Modification of Polypropylene-based Cable Insulation Material

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**Abstract.** Polypropylene (PP) is one of the recyclable and ease processed thermoplastics，and it expected used as the cable insulation material to reply the unrecoverable material crosslinked polyethylene(XLPE). However, isotactic polypropylene which is too stiff with a lower electrical character cannot be directly used into the high voltage cable insulation system. In this paper, several polymers have been blended with polypropylene to improve the properties of the PP blend samples. Electrical and mechanical experiments of the modifying PP blends will also be done. In accordance with the experimental results, blends of PP and propylene-ethylene copolymers (PEC) are potentially more suitable for modifying PP in HV insulation.

1. Introduction

Electrical insulation is the most critical part of a high-voltage cable. The cross-linked polyethylene (XLPE), a type of polyethylene (PE) polymer, is widely employed as insulation materials in high voltage cable. It has high heat resistance which could work at the temperature of 90 ℃, and excellent mechanical properties enables its application in the complex environment of power transmission [1]. However, XLPE is not recyclable because of their cross-linked structure and can therefore not be reprocessed [2]. Meanwhile, several impurities which may affect the electrical properties of insulating materials are introduced in the crosslinking process. In this case, using a recyclable and eco-friendly material to replace XLPE is a promising research direction [3].

Isotactic polypropylene (iPP) refers to a polymer with numerous property to polyethylene that are basically the same, including electrical properties and solution character, as well as with high melting temperature at 170℃, The methyl group in the polypropylene reached mechanical properties and thermal resistance improved, albeit the chemical resistance decreases [4] . Moreover, the relatively lower thermal conductivity of PP compared with metals, makes it could against the change of the external temperature and thus it can be a material applied to insulation material[4]. However, because the long-distance cable needs to be rolled up for transportation and installation in the real application, iPP with low toughness and low breakdown strength is not suited for application as cable insulation materials [5].

Blending PP with other kinds of softer or compatible polymers, including high-density polyethylene (HDPE), low-density polyethylene (LDPE), propylene-ethylene copolymers (PEC) could be a potential method to reduce the limitation of PP application [1,5,6,7].

In this work, in order to observe the dielectric and mechanical characteristics of the PP blends, the following experiments have been developed: i) AC breakdown test. ii) DC conductivity test. iii) Tensile test. Moreover, the enhancement of the iPP could be analysed by comparing the experimental data of PP blends and XLPE reference.

1. Experimental
   1. **Materials**

Sigma-Aldrich produced the fundamental material of isotactic polypropylene. The LDPE used LD100BW was produced by ExxonMobil, the HDPE used HD5813A was from Rigidex, and three kinds of the metallocene-catalyzed propylene-ethylene copolymer (PEC) named VERSIFY™ 2200, VERSIFY™ 2300, VERSIFY™ 2400 were provided by Dow which contains 9mol%,12mol% and 15mol% of ethylene respectively.

* 1. **Sample preparation**

The testing samples were overall prepared by three main steps. First, the film samples were pre-melted at the temperature of 180 ℃ for 2 min in a Micro-Mould Press. Next, 1 tonne of pressure strength was introduced for 1 min to better mix the samples. Second, add 5 tonne of pressure strength for 3 min for the production of film samples to be tested with nearly 150 μm thickness. Third, the extraction operation was performed to guarantee that all samples had the same crystallization rate, which cooled the samples down in the 30s when the samples were extracted. According to the rating voltage and structure of different test instruments, three types of samples with different thickness were prepared for testing. Films with thickness about 50 μm applied for the conductivity test, films with thickness about 75 μm applied for the breakdown test, and films with thickness about 200 μm applied for mechanical testing were pressed by a Graseby Specac hydraulic press system.

* 1. **Breakdown test**

Breakdown of the above film samples was carried out by AC ramp testing methodology which including a two-electrode system and the test process in the silicone fluid [5,8]. The 75 μm thickness film samples were tested by the AC breakdown test instrument including an AC breakdown test box, a controller to set the ramp rate of voltage, a ondoscope, and a meter.

