

Effect of Ethylene-Propylene-Diene Monomer Blending on the Dielectric Properties of Polypropylene for Power Cable Insulation

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Abstract: Thermoplastic materials such as polypropylene (PP) are ideal candidates to be used as power cable insulation materials. However, the mechanical properties of PP, which is brittle and stiff, need to be modified to meet the requirements of power cable insulation applications. To overcome this, ethylene-propylene-diene monomer (EPDM) can be melt blended with PP to improve PP's mechanical properties. In the current work, the effect of blending different contents of EPDM with PP was investigated in terms of the morphology, chemical structure, dielectric response, electrical, and mechanical properties. The results show that the EPDM blended well with the PP as the EPDM dispersed in the PP structure. Meanwhile, the chemical structure and dielectric response of the EPDM and PP blend became slightly different with high contents of EPDM, which subsequently affected the breakdown strength and mechanical properties. Nevertheless, it is worth noting that PP blended with low contents of EPDM has the potential to be used as recyclable power cable insulation materials.

Keywords: Dielectric properties, ethylene-propylene-diene monomer, insulation material, polypropylene, power cable

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1. INTRODUCTION

Polymeric insulation materials have been extensively used in power cable insulation applications since the introduction of cross-linked polyethylene (XLPE) in 1968 [1]. Due to its excellent electrical and thermo-mechanical properties, XLPE has been widely used for power cable insulation for almost seven decades [2]. However, XLPE is difficult to recycle at the end of its lifetime as XLPE becomes a thermosetting material after the cross-linking process of polyethylene. In addition, the presence of by-products fumes during the cross-linking process also affects global environmental pollution. Even though the by-products can be reduced by the degassing process, the process will increase production costs [3]. Therefore, the usage of XLPE has become a concern due to the issue of environmental sustainability. In the past few years, researchers have begun investigating recyclable polymeric materials that can potentially replace XLPE in power cable insulation, which include polypropylene (PP) [4].

PP is said to be an ideal candidate due to its high melting point, excellent electrical insulation, strong heat resistance, and good corrosion resistance [5]. In addition, manufacturing of power cable insulation using PP does not

require the cross-linking process, in which the degassing procedure can be removed during production. This will reduce the production cost, leading to a reduced cost compared to other polymeric insulation materials [6, 7]. Also, PP offers benefits to the development of recyclable power cable insulation materials because it is easier to recycle after its lifetime [8-11].

Regardless of the aforementioned advantages, the usage of traditional PP as power cable insulation materials is quite challenging. The properties of PP itself somehow do not meet the requirements for power cable extrusion. Thus, further investigations are required to improve its mechanical flexibility while maintaining its electrical performance, especially for power cable insulation applications. For several years, PP based materials have been explored to modify the mechanical flexibility of PP. It was found that the most effective way was to blend PP with a toughening rubber. For example, Zhou et al. [8] investigated the blending of PP with ethylene-based elastomers and found the blends to possess improved mechanical properties. In addition, Gao et al. [12] investigated the effect of two types of elastomers as toughening rubbers on the mechanical properties of PP by using internal mixer blending. Both the PP/elastomer

blends revealed excellent improvements in the mechanical properties as the content of elastomer increases from 10 wt% to 30 wt%. In some other investigations [8, 13, 14], improving the PP mechanical properties by blending with rubber elastomers resulted in poor compatibility. Of note, physical defects can exist in the microstructure of PP/elastomer blends, which will cause degraded electrical properties [15-17].

Traditional rubber such as ethylene-propylene-diene monomer (EPDM), which is an ethylene-based elastomer, is often used to improve the mechanical flexibility of PP in wide applications. It has better puncture, tensile strength, weathering, ultraviolet radiation, and microbial attack resistance. Also, EPDM materials have the advantage to be used at a wide range of temperatures, which makes them suitable for sun-grade deviation conditions [18, 19]. In this paper, the influence of EPDM on the properties of PP was investigated. Different contents of EPDM were explored to analyze their effect on the PP blend's mechanical properties. The structure, dielectric response, electrical, and mechanical properties of PP/EPDM blends were analyzed to determine the potential use of PP/EPDM blend in the development of recyclable power cable insulation materials.

