İmalat Teknolojileri ve Uygulamaları
Cilt: 4, No: 3, 2023 (129-136)
Araştırma Makalesi
e-ISSN: 2717-7475

Manufacturing Technologies and Applications
Vol: 4, Issue: 3, 2023 (129-136)
Research Article
e-ISSN: 2717-7475

Investigation of The Effect of Micronized Ethylene-Propylene-Diene Monomer (EDPM) Powder Wastes on Physical and Mechanical Properties in EPDM Mixtures

Çağrı Çokaçar[1], İlker Köprü[1], Hakan Salih Yetgin[2,*]

[1]*Seçil Kauçuk San ve Tic. A.Ş. Tarsus, Mersin, Türkiye*
[2]*Tarsus University, Engineering Faculty, Department of Mechanical Engineering, Tarsus, Mersin, Türkiye*

ARTICLE INFORMATION

Received: *13.10.2023*
Accepted: *22.11.2023*

Keywords:
Micronized EPDM powder
Rheological properties
Physical properties
EPDM rubber

ABSTRACT

In this study, micronized-ethylene-propylene-diene monomer powders (m-EPDM) by the physical crushing method were added to EPDM rubber, and their effects on physical and mechanical properties were examined. Micronized EPDM powders were added at rates of 10-20-30%. EPDM rubber was pulped with micronized EPDM powder using a 1.5-liter laboratory type mini banbury. The obtained paste was vulcanized in the press at 180 °C for 20 min., and test plates were acquired. Rheological and fluidity properties of EPDM rubber with m-EPDM powder added were examined by pre-press rheometer (Moving Die Rheometer_MDR) and Mooney viscosity tests. Tensile and permanent deformation tests were performed to determine mechanical properties density, Shore A hardness tests were performed to determine physical properties and dispersion analysis was performed to distribute the additives in EPDM rubber. As a result of the study, minimum torque (ML) values increased while maximum torque (MH) values decreased depending on the amount of m-EPDM. Mooney viscosity of EPDM/30m-EPDM rubber increased by 40.9%. It was observed that cure rate index (CRI) did not change depending on the amount of m-EPDM. With the addition of 30% m-EPDM, breaking strength decreased by 39.2%, and elongation at break was reduced by 9.87%.

Mikronize Etilen-Propilen-Dien Monomer (EDPM) Atıklarının EPDM Karışımlarındaki Fiziksel ve Mekanik Özelliklere Etkisinin Araştırılması

MAKALE BİLGİSİ

Alınma: *13.10.2023*
Kabul: *22.11.2023*

Anahtar Kelimeler:
Mikronize EPDM tozu
Reolojik özellikler
Fiziksel özellikler
EPDM kauçuğu

ÖZET

Bu çalışmada fiziksel kırma yöntemiyle mikronize edilmiş Etilen Propilen Dien Monomer (m-EPDM) tozları EPDM kauçuğuna ilave edilerek fiziksel ve mekanik özelliklere etkisi incelenmiştir. Mikronize EPDM tozları %10-20-30 oranlarında ilave edilmiştir. 1.5 lt'lik laboratuvar tipi mini banbury kullanılarak mikronize-EPDM tozu ile EPDM kauçuğu hamur haline getirilmiştir. Elde edilen hamur preste 180 ^{o}C ve 20 dk. boyunca vulkanize edilerek test plakaları elde edilmiştir. Pres öncesi reometri (MDR) ve Mooney viskozite testleri ile m-EPDM tozu ilaveli EPDM kauçuğun reolojik ve akışkanlık özellikleri incelenmiştir. Mekanik özelliklerin belirlenmesi için çekme testi ve kalıcı deformasyon testi, fiziksel özelliklerin belirlenmesi için yoğunluk ve Shore A sertlik testleri, EPDM kauçuğunda mevcut olan katkıların dağılımı için ise dispersiyon analizi yapılmıştır. Çalışma sonucunda, m-EPDM miktarına bağlı olarak minimum tork (ML) değerleri artarken maksimum tork (MH) değerleri azalmıştır. EPDM/30m-EPDM kauçuğun mooney viskozitesi %40.9 oranında artmıştır. Kür indeksi (CRI) oranlarının, m-EPDM miktarına bağlı olarak değişmediği gözlenmiştir. %30 m-EPDM ilavesi ile kopma mukavemeti %39.2, kopma uzaması ise %9.87 oranlarında azalmıştır.

