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Research Article

Investigation of using waste banana peels in EPDM as bio-based filler

İsmail Kutlugün Akbay [a] ***and Ahmet Güngör*** [a,*]

[a] *Mersin University, Chemical Engineering Department, 33343, Yenişehir/Mersin*

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ABSTRACT

The rubber and plastics sectors have been developing in both 19th and 20th centuries. The most of the developments have been realized in the 20th century. As a result of this, the need for petrochemical products has increased day by day. Synthetic rubber is used to protect vehicle tires, automotive parts, white goods, insulation materials and especially bridges, highways, viaducts and nuclear facilities requiring high elastic modulus from seismic and acoustical effects. Rubber type materials are preferred in many engineering applications due to due to their sufficient properties such as high elasticity. The importance of the study carried out in the rubber industry in order to improve the performance characteristics of the material as well as to increase the production efficiency and reduce the cost is increasing day by day. In order to remove or reduce the harmful effects of carbon black, which is used as a common filler the effects of additives obtained from some biological wastes such as banana peels instead of carbon black, on the physical and mechanical properties of ethylene propylene diene monomer (EPDM) rubber have been investigated. The results were evaluated considering MDR and Soxhlet extraction test results applied on rubber samples.

1. Introduction

Polymer is a macromolecule that occurs as a result of continuous repeating of small molecules. Small units that formed polymers are named as monomers. A polymer is a large molecule formed via covalent bonding of repeating monomers. Today, polymers and polymer based materials have been used in various areas such as medication, nutrition, communication, container production, clothing, buildings, highways. They are found in all materials utilized in daily life [1].

Rubber is a material that is used in many applications with increasing usage areas in our daily life. The rubbers which are seen as alternatives to metals, has many advantages because it provides ease of use in places where metals cannot be used. Rubber material can be described as a material obtained from the milk essence of some plants in natural state (latex) or synthetically obtained as a result of mixing petroleum and alcohol [2].

The Ethylene propylene diene rubber (EPDM) is an inexpensive rubber that is a widely used raw material in many areas. EPDM has been used for many years in applications such as electricity, sports equipment, footwear, industry, radiation, automobile, etc. In addition, EPDM is easy to access and resistant to aging for a long time. This makes EPDM a preferred rubber type. Low cost, good mechanical properties, good radiative protection, large filler incorporation capacity during compounding and good chemical resistance are the main reasons for the use of many areas [3,4].

The mechanical, thermal and rheological properties of EPDM can vary according to the physical and chemical properties of the content, vulcanization properties and filler. The filler is used in compounding process in different ratios in order to obtain the desired mechanical and thermal properties. In this context, the chemical interactions between EPDM and fillers and the homogeneous dispersion have a significant effect on the mechanical, thermal and rheological properties of the material matrix in the final product [2-6].

Carbon black (CB) is the most important filler for rubber and it is widely used in rubber industry due to its chemical and heat stability. In addition, carbon black is used as a colorant. Despite the use of many areas, with a long exposure, negatively affects the health of the workers in rubber industry. In addition, due to small particle size and easy static electrification, there is

* Corresponding author. Tel.: *+ 90 (324) 3610001 - 7396*.
E-mail addresses: *akbay@mersin.edu.tr*, *ahmet.gungor@mersin.edu.tr*

usually a tendency to agglomerate within the material and this can be a problem in some cases. In order to overcome these disadvantages, the main purpose is to use a non-harmful and environmentally compatible filler material instead of carbon black [7-9].

Although there is no study regarding the use of biofillers obtained from banana peels, many studies have been carried out for the possible use of bio-based fillers in rubber compounding. Karaağaç [10] have studied to utilize of pistachio shells as filler in natural rubber/styrene–butadiene (NR/SBR) rubber composites, characterizing mechanical, thermal, morphological properties of the composite materials. Analysis results showed that mechanical and thermal properties of final material were increased with the addition of pistachio shell. Besides, pistachio shells containing NR/SBR composites have high abrasion resistance. In addition, Botros et al. [11] have reported natural rubber (NR) containing lignin and lignin derivative material composites. Characterization results demonstrated that mechanical and thermal properties of prepared composites were improved with the addition of bio-polymeric filler material. Menon et al. [12] studied vulcanization parameters and physico-mechanical properties of natural rubber (NR) with different cashew nut shell liquid (PCNSL) content. With the increase of PCNSL addition increased of the rheological and tensile properties of NR composites. In a different study, Arayapranee et al. [13] reported NR/EPDM composites with rice husk. The addition of rice husk has decreased the optimum curing time while increasing the rheological properties. Mechanical properties have decreased with the addition of rice husk. Intiya et al. [14] reported natural rubber with filler sludge ash for the gaining an economic purpose. The pyrolysis of sludge was studied and compared with $CaCO_3$ filler. The content of sludge ash was analysed. It was concluded that both filler was activated the vulcanization reactions.
In this study, the effects of some biological wastes such as banana peels on the physical and mechanical properties of ethylene-propylene-diene monomer (EPDM) rubber were investigated. The obtained bio-based filler material was added into EPDM matrix with different ratios. MDR (Moving die rheometer), modulus of elasticity, elongation at break and swelling tests were performed to observe the effects of these wastes on EPDM rubber.

