and morphology. In this study, EPDM waste was recycled via a batch-type thermomechanical process and blended with polypropylene (PP) to prepare peroxide-crosslinked EPDM (PO-EPDM) materials. The recycled EPDM (rEPDM) content was varied from 5 to 25 phr, and its effects on devulcanization behavior, revulcanization characteristics, mechanical properties, compression set, and morphology were investigated. The experimental results are expected to provide a practical basis for selecting an appropriate rEPDM content for PO-EPDM materials.

Experimental

1. Experimental materials and manufacturing methods

The EPDM waste used in this study was supplied by Dong-A Hwaseong Co., Ltd. The manufacturing conditions of the EPDM prior to disposal are presented in Table 1.

The EPDM waste was washed and dried to remove surface contaminants and foreign substances before being used in the process. The EPDM waste was then devulcanized using a batch-type thermomechanical method to restore its reprocessability.

First, the EPDM waste was pulverized into particles of 60 ± 5 mesh using a high-temperature shear pulverizer (2PLS600, Duksan Co., Ltd., Korea). For reproducibility, the pulverizing unit was described as a single rotating shaft equipped with three rows of serrated rotary cutters arranged in a staggered configuration along the shaft, with stationary

Table 1. Manufacturing Conditions of EPDM Prior to Disposal

Component	Amount (phr)
NVE TER 4049*	100
Calcium carbonate	25
N550 carbon black	50
N774 carbon black	30
Paraffinic process oil	20
Polyethylene glycol	1
Zinc oxide	5
Stearic acid	1
Zinc stearate	1
TMTD**	0.6
MBT***	1.2
ZDBC****	0.7
Sulfur	1.2

*Ter 4049 : Ethylene : 55.5, Propylene : 40, ENB : 4.5, Mooney viscosity(-) : 76.

**TMTD : Tetramethylthiuram disulfide.

***MBT : 2-Mercaptobenzothiazole.

****ZDBC : Zinc dibutyldithiocarbamate.

counter-knives mounted on the inner wall of the pulverizing chamber. The nominal clearance between the rotary cutters and stationary counter-knives was set to approximately 0.3 ± 0.1 mm. The rotation speed of the pulverizer was set to 2,000 rpm, and 50 g of EPDM waste was fed every 5 s. The resulting material was subsequently sieved, and only particles passing through a 60-mesh sieve were collected for thermomechanical devulcanization.

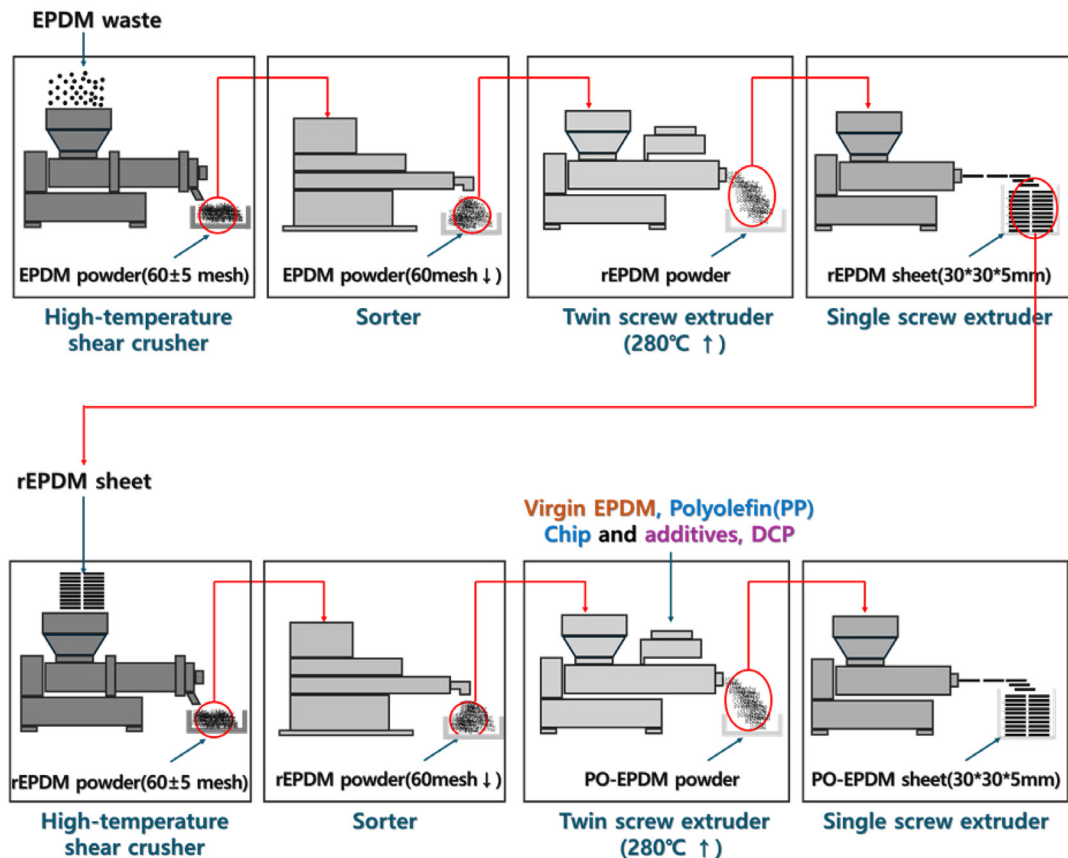
The powdered EPDM waste was then fed into a sieve to collect only particles passing through a 60 mesh sieve. The sieved EPDM powder was subsequently fed into a twin-screw extruder (BA-11, Bautek Co., Ltd., Korea), which constituted the thermomechanical devulcanization step in this study. The twin-screw extruder consists of six heating zones, and the set temperatures for each zone are presented in Table 2. The screw speed was set to 150 rpm. After the twin-screw extrusion step, the material was defined as devulcanized EPDM (rEPDM). Crosslink-density reduction and soluble fraction were evaluated for the E1-E5 extrusion conditions, and the most suitable condition was selected. The rEPDM prepared under the selected condition was then passed through a single-screw extruder (Haake, Thermo Fisher Scientific, USA) at 280°C to form 30 mm × 30 mm × 5 mm sheets. The subsequent second high-temperature shear pulverization was used only to reduce the sheet to 60