**Sorumlu yazar, e-posta: shyetgin@gmail.com*
To cite this article: Ç. Çokaçar, İ. Köprü, H. S. Yetgin Investigation of The Effect of Micronized Ethylene-Propylene-Diene Monomer (EDPM) Powder Wastes on Physical and Mechanical Properties in EPDM Mixtures, Manufacturing Technologies and Applications, 4(3),129-136, 2023.
https://doi.org/10.52795/mateca.1375512,

1. INTRODUCTION (GİRİŞ)

Polymer and polymer composite materials are used in various applications, from simple daily applications to complex high-tech applications. The rapid consumption of polymer materials causes the waste of these materials to increase rapidly. Especially in the automotive industry, used tires and waste generated during production and after use constitute a large portion of polymer material waste [1]. Since rubber-based materials do not decompose quickly, waste disposal has become a major environmental problem for manufacturers and the state. Since most waste contains petroleum-based products, reuse and recycling are essential for protecting non-renewable resources [1-2].

Karaağaç et al. [1] stated that approximately 40% of the world's rubber consumption comes from the use of natural rubber (NR), and the majority of natural rubber is followed by general-purpose rubbers such as styrene-butadiene (SBR) rubber, butyl rubber (IIR) and ethylene-propylene-diene monomer (EPDM) rubber. It has been stated that EPDM rubber, among these rubbers, contributes to approximately 12% of the world's rubber production [3]. EPDM rubber, which was first introduced commercially by the United States in 1962, is the fastest growing general-purpose rubber type with a wide range of applications due to its features such as low density, economical, high filler and process oil capacity, and superior thermal and ozone resistance [1-2, 4-5]. Two-thirds of any automobile's total amount of rubber is used in tires. The remaining rubber-based components include gaskets, sealants, hoses, anti-vibration materials, and plugs. Almost 25-50% of these non-tire rubber products are produced from EPDM rubber. Therefore, it is not surprising that EPDM-based wastes emerge in increasing amounts every day [6-9].

Rubber-based products generally do not deteriorate or melt, so traditional reprocessing techniques are unsuitable for recycling rubber [10]. Two methods are used to prepare natural or synthetic rubbers for recycling: crushing at room temperature and crushing by cooling with liquid nitrogen [1]. In the crushing process at room temperature, the particle size is larger because rubber materials are not rigid; they are as elastic as possible. Although the initial investment and operating costs are relatively high in the liquid nitrogen method, smaller grains can be obtained compared to the crushing process at room temperature. In the cracking process performed by cooling with liquid nitrogen, the embrittlement effect of low temperatures, called cryogenic, on the materials is used [1, 4, 11].

Waste recovery studies in the rubber industry have generally focused on vehicle tire rubber, both academically and industrially. The amount of waste vehicle tires has increased temporarily, reaching around 325 million annually [12]. However, since vehicle tire rubber contains different compositions, there are inefficiencies in the recycling of waste [1]. The waste powders from grinding EPDM rubber have a reinforcement effect with a low elastic modulus due to the processing oil content. High waste powder size can cause stress stresses or poor bonding between waste rubber and untreated rubber due to the smooth surfaces of waste powders [13].

There are many studies on the use of vehicle tire waste and ground EPDM waste in different rubber and polymer formulations [1-2, 4-5, 8, 14-15]. Güngör et al. [5] analyzed the vulcanization process with kinetic parameters by adding waste tire particles to EPDM and Silicone Rubber matrices. As a result of the study, it was stated that adding waste rubber to rubber products would reduce the waste problem. Still, it should be compatible so as not to affect the vulcanization process. Demirer et al. [4] examined the effect of waste EPDM powders on the bending properties of Polypropylene (PP)/EPDM mixtures. EPDM waste ground in different sizes (0-50, 50-75, 75-150, and 150-300µ) and different proportions (0-10-20-30 and 50%) was produced by extrusion and subsequent injection molding methods. As a result of the study, bending strength and bending modulus decreased with increasing amount of waste EPDM. The % elongation value at maximum power has increased. Since the obtained strength values are acceptable, it has been stated that waste EPDM can be used as a recycling material.

