The Study of Recycled Ethylene Propylene Diene Monomer (EPDM-r)/Polypropylene (PP) Polymeric Blends

MH Zulkifli1, MSM Rasidi1,2, NAM Rahim1, L Musa1,2, and Abdul Hakim Masa3

1Faculty of Chemical Engineering, Universiti Malaysia Perlis, Arau, Perlis 02600, Malaysia
2Geopolymer & Green Technology, Centre of Excellence (CEGeoGTech), Universiti Malaysia Perlis (UniMAP), Perlis, Malaysia
3Sino-Thai International Rubber College, Prince of Songkhla University, Hat Yai, Songkhla, 90110 Thailand

Abstract. In this study, recycled ethylene propylene diene monomer (EPDM-r) were blended with polypropylene (PP) by compounding via heated two roll mills with the various ratio of EPDM-r. Certain blends were included with PP-g-MA as a compatibilizer. In tensile tests, the increasing of EPDM-r content in blends resulted in the reduction of tensile strength and Young’s Modulus but increased elongation at break. Furthermore, the presence of compatibilizer in blends enhanced the tensile properties. It was found all the samples with compatibilizer performs better results in tensile strength, Young’s Modulus, and elongation at break than samples without compatibilizer. On the other hand, the aging affected were studied on compatibilized and uncompatibilized blends. It was found that aging affects the samples by decreasing the tensile strength, Young’s Modulus, and elongation at break. The crosslink density had been found higher in the blends with high EPDM-r content as the EPDM-r had the ability to swell. The morphological study related to the structure with the tensile properties. It was confirmed that the presence of a compatibilizer increased the compatibility between EPDM-r and PP matrix.

1 Introduction

Materials that combine the properties of plastic with rubber-like behavior are known as thermoplastic elastomers. Industrial rubbers have to be vulcanized in order to provide useful properties. This is a slow, irreversible process, and heating takes place. However, in thermoplastic elastomers, the transition from a processable melt to solid rubber-like object is fast, reversible, and cooling takes place. In common solvents, certain thermoplastic elastomers dissolved and when the solvent evaporated, they can recover their properties [1]. Since Fischer first proposed dynamically vulcanized thermoplastic elastomers (TPVs), they have been globally used in the rubber and plastic industries [2]. Among them, are those prepared by melt mixing of rubber with polyolefins such as polypropylene which classifies as thermoplastic elastomer has gained more acceptance because of the ease of preparation. Polypropylene (PP), used in a wide scope of applications, is a thermoplastic polymer [3]. It is generated from the monomer propylene through chain-growth polymerization.

1Corresponding author: syahmie@unimap.edu.my
Polypropylene is part of the polyolefin group and is partly crystalline and non-polar. PP has strong mechanical properties but lacks properties of temperature impacts. Mechanical properties and thermal resistance are enhanced by the methyl group, whereas the chemical resistance decreases. Consequently, rubber toughening is often used to improve the impact strength of polypropylene. Improvement in the impact strength of polypropylene has been sought widely by melt blending with elastomer [4].

The synthetic rubber ethylene propylene diene monomer (EPDM) is suitable for use in wide applications. It is resistant to cuts and tears and can compress well and recover completely. EPDM exhibits excellent puncture, high tensile strength, weathering, ultraviolet radiation, and microbial attack resistance [5]. It is also a highly versatile material with a low thermal expansion and contraction coefficient, allowing it to lie flat in a wide range of temperatures and terrains, thus conforming well to the sun-grade irregularities. This is because of its exceptional ability to accept high filling loads and its good resistance to oxygen, ozone, UV, and heat [6]. Some modifications apply to enhancing the polymer's properties such as mixing. The use of EPDM-PP blends has been steadily growing for several decades in various industrial fields.

In some parts of the world, a decrease in usable landfill space and increasing disposal costs have led to the need for alternative solutions. U.S. There is 40 percent of overall landfill waste from building and demolition waste. One-quarter of which is created by materials for roofing. While landfill space is abundant nationwide, some areas, such as the heavily populous East Coast face serious capability problems and have seen the cost of disposal escalate. Over 20-30 years, they decompose slowly. Apart from harmful additives in PP such as lead and cadmium, this case poses major environmental concerns [7]. Dioxins and vinyl chloride, both of which are toxic, can be released by incineration. To solve the problems, blending PP with other materials could reduce the usage it immediately reduce the PP waste. It is estimated that approximately 1.4 billion tires are manufactured globally on an annual basis [8]. Tires disposed of in landfills have associated socio-economic and environmental concerns. As a result of their incompressibility, tires require a high volume of landfill space. In addition, they provide a breeding site for mosquitoes and rodents [9,10]. Due to their tough characteristics, tires are immune to degradation. As all aware, recycling of vulcanized rubber had been a major discussion as it is difficult to be reprocessed and recycled. Currently, the best method to utilize rubber waste is to reuse it. Therefore the waste EPDM is been used as a part of a blend to produce new materials.