Table 2. Zone-specific Set Temperatures in the Twin-screw Extruder

		Sample				
		E1	E2	E3	E4	E5
Temperature(°C)	Zone 1	190	195	200	205	210
	Zone 2	200	205	210	215	220
	Zone 3	210	215	220	225	230
	Zone 4	220	225	230	235	240
	Zone 5	230	235	240	245	250
	Zone 6	240	245	250	255	260

Table 3. PO-EPDM Specimen Preparation Conditions

Sample	rEPDM (phr)	Virgin EPDM (phr)	PP (phr)	Dicumyl peroxide (phr)	Almax-3600 (phr)	Antioxidant (phr)
P0	0	100	50	3	4	2
P5	5	95	50	3	4	2
P10	10	90	50	3	4	2
P15	15	85	50	3	4	2
P20	20	80	50	3	4	2
P25	25	75	50	3	4	2

**Figure 1.** Schematic diagram of PO-EPDM specimen preparation. The twin-screw extrusion step constitutes thermomechanical devulcanization; the subsequent single-screw extrusion and second pulverization are post-devulcanization processing steps.

± 5 mesh particles for compounding and was not considered an additional devulcanization step. The pulverized rEPDM, virgin EPDM, and polypropylene (PP, Dainpolymer Co., Ltd., Korea) were fed into a twin-screw extruder to prepare PO-EPDM. To improve the dispersibility of EPDM, a dispersant additive (Almex-3600, Ilshin Wells Co., Ltd., Korea) was used. Additionally, an antioxidant (RD, Choyang Chemical Co., Ltd., Korea) was incorporated to prevent thermal and oxidative degradation of the material. The composition of the PO-EPDM is presented in Table 3.

The PO-EPDM compounds were processed into rectangular plate specimens with dimensions of 30 mm (width) $\times$ 30 mm (length) $\times$ 5 mm (thickness) via a single-screw extruder at an extrusion temperature of 280°C. The schematic illustration of the specimen preparation procedure is presented in Figure 1.

2. Characterization methods

2.1 Devulcanization analysis

Horikx analysis was performed to evaluate the relative contributions of crosslink scission and polymer main-chain scission during thermomechanical devulcanization. The degree of devulcanization (D_{dev}) was defined from the decrease in crosslink density relative to the untreated EPDM control as $D_{dev} (\%) = [(v_0 - v_d)/v_0] \times 100$, where v_0 is the crosslink density of the untreated EPDM waste and v_d is the crosslink density after devulcanization. This definition follows the crosslink-density-based approach of ASTM D6814.