2. Material & Method

Banana peels, vinegar, EPDM rubber, mineral oil, stearic acid, zinc oxide, carbon black, TAC / 50, peroxide were used in all compound formulations.

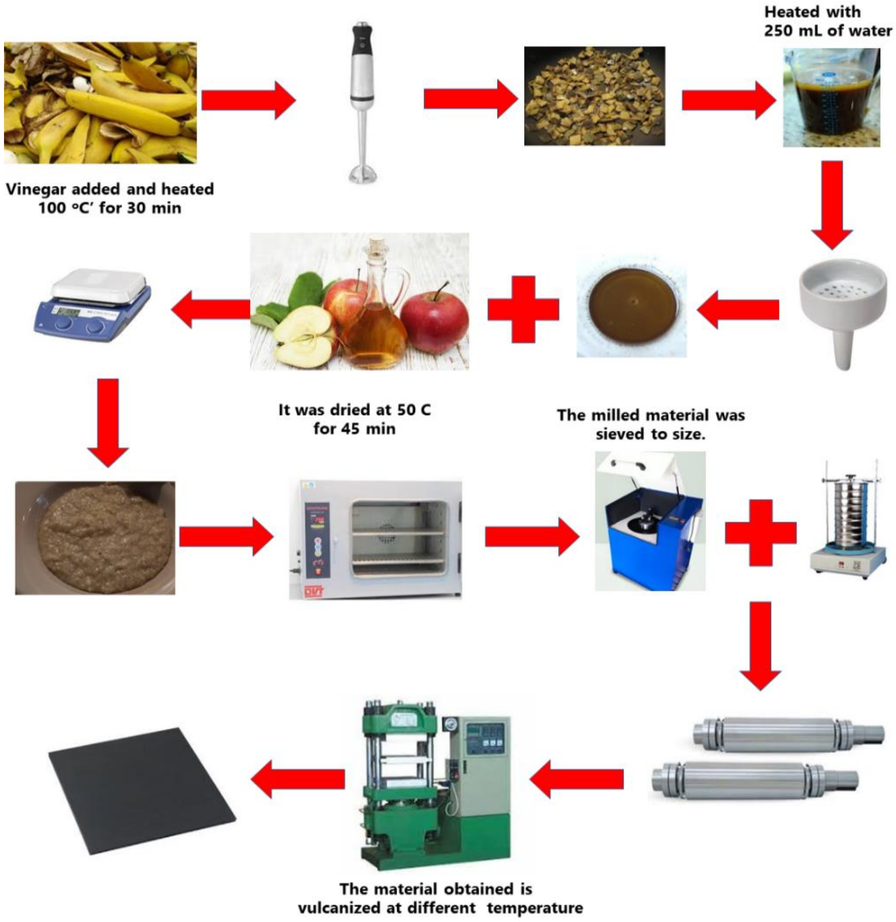

Figure 1. Schematic presentation of composite preparing

The recipes for the EPDM with different biopolymer content are given in Table 1. KELTAN 9650Q was used as the rubber matrix and purchased from Lanxess. Other ingredients in the compounds which are conventional materials (stearic acid, mineral oil, zinc oxide etc.) were used for constituting the vulcanization system. The ingredients were mixed in a Kneader-type internal mixer until they became rubber compound.

The obtained compound was taken from the mixer and peroxide was added in order to obtain crosslinking within EPDM matrix and shaped by passing it through the two roll mill. In this way, a uniform dispersion of peroxide in the rubber compound is ensured. The rubber compound is prepared to be 2 mm in thickness by a two-roll mill. 15x15 cm moulds were placed and vulcanized for 5 minutes at 172.15 ° C in a hydraulic press to have the vulcanized rubber. Schematic presentation of composite preparing is shown in Figure 1.