The main concern of blend two different types of materials is the compatibility and interfacial adhesion between both materials such as compatibility between elastomer and polymer matrix which determines the particle size of an elastomer in the blend and the interfacial interaction between components and the intrinsic characteristic of a matrix which usually determines the energy absorption mode under the load condition. The common strategy is introducing the third polymer component which acts as the compatibilizer to enhance the interfacial adhesion between components [11].

2 Materials and Method

The materials used in this study were EPDM-r which was obtained from NT-Rubber Seals Sdn. Bhd. (Jitra, Kedah), polypropylene type SM 240 which was purchased from Lotte Chemical Titan (M) Sdn. Bhd and PP-g-MA were supplied by Sigma Aldrich (M) Sdn. Bhd.
2.1 Preparation of EPDM-r/PP blend

EPDM-r was crushed using two roll mill machine and was sieve with 125 μm mesh size to obtain a finer course. Then, the EPDM-r/PP blends were compounded with different formulations using heated two roll mills. The formulation were shown in Table 1.

| Specimen number | EPDM-r (php) | PP (php) |
|-----------------|--------------|----------|
| 1               | 100          | 0        |
| 2               | 90           | 10       |
| 3               | 70           | 30       |
| 4               | 50           | 50       |
| 5               | 30           | 70       |
| 6               | 10           | 90       |
| 7               | 0            | 100      |

All specimens were compounded at a 180°C processing temperature and residence time of 2 minutes. After that, all types of samples were sheeted at 3 mm mold by using GT-7014-P GOTECH Plastic Hydraulic Molding Press with compression temperature and pressure of 180°C and 10 tonnes. The pre-heating, compression molding, and cooling times were 6 minutes, 10 minutes, and 7 minutes respectively for all samples preparation. For compatibilized EPDM-r/PP blends, the compatibilizer was added during the mixing process. The formulation of compatibilized EPDM-r/PP blends were shown in Table 2.

| Specimen number | EPDM-r (php) | PP (php) | PP-g-MA (php) |
|-----------------|--------------|----------|---------------|
| 1               | 90           | 10       | 5             |
| 2               | 70           | 30       | 5             |
| 3               | 50           | 50       | 5             |
| 4               | 30           | 70       | 5             |
| 5               | 10           | 90       | 5             |

2.2 Tensile Test

Instron Universal Testing Machine, model 5960, was used to conduct the tensile test which was based on ASTM D412. The cross-head speed of the testing was at 500 mm/min. Five replicates were tested for each sample. The size of each sample was 115 x 6 x 3 mm. The mean values of the tensile strength, elongation at break, and Young’s Modulus were recorded.

2.3 Ageing Characterization

An aging test has conducted another set of both compatibilized and uncompatibilized EPDM-r/PP blends in an electric thermal oven. After aging, the mechanical properties of EPDM-r/PP samples were tested with a tensile test again to study the aging performance. The condition of the aging test was set at 125°C for 72 hours. The samples having aging tests were compared to samples which not having aging tests then to see the differences.
2.4 Swelling Test

A swelling test was performed by immersing the specimens at room temperature with toluene for 24 hours. The samples were removed at the end of the immersion time, using paper towels to dry the sample, and transferred to the weighing bottle to obtain the swollen weight of the samples. Flory-Rehner equations have been used to obtain the crosslink density value of all EPDM-r/PP blends.

2.4 Morphology Test

The morphological study of a tensile fractured surface of the EPDM-r/PP blends was conducted by using a Scanning Electron Microscope (SEM), Model JSM 6260 LE JOEL. The tensile fracture surface of the specimen was mounted with aluminum stubs after sputter-coated with a thin layer of palladium to avoid electrostatic charging while examine. The SEM micrograph at 10 kV was obtained.

3 Results and Discussion

Fig. 1. exemplifies the tensile strength of the EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging. It showed that the tensile strength of both compatibilized and uncompatibilized blends was gradually decreased with increasing EPDM-r content. This happened because the EPDM-r does not promote higher ability against breaking compared to PP. Other than that, it was found that aging compatibilized samples having lower tensile strength compared to non-aging compatibilized samples. The presence of compatibilizer into blends resulted in better tensile strength due to enhanced bonding between EPDM-r and PP matrix. The thermal aging lowered the tensile strength of all aged samples. The oxidation of the polymer, which resulted in chain scissions, was blamed for the loss in tensile strength at a given blend ratio. Scission of the larger molecular chains increased the number of shorter chains of the respective polymer. As a result, there were fewer entanglements and lowering tensile strength [12].

Fig.1. The tensile strength of EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging

Fig. 2. depicted the elongation at break of EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging. It is shown that the samples which having high EPDM-r content resulted higher elongation at the break because the properties of EPDM-r were elastic. Other than that, compatibilized EPDM-r/PP blends yield the highest elongation at break than the aging compatibilized EPDM-r/PP blends followed by uncompatibilized EPDM-r/PP blends. The reason samples with compatibilizer produced better elongation was the bonding between EPDM-r and PP was stronger and having higher resistance against breaking and managed to elongate longer. It was observed that the aging of the rubber phases degenerated the cross-
linked structure of the entire system and could sustain weaker stress during the stretching process and resulted in lower elongation before break compared to the compatibilized EPDM-r/PP blends.