The soluble fraction was measured using Soxhlet extraction with toluene, and each measurement was repeated five times. The gel fraction was calculated as 100 minus the soluble fraction. Crosslink density was evaluated from equilibrium swelling in toluene using the Flory-Rehner equation. Because the original EPDM formulation contained carbon black, the filler contribution to restricted swelling was considered using the Kraus correction before applying the Flory-Rehner relationship. The degree of devulcanization was calculated according to ASTM D6814 as $D_{dev} (\%) = [1 - (v_d/v_0)] \times 100$, where v_d is the crosslink density of the devulcanized sample and v_0 is that of the untreated EPDM control. The untreated EPDM reference had a sol fraction of 3.0%, a gel fraction of 97.0%, and a Kraus-corrected crosslink density of 3.00×10^{-4} mol/cm³; the corresponding crosslink-density values for E1-E5 were calculated consistently with the reported D_{dev} values.

2.2 Revulcanization analysis

The vulcanization behavior of PO-EPDM composites with varying recycled EPDM contents was analyzed according to ASTM D5289.26 Considering the crosslinking conditions, the test was conducted at 170°C for 30 min. The uncured compound after mixing was placed in the chamber, and the change in torque over time was measured to obtain the vulcanization curve. From the vulcanization curve, the minimum torque (ML), maximum torque (MH), scorch time (t_{s2}), and optimum vulcanization time (t_{90}) were determined.

The minimum torque (ML) was used as an indicator of the initial viscosity and processability of the compound. The maximum torque (MH) was interpreted as an indicator related to the stiffness of the crosslinked network formed after vulcanization. In addition, the difference between the maximum and minimum torque (ΔM) was utilized as a measure to evaluate the relative degree of crosslink formation.

The scorch time (t_{s2}) was defined as the time at which the torque increased by 2 mN·m from the minimum torque (ML). This parameter was used as a criterion to assess the risk of premature vulcanization during the mixing and molding processes.

The optimum vulcanization time (t_{90}) was defined as the time required to reach 90% of the maximum torque increase.

The calculated vulcanization parameters were utilized to compare and analyze the changes in processability, scorch safety, vulcanization reactivity, and relative crosslink density of the compounds with increasing rEPDM content. The vulcanization reaction rate was calculated as the cure rate index (CRI) using Equation (1).

$$CRI = 100/(t_{90} - t_{s2}) \quad (1)$$

2.3 Mechanical and morphological characterization

To investigate the effect of recycled EPDM content on the mechanical properties of PO-EPDM composites, tensile strength and tear strength tests were performed. The test specimens were cut from vulcanized sheets prepared under identical conditions. Prior to testing, the specimens were conditioned for at least 24 h at $23 \pm 2^\circ\text{C}$ and a relative humidity of $50 \pm 5\%$.

Tensile tests were conducted in accordance with ASTM D412 using a universal testing machine (UTM).²⁷ Dumbbell-shaped specimens were prepared, and the crosshead speed was set to 500 mm/min.

Tear strength was evaluated according to ASTM D624.²⁸

The test specimens were cut from vulcanized sheets into the specified geometry. The measurement was performed using a universal testing machine at a crosshead speed of 500 mm/min. Tear strength was calculated by dividing the maximum load recorded during the tearing process by the thickness of the specimen.

For each composition, the test was repeated at least five times, and the results were reported as the mean value with standard deviation. The obtained tensile and tear strength values were used to compare and analyze the strength retention, deformation behavior, and crack propagation resistance of PO-EPDM composites with increasing rEPDM content.

Compression set tests were performed to evaluate the elastic recovery and permanent deformation behavior of PO-EPDM composites with varying recycled EPDM content. The test specimens were prepared from vulcanized sheets or molded bodies produced under identical conditions. Prior to testing, the specimens were conditioned for at least 24 h at $23 \pm 2^\circ\text{C}$ and a relative humidity of $50 \pm 5\%$.

The compression set test was conducted in accordance with ASTM D395.29 After the initial thickness of each specimen was measured, the specimen was compressed to 75% of its original thickness using a spacer and held in place with a compression device. The compressed specimens were then maintained at 70°C for 22 h. After the test, the specimens were removed from the compression device, allowed to recover at room temperature for 30 min, and the final thickness was measured. The compression set (CS) was calculated using Equation (2).

$$CS = (T_0 - T_1)/(T_0 - T_s) \times 100 \quad (2)$$

$T_0 T_s T_1$ Where CS is the compression set (%), T_0 is the initial thickness of the specimen before testing (mm), T_1 is the thickness of the spacer during compression (mm), T_s is the final thickness

of the specimen after recovery (mm). The microstructure of the specimens was examined using a scanning electron microscope (SEM) by observing the fracture surfaces of the tensile test specimens. Prior to observation, the specimens were sputter-coated with gold (Au) to impart electrical conductivity. This analysis was conducted to examine the morphological characteristics of rEPDM in the PO-EPDM composites.