Samples were set between the two electrodes containing silicone oil in the test box. An increasing voltage was applied to the samples with a ramp of 100V/s until breakdown.

The time(T) could be generated by the timer when the breakdown occurs. The breakdown voltage () can be defined as:

. (1)

Then the breakdown strength can be calculated by

. (2)

For the breakdown strength data, considering the differences of both technology and structure, the breakdown voltage of the insulation samples varies from one to another. The breakdown voltage and break gradient is considered a random statistic[9]. Weibull distribution is the probability distribution commonly employed to help the reliable analysis of the breakdown strength. The Weibull distribution is usually applied for statistical processing the data of breakdown strength of dielectric[10] which is expressed by the following formula (3):

. (3)

where: is the breakdown probability; is the scale parameter; is the shape parameter

* 1. **Conductivity test**

The three electrode system is the conventional method of conductivity test[11]. The film samples (~50 μm in thickness) were tested by the DC conductivity test system which including a conductivity test box, a voltage source to set the voltage, a picoammeter to measure the value of voltage and current applied to the samples, and a test software based on the LabVIEW 2015.

The parameters were overall set before the test, including the strength of electrical field (), the voltage applied ().

Based on the magnitude of the electric field formula (4),

(4)

The value of the voltages can be calculated by the thickness of the electrical fields which is a fixed value in this test, and the test time based on the relaxation time constant (). When the test ends, the conductivity curves were collected by the picoammeter and the test software.

* 1. **Tensile Test**

The mechanical test commonly uses the universal testing device to stretch rectangle or dumbbell-shaped film samples at a constant extending speed [8,12,13]. In this study, the mechanical tensile test was characterized by the Tinius Olsen H25KS testing machine. Rectangle shaped samples with ~200 μm in thickness, 5 cm length and 1 cm wide for the test. All of the samples were stretched by the grip head at a constant speed of 5 mm/min and the original distension between grips is 30 mm. The results of the tensile test analysed by the stress-strain curves collected by test software. In addition, the tensile performances of the insulating materials could be determined by Young’s modulus(E) [14] which represented by the formula (5) .

(5)

where is Young’s modulus; is tensile stress; is the extensional strain.

1. Results
   1. **Sample preparation**

In terms of polypropylene blends, different vinyl polymers containing 10% wt%, 30% wt% and 50% wt% were used to compare the properties of polypropylene-based blends with different weight percentages in accordance with Ref. [1,5,6,7]. In this paper, the name of the blend copolymers was shown in Table 1. All samples are kept in the sample storage box to keep dry and maintain the quality.

Table 1. Name of PP-blends.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Wt% | LDPE | HDPE | 2200 | 2300 | 2400 |
| 10 | PP-LDPE-10 | PP-HDPE-10 | PP-2200-10 | PP-2300-10 | PP-2300-10 |
| 30 | PP-LDPE-30 | PP-HDPE-30 | PP-2200-30 | PP-2300-30 | PP-2300-30 |
| 50 | PP-LDPE-50 | PP-HDPE-50 | PP-2200-50 | PP-2300-50 | PP-2300-50 |

* 1. **Breakdown test**

Results of the breakdown strength test comply with the thickness of the film samples when other factors are fixed, inclusive of the voltage applied, electrode geometry and temperature. Besides, to enhance reliability of the experimental data, 20 time tests were done on all kinds of sample. The Weibull probability of breakdown strength of the testing samples is shown in Fig. 1.

|  |  |
| --- | --- |
|  |  |
| (a) | (b) |
|  |  |
| (c) | (d) |
|  |  |
| (e) | (f) |

Fig. 1. Weibull plots comparing the breakdown strength of the (a) PP and XLPE (b) PP-LDPE blends (c) PP-HDPE blends (d) PP-2200 blends (e) PP-2300 Blends (f) PP-2400 blends.