Ismail et al. [16] investigated the effect of three different GTR sizes (250–500 µm, 500–710 µm and 710 µm–1 mm) on the mechanical properties of PP/GTR mixtures. Mixtures containing smaller ground tyre rubber (GTR) particles (250-500 μm) caused high friction due to their higher surface area, which resulted in higher balance torque. However, it has been stated that the results are worsened due to the cross-linked structure of GTR particles and their weak adhesion to the PP matrix, which causes easy crack initiation and rapid crack propagation. Sonnier et al. [17] used three different rubber particle sizes (380–1200 μm) in GTR/Low Density Polyethylene (LDPE) compounds. Since no significant difference in the mechanical properties of GTR/LDPE (50/50 wt%) mixtures was obtained, they suggested that controlling the size of GTR particles is not the only parameter to improve mechanical properties. Karaağaç et al. [1] examined the effect of the amount and size of EPDM waste ground up to 35, 40, and 60 mesh at ambient temperature for automobile application. A new trial product consisting of maleated ethylene-propylene rubber (EPM) and bitumen was used to improve the interphase adhesion of waste and untreated EPDM. As a result of the study, it was stated that the reuse of waste EPDM powders caused moderate deterioration in the product's curing properties and mechanical properties. Jacob et al. [2] examined EPDM rubber's mechanical and rheological properties containing ground EPDM vulcanizate. With the addition of ground EPDM, Mooney viscosity, tensile strength, tear strength, and elongation at break were increased while scorch time decreased. Generally speaking, the most critical parameters for reusing waste powder rubbers are the waste amount and the waste powder size [18]. Munirah et al. [19] examined the effect of waste-cured EPDM rubbers added to natural rubber (NR) on mechanical properties. The amount of waste was adjusted to be 0-20-40-60 phr. The best results were obtained by incorporating up to 60 phr of cured EPDM waste into the NR. The best formulation at 60 phr showed the highest tensile strength and elongation at break with moderate hardness, modulus, degree of cracking, and cycling.

This study aims to reuse EPDM powder waste (m-EPDM) by replacing it with untreated EPDM for building and infrastructure applications. For this purpose, EPDM rubber, which was micronized-powdered by the crushing method at room temperature, was added to untreated EPDM rubber at the rates of 10, 20, and 30%, and the effect of m-EPDM powder on mechanical, rheological and physical properties was examined. The study results were evaluated regarding the suitability of EPDM rubbers with m-EPDM addition for use in blind plugs and transmission pipes of solar energy panels used in water heating systems.

2. MATERIAL AND METHOD (MATERYAL VE YÖNTEM)

2.1. Materials (Malzemeler)

In this study, a commercially available EPDM rubber was used. The additives and their ratios used in producing EPDM rubber are given in Table 1 in volumetric form. EPDM powders in the range of 80-100 mesh, obtained from breaking and micronizing the EPDM scraps produced during production at Seçil Kauçuk A.Ş., were used as filling elements. m-EPDM powders were added at rates of 10-20-30%.

Table 1. Formulation of EPDM rubbers specifications (EPDM kauçukların formülasyonu)

	EPDM	EPDM/10m-EPDM	EPDM/20m-EPDM	EPDM/30m-EPDM
EPDM	100	100	100	100
Carbon black	50	50	50	50
White fill	20	20	20	20
Oil	40	40	40	40
Zinc oxide	4	4	4	4
Stearic acid	2	2	2	2
Sulfur	1	1	1	1
MBT	0.5	0.5	0.5	0.5
TMTD	1	1	1	1
m-EPDM	0	25	55	95

2.2. Production Methods (Üretim Yöntemleri)

Untreated EPDM and m-EPDM powder-filled EPDM rubbers were pulped with a 1.5-liter laboratory type mini banbury. Rheometer tests of the produced m-EPDM filled EPDM rubber samples were carried out by the ASTM D5289 standard on the Alpha MDR 2000 brand rheometer device owned by Seçil Kauçuk A.Ş. The test was performed at 200 ºC and 5 min.