Results and Discussion

1. Chemical characterization results.

1.1 Devulcanization characteristics.

Figure 2 shows the Horikx analysis results of the rEPDM samples prepared under the E1-E5 devulcanization conditions, together with the untreated EPDM control as the reference state ($D_{\text{dev}} = 0\%$). The untreated control had a

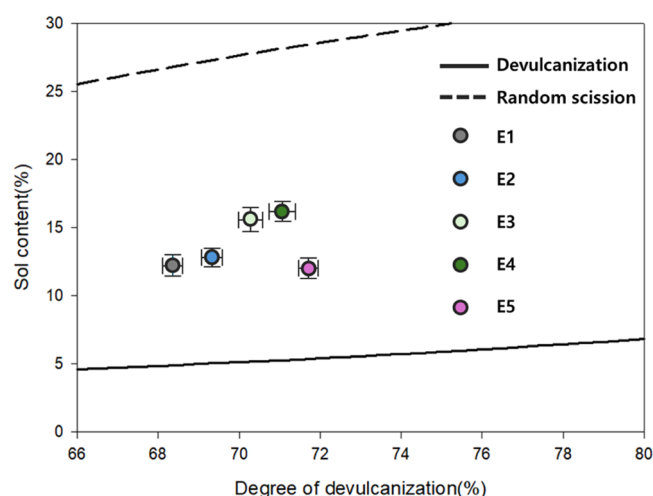


Figure 2. Horikx analysis of untreated EPDM and rEPDM prepared under different thermomechanical devulcanization conditions. The untreated EPDM reference ($D_{\text{dev}} = 0\%$, sol fraction = 3.0%) is indicated for comparison.

Table 4. Sol-gel Characteristics and Crosslink-density Parameters of Untreated and Devulcanized EPDM

Sample	Sol fraction (%)	Gel fraction (%)	Degree of devulcanization (%)	Relative crosslink density, v/v_0	Kraus-corrected crosslink density ($\times 10^{-4}$ mol/cm ³)
Untreated EPDM	3.0	97.0	0.0	1.000	3.00
E1	12.1	87.9	68.4	0.316	0.95
E2	12.8	87.2	69.2	0.308	0.92
E3	15.5	84.5	70.3	0.297	0.89
E4	16.0	84.0	71.0	0.290	0.87
E5	12.0	88.0	71.7	0.283	0.85

sol fraction of 3.0%. In the graph, the solid line represents the ideal devulcanization behavior dominated by selective crosslink scission, whereas the dashed line indicates the case where random polymer main-chain scission is predominant. The sol/gel fractions, degree of devulcanization, relative crosslink density, and Kraus-corrected absolute crosslink-density values are summarized in Table 4.

In general, when the measured data points are closer to the solid line, it can be interpreted that sulfur crosslinks are selectively cleaved, effectively relaxing the three-dimensional rubber network. In contrast, when the data points lie closer to the dashed line, it suggests that significant main-chain scission has occurred, leading to a notable decrease in molecular weight and structural damage to the polymer.

The devulcanized samples exhibited degrees of devulcanization of approximately 68-72% and soluble fractions of approximately 12-16%. Their positions remained substantially below the random-scission theoretical line and closer to the selective-devulcanization trend. Table 4 additionally reports the corresponding gel fractions and Kraus-corrected absolute crosslink densities.

These results indicate that the thermomechanical devulcanization process applied in this study preferentially cleaves the crosslinks in the EPDM network rather than randomly breaking the polymer main chains.

E1 showed a degree of devulcanization of approximately 68.4% and a soluble fraction of about 12.1%. These results indicate that a reduction in crosslink density occurred even under relatively low devulcanization conditions, while the increase in extractable low-molecular-weight components remained limited.

E4 exhibited a degree of devulcanization of approximately

71.0% and a relatively high soluble fraction of about 16.0% compared to the other samples. This suggests that crosslink network cleavage proceeded more actively under this condition.