Three conclusions could be safely drawn from the Fig. 1. First, all of the modifiers for PP blends are intended to enhance the electrical properties of pure PP because of the breakdown strength of PP in lower than the wildly used insulation material XLPE. Second, the scale values of breakdown strength were obviously increased by adding the weight percent of PE polymer including LDPE and HDPE blend with PP. Third, PP-PEC blends have higher breakdown strength than the pure PP. However, the optimization of breakdown strength is not a linear change by the increasing the weight percent of PEC modifiers, and the top value of scale parameter occurred at 10wt% copolymers.

* 1. **Conductivity test**

The conductivity of pure PP and XLPE reference are shown in Fig. 2. The conductivity of XLPE is close to zero and much lower than the pure PP. For PP-blends, their conductivity is expected to be lower than pure PP and are able to approach the performance with XLPE.



Fig. 2. Conductivity-time curves of pure PP and XLPE.

Fig. 3 shows the conductivity testing results of the testing samples. It is clearly seen that the value of conductivity of pure PP is much higher than the PP-blend copolymers when the curves reach to the relatively steady state. The adding of both PE and PEC could reduce the conductivity of insulation materials.



Fig. 3. Conductivity-time curves of all samples.

For all the tests, it is suggested that the start values of different types of insulation materials have verified character. The duration of the steady-state of conductivity current also shows different characteristics from the experimental results. One of the reason is that the element of the resistor (R) and capacitor (C) that affects the relaxation time constant (τ) will have an unpredictable change in the mixed polymer. This will cause the significant crosses in the conductivity curves before they reaching steady states. Using the values of steady-state conductivity for the characterization of certain polymer insulation may not a perfect method, which is also mentioned in the investigation by Ghorbani et al,[15].

* 1. **Tensile test**

Fig. 4 presents the stress-strain curves of the pure PP and XLPE reference. From the viewpoint from the curves, we can safely consider that the pure PP is not suitable for power cable due to its limitation of mechanical characteristic.

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Fig. 4. Stress-strain curves of pure PP and XLPE reference.

The stress-strain curves of PP and the PP blends are shown in Fig. 5. It could be clearly observed that Most of PP blends show the better performances than pure PP sample in tensile strength and elongation at break. It is clearly seen that the pure PP and PP-PE blend samples are broken before they are extended to 50% of their original length. In comparison, the PP-propylene-ethylene copolymer blend could be extended longer than PP and PP-PE blends. In addition, the maximum value of stress on the PP and PP-PE blend samples are higher than PP-propylene-ethylene copolymer samples.

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Fig. 5. Stress-strain curves of pure PP and its blend.

All of the blends could decrease the stiffness by increasing the content of the additional materials except PP-HDPE copolymers. More than 10wt% of HDPE introduced in PP-blend, the copolymer materials will be stiffer due to its higher Young’s modulus.

Results of three main experiments are listed in Table 2, several interesting features could be found.

Table 2. Statistics of all experiments.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Sample | Breakdown Strength [kV/mm] | Conductivity[S/cm] | Young’s Modulus[Mpa] | |
| PP | 203.97 | 5.75E-16 | 245.46 |
| PP-LDPE-10 | 208.74 | 1.03E-17 | 237.24 |
| PP-LDPE-30 | 249.26 | 5.38E-18 | 212.35 |
| PP-LDPE-50 | 277.24 | 2.60E-18 | 134.16 |
| PP-HDPE-10 | 239.97 | 1.32E-16 | 242.43 |
| PP-HDPE-30 | 249.30 | 3.76E-17 | 287.49 |
| PP-HDPE-50 | 277.73 | 3.99E-17 | 424.38 |
| PP-2200-10 | 283.20 | 4.35E-18 | 185.79 |
| PP-2200-30 | 251.28 | 1.51E-17 | 132.91 |
| PP-2200-50 | 244.41 | 1.81E-17 | 83.38 |
| PP-2300-10 | 270.12 | 8.67E-17 | 197.08 |
| PP-2300-30 | 253.77 | 1.69E-17 | 147.20 |
| PP-2300-50 | 244.13 | 3.62E-17 | 48.97 |
| PP-2400-10 | 253.40 | 1.03E-16 | 170.93 |
| PP-2400-30 | 243.90 | 4.95E-17 | 129.61 |
| PP-2400-50 | 222.22 | 7.16E-17 | 108.87 |

