BLENDING PROCESS OF RECYCLED EPDM AND EVA AT VARIOUS RATIO

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ABSTRACT

A blending process of polymers had been conducted using recycled synthetic rubber, EPDM, and copolymer, EVA. EP DM stands for Ethylene Propylene Diene Monomer rubber while EVA stands for Ethylene Vinyl Acetate. The blending process between these two polymers are quite challenging due to cross-linked recycled EPDM and the blending process is between thermosetting and thermoplastic plastics, where the thermosetting plastic is eventually, can not be molded and remolded. The main objective of this experiment is to synthesize a composite using blending process and to investigate the mechanical properties of the composite. The mixture of EPDM and EVA was 0:100, 10:90, 20:80, 30:70, 40:60 and 50:50 wt%. The resultant products were then collected and molded with a hot and cold press machine into desired shapes for mechanical testing namely tensile test and hardness test. During the blending process, the incompatibility of both thermosetting plastic and thermoplastic needed to be solved. Three compatibilization techniques had been performed and the optimum conditions for the blending process obtained are 160°C for the operating temperature, 50 rpm for the speed capacity, total 200.0 g of specimens, 400 Nm for the torque mean, and time duration for the blending process of 30 minutes. The best ratio composition of blending was determined by assessing tensile and hardness testing properties. As a conclusion, 20:80 wt% of recycled EPDM and EVA was found to be the best blending composition based on the best tensile and hardness testing properties compared to other compositions. However, further work can be improved such as to minimize the particle size of EPDM, in order to enhance the rate of the EPDM being heated and being homogeneously blended.

Keywords: blending; EPDM; EVA; mechanical properties.

INTRODUCTION

Rubber has been commonly and widespread used in the world, ranging from the simplest industries such as households to many other important industrial products. Rubber has its own history, since the earliest dating to about 1600 B.C., rubber has been collected for a long time in its native Central America and South America. Since then, rubber has become one of the most important material used widely daily in our lives. Rubber then had been vulcanized into a new product, which is called a synthetic rubber. Natural rubber is often vulcanized, a process by which the rubber is heated and sulfur, peroxide or bisphenol are added to improve resilience and elasticity, and to prevent it from perishing. Vulcanization greatly improved the durability and utility of rubber. The successful development of vulcanization is most closely associated with Charles Goodyear. Carbon black is often used as an additive to rubber to improve its strength, especially in vehicle tires.

Synthetic rubbers have been used widely in our days, especially in automotive industries. However, humans face troubles to treat the rubber waste while the use of large consumption of synthetic rubbers had been possessed in present days. Rubber waste generated from in-process or scrap products has now risen to more than 30 million tons in the world [1]. For the sake of caring and protecting the environment in our world, recycling of rubber waste has become an important issue and study in recent years. However, when the natural rubber is processed into synthetic rubber, which posses for a better use in the industries, the three-dimensional network structure called “crosslinking” is constructed which contributes to rubbery elasticity [2]. This network structure makes processed rubber difficult to recycle and reuse as a raw material. The recycled rubber is found to have a very poor attributes of quality and its productivity as well as low usage. Hence, the study of how to use the recycled synthetic rubber to produce a new useful product is very important and interesting study in polymer fields. One of the most interesting researches, is sulfur cross-linked Ethylene Propylene Diene Monomer (EPDM). Among the synthetic rubbers such as: TPO (EPDM/PP), EPDM/SBR, EPDM/PE and others, EPDM occupies more than 50% of rubber components that are used in vehicles except tires. EPDM can provide longevity; resist the effects of ultraviolet light and costly material [1].

One of the important ways to mix and produce new polymer material is blending process. Blending of polymers is a well-known, economically viable and versatile way which allows to overcome certain deficiencies of the
parent material by incorporating other suitable component and also to develop new structural systems by mixing
different polymers [3]. Polymers blend together to form a new product which has good properties of both
polymers and eliminates any worse properties of the polymers. The blends of elastomers are usually vulcanized
by using conventional chemical methods to improve their mechanical and physical characteristics. For the
reason of environmental friendly and also considering for large quantity of producing a new product which can
be produced from the recycled synthetic rubber EPDM, condition such as low melting temperature, atmospheres
pressure, etc, are more encouraging in order to produce large amount of product in low costs.