2.3. Test Methods (Test Yöntemleri)

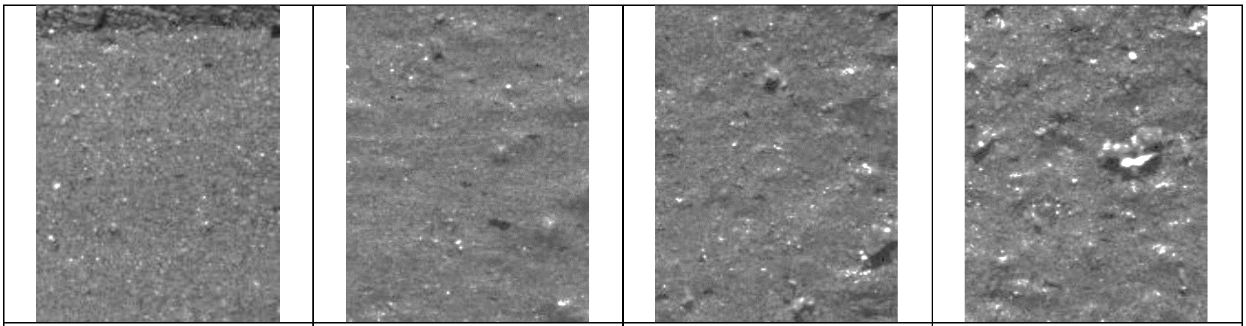
Test plates were obtained by vulcanizing the paste products in the press at 180 ºC and for 20 min. Tensile tests were prepared by ASTM D638 standard. Dumb-bell type test pieces with a length of gauge section of 25mm, a total length of 75mm and an width of ends of 12.5mm were used. The tests were carried out on a Zwick-Z020 model tensile tester. Each tensile test sample was measured at least five times and averaged. Tensile tests were carried out at 200 mm/min tensile speed. Permanent deformation tests were carried out according to the DIN 53517 standard at 100 ºC for 22 hours and 25% compression. Hardness tests were carried out by DIN 53505 ideal, and hardness was measured in Shore A. In hardness measurements, at least ten hardness measurements were made on each test sample. Density tests were carried out using the ISO 1183 standard using the Archimedes principle. Dispersion quality measurements, ranging from 1 to 10 depending on the number and size of agglomeration, were made according to the ISO 11345 standard.

3. EXPERIMENT RESULTS (DENEY SONUÇLARI)

Images of dispersion tests performed with untreated EPDM and EPDM rubber with m-EPDM addition are given in Figure 1. The dispersion test results performed to determine the distribution of carbon black and other filler particles present in EPDM rubbers are shown in Table 2. When the dispersion rates were examined, it was observed that EPDM rubbers were in the 89-93% band. According to the dispersion analysis images in Figure 1 and the test results in Table 2, a 5.5% white area is observed in EPDM/30m-EPDM rubber. This white area allows us to comment on whether the white filling materials in the mixture are mixed homogeneously with other components. The higher the dispersion rate in %, the better all the details in the mix are dispersed. The smaller the white area in % and the smaller the average agglomerate size, the more homogeneously distributed the white fillers in the combination. In line with this information, EPDM/10m-EPDM rubber was the most successful EPDM rubber in dispersion test results with 6.31% white area, 93.69% dispersion, and 2.88 average agglomerate size.

Table 2. Dispersion test results (Dispersiyon test sonuçları)

	X	Y	Z	White area, %	Dispersion, %	Average Agg. Size, [um]	Agg. Size Std. Dev [um]
EPDM	1.50	9.84	75.53	8.57	91.43	3.07	2.93
EPDM/10m-EPDM	2.07	9.63	81.97	6.31	93.69	2.88	3.00
EPDM/20m-EPDM	1.62	9.18	77.32	7.94	92.06	3.19	3.55
EPDM/30m-EPDM	1.12	8.46	68.61	10.99	89.01	3.46	4.21



(a) EPDM	(b) EPDM/10m-EPDM	(c) EPDM/20m-EPDM	(d) EPDM/30m-EPDM

Figure 1. Dispersion images (Dispersiyon görüntüleri)

