Partial Discharge and Breakdown Strength of Plasma Treated Nanosilica/LDPE Nanocomposites

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***Abstract*— Nanocomposites have been actively studied in recent years as an insulating material due to their excellent in electrical, mechanical and thermal properties. Even though, the addition of nanoparticles into polymer matrices showed better performance in relation to partial discharge (PD) and AC breakdown strength tests. However, the introduction of nanoparticles could lead to the formation of agglomeration of the fillers which may nullify the true capabilities of the composites. Therefore, silane coupling agent was introduced for surface functionalization treatment of the nano filler but among the issues associated are toxicity and complexity. In the present study, atmospheric pressure plasma is proposed to enhance the surface functionalization of the nano filler. This proposed method was used to treat the nanosilica (SiO_2) surfaces to enhance the interfacial interaction between the host (LDPE) and nano filler. SiO_2 nano filler was added into the LDPE at weight percentages of 1, 3 and 5%. The phase-resolved PD behaviour and Weibull analysis of AC breakdown strength of untreated and plasma-treated LDPE nanocomposites were measured to evaluate the performance of the samples. As results, the plasma treated LDPE nanocomposites experience apparent increments of the PD resistance and AC breakdown strength as compared to the untreated nanocomposites. It is implied that the plasma treatment of nanosilica has contributed to the enhancement of the filler dispersion and eventually reducing the agglomeration.**

Keywords— Partial discharge, AC breakdown strength, atmospheric pressure plasma; nanosilica; Low Density Polyethylene.

I. INTRODUCTION

Nowadays, polymeric insulating material for example cross-linked polyethylene has attracted much attention due to superior performances especially in power cable application. However, it is eventually subjected to the degradation processes which one of the root causes is electrical discharge. These discharge phenomenon have been considered for an insulation diagnostics and assessment [1]. This drawback of the existing insulating material has opened to a new research area called nanodielectrics. Generally, the nanodielectrics is the combination of host polymer and nanofillers to improve several physical, chemical and electrical properties such as higher PD resistance, higher breakdown strength, reduced space charge accumulation, low tangent delta and reduced permittivity [2]–[4]. Moreover, nanocomposites were controlled by the interfaces between the host polymer and the nanofiller. In addition, the interfaces determine to the surface compatibility between both polymer matrices and the nano fillers. Many methods have been introduced to improve the surface compatibility such as silane coupling agent. This method was identified to produce toxic and inappropriate for mass production [5].

There is an alternative method namely atmospheric pressure plasma treatment method to treat the surface of the nanoparticles. This treatment method was able to enhance the interfacial bonding between the nano fillers and polymer matrices [6][7]. In this study, plasma treatment was used to modify the surface of SiO_2 nanoparticles and the modified nanosilica was augmented into the LDPE polymer to produce plasma treated nanocomposites. Thus, the produced nanocomposites sample were then tested in the laboratory to investigate their PD characteristics and AC breakdown strengths. In addition, the different weight percentages of the treated nanosilica augmented into the LDPE was studied in order to find the optimum formulation. It was believed that the limitation of the existing polymer can be solved using the plasma treated nanocomposites as the solution to enhance the PD resistant insulation for high voltage application.

II. EXPERIMENTAL

A. Materials

Low density polyethylene (LDPE) was used as the host polymer in this study. LDPE was produced by Titan Chemical, Malaysia with a density of 0.922 g/cm^3 and the melting index of 25 g/10min. Silicon dioxide (SiO_2) was used as nanofiller was supplied by Sigma Aldrich with an average particle size of 12 nm.

B. Plasma Treatment

A plasma chamber was designed for surface treatment of nanoparticles having a dimension of 180 mm ×180 mm ×100 mm using two circular plane stainless-steel electrodes with 90 mm × 10 mm in diameter. This arrangement of plasma chamber was applied the dielectric barrier discharge (DBD) concept of electrodes configuration. Both the electrodes were covered by a quartz glass material with thickness of 1mm used as DBD to avoid from flashover. The glass material provided as DBD to maintain the plasma from the occurrences of micro discharge.