In this work, the blends of Ethylene Vinyl Acetate (EVA) with recycled synthetic rubber, EPDM, are of special
interest because incorporation of suitable amount of recycled EPDM in EVA is expected to impart significant
heat and ozone resistance to EVA matrix. On the other hand, EPDM has weak adhesion property and the
products based on sulfur vulcanized EPDM have poor tear strength. Hence, the blending process in different
aspect ratio to the induced vulcanization of recycle EPDM/EVA blends would combine their desired properties
including high mechanical strength.

MATERIALS AND METHODS

In this experiment, the properties of resulted product from the blending process of recycled EPDM and EVA
will be investigated. The methods are divided into four stages, which is blending process, blending with
different weight ratios of recycled EPDM and EVA, hot and cold pressing, and also, mechanical testing.

Blending of Recycled EPDM and EVA

Three different methods of blending were performed to determine the best blending that can produce good
interactions, links between EPDM and EVA and good surface of the blending mixtures.

1. Operating temperature is 190°C, mixing duration period of time is 10 minutes, rotor speed is 50rpm,
   and the sample (EVA and recycled EPDM) was put into the Brabender at the same time, without
   mixing them heterogeneously first.

2. Operating temperature is 190°C, mixing duration period of time is 10 minutes, rotor speed is 50rpm,
   and the EVA is put into the Brabender for the first 2 minutes for preheating then followed by the
   recycled EPDM.

3. Operating temperature is reduced to 160°C, mixing duration period of time is extended to 30 minutes,
   rotor speed still remain as 50rpm, and the sample (EVA and recycled EPDM) is mixed heterogeneously
   before put into the Brabender.

After 20 minutes, the sample was taken out, and being cut into small pieces while the sample was still hot. This
step must be completed in a very short period of time because the sample would hardened and difficult to cut off
after it was cooled down. The sample was then collected and labelled.

Blending with Different Ratios of Recycled EPDM and EVA

The best blending method chosen from the first part will be used with different weight ratios of recycled EPDM
and EVA namely 0:100, 10:90, 20:80, 30:70, 40:60 and 50:50.

Hot and Cold Press

Different amount of samples were needed for different mechanical test. For hardness test, the exact amount to
produce one rectangular block of sample is equal to dimension x EVA’s density, and it is found that 55.8g of
sample is needed for each time press (10cm x 10cm x 0.6cm x 0.93g/m³). Whereas for tensile test, the amount of
sample needed was 20.925g to obtain a size of 15cm x 15cm x 0.1cm specimen (15cm x 15cm x 0.1cm x
0.93g.m⁻³). This step is important to make sure the sample after the hot and cold press is in a prefect rectangular
form and no air bubble per piece sample for each time press. The step is considered failure if the sample piece is
found with air bubbles within, or does not fully occupy the mold.

The weighed sample was put between 2 thin plates before put into the hot and cold press machine. The sample
was heated and pressed for 5 minutes, and followed by cooling press for 2 minutes at 125°C. Lastly, the sample
in block shape was taken out.
Tensile test

The tensile testing specimens were cut into dumbbell shape and the tests were conducted according to ASTM 1822L using INSTRON (Model 4301) Universal Testing Machine with load cell of 1.0KN, using a crosshead speed of 50mm/min. The cross-head speed during the tension was 50mm/min with load cell of 1.0KN, and each test was performed until tensile failure occurred.

Hardness Test

The Rockwell hardness tester (Model HA-101) was used for measuring the relative hardness of soft materials. The test was done according to ASTM standard D2240. Rockwell hardness was measured using a 12.7 mm ball and 60 Kgf (588.4N) of indent force (Rockwell scale L). Specimens were conditioned at 25°C ±50 % relative humidity (RH) prior to hardness testing. Seven specimens of each type were tested and at least five replicate specimens were presented as an average of tested specimens.

