Partial Discharge Characteristics with Morphological Analysis and Tensile Properties of Linear Low-density Polyethylene-Natural Rubber Blends

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Abstract: A series of linear low-density polyethylene (LLDPE)-natural rubber (NR) blends of composition 80/10, 70/20, 60/30, 50/40 and 40/50 containing nano-sized fillers montmorillonite (MMT) and Titanium(IV) Oxide (TiO_2) were produced by a twin-screw extruder with maleic anhydride grafted linear low-density polyethylene (LLDPE-g-MAH) of 10 wt% as a compatibilizer. An electrical performance test through partial discharge (PD) characteristics using CIGRE Method II test was conducted to study the electrical performance of the samples. Applied voltage was set on 7kVrms for 1 hour. The discharge characteristics were observed using picoscope™ and LabView™ programming. Morphological analysis using scanning electron microscope (SEM) was also conducted after the sample of composite was subjected to high voltage stress. The degradation of composite surface was analyzed. Then tensile test carried out to investigate the mechanical performance of the composites. The combine results of PD characteristics and tensile properties described the insulating performance of the composites. The results revealed that total PD numbers decrease with increasing of weight percentages of natural rubber in the composition of the composite without any filler. In addition, it is found that total PD numbers significantly decreased on sample with MMT filler compared to TiO_2 one.

Keywords: Partial discharge (PD), LLDPE-natural rubber (NR) blends, CIGRE Method II, montmorillonite (MMT), Titanium(IV) Oxide (TiO_2), scanning electron microscope (SEM)

1. Introduction

Natural rubber (NR) have been used as an insulating material for many electrical application such as tapes, rubber pole insulators and gloves [1]. According to E. G. Kimmich [2], one of the most distinguished characteristics of rubber compared to those of metals, concrete, wood, and ceramics is its great extensibility and deformability. After many decades, there are previous researches which showed that the combination of polyethylene (PE) and NR can produce a type of composite which has an advantage to material industrial technology. Therefore, it becomes relevant to conduct an experimental study to investigate the insulating performance of PE-NR blends to figure out the new material of insulation.

Recent works show that the combination of polyethylene (PE) and natural rubber (NR) produced a type of composite which has an advantage to material industrial technology [3-4]. Among the PE material, linear low density polyethylene (LLDPE) was found as most compatible while compounding with NR [5]. According to a previous study, the introduction of nanofiller in the structure of a composite can develop the interaction strength between the polymer matrix and nanometric fillers, which can increase significantly the electrical, thermal, and mechanical properties [6-7].

However, there is no extensive research on the electrical properties for LLDPE-NR composite especially in the application of high voltage insulation field. Thus, the contribution

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of this work is to investigate the insulating performance of LLDPE-NR blends by studying partial discharge characteristics under high voltage stress. In addition, tensile test was also carried out to explore more mechanical properties of the composite to be applied in the insulation system.

Thus the experimental study of PD degradation on the composites is of interest in the past of 10 years. This is due to its effect on the insulation quality [8]. Therefore, it is very important to study the PD on the LLDPE-NR blends in order to discover its effects before the samples are proposed as new insulating materials in the future.

2. Experimental Setup and Test Procedures

A. Samples Preparation

The linear low density polyethylene (LLDPE) and maleic anhydride grafted linear low-density polyethylene (LLDPE-g-MAH) was supplied by Etilinas LL0220SA from Polyethylene Malaysia Sdn. Bhd while the natural rubber (NR) used in this study was a Standard Malaysian Rubber (SMR).

Table 1. Compound formulation and coding of LLDPE-NR blends

Sample	**Blend component**				
	LLDPE	**NR**	**LLDPE-g-MAH**	**MMT***	**TiO_2***
A1	80	10	10	-	-
A2	70	20	10	-	-
A3	60	30	10	-	-
A4	50	40	10	-	-
A5	40	50	10	-	-
B1	70	20	10	2	-
B2	70	20	10	4	-
C1	70	20	10	-	2
C2	70	20	10	-	4

*phr (part per hundred) of LLDPE-NR weight

The extending filler [9] used for improving electrical performance through partial discharge (PD) resistance of the blend was nano-sized fillers montmorillonite (MMT) and Titanium(IV) Oxide (TiO_2). Table 1 shows the three groups of blending formulation that were prepared based on different weight ratios of the base polymers and different levels of nano-size fillers. In the second stage, each blend was compression moulded into slabs which is of 1mm thickness to produce a sheet of sample.