A carrier gas moves between both electrodes and it was ionized to create plasma discharge. Fine wire mesh was inserted between high voltage and ground electrode in order to obtain a stable glow discharge. The gap spacing was kept

constant at 3 mm. Fig. 1 shows a schematic diagram of the setup for the plasma treatment chamber of the nanosilica nanoparticles Helium gas with a flow rate of 1 L/min used as the working gas for discharge was supplied by Airgas Sdn. Bhd. The applied voltage between 7 to 8.5 kV_{rms} at a frequency 50 Hz was used as a power supply to the plasma chamber. The power consumption of plasma discharge was kept constant between 3 to 15 W.

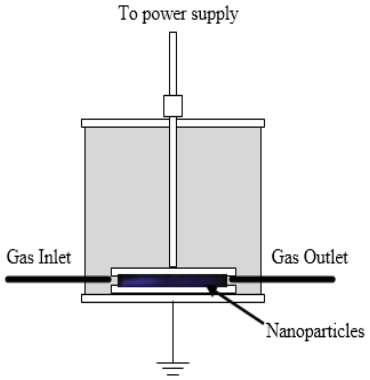


Fig. 1. Schematic diagram of a setup for plasma treatment

The occurrences of filamentary discharge during plasma treatment have been concerned in order to get a homogenous plasma treatment of nanoparticles. To obtain a homogenous and uniform plasma, the nanoparticles were first treated by the atmospheric pressure plasma for 5 minutes and then the nanoparticles were stirred for 30 seconds. This treatment process was repeated 6 times to get the total treatment time of the nanoparticles was 30 minutes [6].

C. Sample Preparation

For the preparation of nanocomposites, the LDPE and SiO_2 nanofiller was weighted using Radwag, ASX 220 analytical balance to ensure that 1005 of LDPE and 1wt%, 3wt% and 5wt% of the total weight of SiO_2. Then, the compounding process of nanocomposites using the Brabender mixer. LDPE nanocomposites were prepared by melt mixing at 165 °C with a rotational speed of 35 rpm. The mixing time was taken for 2 minutes for each sample. Table I shows the sample code and composition of each sample.

TABLE I. CODE AND SAMPLE COMPOSITION OF EACH SAMPLE

Sample code	Composition		
	LDPE (wt%)	*SiO_2 (wt%)*	*Time treatment (minutes)*
A0	100	0	0
B1	100	1	0
B3	100	3	0
B5	100	5	0
C1	100	1	30
C3	100	3	30
C5	100	5	30

After that, LDPE nanocomposites sample had undergone compression process to get the thickness of 100 µm using the Carver hydraulic laboratory press at a temperature of 160 °C. Preheating process was done in this process for 3 minutes followed by 3 minutes of compression process using 2.5 ton pressure. Table I shows the sample code and composition of each sample. Preheating process was done in this process for 3 minutes followed by 3 minutes of compression process using 2.5 ton pressure.

D. AC Breakdown Measurements

The experimental test of AC breakdown voltage were conducted The AC breakdown tests were conducted to observe the breakdown strength of the LDPE containing 1, 3 and 5wt% of untreated and plasma treated SiO_2 nanofiller. The AC breakdown test was based on the American Standard for Testing Materials (ASTM) D149. The measurements of AC breakdown voltage were performed by placing the nanocomposites sample between two 6.3 mm diameter steel ball bearing electrodes immersed in mineral oil in order to avoid from flashover. AC breakdown measurement having an AC voltage with a ramp rate voltage of 1 kV for every 20 seconds was applied to the sample until the breakdown occurred. The totals of 15 breakdown test points were measured on each sample. Fig. 2 shows the experimental setup of AC breakdown tests.