Based on the statistics of testing results, the performance of most PP-blends is close to or greater than XLPE. Compared with polyethylene polymer, VERSIFY™ series of elastomers have good performances for improving the electrical properties and mechanical properties of PP, which incorporates VERSIFY™ 2200-PP-blend with the most electrical performance in all PP-PEC blends. The sample of PP-2200-10 has the largest breakdown strength reaching 283.20kV/mm, which is even large than that of XLPE reference, reaching the lowest conductivity to S/cm. Besides, for mechanical properties, its Young's modulus also reaching 185.79Mpa, almost two-third of that of pure polypropylene. Therefore, comprehensive comparison of all the experimental data and taking into account the electrical performance and mechanical properties of the balance. PP-2200-10 has the optimal performance in the experiment.

1. Discussions
   1. **Ratio of modifier**

According to the experimental results indicated in Table 2, it can be obtained that, the change of breakdown strength of PP-PEC blends is not a linear change. The highest breakdown strength occurs at the 10wt% modifiers then it reduces when the modifiers present increase to 30wt%. The ratio of modifiers is a significant parameter modify the properties of the PP blends. Thus, to determine the best ratio to find a better performance blend, looking to find the properties between 10wt% and 30wt% is necessary. An additional sample, 20wt%content of additional modifiers in PP-blend, was made. Effect of composition on breakdown strength for this additional PP blends is shown in Fig.6.

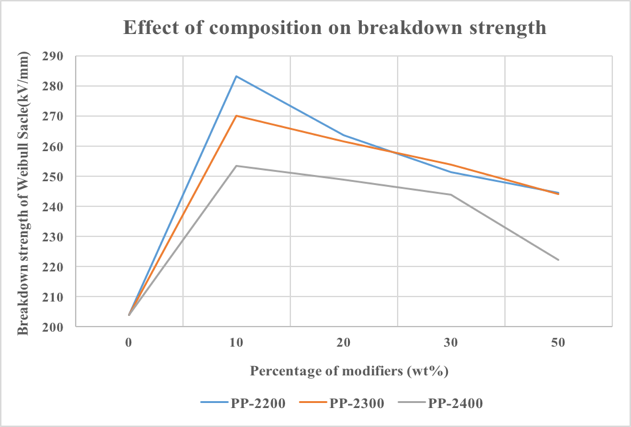


Fig. 6. Effect of composition on breakdown strength for PP blends.

The scale for breakdown strength of all the 20wt% copolymers was lower than 10wt% copolymers but higher than 30wt% copolymers. This suggests that the electrical strength may reach the peak at 10wt% of modifiers and adding more propylene-ethylene copolymer to the PP-blend did not benefit its electrical performance. Meanwhile, from the above analysis, it can be seen that a more precise blending ratio can be used to find a clearer blending ratio scheme for cable materials, which will be considered in future research.

1. Summary

Based on the breakdown strength, conductivity and mechanical tensile, the properties of isotactic PP modifying blends for recyclable high voltage cable insulation are studied experimentally. All of the new PP-PEC blends are potentially suitable for high voltage cable insulation based on their characteristics. The optimal compositional, considering both electrical and mechanical factors, was found the polypropylene-based blend with 10wt% of propylene-ethylene copolymers (VERSIFY™ 2200). These PP-PEC blend with good electrical and mechanical properties and similar to that of XLPE which means this PP blend is a new type of recyclable insulation materials for high voltage cable.

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