B. PD and Tensile Test Procedures

To investigate the electrical performance of the LLDPE/NR blends conducted with the sheet of samples cut into spherical form with 40mm diameter, a standard assembling methodology (CIGRE Method II) for experimental of PD characteristics is applied [10].

Figure 1 shows the picture of the real experimental setup for PD measurement where the high voltage alternating current (HVAC) 50Hz, is applied to the upper CIGRE Method II electrode. The applied voltage was set on 7kVrms for 1 hour ageing time. Induced PD current on the test samples is detected by a designed RC detector device (R=1000Ω, C=1000pF) with simultaneous noise reduction by a high pass filter (HPF) system [11]. In order to acquire the PD signal, the detected signal is transmitted to Picoscope™ and visualized on a personal computer (PC). An acquisition data system was developed using LabVIEW™ to record automatically every number of PD occurred during the ageing time.

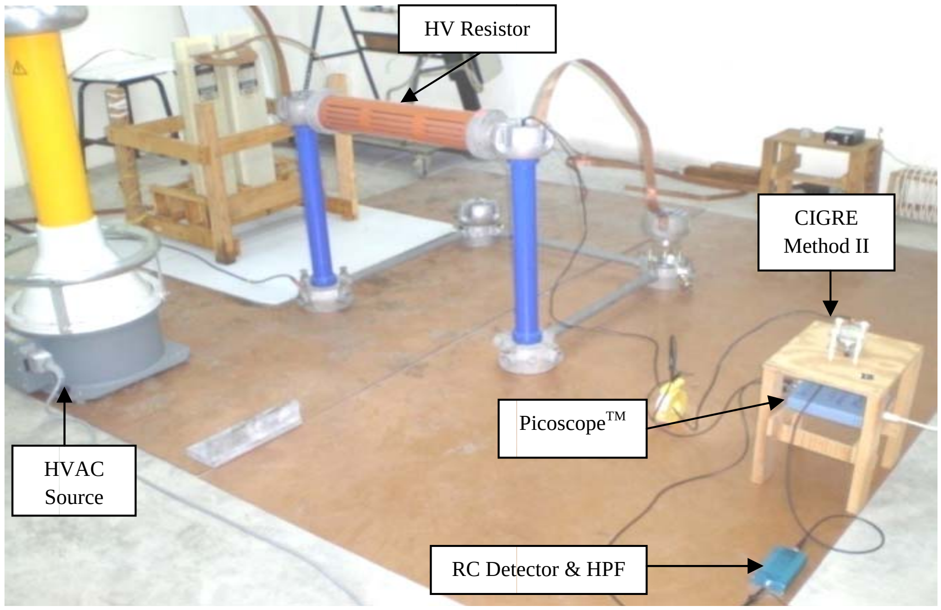


Figure 1. Experimental setup for PD measurement

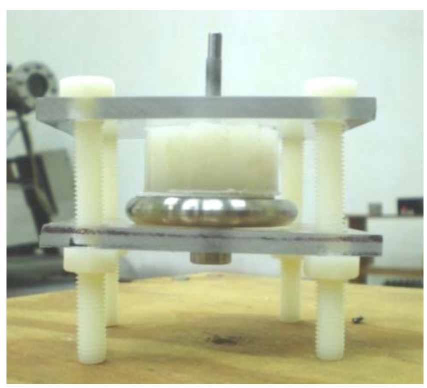


Figure 2. A test cell and sample arrangement of CIGRE Method II

Figure 2 shows a test cell and sample arrangement of CIGRE Method II. This electrode system is an improved version of the CIGRE Method I featured as PD activity is concentrated in a specified area due to geometrical space of void which is 100 times larger than the previous version [12]. For this case, thickness of LLDPE-NR samples that is being used is 10mm and 40mm diameter.

Although the blends of LLDPE-NR have a good mechanical properties [13], tensile test was still conducted for each group of formulation blending. Tensile strength, elongation at break, and modulus properties were measured on dumbell-shape sample using a tensile tester (EZ 20KN LLYORD INSTRUMENT) with a cross-head speed 50 mm/min and 10N torque at room temperature as shown in figure 2.

