

# Enhancement of Partial Discharge Resistance and Breakdown Strength Characteristics of Low-Density Polyethylene Nanocomposites Using Plasma Treatment Method

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*Abstract:* Insulations in the power cable system are prone to ageing and degradation, eventually leading to a complete breakdown. One of the solutions to reduce insulation breakdown in polymeric insulation is by adding nanofillers into the polymer matrices of the insulation to form polymer nanocomposites. However, the addition of the nanofiller into the polymer usually results in agglomeration inside the nanocomposites. Recently, atmospheric pressure plasma (APP) has been introduced by adopting the nanofiller's surface modification method to hinder agglomeration formation. The aims of using APP are to enhance the nanofiller-polymer interfaces and improve the dielectric properties, emphasizing partial discharge (PD) resistance and AC breakdown strength. In this study, APP has been used to treat boron nitride (BN) and silicon dioxide (SiO<sub>2</sub>) nanoparticle surfaces for the purpose of enhancing the compatibility with low-density polyethylene (LDPE) matrices. Untreated and plasma-treated nanoparticles have been added into LDPE with different filler loading of 1 wt%, 3 wt% and 5 wt% via the direct compounding method. Compared with untreated nanocomposites, the 30-minutes plasma-treated nanocomposites could improve the PD resistance by reducing the PD magnitude up to 513 pC and reducing the PD number to 11661. Moreover, the AC breakdown strength of the plasma-treated nanocomposites had increased from 0.53 kV/mm to 26.65 kV/mm. If compared to LDPE/BN nanocomposites, it was discovered that the LDPE/SiO<sub>2</sub> nanocomposites displayed significantly better dielectric characteristics. In addition, plasma treatment of the nanoparticles could produce nanocomposites with better formulation stability and promising dielectric performance, which can prolong the insulation's lifetime and ensure the reliability of the power supply.

*Keywords:* Polymer nanocomposites; low-density polyethylene; partial discharge; breakdown strength; atmospheric pressure plasma treatment.

## 1. Introduction

Polymeric insulating materials have been widely used in high voltage equipment, particularly power cables, as an insulating material, due to their excellent performances. However, the material would eventually be subjected to degradation, one of the root causes of electrical discharge. Electrical discharge phenomenon such as partial discharge occurs primarily in microvoids, defects or protrusions that could be produced during the manufacturing process or other external activities [1]. Since the discharge phenomenon only occurs inside the small defects, an immediate complete breakdown of the insulation would not have occurred. This is because the surrounding healthy insulation is strong enough to prevent a complete breakdown of the insulating material. As a result, the drawbacks of the existing insulating materials have opened up a new research area, so-called nanodielectrics or nanocomposites.

In order to enhance the electrical properties of the nanocomposites and lower maintenance costs, numerous researchers have worked intensively over time to create a good insulating

material. Polymer nanocomposites have become one of the main topics of many publications as it has been reported to improve several physical, chemical and electrical properties; such as higher partial discharge resistance, better electrical treeing suppression, higher breakdown strength, reduced space charge accumulation, and lower dielectric loss [2]–[4]. Widespread interest in the use of nanocomposites as high voltage insulating materials has been generated by research on the subject. Typically, host polymer and nanofillers are combined to create nanodielectrics. These have been made by adding nanometer-sized fillers and dispersed homogeneously into the polymer matrices by several weight percentages (wt%).

Because of its low cost, superior electrical qualities, resistance to moisture and chemicals, flexibility at low temperatures, and low density, polyethylene has been used as a novel insulating material in cable insulation systems [5]. Numerous studies discovered that adding nanofillers to nanocomposites enhanced the polymeric insulator's dielectric properties [6]–[10]. Silicon dioxide ( $\text{SiO}_2$ ) has been utilized as a nanofiller because it has good electrical insulation, high electrical resistivity and high thermal stability [11]. In contrast, boron nitride (BN) has been used as a filler as it exhibits high resistivity, high breakdown strength, and low relative permittivity [12].

Studies have been carried out [13]–[16] on adding different weight percentages of nanofillers into polymeric matrices because of their potential in improving electrical, mechanical, chemical and thermal properties. However, previous research reported that nanofiller would not give better results to the dielectric performance because of the formation of agglomeration, which diminishes the enormous surface area per unit weight of the nanoparticles and thereby nullifying the enhancement of the properties of the nanocomposites [6], [9], [10], [17]. Unfortunately, agglomeration of the filler may alter the insulation properties and causes worse degradation of polymer insulation because the nanofiller is not tightly bonded to its polymer matrix.

To improve the compatibility between the nanofiller and the polymer matrix, and so help reduce agglomeration for the creation of nanosized particles, these nanofillers must therefore be treated and their surfaces modified. The interactions area in the middle of the host polymer and the nanofiller were responsible for improving the dielectric performances of nanocomposites. The surface interaction between polymer matrices and silica nanoparticles is generally connected to the interfaces. The surface of the nanofiller was previously altered with a silane coupling agent, improving the adherence of the matrices and the filler particles. Many scientists have employed silane coupling agent, a chemical-based solution, extensively. Nevertheless, this age-old technique has some drawbacks, such as toxicity and a complicated preparation process. Consequently, unsuitable for mass production. Therefore, a clean, safe electrical approach that can be used in mass production is needed to improve the bonding between the nanofillers and the polymer matrix.

Researchers eliminated this breakdown problem by introducing an APP method. APP has been proposed as it overcomes the drawbacks of silane coupling agents. The plasma treatment method improved the interfacial bonding of the nanofillers and polymer matrices [18], [19]. The main advantages of this method are the elimination of vacuum systems used in low-pressure plasma, reduction of costs, and the possibility for a continuous system and treatment of materials. Plasma treatment is a clean, safe, and efficient electrical-based approach system for surface modification that can be used on a large scale. The technique has been widely employed in a variety of fields, particularly the pharmaceutical and medical ones. However, the method is yet to be applied comprehensively in the high voltage insulation study.

Previous studies [1], [2], [18]–[20] have been carried out on the effect of plasma-treated  $\text{SiO}_2$  nanofiller using atmospheric pressure plasma for electrical performance in insulating material. Among the issues studied were functional groups or the untreated and plasma-treated chemical composition for each sample. Yan *et al.* [18], [21] and Musa *et al.* [1], [20] applied the APP treatment method in their studies to modify the surface of the  $\text{SiO}_2$  nanofiller for the purpose of retarding the electrical treeing, increasing the breakdown strength, resisting the PD, mitigating the space charge distribution and improving the dielectric losses of the nanocomposite materials. Interestingly, they have successfully designed a new dielectric material with modified  $\text{SiO}_2$  nanoparticles augmented into epoxy resin and silicone rubber (SiR) with improved dielectric

performances. However, their studies are limited to certain parameters and materials, which the gaps are filled in this study.

The current study has improved the LDPE properties, an insulating material commonly used as high voltage insulation. Polyethylene has been chosen in this study because of its application in the power cable industry as insulation for medium and high voltage cables. Besides, BN was used as a nanofiller due to its superior dielectric properties over other types of nanofillers. Yet, according to the literature review study, there is none of research has been carried out on LDPE nanocomposites augmented with surface-treated silicon dioxide and boron nitride nanoparticles using the APP method focusing on PD and AC breakdown voltage study. As a result, this study is certain that plasma-treated nanocomposites can be used as a solution to improve PD resistance, breakdown strength, and dielectric characteristics for high voltage applications.