Table 3 shows the results obtained after rheological and physical tests of EPDM rubbers with different amounts of m-EPDM powder added. When the table is examined, it is determined that the minimum torque (ML) values increase with increasing m-EPDM amount while the maximum torque (MH) values decrease. The initial baking time of the pulp (t_{s2}) and the time when the dough reached ultimate baking (t_{90}) decreased with the addition of m-EPDM. When Shore A hardness results were evaluated, it was determined that the hardness values did not change when m-EPDM was added. In general, hardness values of EPDM rubber samples were obtained between 56-58 Shore A. When tear strength and elasticity values were examined, it was stated that tear strength and elasticity decreased significantly with m-EPDM and increasing m-EPDM amount. The study conducted by Jacob et al. [2] stated that as the amount of waste EPDM increased, the elasticity ratio decreased due to the increase in carbon black content.

Table 3. Rheological and physical test results (Reolojik ve fiziksel test sonuçları)

	EPDM	**EPDM/10m-EPDM**	**EPDM/20m-EPDM**	**EPDM/30m-EPDM**
ML (dNm)	0.96	1.17	1.51	1.81
MH (dNm)	13.31	12.88	12.09	11.43
t_{s2} (min)	0.43	0.40	0.41	0.42
t_{90} (min)	1.51	1.37	1.48	1.48
Tear strength, N/mm	79.3	56.0	62.2	53.5
Density (g/cm^3)	1.086	1.098	1.102	1.122
Hardness (Shore A)	58	58	57	56
Elasticity, %	61.22	58.84	57.88	57.38

Figure 2 shows the EPDM rubber's breaking strength and elongation at break graph with m-EPDM powder added. As seen in the figure, breaking strength and elongation at break decreased with the addition of m-EPDM to EPDM rubber and the increasing amount of m-EPDM. Compared to untreated EPDM, with the addition of 30% m-EPDM, the breaking strength decreased by 39.2%, and the elongation at breaking reduced by 9.87%. Similar results were obtained in the study conducted by Karaagaç et al. [1].

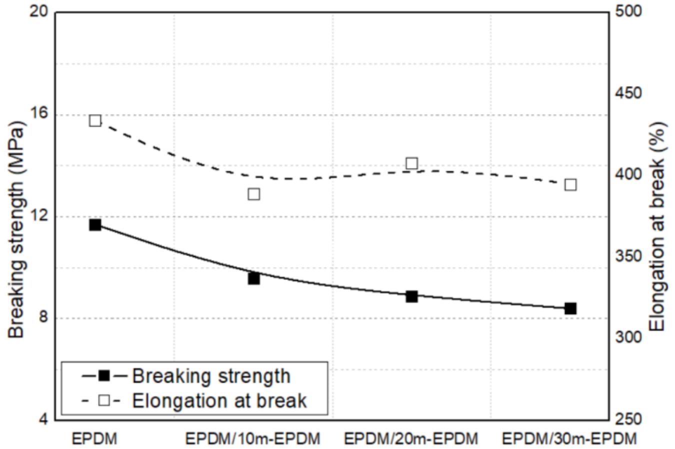


Figure 1. Breaking strength and elongation at break change (Kopma mukavemeti ve kopma uzaması değişimi)

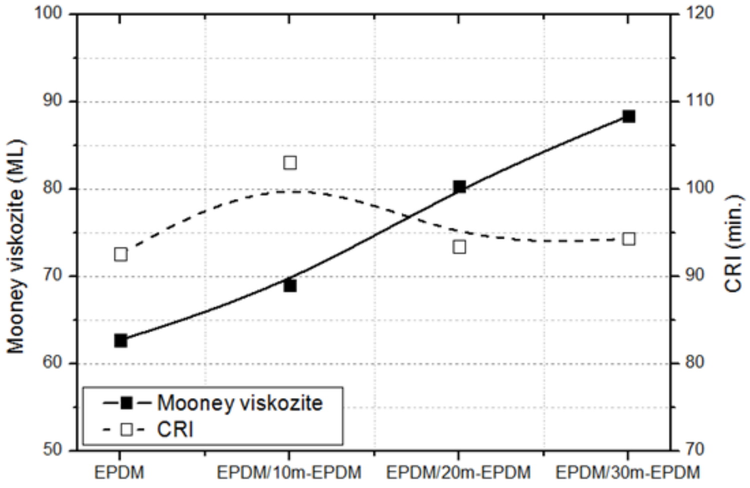


Figure 3. Breaking strength and breaking elongation change (Kopma mukavemeti ve kopma uzaması değişimi)