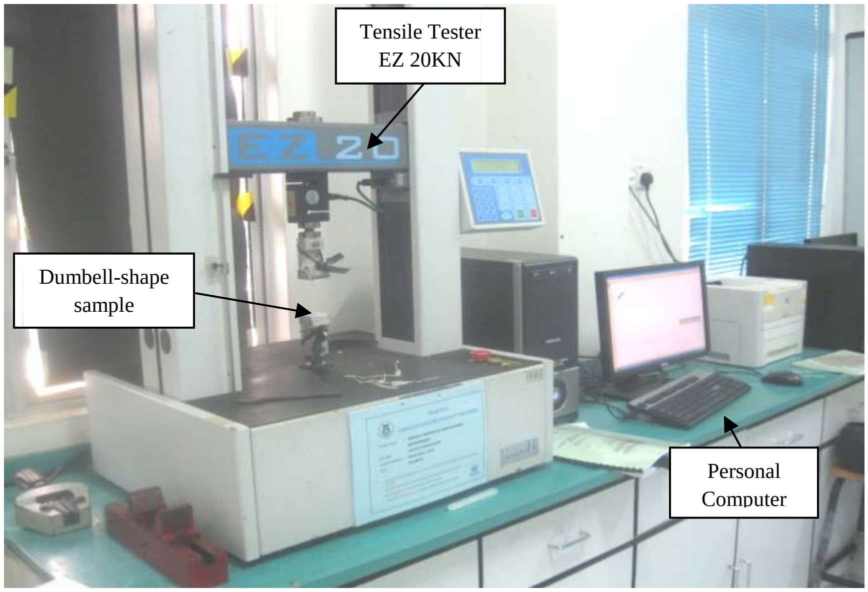
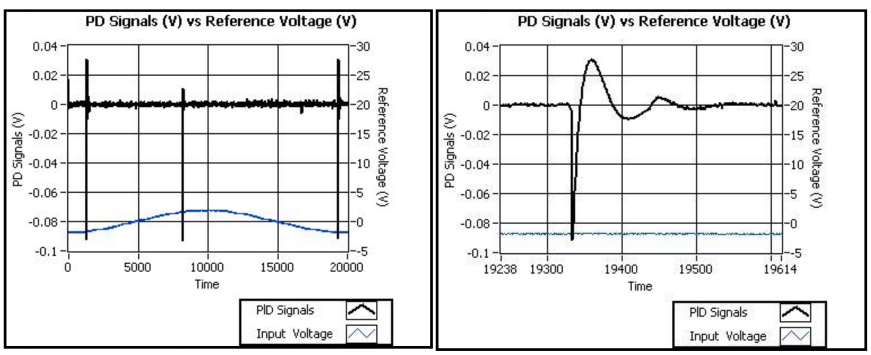


Figure 3. Experimental setup for tensile test

3. Results and Discussion

A. PD Numbers and Characteristics

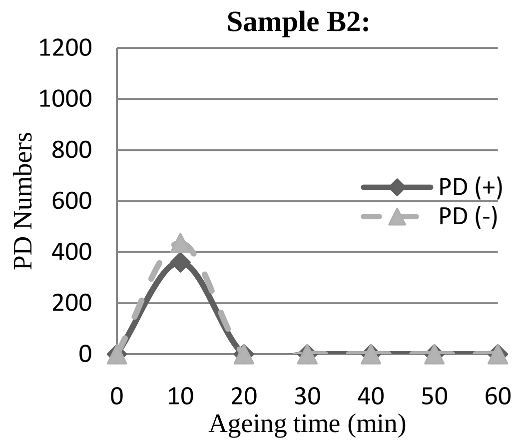


(a) (b)

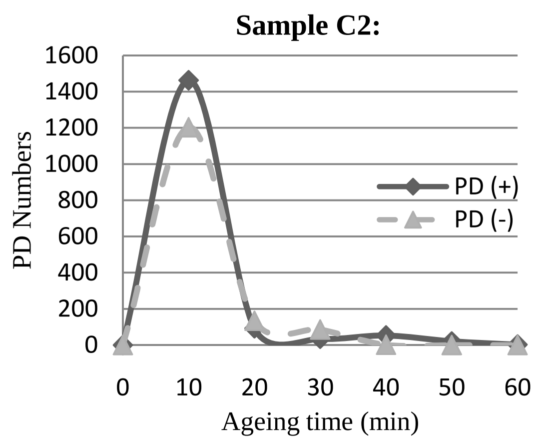
Figure 4. Typical PD waveforms: (a) PD signals (mV) for 1 cycle input voltage, (b) Zoom in on one PD signal waveforms obtained from (a)

Figure 4 shows the typical PD waveforms. Figure 3 (a) is the PD signals and numbers accompanied for 1 cycle waveform from HVAC input. Then, figure 3 (b) shows 1 PD pulse after zoomed in from figure 3 (a). The magnitude of PD pulse corresponds to 90mV.