In contrast, although E5 achieved a high degree of devulcanization of approximately 71.7%, it showed a relatively low soluble fraction of about 12.0%. This implies that while the crosslink density was effectively reduced, the increase in soluble fraction associated with main-chain scission was relatively suppressed.

Therefore, the E5 condition was considered to exhibit the most favorable crosslink scission characteristics among the samples prepared in this study.

The thermomechanical devulcanization process employed in this study effectively relaxed the three-dimensional cross-linked network of EPDM waste while relatively suppressing random main-chain scission of the polymer.

In particular, the E5 condition simultaneously exhibited a high degree of devulcanization and a low soluble fraction, suggesting that E5 was the most favorable devulcanization condition for maintaining rubber-like properties in subsequent reprocessing and revulcanization processes.

1.2 Revulcanization characteristics.

The revulcanization behavior of PO-EPDM composites with varying recycled EPDM contents was evaluated through vulcanization curve analysis, and the results are presented in Figure 3 and Table 5. All samples exhibited a rapid increase in torque after 0.5 min, followed by a gradual increase toward the maximum torque region.

This behavior is attributed to the fact that the uncured compounds first became fluidized under heating and shear conditions, after which the vulcanization reaction proceeded

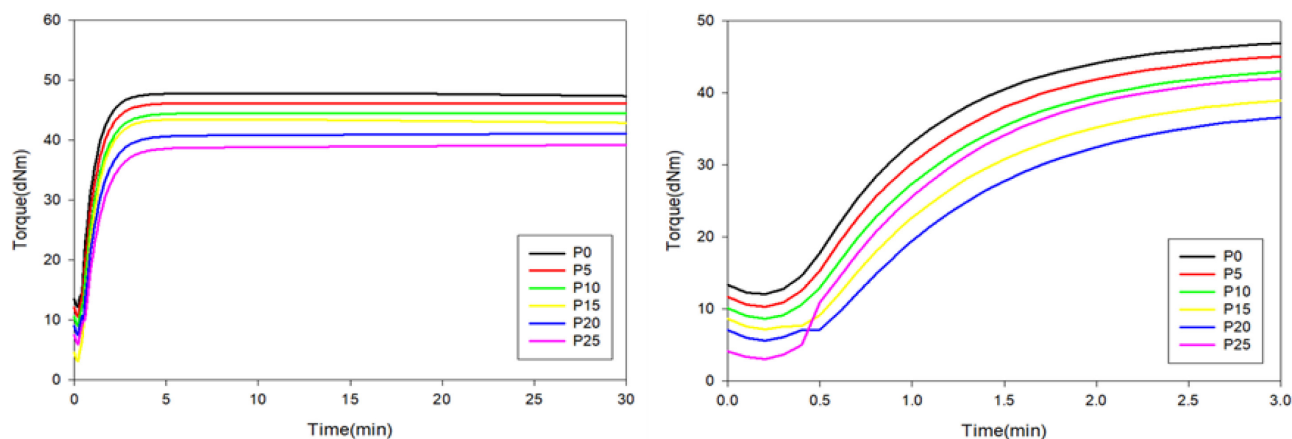


Figure 3. Vulcanization curve according to PO-EPDM manufacturing conditions.

Table 5. Revulcanization Analysis Results According to PO-EPDM Manufacturing Conditions

	ML (dN·m)	MH (dN·m)	t_{s2} (min)	t_{90} (min)	CRI(min ⁻¹)
P0	12.7	47.6	0.35	1.89	64.9
P5	10.9	46.1	0.33	2.10	56.5
P10	9.3	44.5	0.33	2.22	52.9
P15	3.4	43.0	0.34	2.02	59.5
P20	7.8	41.1	0.34	2.44	47.6
P25	6.3	39.2	0.36	2.60	44.6

to form a three-dimensional crosslinked network.

The minimum torque (ML) was highest for P0. The ML values decreased from P0 to P15, increased at P20, and then decreased again at P25; therefore, ML did not decrease monotonically with increasing rEPDM content. Nevertheless, all rEPDM-containing formulations exhibited lower ML values than P0, indicating lower initial viscosity and improved processability relative to the virgin-reference formulation.

This is attributed to the increased chain mobility of the devulcanized EPDM due to partial cleavage of the existing crosslink structure, which resulted in a softening effect within the compound.

The maximum torque (MH) was also highest for P0 and exhibited a gradual decreasing trend as the recycled EPDM content increased. Since the maximum torque is an indicator associated with the stiffness of the crosslinked network and the relative crosslink density after vulcanization, this indicates that P0 formed the densest and strongest crosslinked network.