Figure 3 shows the change in Mooney viscosity and CRI values of untreated EPDM and EPDM rubber with m-EPDM powder added. As seen in the figure, it was determined that Mooney viscosity values increased depending on the amount of m-EPDM. At the same time, untreated EPDM rubber had a viscosity value of 62.7 MU; with the addition of m-EPDM and the increasing amount of m-EPDM, the Mooney viscosity value increased by 40.9% and reached 88.4 MU. When the CRI results defining the difference between t_{90} and t_{s2} were evaluated, it was determined that the CRI values were between 92.5 and 103.0 min, depending on the amount of m-EPDM.

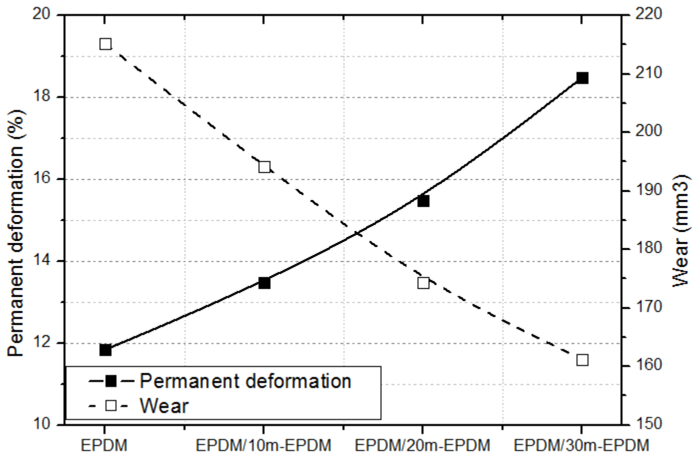


Figure 4. Change in permanent deformation and wear amounts (Kalıcı deformasyon ve aşınma miktarlarındaki değişim)

Figure 4 shows the change in permanent deformation and wear amounts of untreated EPDM and EPDM rubbers with m-EPDM powder added. As seen in the figure, the permanent deformation rate of untreated EPDM rubber, which was 11.8%, increased by 13.8% with the addition of 10% m-EPDM. Similarly, adding m-EPDM to 20% and 30%, permanent deformation rates increased by 30.7% and 56.1%, respectively. When the wear results are evaluated, the wear amounts of EPDM rubbers with m-EPDM powder added have decreased. This reduction was achieved at 9.76%, 18.99%, and 25.07% for EPDM/10m-EPDM, EPDM/20m-EPDM, and EPDM/30m-EPDM rubbers, respectively. This shows that wear resistance increases with the addition of m-EPDM.

4. CONCLUSIONS (SONUÇLAR)

As a result of the study examining the rheological and mechanical properties of EPDM rubber with the addition of EPDM powder micronized by physical crushing method.

When the MDR test results were evaluated, the minimum torque (ML) values increased, and the maximum torque (MH) values decreased depending on the amount of m-EPDM. Compared to untreated EPDM, the Mooney viscosity of EPDM/30m-EPDM rubber increased by 40.9%. It has been observed that CRI rates do not change depending on the amount of m-EPDM. Compared to untreated EPDM, with the addition of 30% m-EPDM, tensile strength decreased by 39.2%, and elongation at break reduced by 9.87%. Depending on the amount of m-EPDM, permanent deformations decreased, and wear resistance increased.

When the results are evaluated, it is thought that EPDM rubbers with m-EPDM addition will be suitable for use in blind plugs and transmission pipes of solar energy panels used in water heating systems.