RESULTS AND DISCUSSION

Selection Criteria for the Blending Process

The major challenge to blending recycled EPDM with EVA is the incompatibility between the two materials; a design of experiments based on the processing conditions and variables was performed to determine the optimum processing conditions [2]. From the observation of the product, the blending outcome always non-homogeneous that recycled EPDM particle embedded in EVA matrix.

By comparing all three outcome samples with different blending parameters, the third method was considered as the best result. This is because the sample after the blending process seemed to be more “compatible” and “homogenous”. More “homogenous” had been stated because of the observation on the colour of the sample in the third method which was darker than the other two. Since the pure recycled EPDM is black in colour, whereas EVA is white in colour, from the observation, the recycled EPDM was “dissolved” more in EVA in the third method of blending process. So, the parameters in the third method were chosen for the rest of the blending process.

Figure 1 shows the picture of blending sample after the blending process using method 1, method 2 and method 3.

![Sample of method 1](image1.png)  ![Sample of method 2](image2.png)  ![Sample of method 3](image3.png)

Figure 1: Photos of Blending Samples at Different Blending Processes

Rheological properties of elastomer modified polymer blends are of prime importance as their understanding would help in optimizing the commercial production of the blends [3]. The parameters such as various grades of EVA and EPDM, their compositions, mixing time, mixing temperature and rotor speeds have contributed significant effects on the rheological properties of the blends [4]. So, it is important to use suitable conditions for optimizing the blending process.
These conditions are based on the general knowledge on properties of recycled EPDM and EVA; EVA is melting at 80°C, degradation will occur at temperature higher than 200°C, and the mixing time should not be too long [4].

Based on the torque–time curves (Figure 2) obtained in blending at 50rpm for the three different methods, the torque shown in the figure are initially the highest at the early state of blending. While the time increased, the torque started to decrease until it reached to a point and remain constant throughout the mixing time. By comparing the results from the graph, the torque in the third method possessed the highest point among two other methods, following by first method and lastly is the second method, in the beginning of blending process time. The torque was high in the beginning because during the early state due the mixture of EVA and EPDM had not melted well, thus, the blending machine needed more energy to move the blade to blend the mixtures. This is because the EVA took some times to completely melt and the recycled EPDM could not melt well because of its cross-linked and thermosetting plastic characteristic. As the time goes on, the mixtures begin to melt. This shows that the mixture melted not only proportional to the temperature, it also could be related to the time of heating. This is because the particle which was initially cool needed time to be heat, until it reached its melting point. However, since no strong evidence states that the recycled EPDM could not 100% be molten, the effects of the recycled EPDM in the chamber of Brabender blending machine should be taken into consideration. The high value of torque at early state mixing process was caused by resistance exerted by the recycled EPDM against the rotor. As the recycled EPDM became melted and subjected to mechanical shearing [4], the temperature inside the chamber increased, resulting in the reduction of torque value, and became constant at last.

In other specs of view, the lines between first method and the second method in the torque-time graph could be compared and it seemed like the sequence of step to put the sample into the chamber of the Brabender would cause another result of graph patents. In the first method, the sample which consists of both material, recycled EPDM and EVA which was not yet mixed well (rather just put in a beaker, the upper part is recycled EPDM and the lower part is EVA) were inserted together into the Brabender at the early stage. What happened inside the chamber was, there was high resistance between recycled EPDM and rotor, and reduced abruptly with time, longer time that was needed to become constant compared to the second method. Whereas for the second method, which inserted the recycled EPDM at first for 2 minutes to melt and after that followed by EVA in the later. A lower torque graph line was determined for mixing, and the torque value become constant faster. This can be conclude that different way or sequence the sample was put in to Brabender would lead to a different shape of curve in torque-time graph.