It was found that the properties of the blends decreased gradually with increasing the period of aging time and temperature, possibly due to the degradation of the polymers [13]. The hardness of the aged blends was higher than that of the unaged. This may be attributed to the crosslink density which increased due to the thermal aging, indicating the post-vulcanization reaction during aging [13]. However, it was confirmed by the increase of rebounding value after thermal aging.

**Fig. 2.** The elongation at break of EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging.

Fig. 3. shows the Young’s Modulus of EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging. The Young’s Modulus of samples was found to decrease with increasing in EPDM-r content. This was because as in the case of EPDM-r required a small amount of force to produce large deformation while in the case of PP need more force to produce even a slight deformation. Moreover, the Young’s Modulus of compatibilized samples were highest while aging compatibilized samples and controlled samples were about to same. The compatibilized samples were stiffer than uncompatibilized samples. However, with the aging process, the samples became softer because of the temperature in the aging process. The effect of thermal aging reduces the modulus strength [14] and thus reducing Young’s Modulus value.

**Fig. 3.** The Young’s Modulus of EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging.

Fig. 4. shows the crosslink density of the EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging. The crosslink density of EPDM-r/PP blends was higher at higher EPDM-r content due to solvent likely to absorb into EPDM-r. EPDM-r molecules eventually became insoluble in toluene because chemical bonds have to be raptured to break the rubber when stretched. Other than that, compared to every type of sample, uncompatibilized blends
had the highest crosslink density followed by compatibilized blends and aging compatibilized blends overall. The presence of compatibilizer between EPDM-r and PP molecules reducing ‘space’ between molecules which makes these blends bonded uniformly and resulted in the blends swell lesser. The effect of aging can be seen as reduced crosslink density because thermal cracking happened on the both EPDM-r and PP matrix. It happened due to the increased diffusion rate of oxygen and activated oxidation reaction. The crosslink densities of the blends increased with a further increase in the concentration of EPDM after aging. This phenomenon is possible because of the crosslink precursor and carbon black content of EPDM, which led to a reduction in the free volume between the rubber matrix molecules and therefore led to high solvent swelling [15].

![Graph showing crosslink density vs EPDM-r content](image)

**Fig. 4.** The crosslink density of EPDM-r/PP blend, affected by EPDM-r contents, compatibilizer, and aging

Fig. 5. and Fig. 6. show the SEM micrograph between uncompatibilized and compatibilized EPDM-r/PP with the same EPDM-r/PP ratio at 90:10. It was observed surface of the uncompatibilized blend seems shredded parts of EPDM-r were separately mixed with PP matrix. Unlike compatibilized blend, the shredded parts of EPDM-r were ‘engulfed’ in PP matrix which resulted in the surface of compatibilized blend look smoother than uncompatibilized blend. According to Fig. 1., the tensile strength between compatibilized and uncompatibilized blends was closely related to the blends morphological. It was known that a compatible matrix relationship will yield better performance against breaking. Fig. 7. shows the uncompatibilized EPDM-r/PP blend at the lowest content of EPDM-r (10%). As seen in the figure, the EPDM-r parts were barely to be noticed as they were embedded into PP during the sheeting process. Compared to Figure 5, EPDM-r was more observable as the content of EPDM-r was higher (90%). Figure 8 shows a micrograph of the compatibilized EPDM-r/PP blend at 10% of EPDM-r. Comparing both Figure 8 and 5, the surface of the lower content of EPDM-r seems smoother. This happened was due to the ability of PP to re-melt at its melting temperature but the EPDM-r were not. The vulcanized EPDM rubber cannot re-melt as PP does. This resulted in a smoother surface on a higher PP content blend. Figure 9, shows a micrograph of aging compatibilized EPDM-r/PP blend. The effect of aging slightly changed the surface texture of the blend. Referring to Figure 6, the surface of the blend was smoother than in Figure 9. In Figure 9, the aging process slightly affects the surface by making some unconditional random pores.
4 Conclusion

The research showed the mechanical properties and crosslink density affected by the increasing of EPDM-r content in EPDM-r/PP blends. The result showed the elongation at break and crosslink density increased with increasing EPDM-r content in blends but tensile strength and Young’s Modulus decreased. The presence of a compatibilizer enhanced toughness and resistance over breaking the samples. The compatibilized EPDM-r/PP blends showed higher tensile strength, elongation at break, and Young’s Modulus compared to uncompatibilized EPDM-r/PP blends but for the crosslink density, uncompatibilized EPDM-
r/PP was higher. The incorporation of EPDM-r and PP proved to be effective in enhancing the compatibility of a system comprising of hydrophilic and hydrophobic interaction. It was confirmed by a morphological study in which the EPDM-r matrix and PP matrix finely bonded together with the presence of a compatibilizer. The introduction of the aging test to EPDM-r/PP blends resulted in the degradation of their mechanical properties and crosslink density. It was found that the tensile strength, elongation at break, Young’s Modulus, and crosslink density of compatibilized EPDM-r/PP blends were decreased due to chain scissions between the EPDM-r and PP matrix. As seen in the SEM micrograph, the surface of blends with the aging test was slightly different from the unaged one.

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