In contrast, P25 showed a lower maximum torque than the other samples and a relatively slower rate of torque increase. This is attributed to the reduced number of active sites available for the vulcanization reaction and the interference of devulcanized rubber particles with the formation of a uniform crosslinked network as the recycled EPDM content increased.

Although P25 exhibited a low ML value, the lowest ML was observed for P15 (Table 5). The maximum torque (MH) of P25 remained lower than those of P0, P5, and P10, indicating that the crosslinked network formed at the highest rEPDM content was less developed than in the lower-rEPDM formulations.

Therefore, although the P25 condition may be advantageous in terms of processability, it is expected to have limitations in final mechanical strength and elastic recovery properties.

As the rEPDM content increased, MH showed a continuous

decreasing trend, whereas ML showed an overall reduction relative to P0 but with non-monotonic variation among the rEPDM-containing formulations. These results suggest that incorporation of devulcanized EPDM generally enhances initial flowability while reducing the stiffness and relative density of the effective crosslinked network formed after revulcanization.

In particular, at high recycled EPDM contents, the formation of a uniform crosslinked structure is likely to be limited due to residual crosslinked particles, decreased interfacial adhesion, and chain scission.

Recycled EPDM can participate to a certain extent in the revulcanization reaction within the PO-EPDM composites. However, as its content increases, the post-cure torque and the relative degree of crosslink formation tend to decrease.

These results suggest that an increase in recycled EPDM content may lead to a reduction in tensile strength and tear strength, along with an increase in compression set.

2. Physical and Morphological Characterization Results.

The tensile strength and tear strength of PO-EPDM composites with varying recycled EPDM content are shown in Figure 4. The tensile strength of P0 was approximately 11.5 MPa, and it remained almost unchanged at about 11.3 MPa even when the recycled EPDM content was increased up to 15 phr. This indicates that recycled EPDM was relatively well dispersed in the rubber matrix at loadings of 15 phr or less, without causing a significant decrease in tensile properties.

In particular, the P5, P10, and P15 samples exhibited only minimal reductions in tensile strength compared to P0. These results suggest that the addition of recycled EPDM up to a moderate level can be considered applicable in terms of maintaining mechanical strength.

When the recycled EPDM content increased beyond 20

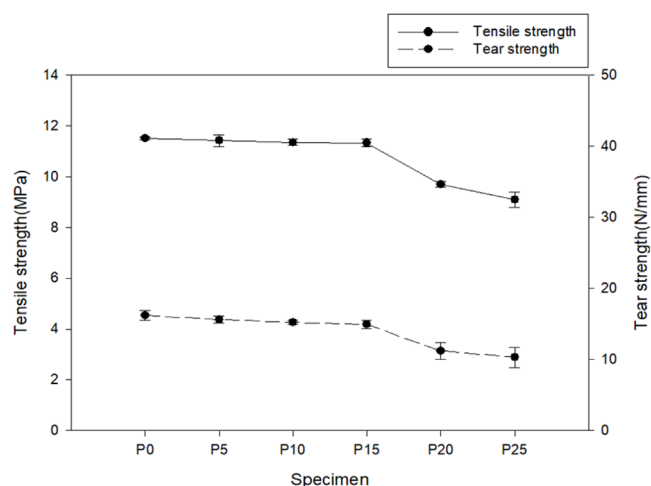


Figure 4. Analysis results of tensile strength and tear strength according to recycled EPDM content.

phr, the tensile strength decreased markedly. The P20 sample exhibited a tensile strength of approximately 9.7 MPa, while the P25 sample showed about 9.1 MPa, corresponding to decrease of approximately 15.7% and 20.9%, respectively, compared to the P0 sample.

This reduction is attributed to the deterioration of interfacial adhesion in the rubber matrix and the decreased dispersibility of recycled rubber particles with increasing rEPDM content, which resulted in lower stress transfer efficiency. In addition, since recycled EPDM already contains partially crosslinked structures, its revulcanization efficiency with virgin EPDM or the rubber matrix tends to be lower. Consequently, the tensile properties deteriorated at high rEPDM loadings.

The tear strength also showed an overall decreasing trend with increasing recycled EPDM content. The P0 sample exhibited a tear strength of approximately 16 N/mm, while the P5, P10, and P15 samples maintained values around 15 N/mm. This indicates that the resistance to crack propagation remained relatively stable at recycled EPDM loadings of 15 phr or less.