## 2. Methodology

### A. Sample Preparation

Two types of nanofillers, BN and SiO<sub>2</sub>, and LDPE as a polymer host were the materials used in this investigation. Lotte Chemical Titan Malaysia provided the LDPE pellets that were used. The BN and SiO<sub>2</sub> nanoparticles, in contrast, were acquired from Nanostructured and Amorphous Materials and Sigma Aldrich, respectively, and had an average size of 12 nm. The weight percentage of untreated and plasma-treated nanofillers that dispersed into the LDPE matrix was varied into 1, 3 and 5 wt% of concentration. LDPE/BN and LDPE/SiO<sub>2</sub> were compounded using a Brabender mixer with a 50 cm<sup>3</sup> chamber by melting the mixtures together at 165 °C. The high shear force electrode speed mixer was set at a speed of 35 rpm to make sure the mixing process combined the polymer and the nanofillers uniformly. The LDPE nanocomposites mixing time was maintained at 2 minutes for each sample.

Table 1. Code and composition of each sample

| Sample                                 | Composition, (wt%) |            | Plasma treatment, (mins) | Sample code |
|--|--------------------|------------|--------------------------|-------------|
|  | LDPE               | Nanofiller |                          |             |
| LDPE                                   | 100                | -          | -                        | A0          |
| LDPE / BN nanocomposites               | 99                 | 1          | -                        | B1          |
|  | 97                 | 3          | -                        | B3          |
|  | 95                 | 5          | -                        | B5          |
|  | 99                 | 1          | 15                       | BA1         |
|  | 97                 | 3          | 15                       | BA3         |
|  | 95                 | 5          | 15                       | BA5         |
|  | 99                 | 1          | 30                       | BB1         |
|  | 97                 | 3          | 30                       | BB3         |
|  | 95                 | 5          | 30                       | BB5         |
| LDPE / SiO <sub>2</sub> nanocomposites | 99                 | 1          | -                        | C1          |
|  | 97                 | 3          | -                        | C3          |
|  | 95                 | 5          | -                        | C5          |
|  | 99                 | 1          | 15                       | CA1         |
|  | 97                 | 3          | 15                       | CA3         |
|  | 95                 | 5          | 15                       | CA5         |
|  | 99                 | 1          | 30                       | CB1         |
|  | 97                 | 3          | 30                       | CB3         |
|  | 95                 | 5          | 30                       | CB5         |

The sample of LDPE nanocomposites for the partial discharge and AC breakdown strength measurements were prepared with a thickness of  $100 \pm 0.5$   $\mu\text{m}$  and a diameter of 80 mm of each sample by a hydraulic laboratory press machine. The hot press was set at 160°C for both upper and lower plates and a load of 3 tons. A stainless-steel mould with dimensions of 100 mm x 100 mm was used. Three minutes of compression were used for the pre-heating process. The moulded

sample was then maintained for 3 minutes to allow for cooling. Table 1 shows the sample code and content for each sample.

*B. Process of Nanofillers Surface Modification Using Plasma Treatment*

The filler distribution of nanoparticles in the polymer matrices was enhanced in this work by surface modification. Both nanofillers experienced plasma treatment in the plasma chamber, which utilized the DBD concept. The dimension of the plasma chamber in this study was 180 mm × 180 mm × 100 mm using a plane-to-plane electrode configuration. In the middle of the upper electrode and the glass was placed a stainless steel electrode attached with the fine wire mesh measuring 90 mm by 10 mm. The fine wire mesh was used to obtain a stable filamentary discharge. The high voltage electrode was separated from the plasma by two quartz glasses that were each 1 mm thick and served as a dielectric barrier to allow the charge to build up [22].

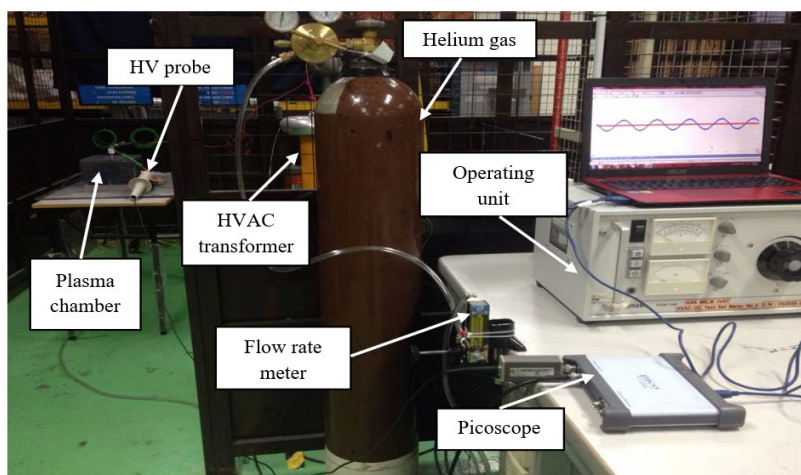
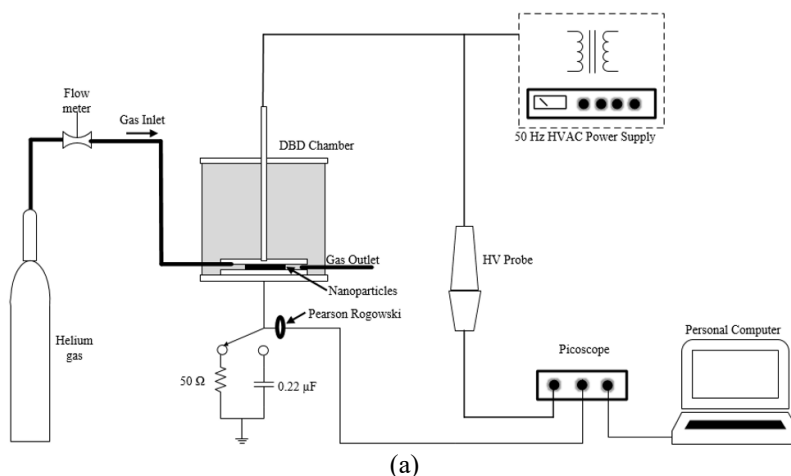


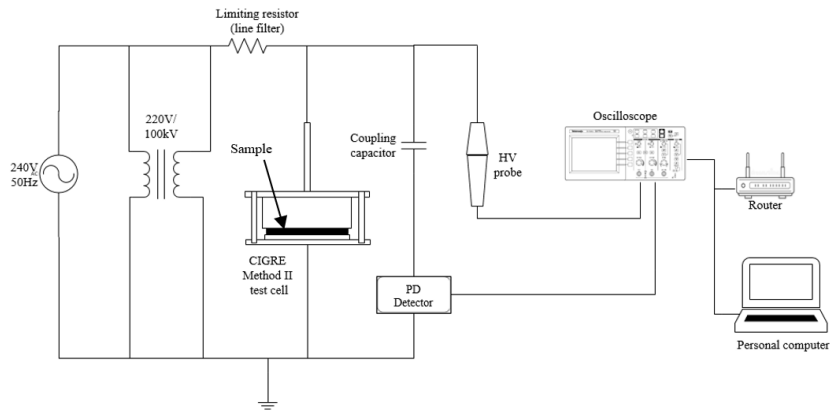
Figure 1. The schematic diagrams of plasma treatment experimental setup (a) Schematic diagram and (b) Pictorial view

The gap distance was maintained at 3 mm. A high voltage probe was used to measure the voltage applied to the electrodes (Tektronix, P6015A). The plasma chamber received a voltage of 7 kVrms at a frequency of 50 Hz. Working gas for the discharge was helium gas, delivered at a flow rate of 1 L/min. The plasma power was used at 15 W. Then, a 50 Ω resistor linked in