Table 1. Recipes of EPDM composites

Compound	Amount (phr)	Function
EPDM Rubber	100	Raw Material
Carbon Black	100	Filler Material
Mineral Oil	82	Processing aid
Zinc Oxide	5	Activator
Stearic Acid	1,5	Co-activator
TAC/50	2	Co-agent
Peroxide	7	Crosslink agent
Banana peels*	5, 7.5, 10% (w/w)	Bio-based filler material (MP)

** Cellulose polymer obtained from waste Banana Peels*

2.1 Obtaining Bio-Based Filler from Banana Peels

Banana peels are cut into small pieces. The small pieces of banana peel were then placed in a beaker and 250 ml of water was added and heated at 80 ° C for 5 minutes in a magnetic stirrer. When the boiling started, the heating process was terminated and 250 ml of water was added again. After the addition of water, 40 gr was weighed from the remaining precipitate by filtration and heating was continued by adding 20 ml of vinegar. After this process, the sample was taken and dried at 50 ° C for 45 minutes. After drying, the sample was milled in a ball mill. The completely powdered samples were sieved with a size of 250 μm.

2.2 Characterization

The effect of bio-based filler material on EPDM was investigated. For this purpose, Neat EPDM (Control sample) and EPDM with different bio-based filler content (5, 7.5 and 10% MP) were characterized in terms of mechanical and rheological properties.

Tensile test was performed on samples cut into 'Dogbone' according to ASTM D412 standards by applying a force of 50 mm/min in Shimadzu AGS-X Tensile Testing Machine tester. The rheological properties of the samples containing cellulose polymer obtained from waste banana peels at various content were determined by vulcanization at MDR 3000 Basic at 172,15 ° C for 5 and 10 minutes. In order to examine the MDR results, the Cure rate index formula was used.

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

In Equation 1, t_{90} defines the optimum vulcanization time and ts_2 defines the scorch time.

The crosslink density of composites prepared is determined by Soxhlet Extraction method as given in Equation. In the Soxhlet Extraction analysis, it is intended to dissolve the non-crosslinked moieties of the suspended materials in the solvent (hexane), measure the amount of remaining mass, and thus find the crosslinking percentage.

$$\text{Gel Content} = (W_f/W_i) * 100 \quad (2)$$

In Equation 2, W_f represents the final weight and W_i represents the initial weight.

3. Results and Discussions

3.1 MDR Test Results

The vulcanization parameters of the EPDM samples prepared using the MP at different ratios are given in Table 3 and the torque curves are demonstrated in Figure 2. As seen in Table 2, It is seen that the Neat EPDM (Control Sample) and EPDM with different MP content have similar rheological properties. When the CRI values are examined, it is once again understood that the optimum value of EPDM with 7.5 MP, which is close to each other. This result is compatible with both the mechanical analysis results and the crosslink density obtained from Soxhlet extraction.

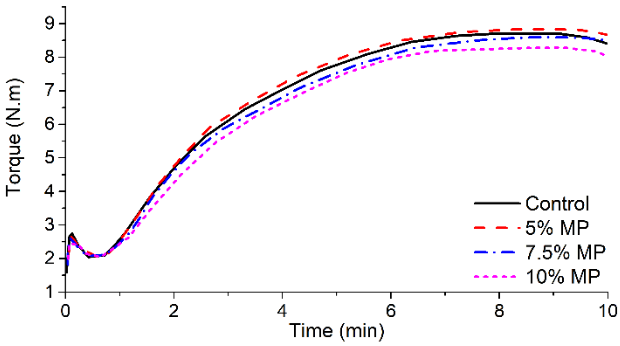

Figure 2. Torque curves of composite materials

Table 2. Vulcanization parameters of EPDM and EPDM with different MP content

Sample Name	t_{s2} (min)	t_{90} (min)	CRI
Control	1.01	7.85	14.62
5% MP	1.06	7.98	14.45
7.5% MP	1.11	7.88	14.77
10% MP	1.06	8.25	13.91

3.2. Soxhlet Analysis Result

Gel content of the Neat EPDM (Control Sample) and EPDM with different bio-based filler content is given in Table 3. It is observed that the cellulose polymer obtained from the banana peels (MP) slightly contributed the crosslinking in the EPDM composites. It has been understood that when the MP ratio exceeds 7.5%, it affects the gel content negatively. According to the obtained results of the Soxhlet Extraction, it is considered that this could be possible of the decrease of the interaction between the surfaces.

Table 3. Soxhlet extraction results

Compound	Gel Content (%)
Control Sample	79.21
5% MP	79.14
7.5% MP	80.26
10% MP	78.89

3.3 Mechanical Analysis Results

Mechanical test results of samples are given in Figure 3. When the modulus of elasticity analysis result was examined, the modulus of elasticity values increased as the MP addition increased. However, modulus of elasticity value was decreased for 10% MP case. As a result, it was understood that the biopolymer used was compatible with the EPDM matrix and increased the modulus of elasticity values.