2. EXPERIMENTAL

2.1 Materials

The 6531M PP homopolymer was supplied by the Lotte Chemical Titan and used as the matrix material. The Vistalon 722 EPDM was obtained from Exxon Mobil Chemicals and used as a toughening agent. The blending of the PP and EPDM materials was done according to Table 1.

2.2 Characterization

The samples' morphologies were observed using a field emission scanning electron microscopy (FESEM) (JEOL JSM-IT 500HR). The cross-section of the samples was gold-coated using an auto fine coater spray (JEOL JEC3000FC) for FESEM observation.

The chemical spectra of the samples were collected by using a Fourier transform infrared (FTIR) spectrometer (Shimadzu IRAFFINITY-1). The scan spectra were collected from 550 to 4000 cm^{-1} .

The dielectric spectroscopy measurements were carried out by Gamry Instruments Interface 1000TM with Tettex's 2914 solid test cell. The permittivity of the samples was recorded in the range of 10^2 - 10^6 Hz over 20 points per decade.

AC breakdown testing of the samples was performed by using a high voltage test (BAUR PGK 110B AC/DC) according to the American Society for Testing and Materials (ASTM) D149 standard. The samples were placed between 6.3 mm diameter ball-to-ball electrodes and immersed in silicone oil for breakdown testing. 15 breakdown tests were carried out for each sample. The calculation of sample breakdown strength was done by dividing the breakdown voltage over the sample thickness. The two-parameter Weibull statistical distribution method was used for breakdown strength analysis.

Tensile tests were done with the mechanical strength tester (Shimadzu AGS-X Series Universal Testing Machine) at a crosshead speed rate of 25 mm/min. Dumbbell-shaped samples with a cross-section of 0.5 x 4 mm and a gauge length of 20 mm were used to determine the stress-strain curves based on the ASTM D882 standard [12]. Five measurements were repeated during mechanical strength tests.

Table 1. Sample designation.

Group	PP content (wt%)	EPDM content (wt%)
PP	100	0
EPDM10	90	10
EPDM20	80	20
EPDM30	70	30

3. RESULTS AND DISCUSSION

3.1 Microscopic Morphology

Figure 1 illustrates the microscopic morphology of the PP and PP/EPDM blends. It can be seen from Figure 1(a) that the cross-section morphology of PP presents a lamellar texture with no obvious phases.

The morphology of PP/EPDM blends containing 10 wt%, 20 wt% and 30 wt% are shown in Figure 1(b), Figure 1(c), and Figure 1(d), respectively. The morphology of PP containing 10 wt% of EPDM elastomer is similar to the morphology of the pure PP. The addition of EPDM elastomer at 20 wt% causes the formation of morphology with the presence of a typical sea-island structure as shown in red circles in Figure 1(c). As can be seen in Figure 1(d), the formation of these structures become more apparent as the content of EPDM increases to 30 wt%.

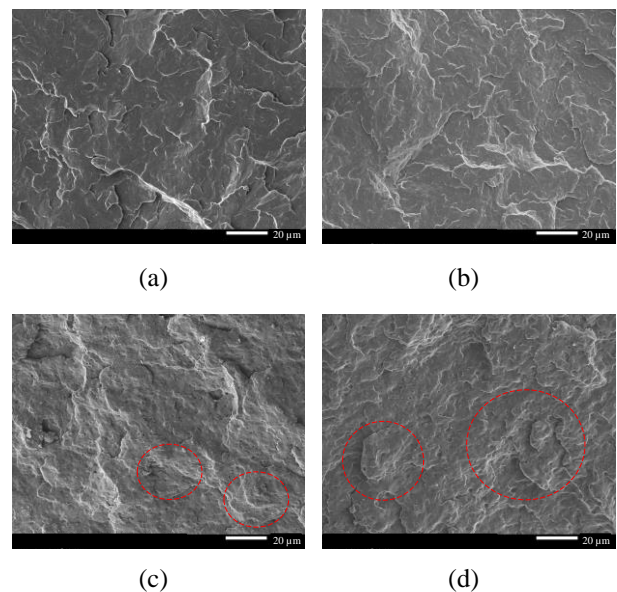


Figure 1. FESEM images showing (a) pure PP, (b) PP containing 10 wt% of EPDM, (c) PP containing 20 wt% of EPDM, and (d) PP containing 30 wt% of EPDM.