While in the initial stage (10 minute of ageing time), PD activities show the most number of occurrences and then after the sample approaches to the end of ageing time (60 min), a reduction number of detected discharges is clearly observed. The characteristics of the number of PD along ageing time are reported in Figure 5 for 4 phr of MMT and TiO_2 nano-sized fillers.



(a) MMT filler



(b) TiO_2 filler

Figure 5. Time dependence of PD of positive (PD +) and negative (PD -) discharge numbers for 4 phr of MMT (sample B2) and TiO_2 (sample C2) nano-sized fillers.

After 20 minutes of ageing time, PD activity always change to a very small PD pulses known as swarming pulsive microdischarges (SPMD) [14] with discharges amplitude under the detection system sensibility.

Figure 6 shows the PD numbers bar graph of all LLDPE-NR blends formulations. The results are based on a sample tested for 1 hour of ageing time and 7kVrms of applied voltage.

Electrical insulating performance of each LLDPE-NR blends is determined from the total PD numbers occurring during the experimental session. Therefore, figure 4 displays the bars of the positive, negative and total PD pulse numbers for all LLDPE-NR blends formulations. It is observed that the peak value for the case of LLDPE-NR blends without filler (sample; A1, A2, A3, A4) is higher than those of LLDPE-NR blends with filler (sample; B1, B2, C1, C2).

The result also indicates that LLDPE-NR without filler decreasing in PD activities as increasing of NR compounding ratio as shown by sample A5 comparing with LLDPE-NR with filler (Sample C1 and C2). Moreover, it is clearly found that the total PD numbers significantly decreases on sample with MMT filler (sample; B1 and B2) compared to TiO_2 filler (sample;

C1 and C2). Thus, the addition of 4 phr of MMT nano-sized filler resulted in the general improvement of the PD characteristics of LLDPE-NR blends.

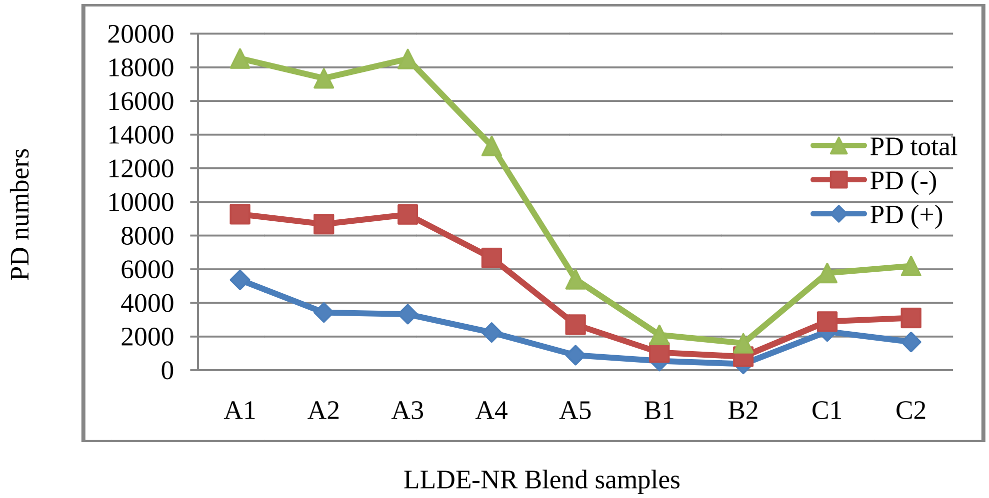
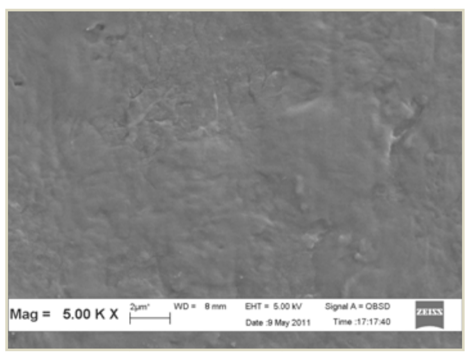


Figure 6. PD numbers for all LLDPE/NR blends formulation

B. Morphological Analysis

The studies were used to reveal information on how PD activities play its role in the degradation on dielectric. Two kinds of comparison are applied using a scanning electron microscope (SEM), the sample of LLDPE-NR before PD test and after the PD test.