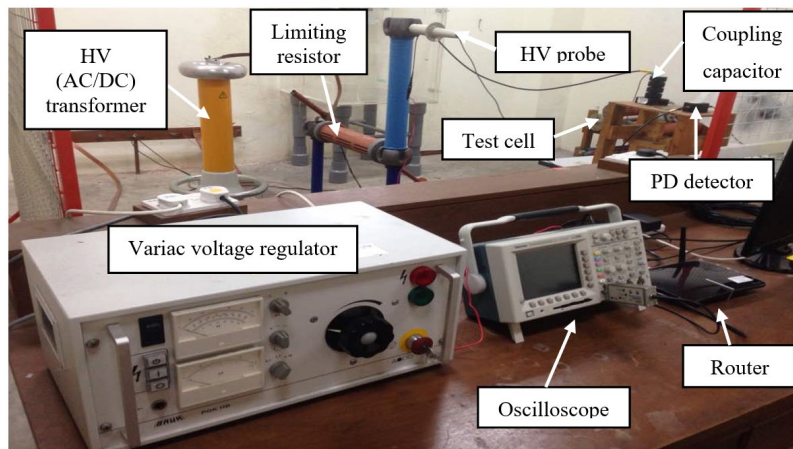
series to the ground was employed to measure the discharge current of the plasma treatment using the Pearson's Rogowski current transformer. A PicoScope 5244B was used to record the waveforms of the applied voltage and discharge current. Figure 1 shows the schematic diagram and pictorial view of the nanoparticles' setup for the plasma treatment process. The treatment was lasted for 15 and 30 minutes, respectively. To achieve a uniform exposure to the plasma irradiation, it was repeated every five minutes and swirled for 30 seconds on alternate occasions.

*C. The Measurement of Partial Discharge*

Figure 2 shows the PD measurement setup's schematic diagram and pictorial view. The IEC 60270 standard, which specified that the AC voltage of a 50 Hz power supply needs to be applied onto the solid insulation, was the basis for the PD measurement, which also included the preparation of measurement equipment as a standard of solid insulating material. This standard was established to indicate an insulating material's capacity to avoid partial discharges from starting when it is subjected to intense electrical stress. A high voltage probe was linked to the output side of the high voltage transformer in a 1:1000 volts ratio. Due to the LAN communication ports utilized to interface with the LabVIEW™ application, an oscilloscope (Tektronix, TDS 3034B) is compulsory in this PD arrangement. According to the IEC60270 standard, a 1 nF coupling capacitor and a measuring impedance were attached to the test item for the PD measurement. Each sample was aged for a period of one hour at a voltage that kept up to 4 kVrms.



(a)

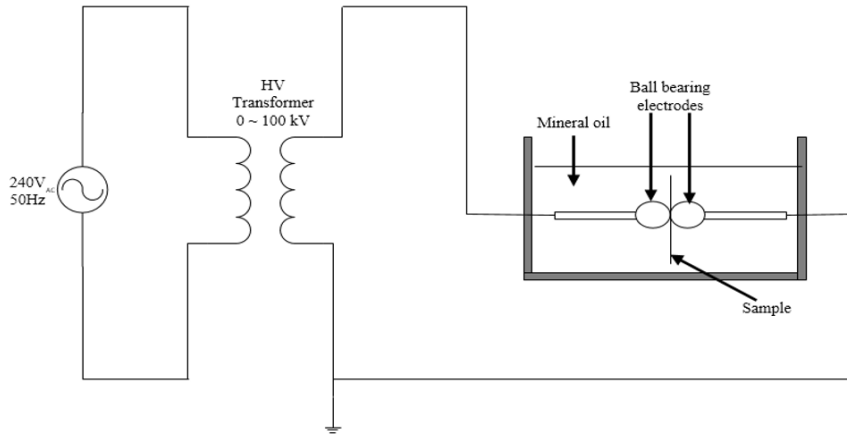


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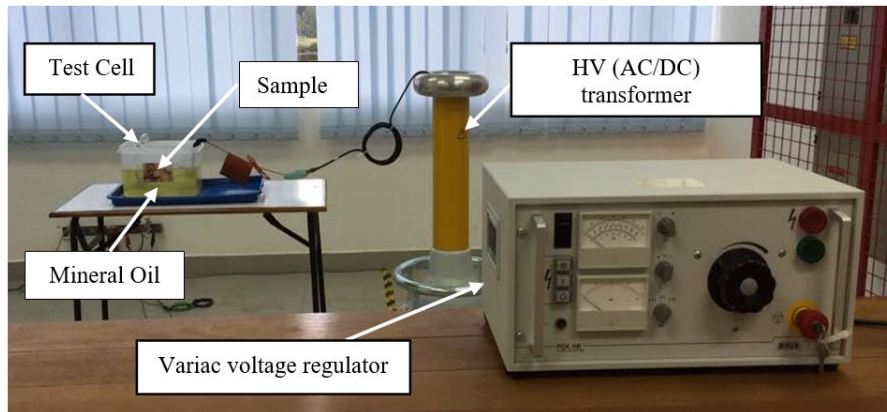
Figure 2. Schematic diagram of partial discharge measuring system (a) Schematic diagram and (b) Pictorial view

#### D. The Measurement of AC Breakdown Strength

The ASTM D149 standard served as the basis for the AC breakdown tests. As illustrated in Figure 3, the LDPE nanocomposites sample was placed between two 6.3 mm diameter steel ball-bearing electrodes that were submerged in mineral oil to minimize flashover in order to evaluate the breakdown voltage of the material. The sample was subjected to a 50 Hz HVAC voltage under a step voltage test that applied 1 kV every 20 seconds until it failed. Each sample had a total of 15 measurements taken at test points. All the data were analyzed using Weibull analysis with a two-parameter function [14].



(a)



(b)

Figure 3. The setup of AC breakdown strength measurement (a) Schematic diagram and (b) Pictorial view

### 3. Results and Analysis

#### A. Analysis of Partial Discharge Performances

The PD data in the following section show the results of partial discharge measurements of LDPE nanocomposites samples. The phase-resolved partial discharge (PRPD) patterns, the maximum PD magnitude, and the overall PD number are the PD features that are discussed in this section.

##### I. Phase-resolved Partial Discharge Patterns

The apparent charge connected and detected at the test object's terminal during partial discharge operations over the supply voltage phase is shown by phase-resolved partial discharge (PRPD), although its density is dependent on the length of the test. Figure 4 demonstrates the

PRPD pattern of the pure LDPE sample. Figures 5 and 6 display the PRPD patterns of LDPE nanocomposites containing 1, 3, and 5 wt% of SiO<sub>2</sub> and BN nanoparticles that have undergone 15-minute and 30-minute plasma treatments, respectively. It is necessary to perform a PD analysis based on the PRPD pattern to determine the kind of discharge occurred and the highest magnitude of positive and negative PD. In Accordance with the PRPD patterns that appeared on all samples, the third quadrants illustrate the phase angle from 180° to 270°, while the PD pulses typically existed in the first quadrants between 0° and 90° of phase angle.