Generally, the EPDM was homogenously dispersed in the PP matrix. This indicates that the EPDM elastomer has a certain grade of compatibility with the PP matrix. When the content of the EPDM is at 20 wt% and 30wt%, the EPDM structures can be observed clearly. The dispersion of agglomeration into an “island form” in the PP matrix as a red circle in Figure 1(c) and Figure 1(d) is attributed to the ethylene-propylene phase of the EPDM [20, 21]. Thus, the structure of the PP matrix was changed drastically as the boundaries between the PP and the EPDM phases could be seen clearly. With a low content of the EPDM, the EPDM less affected the structure of the PP matrix.

3.2 Chemical Analysis

The FTIR spectra of the pure PP and the PP/EPDM blend with different contents of EPDM elastomer are shown in Figure 2. The characteristic absorption bands between 2835 cm^{-1} and 2930 cm^{-1} represent methyl and methylene groups. In addition, the absorption bands from 1373 cm^{-1} to 1454 cm^{-1} also indicate the bending vibration of the methyl and methylene groups. Based on the evidence from the FTIR spectra, the PP materials were in the polypropylene group [22, 23].

By adding 10 wt% of the EPDM to the PP, an additional band at 725 cm^{-1} can be observed; this is indicative of the C-H₂ group vibration, which is attributed to the ethylene group of the EPDM. The absorption peak between 1454 cm^{-1} and 2017 cm^{-1} reflects the out-of-plane vibration of the C-H bond that belongs to ENB (5-ethylidene-2-norbornene) contents of EPDM group. For the C-H₃ and C-H₂ groups of the polypropylene segments in EPDM, their absorption peaks appear between 1373 cm^{-1} and 1454 cm^{-1} [24, 25], similar to that of the pure PP. With increasing amounts of the EPDM in the PP, the intensity of the FTIR spectra represented by the ethylene and propylene segments of EPDM slightly increases. Consequently, the PP/EPDM blend possesses slight changes in the FTIR spectra with different contents of EPDM. This shows that the EPDM has a good compatibility with the PP since it has a similar spectral group with the PP matrix.

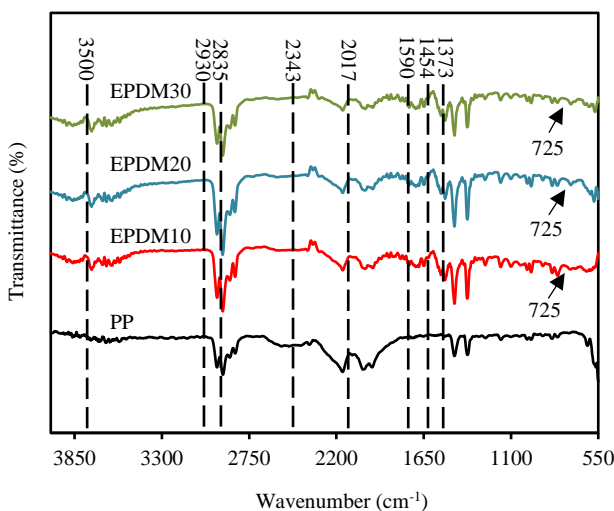


Figure 2. FTIR spectra comparing pure PP and PP/EPDM blend.

3.3 Dielectric Response

Figure 3 depicts the real part of the relative permittivity of pure PP and PP/EPDM blend containing 10 wt%, 20 wt%, and 30 wt% of EPDM. Throughout the frequency range between 10^2 and 10^6 Hz, the real permittivity of the pure PP is approximately 2.61.

The permittivity of the PP/EPDM blend containing 10 wt% of EPDM shows a slight increment of up to 2.7 compared to the pure PP. Significantly, the permittivity of the PP blend containing 20 wt% and 30 wt% of EPDM elastomer is much higher compared to the pure PP. The permittivity increases to approximately 2.95 and 3.06 for PP/EPDM blend containing 20 wt% and 30 wt% of EPDM, respectively.

The relative permittivity of the PP/EPDM blend increases significantly around to 17 % with the presence of EPDM in PP especially at high content as compared with the pure PP. Referring to the FTIR spectrum of the PP blends, the chemical structure slightly changes since the PP blended with different contents of EPDM has a different group of spectra. This is attributed to the influence of the larger formation of EPDM elastomer microstructure in PP blends with a higher content of EPDM. In short, different contents of EPDM elastomer that contributed to the different relative permittivity value of PP blends were affected by the formation of defect interfaces due to bad compatibility. This may lead to affecting the orientational polarization caused by the PP and EPDM, which may serve to reduce the breakdown strength.