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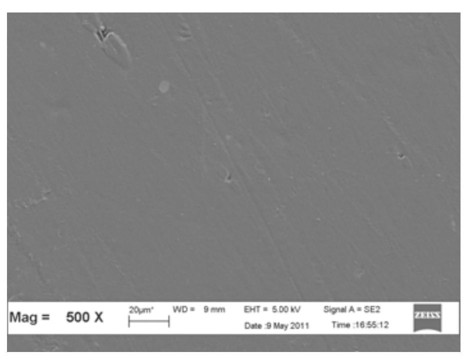

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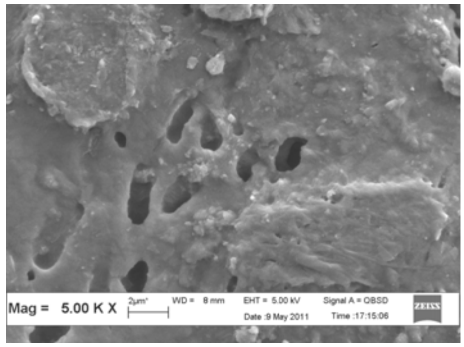

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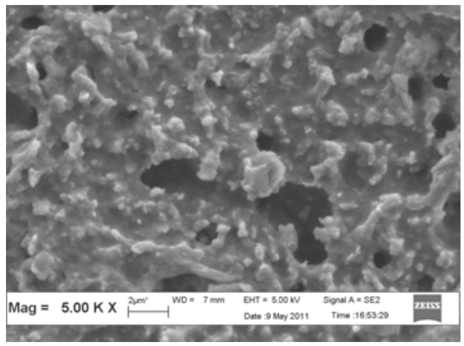

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Figure 7. SEM micrographs of sample LLDPE-NR before and after PD test. 7a-i. Sample A2 (20/70/10, NR/LLDPE/LLDPE-g-MAH) – before test; 7a-ii. Sample B1 (20/70/10, NR/LLDPE/LLDPE-g-MAH and 2phr MMT) – before test; 7b-i. Sample A2 – after test; 7b-ii. Sample B1 – after test;

Reference [15] has reported that the degradation of composites due to PD can be studied through SEM observation. Surface micrographs of sample A2 (20/70/10, NR/LLDPE/LLDPE-g-MAH) and B1 (20/70/10, NR/LLDPE/LLDPE-g-MAH and 2phr MMT) will be discussed to illustrate compatibility of the blends and the degradation caused by PD.

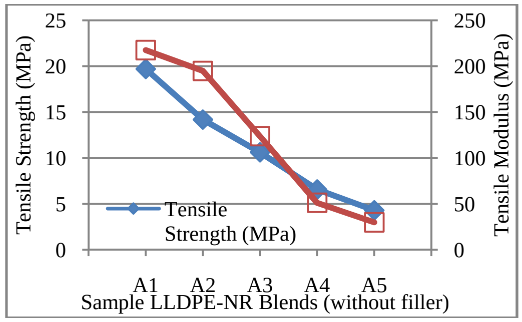
Basically the basic components in both samples are homogenously dispersed as shown in Figure 7a-i and Figure 7a-ii. For the sample filled with 2phr MMT (B1), there is only a small agglomeration of the fillers occurring as presented in Figure 7a-ii.

It shows that the interaction between fillers and the polymer matrix is strong. When the samples are applied with high voltage stress along 1 hour ageing time, the surface structure become porous and degradation appeared as shown in Figure 7b-i and 7b-ii.

The degree of surface damage depends on the periodic of ageing time as well as the PD characteristics. In the case of sample with nano-MMT filler (B1), it is clearly seen that a lot of aggregates of nanofillers appear on eroded surface of composite material. Therefore, the surface morphology observation which is presented in the Figure 7, confirms that the observation of degradation due to PD activity on sample A1 and B1.