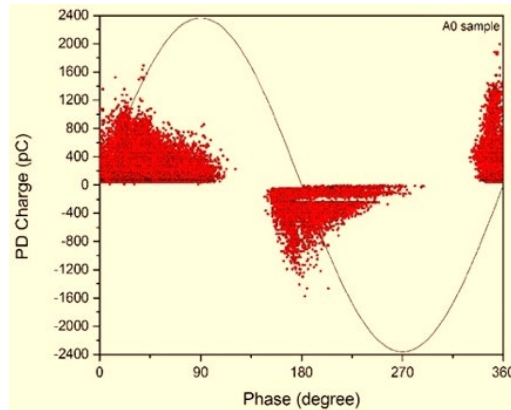
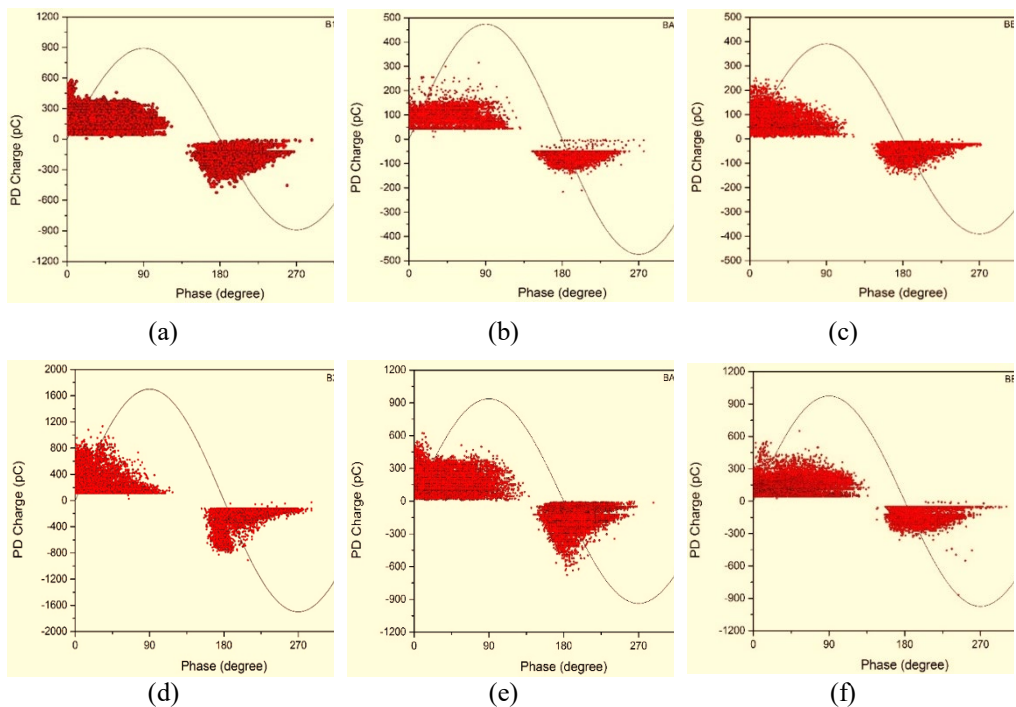


Figure 4. The PRPD pattern of the pure LDPE sample



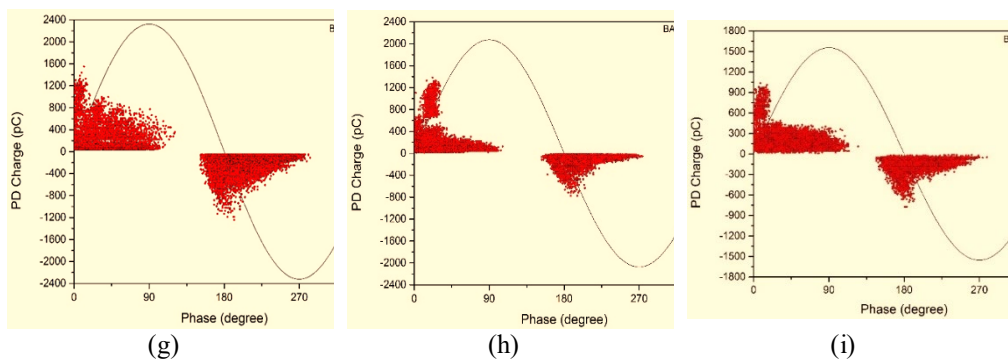
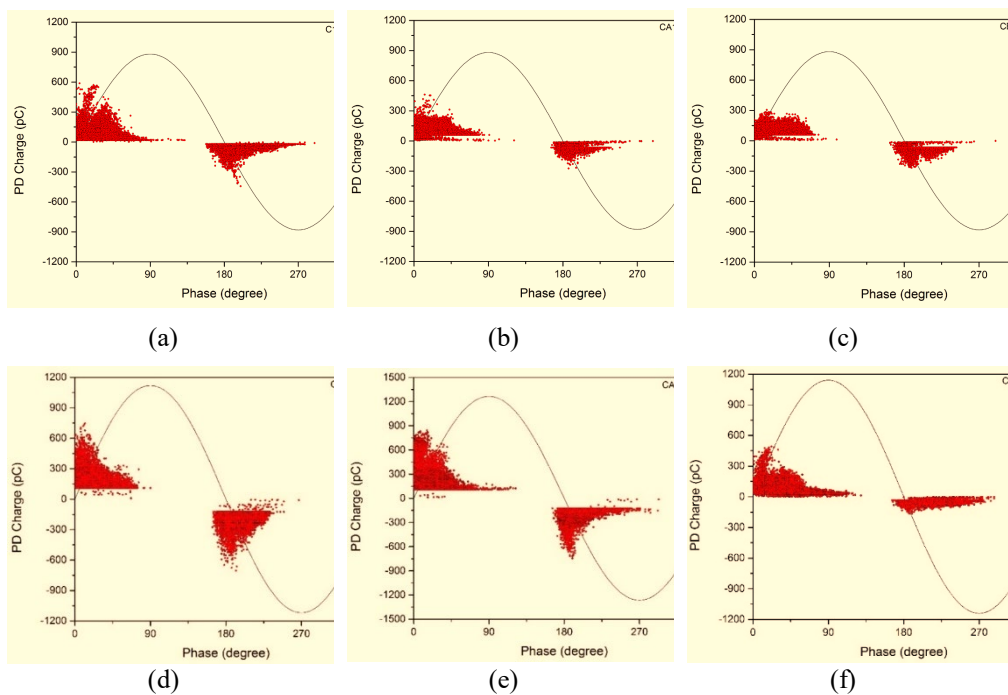


Figure 5. The PRPD patterns of LDPE nanocomposites: (a) B1, (b) BA1, (c) BB1, (d) B3, (e) BA3, (f) BB3, (g) B5, (h) BA5, and (i) BB5

Importantly, the positive and negative half-cycles' PD behaviors were essentially symmetrical since the void was in the center of the sample. As a result, internal discharges are the type of discharge seen in all samples. The Townsend's and streamer theories describe the fundamental mechanism of the partial discharges, it claims that the cathode discharge was caused by electrons produced from the cathode colliding with gas molecules which is exist inside the cavity after gaining enough energy in the electric field. The gas molecules would be ionized by this efficient impact, increasing the amount of charged particles [11]. Due to the presence of gases in the void, the dielectric constant of the void is often smaller than the solid dielectric around it (LDPE nanocomposites). So, compared to the sample of LDPE nanocomposites, the electric field in the void is larger.