3.4 Breakdown Strength

Figure 4 compares the AC breakdown strength based on the Weibull probability distribution plot of the PP and PP/EPDM blend containing 10 wt%, 20 wt%, and 30 wt% of EPDM. Meanwhile, Table 2 lists the Weibull parameters for the PP and PP/EPDM blend, where the scale parameter is the breakdown strength value at 63.2 % based on Weibull cumulative failure probability and shape parameter is the range of the breakdown strength based on the experiment. Based on the Weibull probability plot from Figure 4, the AC breakdown strength of the PP is 174 kV mm^{-1} . This reveals that the PP breakdown strength value is higher than XLPE where the AC breakdown strength was reported around $130\text{--}150\text{ kV mm}^{-1}$ [26, 27].

As seen in Figure 4, the presence of 10 wt% of EPDM in the PP reduces the AC breakdown strength of PP by about 5% to 165 kV mm^{-1} . For the sample containing 20 wt% of EPDM, the AC breakdown strength decreases to 158 kV mm^{-1} . The AC breakdown strength further decrease to 138 kV mm^{-1} with the addition of 30 wt% of the EPDM to the PP.

The presence of EPDM has a negative effect on the breakdown performance of PP/EPDM blend. Although the AC breakdown strength for all three PP/EPDM blends is lower than the PP, the AC breakdown strength is still acceptable as compared to XLPE (about $130\text{ to }150\text{ kV mm}^{-1}$ [26, 27]). Of note, EPDM/10 exhibits higher breakdown strength compared to other PP/EPDM blends.

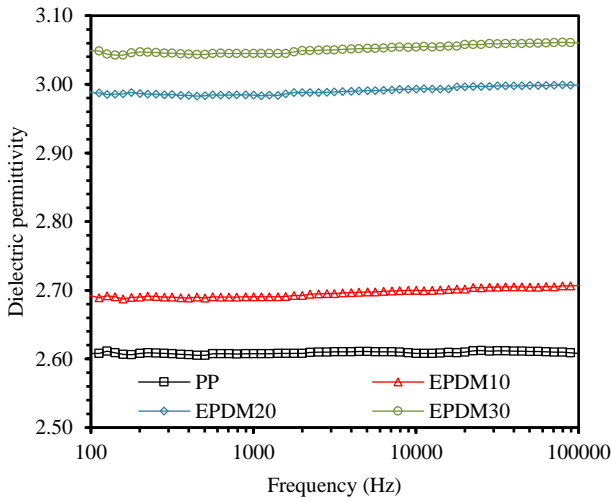


Figure 3. Real permittivity comparing pure PP and PP/EPDM blend.

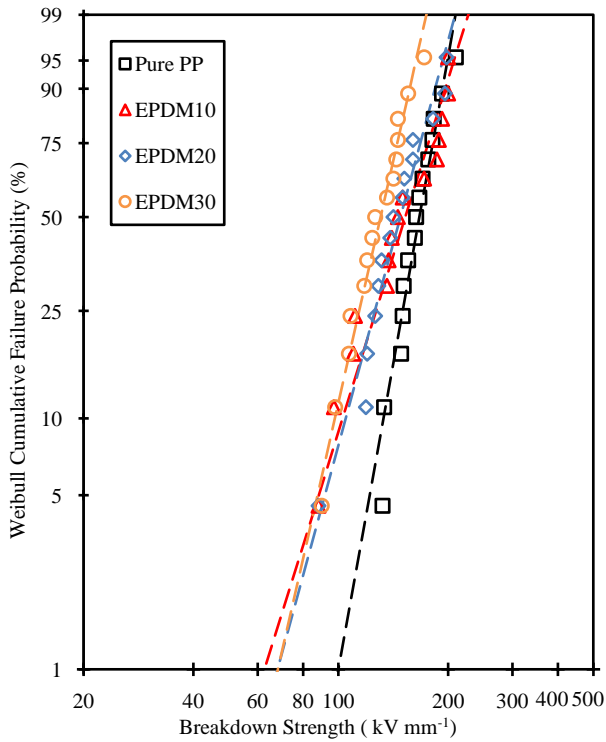


Figure 4. Comparison of the Weibull plots for pure PP and PP/EPDM blend.