REFERENCES (KAYNAKLAR)

1. B. Karaağaç, H.O. Turan, D.D. Oral, Use of ground EPDM wastes in EPDM-based rubber compounds: With and without compatibilization, Journal of Elastomers & Plastics. 47(2):117-135, 2015.
2. C. Jacob, A. K. Bhowmick, P.P. De, S.K. De, Recycling of EPDM waste. I. Effect of ground EPDM vulcanizate on properties of EPDM rubber, Journal of Applied Polymer Science, 82(13):3293-3303, 2001.
3. P. Sutanto, F.L. Laksmana, F. Picchioni, L.P.B.M. Janssen, Modeling on the kinetics of an EPDM devulcanization in an internal batch mixer using an amine as the devulcanizing agent, Chemical Engineering Science, 61(19):6442-6453, 2006.
4. H. Demirer, Y. Tezcan, K. Büyükkaya, İ. Kartal, Effect of addition of recycled EPDM size on the bending properties of Polypropylene, European Journal of Science and Technology, 15:381-386, 2019.
5. A. Güngör, İ.K. Akbay, T. Özdemir, Accumulation of recycled waste tyre within different rubbers: A kinetic study, Sigma Journal of Engineering and Natural Sciences, 37(4):1169-1179, 2019.
6. J. Yun, V.V. Yashin, A.I. Isayev, Ultrasonic devulcanization of carbon black-filled ethylene propylene diene monomer rubber, Journal of Applied Polymer Science, 91(3):1646-1656, 2004.
7. D.Z. Pirityi, K. Pölöskei, Thermomechanical devulcanisation of ethylene propylene diene monomer (EPDM) rubber and its subsequent reintegration into virgin Rubber, Polymers, 13(7):1116, 2021.
8. M. Sabzekar, G. Zohuri, M.P. Chenar, S.M. Mortazavi, M. Kariminejad, S. Asadi, A new approach for reclaiming of waste automotive EPDM rubber using waste oil, Polymer Degradation and Stability, 129:56-62, 2016.
9. A. Fazli, D. Rodrigue, Waste rubber recycling: A review on the evolution and properties of thermoplastic elastomers, Materials, 13(3):782, 2020.
10. M. Forrest, Overview of the world rubber recycling market. In Recycling and Reuse of Waste Rubber, 1st ed.; Potter, D., (EdS.) Smithers Rapra: Shawbury, UK, 1:17-30, 2014.
11. K. Ünlü, EPDM Atıklarının Otomotiv Endüstrisinde Degerlendirilmesi Üzerine Bir inceleme, Yüksek Lisans Tezi, Marmara Üniversitesi Fen Bilimleri Enstitüsü, İstanbul, Türkiye, 2007.
12. İ. K. Akbay, A. Güngör, T. Özdemir, Optimization of the vulcanization parameters for ethylene–propylene–diene termonomer (EPDM)/ground waste tyre composite using response surface methodology, Polymer Bulletin, 74:5095-5109, 2017.
13. C. Jacob, A.K. Bhowmick, P.P. De, S.K. De, Utilization of powdered EPDM scrap in EPDM compound, Rubber Chemistry and Technology, 76(1): 36, 2003.
14. A.M. Lepadatu, S. Asaftei, N. Vennemann, Recycling of EPDM rubber waste powder by activation with liquid polymers, Rohstoffe Und Anwendungen Raw Materials and Applications, 41-47:2014.
15. A.B. Habieb, F. Milani, G. Milani, R. Cerchiaro, Rubber compounds made of reactivated EPDM for fiber-reinforced elastomeric isolators: an experimental study, Iranian Polymer Journal, 29:1031-1043, 2020.
16. H. Ismail, M. Awang, M. Hazizan, Effect of waste tire dust (WTD) size on the mechanical and morphological properties of polypropylene/waste tire dust (PP/WTD) blends, Polymer-Plastics Technology and Engineering, 45:463-468, 2006.
17. R. Sonnier, E. Leroy, L. Clerc, A. Bergeret, J. Lopez-Cuesta, Polyethylene/ground tyre rubber blends: Influence of particle morphology and oxidation on mechanical properties, Polymer Testing, 26:274–281, 2007.

18. C. Jacob, A.K. Bhattacharya, A.K. Bhowmick, P.P. De, S.K. De, Recycling of eyhylene propylene diene monomer (EPDM) waste. III. Processability of EPDM rubber compound containing ground EPDM vulcanizates, Journal of Applied Polymer Science, 87:2204, 2003.
19. O. Munirah, M.S. Mustafa, Z. Ahmad, N.M.A. Wahab, Performance evaluation of natural rubber filled cured ethylene propylene diene rubber (EPDM) waste reinforcement, Solid State Science and Technology, 26(1):186-193, 2018.