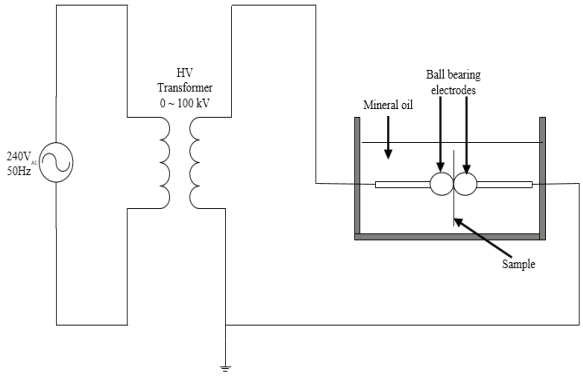


Fig. 2. Experimental setup of AC breakdown tests

E. Partial Discharge Measurements

Fig. 3 depicts the experimental setup for partial discharge testing. CIGRE Method II test cell was used in this research work to generate PD characteristics in solid insulation. PD measurement has followed the IEC 60270: 2000 standard [8] including preparation of measurement tools as a standard of solid insulating material which stated that the AC voltage of 50 Hz power supply needed to be injected in the solid insulation. The output high voltage transformer was connected to high voltage probe with a ratio of 1:1000 volts. An oscilloscope (Model Tektronix TDS 3034B) requires in this PD setup due to the LAN communication ports availability used to connect with LabVIEW™ program. A 1 nF coupling capacitor was used as a voltage divider to make sure the voltage does not rise on the impedance of PD signals. PD data were analyzed by using a PD program which developed based on the LABVIEW™ software. The voltage was applied up to 4 kV_{rms} for 1-hour ageing time of each sample.

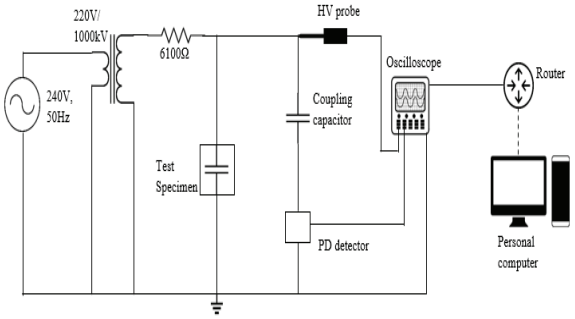


Fig.3. Experimental setup of partial discharge measurement studies

III. RESULT AND DISCUSSION

A. AC Breakdown Strength

Fig. 4 represents the summary of AC breakdown strength for LDPE containing 1wt%, 3wt% and 5wt% of untreated and plasma treated nanosilica. With the addition amount of nanosilica concentration in LDPE, the AC breakdown strength values increased for both untreated and plasma treated samples compared to pure LDPE sample which were 160.15 kV/mm, 159.78 kV/mm, and 170.71 kV/mm for B1, B3 and B5 samples respectively. For A0 sample, the AC breakdown strength was 155.47 kV/mm. However, AC breakdown strength for the B3 sample almost similar with the B1 sample; it may be caused by the agglomeration of nanofiller that exists in the polymer matrices after blending process. However, it can be clearly noticed that the AC breakdown strength of the plasma treated nanosilica was slightly higher compared to the untreated samples having results of 173.01 kV/mm, 174.70 kV/mm and 176.44 kV/mm for C1, C3 and C5 samples respectively. The highest value of AC breakdown strength was recorded for C5 sample compared to other samples.

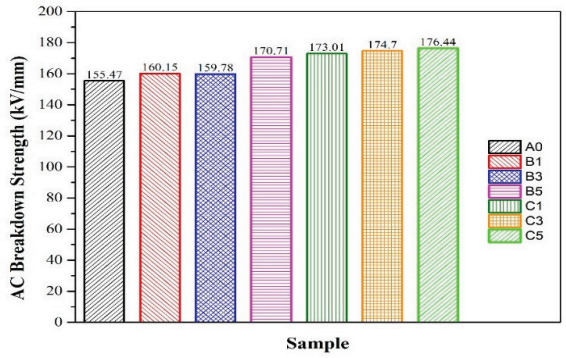


Fig. 4. Comparison of AC breakdown strength for untreated and plasma treated SiO_2 nanofiller

B. Phase-resolved Partial Discharge Patterns

The phase-resolved partial discharge (PRPD) patterns of internal discharge using CIGRE Method II at 4 kV_{rms} AC sinusoidal voltage are presented in Fig. 5. Each dot points represent the maximum PD magnitude of all the PD pulses that occur within the first and third quadrants of the voltage waveform. Fig. 5(a) shows the unsymmetrical PRPD pattern of the pure LDPE sample because the positive PD pulse count is higher than negative pulse count for A0 sample and it has the highest PD magnitude was recorded up to 1800 pC. This is due to the unbalanced electric field at the LDPE nanocomposites sample surface of the positive and negative cycles during PD measurements [9][10].