When the elongation at break is taken into account, the best result is for EPDM with 5% MP (approx. 450%). The second best result is EPDM with 7.5% MP. When both analysis results were examined, it was selected that the optimum bio-based filler content was EPDM sample containing 7.5 MP according to tensile test results.

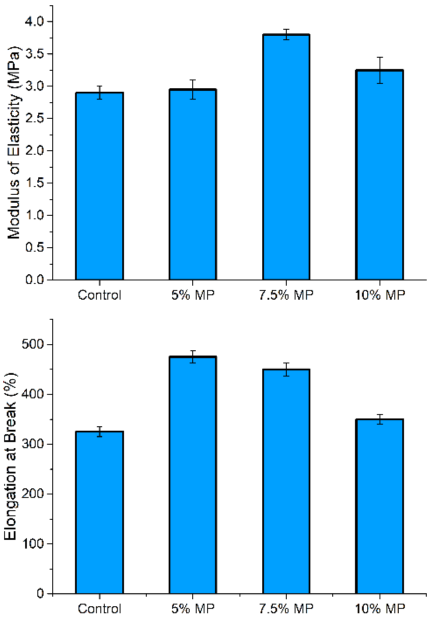

Figure 3. Mechanical properties of the samples

3.4 Investigation of the Use of Cellulose Polymer Produced from Waste Banana Peels as a Filler Material Instead of Carbon Black

In this part of the work the availability of cellulose polymer produced from banana peels instead of carbon black material with cancerogenic effects has been examined and analyses results are given Figure 4 and Table 4. During the study, the amount of carbon black used was 30 phr as given in Table 1. At this stage, MP was gradually added to the EPDM instead of carbon black as the total amount of filler material was 30 phr. When Figure 4 and Table 4 are taken into consideration, it is seen that the vulcanization parameters of EPDM containing MP composites give close results with the control sample. As the amount of added MP increases, the value of t_{90} decreases but the value of t_{s2} increases, which is a desirable result. In addition, CRI results also a similar trend. For this reason, EPDM rubber with 20 phr CB + 10 phr MP was selected as the optimum compound according to high CRI value and smaller t_{90} value compared with control sample. In the light of these results, it is understood that MP could be used instead of carbon black.

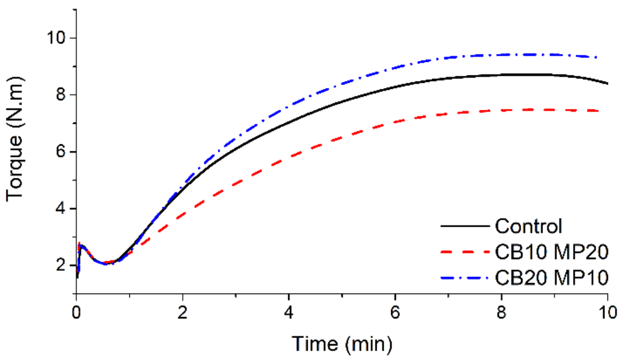

Figure 4. Effect of carbon black change on EPDM torque curves

Table 4. Effect of carbon black change on rheological properties

Sample	t_{s2} (min)	t_{90} (min)	CRI
Control Sample	1.01	7.85	14.62
CB20 MP10	1.03	7.42	15.65
CB10 MP20	1.08	8.03	14.39

3. Conclusions

In this study, the waste banana peel was used for the disposal of wastes as well as for reducing the carbon content in EPDM rubber. Firstly, the biopolymer was synthesized from banana peel wastes and then synthesized biopolymer as a filler material was added EPDM rubber at different ratios. Mechanical, chemical and rheological properties of Neat EPDM and EPDM with different biopolymer filler content were investigated and optimum parameters were chosen. Mechanical tests results showed that with the increase of biopolymer content was increased the mechanical properties of the final material. Additionally, Soxhlet extraction test result indicated that the crosslink density of the samples containing MP up to 7.5% was increased while the crosslink density after 7.5% was decreased, which this result is compatible with mechanical analysis. In the MDR results, the rheological properties were improved by the addition of the polymer obtained from the banana shell. Furthermore, MDR analysis results showed that EPDM samples with 7.5% MP content are selected the optimum composite material owing to has higher CRI and t_{s2} value and smaller t_{90} values. It was understood that the biopolymer obtained from the banana peels could be used instead of the carbon black within the EPDM rubber and thus, the negative effect of carbon black on human health will be reduced. Consequently, it was concluded that the biopolymer obtained from banana peels is an effective filler material along with bio-based, biodegradable and environmental friendliness.

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Abbreviations

MP : Cellulose polymer derived from banana peel

CB : Carbon Black

phr : Parts per Hundred Rubber

CRI : Cure Rate Index

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