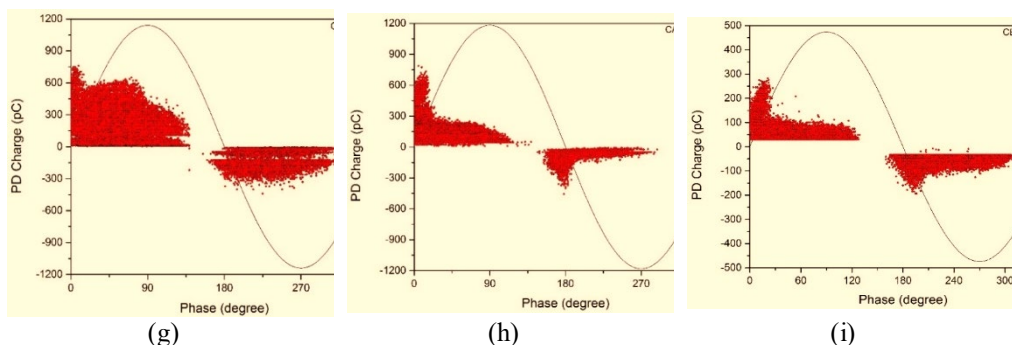


Figure 6. The PRPD patterns of LDPE nanocomposites: (a) C1, (b) CA1, (c) CB1, (d) C3, (e) CA3, (f) CB3, (g) C5, (h) CA5, and (i) CB5

Ionization will occur for the gas molecules in the void when the gas molecules come into contact with the freed electron from the cathode as it accelerates across it to the anode. An avalanche process is another name for this. On the other hand, because not every collision ionizes the gas, the discharge process is a stochastic phenomenon. This is to ensure that, upon impact, the electron may not be able to knock out another electron if its kinetic energy is insufficient. Electrons with negative charges will build up on the void walls close to the anode. On the other hand, the positively charged ionized molecule will build up on the empty walls close to the cathode.

The aggregation of electron and positive ion molecules causes the emergence of two streamer channels with opposing charge polarities. The transferred charge will create a channel in this streamer that will oppose and distort the local electric field of the external supply. Until the electric field generated by the electron and positive ion in the void exceeds the specific value of the extinction electric field, this action happens quickly and without interruption. Following the partial discharge, the LDPE nanocomposites dielectric in the middle of the two conduction sections is partially bridged by quenching. The strongest PD magnitude is then translated into dot points, as in the PRPD patterns, once the PD signal is first detected as PD pulses.

## II. The Magnitude of Partial Discharge

From the PRPD plots, the maximum PD magnitudes of pure LDPE and LDPE nanocomposites containing BN and SiO<sub>2</sub> nanoparticles were recovered and presented in Figures 7 and 8, for positive and negative PD magnitudes, respectively. The largest PD magnitude was found in the pure LDPE sample (A0), which had positive of 1992 pC and negative magnitudes of 1577 pC. Meanwhile, it can be shown that the positive as well as the negative PD magnitude of the LDPE nanocomposites sample was below 1600 pC for those that contained BN nanoparticles. By inserting BN nanofillers into LDPE at a rate of 1 wt% (B1 sample) resulted to reducing the PD magnitude to 580 pC. The addition of untreated BN nanofiller increased for 3 wt% (B3 samples) and 5 wt% (B5 samples), has led to a significant rise in the PD magnitude recorded at 1133 pC and 1549 pC, respectively.

After 15 minutes of plasma treatment, the BN nanocomposites of the BA1, BA3, and BA5 samples obtained PD magnitudes of 316 pC, 625 pC, and 1382 pC, respectively. In comparison to untreated BN nanofillers and A0 samples, the PD magnitude of the nanocomposites with the 15-minute plasma-treated BN dramatically decreased. However, for nanocomposites containing BN that were plasma-treated for 30 minutes (BB1, BB3, and BB5 samples), the observed difference in the filler loading levels' effects on PD magnitude was 249 pC, 650 pC, and 1036 pC, respectively. As the filler loading levels were increased up to 5 wt%, the positive and negative PD magnitude exhibited a slight increase of LDPE containing untreated BN and plasma-treated BN nanoparticles. However, the BB3 sample increased the negative PD magnitude compared to the BA3 sample, and it became slightly decreased at higher filler loading.

The outcome demonstrates that when related to LDPE with untreated BN and 15 minutes of plasma-treated BN nanoparticles, the LDPE filled with 30 minutes of plasma-treated BN nanoparticles lowered their positive as well as negative PD magnitude. These findings concur with Yan *et al.* [19] who found that the larger amount of oxygen atoms linked to the surface of plasma-treated nanoparticles following the treatment method also contributed to a reduction in the PD magnitude.

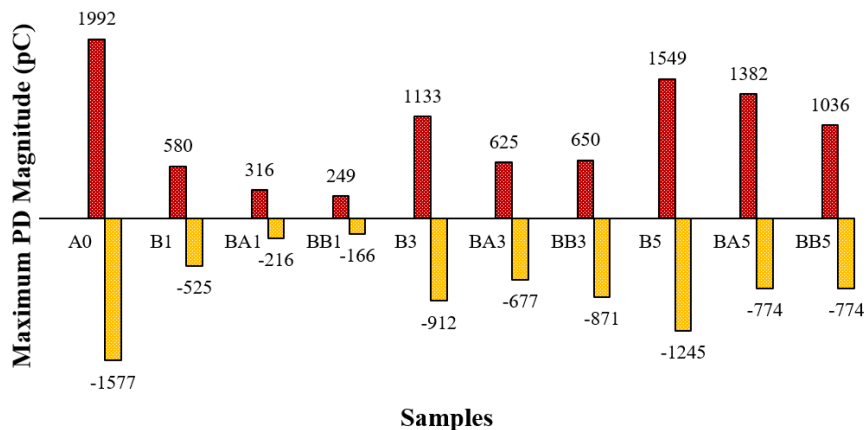


Figure 7. Comparison of the maximum PD magnitude of LDPE nanocomposites containing 1 wt%, 3 wt% and 5 wt% untreated and plasma-treated BN nanofillers

It can be shown that the PD magnitude trends for LDPE with SiO<sub>2</sub> nanofiller and BN nanofiller were similar. According to the results, related to the A0 sample, the PD magnitude has decreased since the addition of a tiny amount of SiO<sub>2</sub> nanofiller. In contrast to the nanocomposites filled with BN nanoparticles sample, PD pulse occurrences in LDPE treated with plasma or left untreated with SiO<sub>2</sub> nanofiller were a little bit less frequent for both positive and negative polarities. The PD magnitude values for nanocomposites with 1 wt%, 3 wt%, and 5 wt% of untreated SiO<sub>2</sub> nanofiller into LDPE (C1, C3, and C5 samples) were 587 pC, 746 pC, and 760 pC, respectively. Compared to the A0 sample, these PD magnitudes were less.

The C5 sample had the highest PD pulses among the C1 and C3 samples due to the higher filler concentration, which contributed to space charge trapping [23]. The PD magnitudes of LDPE containing SiO<sub>2</sub> nanoparticles treated with plasma for 15 minutes were around 461 pC, 843 pC, and 787 pC, respectively. Additionally, the 30-minute plasma-treated SiO<sub>2</sub> nanoparticle additions to LDPE nanocomposites at 1%, 3%, and 5% resulted in PD magnitudes of 305 pC, 491 pC, and 283 pC, respectively. However, among all the plasma-treated samples, the CA3 and CB3 sample with 3 wt% plasma-treated SiO<sub>2</sub> nanoparticles displayed the highest PD magnitude. At 1 wt% to 5 wt% of untreated nanocomposite samples, the positive and negative PD magnitudes can be seen to have marginally increased, and the negative PD magnitude has an abruptly decreased pattern at 5 wt% filler loading of the C5 sample. The CB5 sample had the smallest positive PD magnitude when compared to other samples and the plasma-treated nanocomposites. In comparison to the nanocomposite sample containing untreated SiO<sub>2</sub> and 15 minutes of plasma-treated SiO<sub>2</sub> nanoparticles, the longer the treatment duration, the lower the positive and negative PD magnitudes. It was found that the positive and negative PD magnitudes of CA3 and CA5 samples were greater than the untreated SiO<sub>2</sub> and 30-minutes plasma-treated SiO<sub>2</sub> nanofillers.