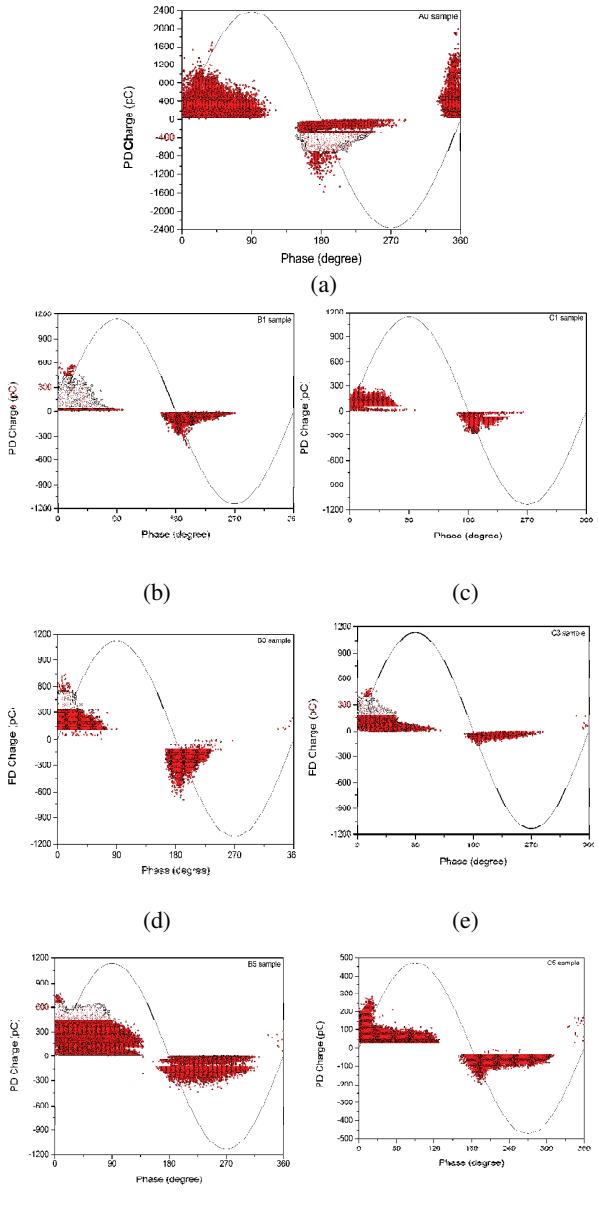


Fig. 5. PD patterns for untreated and plasma treated SiO_2 nanofiller of (a) A0 (b) B1 (c) C1 (d) B3 (e) C3 (f) B5 and (g) C5 samples under 4 kVrms of applied voltage level for one hour ageing time.

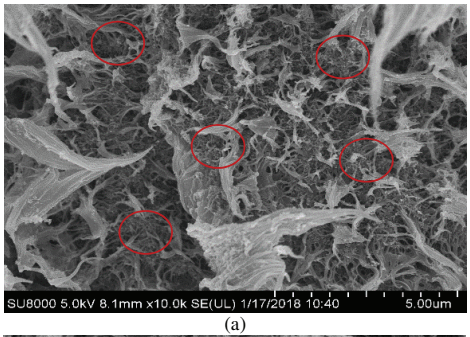
In general, the samples containing treated nanosilica show better PD resistance which resulting in lower PD magnitude compared to the A0 sample. For nanocomposites containing 1 wt%, 3 wt% and 5 wt% of untreated SiO2 nanofiller into LDPE (C1, C3 and C5 samples), the PD magnitude values are 600 pC, 750 pC and 800 pC. Despite this, the PD magnitude are lower than A0 sample. However, B5 sample depicts the higher PD magnitude among B1 and B3 samples; this is due to the higher filler concentration which contributes to the

space charge trapping [11] also the formation of agglomeration in LDPE nanocomposites sample. It can be seen in Fig. 5(c) and 5(g), the plasma treated nanosilica exhibited the lower PD magnitude rather than untreated nanosilica were 300 pC, 700 pC and 300 pC for C1, C3 and C5 samples; it may be caused by the enhanced the interfacial bonding between nanofiller and polymer matrices after plasma treatment of nanosilica. Nonetheless, for the C3 sample the PD magnitude was larger than C1 and C5 sample. It may be associated with the poor dispersion of the nanosilica particles and incompatible interfaces between nanoparticles and polymer matrices which eventually led to the larger PD magnitude.