C. Tensile Test and Properties

The study of tensile properties was used to investigate the mechanical performance of the composites as one of the major considerations for designing the insulation material [16]. The general mechanical performances of LLDPE-NR blends determined by this test are tensile strength, tensile modulus and elongation at break, respectively. The results of the tensile tests are summarized in figure 8.



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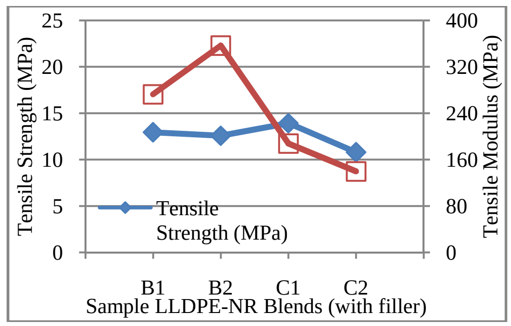

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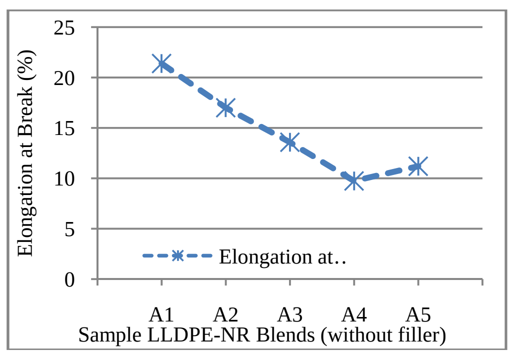

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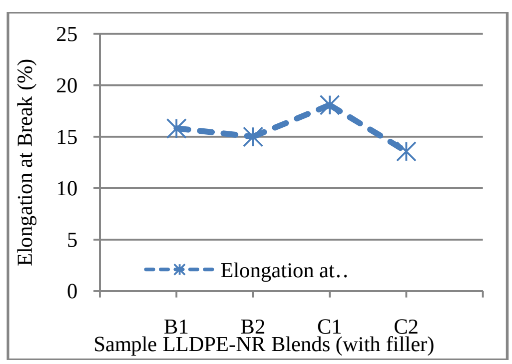

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Figure 8. Graph of tensile properties of LLDPE-NR samples after tensile test. 8a-i. Tensile strength and tensile modulus of LLDPE-NR samples without filler (A1,A2,A3,A4,A5); 8a-ii. Tensile strength and tensile modulus of LLDPE-NR samples with filler (B1,B2,C1,C2); 8b-i. Elongation at break of LLDPE-NR samples without filler (A1,A2,A3,A4,A5); 8b-ii. Elongation at break of LLDPE-NR samples with filler (B1,B2,C1,C2).

According to Figure 8, the typical tensile strength, tensile modulus and elongation at break of LLDPE-NR contain with and without nano-sized fillers are shown. In the case of Figure 8a-i, the tensile strength and tensile modulus of LLDPE-NR without filler (A1,A2,A3,A4,A5) decrease with increasing of NR ratio contains. While in Figure 8a-ii, the tensile strength and tensile modulus of the composite with nano-sized filler increased after the addition of TiO_2 and MMT filler content at 4phr and 2phr (B2,C1).

Also as an interesting observation from Figure 8b-ii is the increasing behavior of LLDPE-NR in the elongation at break after the addition of MMT 2phr (C1). It was believed that the possible factor of increasing the tensile strength and tensile modulus of LLDPE-NR composite cause by the incorporation between the polymer matrix and filler-filler interaction [17]. It also was reported by Ibrahim et al. [5] and Dahlan [3], the presence of LNR increased the tensile strength and the elongation at break of LLDPE-NR blend.

Thus the overall results present that the addition of MMT and TiO_2 nano-sized filler at the suitable quantities will enhance the tensile properties of LLDPE-NR composites.

Conclusions

The insulating performance of LLDPE-NR blends under electrical stress and mechanical strain is investigated by analyzing PD characteristics and tensile properties. Generally, the composite filled with 4 phr nano-sized filler (sample B2) tend to suppress PD activities during ageing time and at the same time to be the best composite based on high tensile modulus result. By considering the PD characteristics and tensile properties, it will explore a correlation and consideration to be applied as electrical insulating material from LLDPE-NR blends composite in future.

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