For a one-hour PD measurement, the maximum PD magnitude results were below 2000 pC. Pure LDPE, the A0 sample, was shown to have the largest PD magnitude compared to samples made from LDPE/BN and LDPE/SiO<sub>2</sub> nanocomposites. The results showed that the samples of B5 and BB1 have the highest and lowest PD magnitude per cycle, respectively. It can be observed that the PD magnitude for the nanocomposites sample was below 900 pC. The highest PD

magnitude was 843 pC for the CA3 sample, except for the A0 sample, and CB5 was the lowest magnitude among others.

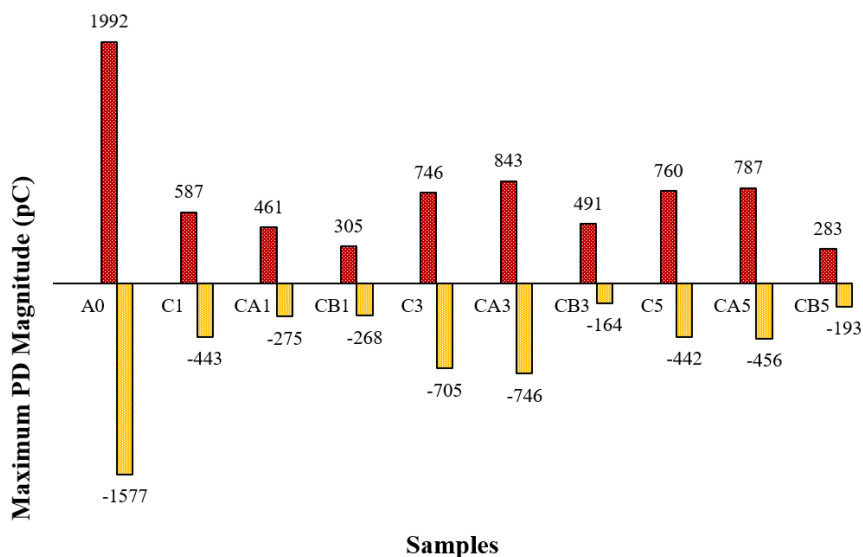


Figure 8. Comparison of the maximum PD magnitude of LDPE nanocomposites containing 1 wt%, 3 wt% and 5 wt% of untreated and plasma-treated SiO<sub>2</sub> nanofillers

In comparison to samples of LDPE with SiO<sub>2</sub> nanofiller, those with BN nanofiller had the highest PD magnitude. The pure LDPE sample, however, showed the highest PD magnitude when compared to the other samples. By arranging nanosized fillers in the host polymer, the addition of nanofillers to LDPE composites would generate a wall. During the electrical stress, nanofillers served as a resistance to the flow of electrons in the middle of two electrodes. The LDPE sample with nanofillers was therefore shown to be a more PD-resistant insulator than the LDPE sample without nanofillers. The polymer matrix's nanofillers act as a defense against PD attack. Hilmey *et al.* [24] and Arief *et al.* [25] provided evidence in support of this claim by confirming that the addition of nanofillers improved the ability of LDPE nanocomposites to tolerate the surface degradation caused by PD. The PRPD results show that PD pulses have slightly reduced for the LDPE with SiO<sub>2</sub> samples for both half cycles compared to the BN samples. Due to the surface modification of the nanoparticles, the plasma-treated BN and SiO<sub>2</sub> nanofillers showed a smaller PD magnitude than untreated samples, which may be a result of the stronger chemical interactions between the nanofiller and polymer matrix [18].

### III. Number of Partial Discharge

Figure 9 displays the total number of PD pulses for various samples of LDPE containing 1 wt%, 3 wt%, and 5 wt% of untreated, 15 minutes, and 30 minutes plasma-treated BN nanoparticles after one hour of high voltage stress. Upon completion of an hour of high voltage stress, the total PD pulse number of the A0 sample is 42865. The overall number of PD pulses was slightly lower in the LDPE sample with a tiny amount of BN nanoparticles added compared to the A0 sample. The total PD values for untreated BN nanoparticles increased as the filler loading was increased from 1 wt% to 5 wt% of loading levels. These values varied from 29497 for the B1 sample to 31471 for the B5 sample. When plasma was applied to the nanoparticles, the frequency of PD pulses decreased.

In comparison to the untreated and pure LDPE samples, the total PD pulses of the BA1, BA3, and BA5 samples as well as the samples with 15 minutes of plasma-treated BN nanoparticles had a reduced PD number. As a result, a significant increase in the total number of PD pulse counts was seen for the LDPE nanocomposites with 30-minute plasma-treated BN nanoparticles

for the BB1, BB3, and BB5 samples in comparison to LDPE with untreated and the 15-minute plasma-treated BN nanoparticles sample. The results generally indicated that the overall PD pulse counts were below 32000, with the exception of the BB5 sample, which had the highest total pulse numbers at 39441. Overall, the BA3 sample generated the fewest PD pulses, 23713.

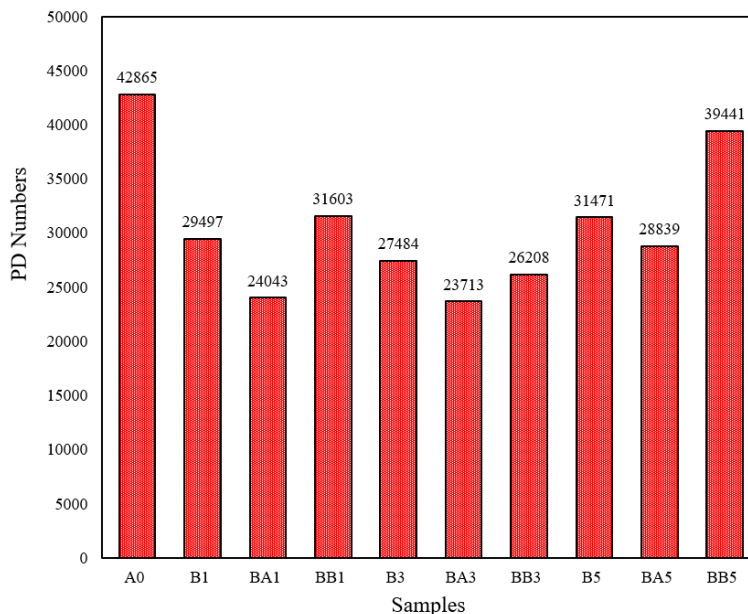


Figure 9. The total PD pulse number of pure LDPE and LDPE nanocomposites containing 1 wt%, 3 wt% and 5 wt% untreated and plasma-treated BN nanoparticles