As a logarithmic of torque (log (τa/S)) as a function of inverse of temperature (1/T(K⁻¹)) graph was plotted using the torques taken after 10 and 30 minutes of blending process, a relationship was found, which is similar to that of Arrhenius. A linear relationship was observed for the data corresponding to 10 minutes of mixing for both first and second methods, which might be due to the insignificant changes that had occurred at such low residence time and temperatures [4]. But for the 30 minutes of blending process (method 3), a non-linear graph patent was observed. Tangents were drawn on the non-linear sections which gave different slope values,
indicating the different flow of activation energies. In this case, those varies of the graph might be due to some morphological transitions in the polymer [4]. Thus, beyond all these reasons, the third method was the best method to blend the recycled EPDM and EVA.

Besides the abovementioned factors which affects to the compatibility of the blending process between EPDM and EVA, the particle size of the rubber must also be considered. Rubber particle size is known to affect the mechanical properties of recycled rubber/plastic blends, thus, this has been attributed to a reduction in the inherent flaw size of the resultant composite or to better interfacial contact [2]. This is very important because, when the size of the particles of the EPDM is smaller, the total surface that contact with the heater in the chamber become higher. The recycled EPDM will absorb more heat, and also the recycled EPDM could be blended more thoroughly, hence, the kinetic of the reaction of EPDM would also increase. But, unfortunately, in this case, the minimum size of the particle used in this research is it still considered big.

From the observation based on Figures 2 and 3, although the best method (method 3) was chosen to set up the optimum condition for the blending process, 100% homogenously blended product sample could not be obtained. The collected product samples were in a composite form, but could not be considered as a new homogeneously blended polymer product (see Figure 1).

**Blending Composition**

The blending composition between the recycled EPDM and EVA is the main focus of this report. The blending composition possesses the important factor which affects to the properties of the product samples. In the lab, the sample of recycled EPDM and EVA was blended at different weight ratios of 0:100, 10:90, 20:80, 30:70, 40:60 and 50:50. The composition of EPDM in the composite will give a various physical properties effect to the composite.

**Tensile Test**

From the tensile test, the physical characteristic of the polymer in three aspects namely Young Modulus, Elongation Break and Tensile Strength were obtained.

**Young Modulus**

In scientific words, Young Modulus represents the stiffness of a material. From Figure 4, the Young Modulus increased as the percentage composition of EPDM increased, for the percentage composition from 0% to 20%. However, after 20% of composition of EPDM in the mixture, the Young Modulus decreased until 40%, and it
rose again at 50%. To describe this phenomena, the graph was divided into three regions, which are A, B and C, as shown in Figure 4. What happened in A is, increasing the percentage of EPDM, increased the Young Modulus, and also increased the stiffness of the polymer. This was happened, because the EVA itself had high degree of crystallinity, which had the properties of high stiffness. When small amount of EPDM added to EVA, yet contained of EVA still remained as majority in the composite, leading to the increment of the stiffness of the mixtures. As described earlier, EPDM has permanent elastic properties, which can slightly increase the stiffness property of the mixture. However, as in region B, the Young Modulus had been decreased as the composition of EPDM increased. This is because the composition of EVA had become less in region B, thus, the stiffness properties of EVA became less influenced. However, in region C, the Young Modulus increased again. As the composition of EPDM in the mixture increased until 50%, the effect of the properties of EPDM increased. However, the stiffness of EPDM was less than the stiffness properties of EVA. Hence, the highest point of Young Modulus obtained in region C was less than what is in region A.

Figure 4: Young Modulus at Different Composition of Recycled EPDM

Elongation Break

As shown Figure 5, elongation break of the product sample decreased dramatically as the percentage of composite of EPDM increased. Theoretically, elastic behavior of EPDM was expected to increase the elongation break of the product sample. The higher the elastic properties of the material, the more energy required to break it. EPDM, a synthetic rubber, is a material which is capable of recovering from large deformations quickly and forcibly. A rubber will react, within one minute, to less than 1.5 times its original length, after being stretched to twice its length and held for one minute before release [5]. However, the product sample (matrix) obtained was not fully homogenously blended. Hence, the characteristic of the EPDM could not fully perform in the composition; inversely, it became a deflection to the composition of the product sample. As the experiment went on, the place that the elongation break occurred was the point that the particle of EPDM took place. Hence, for the elongation break, the result was not satisfied with the early expectation. However, if the size of the particles of the EPDM could be reduced, most probably the displacement of elongation break of the product sample would be decreased. But somehow, new but economic technology should be developed to minimize and reduce the size of the recycled EPDM to achieve the target of homogeneously blended composite.