However, for the P20 and P25 samples, the tear strength decreased significantly to approximately 11 N/mm and 10 N/mm, respectively. These results indicate that the tear resistance drops sharply when the recycled EPDM content exceeds 20 phr.

These results suggest that the increase in recycled EPDM content leads to higher levels of micro-defects, interfacial inhomogeneity, and uneven crosslink density within the specimens. Tear strength is highly sensitive to crack initiation and propagation. When the interfacial adhesion between the

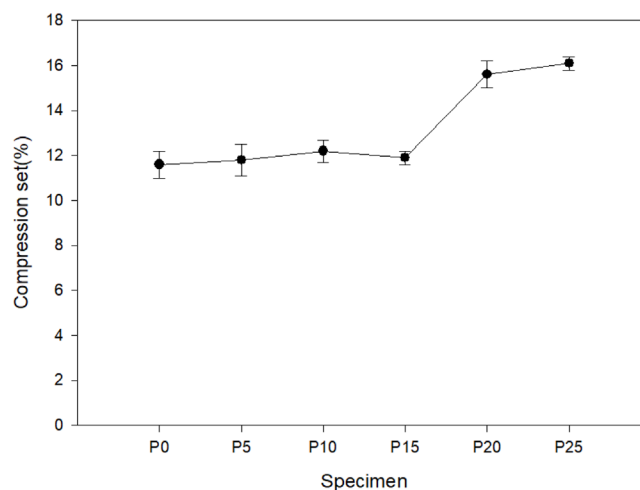


Figure 5. Analysis results of compression set according to recycled EPDM content.

recycled EPDM particles and the rubber matrix is insufficient, cracks can propagate more easily, resulting in reduced tear resistance.

It was observed that the deterioration in tear strength with increasing rEPDM content was more pronounced than that in tensile strength.

The change in compression set with varying recycled EPDM content is shown in Figure 5. The compression set of the P0 sample was approximately 11.6%. The P5 and P10 samples, containing 5 phr and 10 phr of recycled EPDM, exhibited compression set values of approximately 11.8% and 12.2%, respectively. These values showed only a slight increase compared to P0, with no significant difference observed. The P15 sample also displayed a similar value of approximately 11.9%.

These results indicate that the addition of recycled EPDM up to 15 phr has a limited effect on compression set, and the compression recovery characteristics remain relatively stable.

In contrast, the compression set increased markedly when the recycled EPDM content exceeded 20 phr. The P20 sample exhibited a compression set of approximately 15.6%, while the P25 sample showed the highest value of approximately 16.1%. These values represent increases of about 34.5% and 38.8%, respectively, compared to the P0 sample. This result indicates that the incorporation of high rEPDM content can significantly deteriorate the recovery after compressive deformation.

This behavior is considered to be related to the increase in interfacial inhomogeneity and the formation of an uneven crosslinked structure within the rubber matrix as the recycled