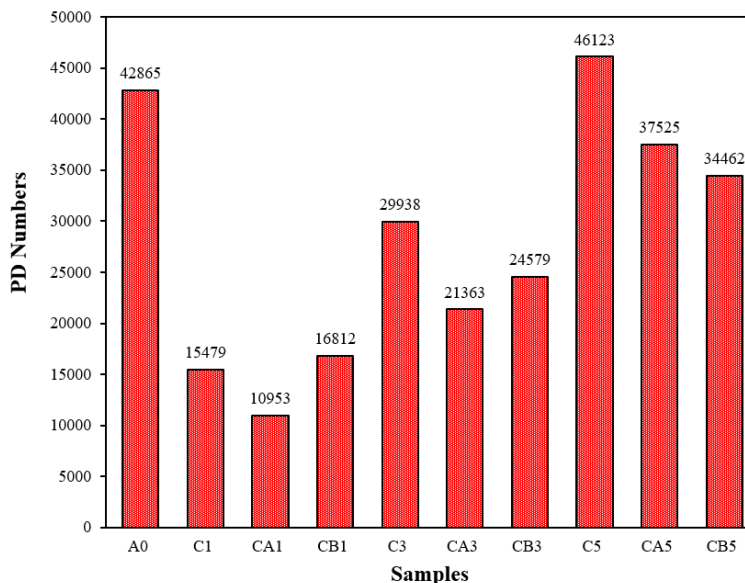


Figure 10. The total PD pulse number of pure LDPE and LDPE nanocomposites containing 1 wt%, 3 wt% and 5 wt% untreated and plasma-treated SiO<sub>2</sub>

Figure 10 shows the total PD pulse number for different samples of LDPE nanocomposites added with SiO<sub>2</sub> nanofillers, respectively. With the addition of 1 wt% and 3 wt% untreated nanoparticles, the total PD pulse number was lower than the A0 sample. Meanwhile, the total

PD numbers of samples with untreated SiO<sub>2</sub> nanofillers (C1, C3 and C5), samples with 15-minutes plasma-treated SiO<sub>2</sub> nanofillers (CA1, CA3 and CA5) and samples with 30-minutes plasma-treated SiO<sub>2</sub> nanofillers (CB1, CB3 and CB5) had slightly increased with the increase in the filler loading levels. With the addition of 5 wt% untreated SiO<sub>2</sub>, the C5 sample exhibited a higher total PD number, which was followed by the CA5 and CB5 samples, respectively. Additionally, the total PD number for the CA3 and CA5 samples significantly decreased. Total PD values for samples CA1 and CB3 were somewhat greater than for sample C1.

The nanocomposites with SiO<sub>2</sub> samples had higher PD values at the highest filler loading level when compared to the LDPE containing BN nanoparticles. These findings support Sayuti's [26], claim that SiO<sub>2</sub> nanofiller added to LDPE had a greater effect. The concentration of nanoparticles and the frequency of PD pulses tend to increase with increased filler loading in LDPE. Conversely, it was noted from the nanocomposite samples with plasma-treated nanoparticles; the detected PD pulse number was slightly lower compared to the untreated nanoparticles and pure LDPE sample. However, BB1 and BB5 samples and CB1, when the rise in time treatment significantly increased the total PD pulse numbers compared to untreated and 15-minutes plasma-treated nanoparticles. This could be due to the plasma discharge containing many active species, which led to the formation of radicals when the longest treatments were made during plasma treatment at atmospheric conditions. The result agrees with Arief *et al.* [25], who observed that the enhanced electric field intensity caused the electrons to move more quickly and with enough kinetic energy to push against one another, leading to a rise in collision, ionization, and avalanche events.

### B. Analysis of AC Breakdown Strength

Figure 11 and Table 2 summarize the AC breakdown strength trends of pure LDPE and nanocomposites containing untreated and plasma-treated BN nanoparticles. The AC breakdown strength of the LDPE with untreated BN nanoparticles showed a significant reduction in results compared to the pure LDPE (A0 sample), which were 145.01 kV/mm, 132.96 kV/mm, and 130.07 kV/mm for 1, 3, and 5 wt% (B1, B3, and B5 samples), respectively. The highest AC breakdown strength of 155.47 kV/mm was recorded for the A0 sample. Conversely, when the amount of filler loading increased to 5 wt%, the B5 sample had the lowest results of breakdown strength compared to other samples; the value was reduced to about 25.4 kV/mm lower than that of the A0 sample. The results align with the previous researchers, Qiang *et al.* [16], who experimented with the reduced AC breakdown strength of epoxy nanocomposites with untreated BN nanoparticles.

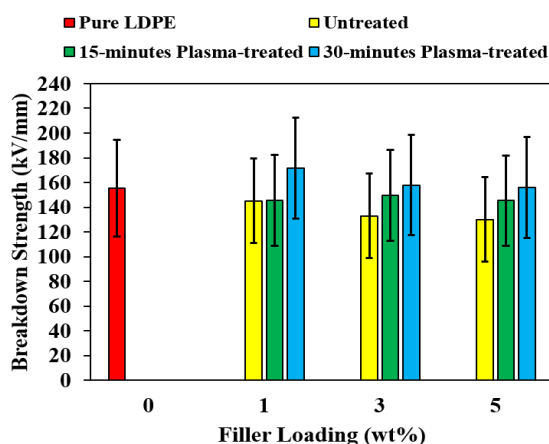


Figure 11. Summary of AC breakdown strength obtained from Weibull Analysis of pure LDPE and nanocomposites containing 1 wt%, 3 wt% and 5 wt% untreated BN and plasma-treated BN nanoparticles

Compared with untreated samples, LDPE with 15 minutes plasma-treated BN nanoparticles showed an increment trend in AC breakdown strength if related to untreated BN nanoparticles samples, 145.54 kV/mm, 149.66 kV/mm, and 145.32 kV/mm for BA1, BA3, and BA5 samples respectively. However, in contrast to the BA1 and BA5, the BA3 sample exhibited the highest breakdown strength value. This surprising result may be attributable to the stronger chemical bonds between the nanofiller and polymer matrices following plasma treatment [18]. That might be the cause of the samples of LDPE nanocomposites' increased AC breakdown strength. There is a clear increment trend in the AC breakdown voltage of all samples with the introduction of 30-minutes plasma-treated BN nanoparticles. The AC breakdown voltage of BB1 significantly increased as the concentration of BN nanofiller increased to 1 wt%. After adding 3 wt% of BN nanofiller, the AC breakdown voltage showed a slight decrease from 171.66 kV/mm to 157.96 kV/mm. Meanwhile, the AC breakdown voltage for BB5 and A0 samples was similar, ranging from 155.47 kV/mm to 155.93 kV/mm. When the plasma treatment time was raised to 30 minutes, the AC breakdown strength marginally increased.

Table 2. AC breakdown strength results from Weibull analysis of the pure LDPE, LDPE containing untreated BN and plasma-treated BN nanofillers

| Samples | Weibull distribution parameter       |                               |
|---------|--------------------------------------|-------------------------------|
|         | Breakdown strength (kV/mm), $\alpha$ | Failure distribution, $\beta$ |
| A0      | 155.47                               | 10.11                         |
| B1      | 145.01                               | 11.19                         |
| B3      | 132.96                               | 18.41                         |
| B5      | 130.07                               | 12.92                         |
| BA1     | 145.54                               | 16.02                         |
| BA3     | 149.66                               | 13.01                         |
| BA5     | 145.32                               | 9.66                          |
| BB1     | 171.66                               | 14.03                         |
| BB3     | 157.96                               | 17.43                         |
| BB5     | 155.93                               | 17.77                         |

For LDPE, the addition of the SiO<sub>2</sub> nanofiller may alter the AC breakdown strength. The AC breakdown strength trends of pure LDPE and nanocomposites containing untreated and plasma-treated SiO<sub>2</sub> nanoparticles are summarised in Figure 12 and Table 3. For untreated SiO<sub>2</sub> nanoparticles, the addition of 1 wt% of untreated SiO<sub>2</sub> increased the AC breakdown strength from 155.47 kV/mm of the A0 sample to 160.15 kV/mm for the C1 sample. Meanwhile, LDPE's AC breakdown strength performance decreased to 159.78 kV/mm once SiO<sub>2</sub> was increased to 3 wt% for the C3 sample. The highest value of AC breakdown strength for the untreated SiO<sub>2</sub> sample was observed at the higher loading level of 5 wt%, having 170.71 kV/mm compared to the A0 sample.