Table 2. Comparison of the Weibull parameters for pure PP and PP/EPDM blend.

Group	Weibull parameter	
	Scale parameter, α (kV mm ⁻¹)	Shape parameter, β
PP	174 ± 10	8 ± 3
EPDM10	165 ± 16	5 ± 2
EPDM20	158 ± 14	5 ± 2
EPDM30	138 ± 10	7 ± 3

The breakdown strength of the polymeric blend can be impacted by many factors including the formation of structural defects. With the presence of high contents of the EPDM, the distribution of the ethylene-propylene rubber phase dispersed in the PP matrix can be seen obviously. The dispersion of the rubber phase of the EPDM at 30 wt% becomes more apparent as shown in Figure 1(d). It is therefore believed that the microstructure of PP/EPDM blend changes with the formation of the ethylene-propylene rubber phase of EPDM elastomer and result in partial defects of the polymer molecular chains. Thus, the formation of these structure defects affects the lamella thickness of PP and contributed to the decrement of breakdown strength. Therefore, the decrement of the breakdown strength with different contents of EPDM is related to the physical microstructural changes in the PP/EPDM blend. For EPDM/10, the physical microstructure does not change significantly, so the breakdown strength is less affected [28].

3.5 Mechanical Strength

Figure 5 presents the stress-strain curves of the PP and PP/EPDM blend containing 10 wt%, 20 wt%, and 30 wt% of EPDM. Based on the stress-strain curves, the PP has high tensile yield strength of up to 25 MPa. Compared with the PP, the addition of the EPDM to the PP results on a lower tensile yield strength and higher elongation at break. However, for higher contents of EPDM, i.e., EPDM/20 and EPDM/30, the elongation at break is lower compared to EPDM/10. This indicates that the tensile yield strength of the PP/EPDM blend decreases due to the low tensile yield strength of the EPDM. On the other hand, the microstructural changes in the PP caused by the EPDM result in a large effect on the yield strength, indicating that high contents of EPDM has changed the brittle-ductile behavior of the PP/EPDM blend [8].

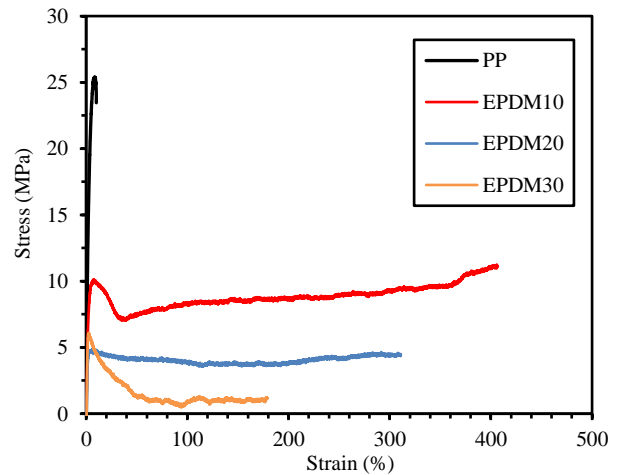


Figure 5. Tensile stress-strain curves comparing pure PP and PP/EPDM blend.

4. CONCLUSION

In this paper, the effect of the presence of different contents of EPDM in PP is reported. The morphology, chemical structure, dielectric response, electrical, and mechanical

properties of the materials is presented. The dispersion of the EPDM structure within the PP indicates that the blend between PP and EPDM has certain compatibility. The real permittivity of the PP/EPDM blend is stable over the frequency range, albeit that EPDM/20 and EPDM/30 has much higher real permittivity. Although the breakdown strength of all the PP/EPDM blends decreases compared to the PP, the EPDM/10 shows the highest breakdown strength that is still favorable for dielectric applications. The PP/EPDM blend possesses excellent mechanical properties, where the elongation at break of the PP improves with the presence of the EPDM. Based on the results of this paper, PP when blended with EPDM at 10 wt%, possesses improved mechanical properties and acceptable electrical properties. The PP/EPDM blend can therefore be considered for use in the development of future power cable insulation.

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