Tensile Strength

Tensile strength of a polymer is the maximum or ultimate tensile stress in order to cause fracture or permanent deformation to the polymer. Hence, tensile strength is closely related to the displacement of elongation break of the polymer. The higher the tensile strength, the higher the displacement of elongation breaks of the polymer. The inhomogeneous of the composite recycled EPDM and EVA due to the incompatibility of the recycled EPDM (cross-linked, hardly melt), caused deflection to the product sample.

The particles that did not melt became the deflection point to the product sample. The concept is just like putting impurities into a rubber, which cause the decrease of internal elasticity of the material.
Hardness Test

Hardness test describes the resistance of a material to localized permanent (plastic) deformation. Based on the observation from Figure 7, the hardness of the product sample decreased and is inversely proportional to the percentage of composition of the recycled EPDM in the product sample. The more recycled EPDM in the product sample, the softer the product sample. This is because the recycled EPDM behave like elastomers, which is not hard, but elastic. Besides, the presence of the recycled EPDM in the product sample would decrease the existing of the EVA. In other words, more recycled EPDM would cause less amount of EVA present in the composite product as the total amount of initial sample before blending remained at 200.0g. The lesser the presence of EVA in the product, the hardness of the product would be reduced.

However, there is slightly change of the graph pattern in the 50% composition of recycled EPDM. This is because the elastomers crystallinity still has little effect on the surface hardness [5, 6]. When more EPDM present in the product, it actually would increase small changes to the hardness property. The increment did not apparently and significant, but still could be observed when large quantity of recycled EDPM present. Hence, the increment of the hardness from the contribution of the recycled EPDM could be observed when the quantity of recycled EPDM was added unto a certain level.
CONCLUSIONS

During the past few years, there has been substantial progress in the recycling of polymeric materials. Unfortunately, progress in the area of recycling thermosetting polymers, such as rubbers, has not been as successful, since these materials cannot be reformed once they have been cross-linked. As a result, other methods to recycle the rubber must be found. One of the methods to recycle a thermosetting plastic is grinding. The ground rubber can be used alone or mixed with thermoplastics to achieve the desired properties, such as impact modification.

In this research, the best method to obtain the best compatibility and homogeneous mixture of recycled EPDM and EVA is as follows:

- Operating temperature = 160 °C
- Operating speed capacity = 50 rpm
- Operating hours = 30 minutes
- Special condition = recycled EPDM and EVA must be mixed together initially before feeding into the Brabender chamber

This research also found that as higher percentage composition of recycled EPDM, more energy needed in the blending process due to difficulty of melting the recycled EPDM. Based on the finding from the tensile testing, the characteristic of recycled EPDM and EVA affected each other accordingly. An optimum Young Modulus was determined at 15:85 wt% of recycled EPDM to EVA. Furthermore, the elongation break and tensile strength of the product were very poor. This might be due to the defect of the particles which did not melt during the blending process. Consequently, the blended product was too easy to be broken. In addition, the hardness was inversely proportional to the percentage composition of the recycled EPDM in the product sample. However, at 50% recycled EPDM in the blended mixture, a noticeable effect of the recycled EPDM was occurred leading to a small increase to the hardness property of the mixture product.

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REFERENCES


NOMENCLATURE

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<tr>
<td>EPDM</td>
<td>Ethylene propylene diene monomer</td>
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<td>EVA</td>
<td>Ethylene vinyl acetate</td>
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<tr>
<td>rpm</td>
<td>Revolution per minute</td>
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<td>TPO</td>
<td>Polyolefin</td>
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<td>PP</td>
<td>Polypropylene</td>
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<td>PE</td>
<td>Polyethylene</td>
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<td>S</td>
<td>Rotor speed, rpm</td>
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<tr>
<td>SBR</td>
<td>Styrene butadiene rubber</td>
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<td>τa</td>
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