The addition of a small amount of SiO<sub>2</sub> filler loading increased the AC breakdown value. As a result of electrical stress that turn fast electrons into slow, negatively charged nanoparticles, it is implied that the nanoparticles are conductive materials that serve as electron scavengers in the insulation material. It can increase the streamer propagation process to the breakdown under electrical stress for the polymer-containing nanoparticles because of the electron scavenger mechanism [27]–[29]. The improvement of breakdown strength was attributed to the ability to

suppress an electron avalanche by trapping the electrons of the nanoparticles. The presence of a large number of interfaces between the nanofiller and polymer contributed to the increase in the trapping of charge carriers. The increase of trapped charge carriers around the boundary of the polymer chains led to a reduction in the mobility of the charge carriers in the insulation material. The mobility reduction would have enhanced the dielectric strength because more time and energy would be needed to reduce the charge carriers for conduction [23], [30], [31]. Therefore, the increase in filler loading increased the AC breakdown strength due to the trapping of charge carriers. The trapping of charge carriers increased due to the inclusion of nanoparticles.

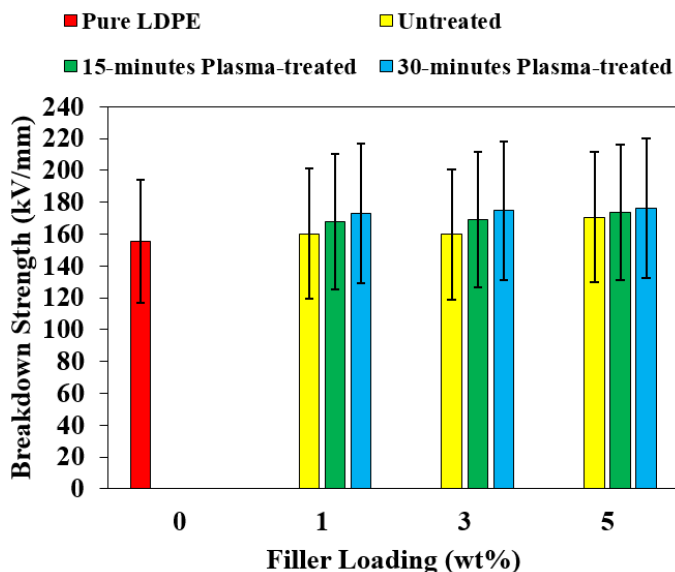


Figure 12. Summary of AC breakdown strength obtained from Weibull Analysis of pure LDPE and nanocomposites containing 1 wt%, 3 wt% and 5 wt% untreated SiO<sub>2</sub> and plasma-treated of SiO<sub>2</sub> nanoparticles

Table 3. AC breakdown strength results from Weibull analysis of the pure LDPE, LDPE containing untreated and plasma-treated SiO<sub>2</sub> nanofillers

| Samples | Weibull distribution parameter       |                               |
|---------|--------------------------------------|-------------------------------|
|         | Breakdown strength (kV/mm), $\alpha$ | Failure distribution, $\beta$ |
| A0      | 155.47                               | 10.11                         |
| C1      | 160.15                               | 12.45                         |
| C3      | 159.78                               | 12.77                         |
| C5      | 170.71                               | 19.11                         |
| CA1     | 167.80                               | 17.19                         |
| CA3     | 168.99                               | 24.66                         |
| CA5     | 173.65                               | 18.93                         |
| CB1     | 173.01                               | 17.62                         |
| CB3     | 174.70                               | 21.56                         |
| CB5     | 176.44                               | 31.04                         |

The AC breakdown strength for nanocomposites containing plasma treated SiO<sub>2</sub> nanoparticles, the AC breakdown strength led to a significant improvement than that of the nanocomposites containing untreated SiO<sub>2</sub> samples. With the presence of 15-minutes plasma-treated SiO<sub>2</sub> nanoparticles within the LDPE matrix, the AC breakdown voltage of the CA1 sample increased from 155.47 kV/mm to 167.80 kV/mm once 1 wt% of SiO<sub>2</sub> is added. After adding 3 wt% and 5 wt% of SiO<sub>2</sub>, the AC breakdown voltage of CA3 and CA5 samples showed a steady increment of 168.99 kV/mm and 173.65 kV/mm, respectively. The AC breakdown strength of the plasma-treated SiO<sub>2</sub> nanoparticles was, however, somewhat higher than that of the untreated samples, with values for the CB1, CB3, and CB5 samples of 173.01 kV/mm, 174.70 kV/mm, and 176.44 kV/mm, respectively. When compared to other samples, the CB5 sample had the highest AC breakdown strength and form parameter value.

In contrast, the breakdown strength falls with an increase in the number of untreated BN nanoparticles. This is because bigger BN flakes have a very high density of defects, but small BN flakes may have a higher magnitude of local internal fields [32]. However, the plasma-treated BN nanoparticles sample marginally increased than untreated samples, but it was still lower than the base material as the amount of nanofiller increased. Moreover, the AC breakdown strength results were slightly higher for LDPE-containing untreated and plasma-treated SiO<sub>2</sub> nanoparticles, the base material when filler loading increased. These two nanofillers showed that the nanocomposites containing SiO<sub>2</sub> exhibited higher breakdown strength than BN nanoparticles.

Overall results showed that the plasma treatment enhanced the breakdown strength. The results agree with the findings reported by Yan *et al.* [18], who stated that plasma-treated nanocomposites increased the breakdown strength. With the plasma treatment of nanoparticles, the size of agglomeration can be reduced in the sample containing plasma-treated nanoparticles compared with the nanocomposites containing the untreated nanoparticles [18]. Therefore, it can be implied that the surface treatment has contributed to improving the AC breakdown strength. The best formulation of breakdown strength performance has been seen at about 1 wt% of BN and 5 wt% of SiO<sub>2</sub> nanofiller loading.

#### 4. Conclusion

The partial discharge and AC breakdown strength of pure LDPE, untreated, 15 minutes and 30-minutes plasma-treated LDPE/SiO<sub>2</sub> and LDPE/BN nanocomposites have been successfully investigated and analyzed. The outcomes demonstrate that the APP method's treatment of SiO<sub>2</sub> and BN nanoparticles enhanced the partial discharge resistance and AC breakdown strength. For LDPE/SiO<sub>2</sub> nanocomposites, the increase in filler loading levels resulted in the increasing trend of the AC breakdown strength for both nanocomposites filled with untreated and plasma-treated nanofillers. In addition, LDPE/5 wt% 30-minutes plasma-treated SiO<sub>2</sub> sample demonstrated the highest AC breakdown strength and the lowest PD magnitude among the samples. In contrast, the untreated LDPE/BN nanocomposites sample had an increased filler loading level but came into a decrease in AC breakdown strength and increased PD magnitude. Still, the plasma treatment has contributed to the increase in the AC breakdown strength and reduced the PD magnitude and the PD number. The LDPE/1 wt% 30-minutes plasma-treated BN sample had the highest AC breakdown strength and the lowest PD magnitude, therefore, contributed as the optimum formulation based on the outcome of this study.

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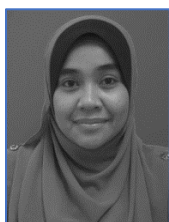
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