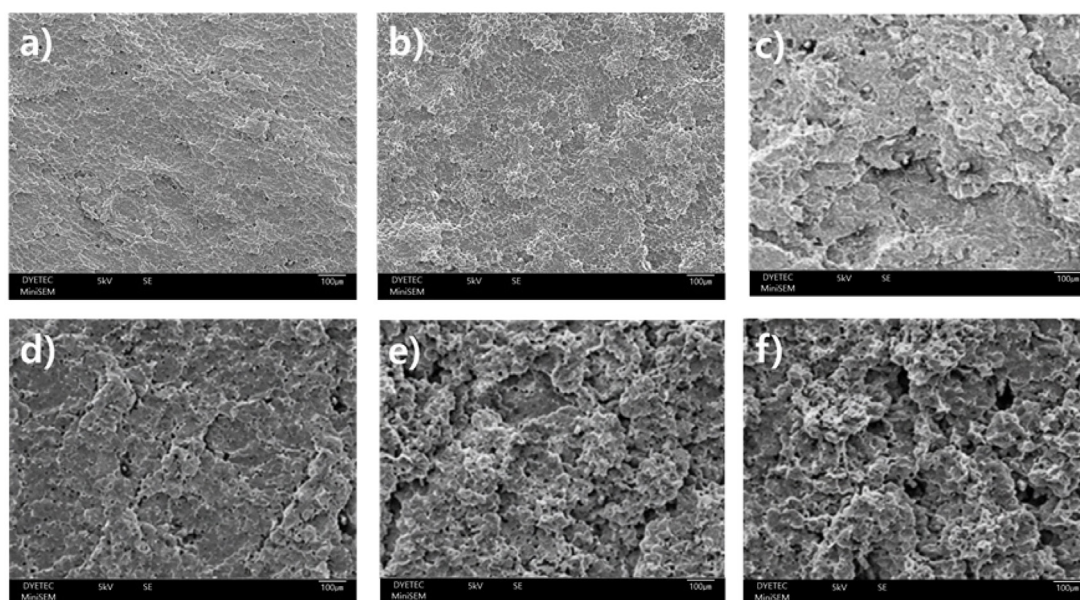


Figure 6. Analysis of morphological characteristics according to recycled EPDM content: (a) P0, (b) P5, (c) P10, (d) P15, (e) P20, (f) P25.

EPDM content increases. Since recycled EPDM already contains pre-existing crosslink structures, its revulcanization reaction and interfacial bonding with the new rubber matrix are likely to be limited.

In particular, at high loadings of 20 phr or more, the compatibility between the recycled EPDM particles and the matrix tends to decrease, leading to poor dispersion and stress concentration. These factors are believed to reduce the ability of the material to recover its original shape after compressive deformation.

The results indicate that recycled EPDM can be applied at loadings of 15 phr or less without significant deterioration of physical properties. However, at loadings of 20 phr or higher, recycled EPDM negatively affects not only compression recovery but also tensile and tear properties.

The morphological characteristics of PO-EPDM samples with varying recycled EPDM content are shown in Figure 6. The P0 sample, which did not contain recycled EPDM, exhibited a relatively uniform and dense fracture surface. Although fine fracture traces were observed on the surface, large pores or particle agglomerates were rarely observed.

The P5 sample containing 5 phr of recycled EPDM also showed a relatively uniform fracture surface similar to that of P0. However, the surface roughness slightly increased, and some minor inhomogeneous regions were observed. This is attributed to the formation of localized interfacial regions resulting from the incorporation of small amounts of recycled

EPDM particles into the rubber matrix.

In the P20 sample with 20 phr of recycled EPDM, the roughness of the fracture surface and the porous structure increased significantly. Irregular agglomerates and distinct porous structures were clearly observed on the surface, suggesting an increase in regions with insufficient interfacial bonding between the recycled EPDM particles and the rubber matrix.

These morphological features may serve as crack initiation sites under applied external stress, thereby promoting crack propagation and contributing to the observed decreases in tensile strength and tear strength.

Conclusions

In this study, the recycled EPDM content was varied from 0 to 25 phr at 5 phr intervals to evaluate the applicability of devulcanized recycled EPDM. The vulcanization characteristics, revulcanization behavior, physical properties, and morphological characteristics were comprehensively analyzed.

The devulcanized recycled EPDM was transformed into a reprocessable state through the partial cleavage of its pre-existing crosslink structures, confirming that it can be effectively reused as a secondary material in new EPDM formulations. However, due to the residual crosslink structures and inhomogeneous molecular chains, increasing the recycled EPDM content was found to negatively influence

the revulcanization reactivity and dispersibility within the rubber matrix.

In terms of revulcanization characteristics, the maximum torque (MH) decreased and the optimum vulcanization time (t_{90}) increased with increasing recycled EPDM content. Consequently, the cure rate index (CRI) showed an overall decreasing trend, with a noticeable decline in revulcanization efficiency observed particularly at loadings of 20 phr or higher.

The physical property analysis revealed that tensile strength and tear strength were maintained at levels similar to those of the control sample (P0) when recycled EPDM was added at 15 phr or less. However, at loadings of 20 phr or higher, both properties decreased sharply, and the compression set also increased, indicating a deterioration in compression recovery performance.

SEM analysis further showed relatively uniform fracture surfaces at recycled EPDM contents of 15 phr or less. In contrast, at 20 phr or above, distinct particle agglomeration, increased porosity, and interfacial inhomogeneity were clearly observed. These morphological defects are considered to reduce stress transfer efficiency, thereby contributing to the primary cause of the observed decline in mechanical properties.

It was confirmed that devulcanized recycled EPDM is a viable material for incorporation into new EPDM formulations at appropriate loadings. Under the conditions of this study, the optimal addition range that balances material properties and recycling efficiency was determined to be 15 phr or less. For future applications at higher loadings, further research on particle size control, surface modification, compatibilizer application, and optimization of the crosslinking system will be necessary.

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Conflict of Interest: The authors declare that there is no conflict of interest.

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