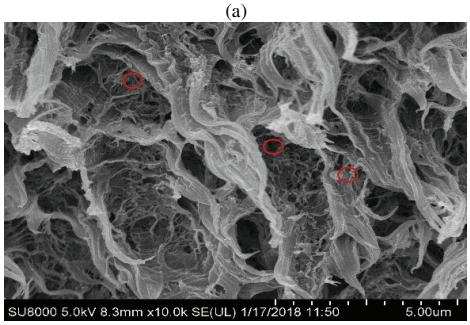
C. *Characterization of Nanoparticles*

The morphological analysis of Field Emission Scanning Emission Microscope (FESEM) was carried out to analyze the filler dispersion of SiO_2 nanoparticles inside the host polymer and the results were depicts in Fig. 6. Observation shows that there are many severe agglomerations were found in the B5 nanocomposite sample compared to the C5 sample as marked as the red circles.

Fig. 6(b) shows the sizes of agglomeration are relatively smaller than that in the nanocomposites with the untreated SiO_2 nanoparticles in Fig. 6(a) which can be the reason for the improvement in AC breakdown strength and PD suppression. In general, the dispersion was improved when the nanofiller was treated with the atmospheric pressure plasma.



(a)



(b)

Fig. 6. FESEM images of cross-section of (a) B5 and (b) C5 nanocomposites samples

IV. CONCLUSION

The partial discharge and AC breakdown strength characteristics of LDPE nanocomposites with different filler loading have been experimentally investigated. Outcomes from this study; the partial discharge resistance and AC breakdown strength have been improved when SiO_2 nanoparticles were treated with the atmospheric pressure plasma. Higher the nanosilica concentration, higher the AC breakdown strength for both untreated and plasma treated nanocomposites. In addition, C5 sample has the highest AC breakdown strength among the samples. In addition, the PRPD pattern of C5 sample has been plotted and discussed. It showed that lower PD magnitude as compared with B5 sample. The cross-sectional FESEM images clearly showed that the treated sample has better filler dispersion as compared to untreated sample. It is implied that the plasma treatment method is appropriate to be applied in producing nanocomposites as well as improving some of the electrical properties.

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REFERENCES

[1] F. N. Musa, N. Bashir, and M. H. Ahmad, "Electrical treeing performance of plasma-treated silicone rubber based nanocomposites," in *ICHVE 2016 - 2016 IEEE International Conference on High Voltage Engineering and Application*, 2016.

[2] Wei Yan, B. T. Phung, Zhaojun Han, and K. Ostrikov, "Reinforced insulation properties of epoxy resin/SiO_2 nanocomposites by atmospheric pressure plasma modification," in *2012 IEEE International Power Modulator and High Voltage Conference (IPMHVC)*, 2012, pp. 391–394.

[3] M. Musa, Y. Z. Arief, and M. H. Ahmad, "Influence of Nano-Titanium Dioxide (TiO_2) on Electrical Tree Characteristics in Silicone Rubber Based Nanocomposite," in *In Electrical Insulation and Dielectric Phenomena (CEIDP)*, 2013, pp. 498–501.

[4] M. Ahmad *et al.*, "A New Statistical Approach for Analysis of Tree Inception Voltage of Silicone Rubber and Epoxy Resin under AC Ramp Voltage," *Int. J. Electr. Eng. Informatics*, vol. 4, no. 1, pp. 27–39, 2012.

[5] F. Musa, N. Bashir, M. Ahmad, Z. Buntat, and M. Piah, "Investigating the Influence of Plasma-Treated SiO2 Nanofillers on the Electrical Treeing Performance of Silicone-Rubber," *Appl. Sci.*, vol. 6, no. 11, p. 348, 2016.

[6] W. Yan and Z. Han, "Silica Nanoparticles Treated by Cold Atmospheric-Pressure Plasmas Improve the Dielectric Performance of Organic–Inorganic Nanocomposites," *ACS Appl. Mater. Interfaces*, vol. 4, pp. 2637–2642, 2012.

[7] W. Yan, B. T. Phung, Z. J. Han, K. Ostrikov, and T. R. Blackburn, "Partial discharge characteristics of epoxy resin-based nanocomposites fabricated with atomospheric plasma treated SiO_2 nanoparticles," in *Proceedings of 2011 International Symposium on Electrical Insulating Materials*,2011, pp.353–357.

[8] W. Hauschild and E. Lemke, *High-Voltage Test and Measuring Techniques*.2014.

[9] M. Z. H. Makmud, H. A. Illias, and C. Y. Chee, "Partial Discharge Behaviour within Palm Oil-based Fe_2O_3 Nanofluids under AC Voltage," in *IOP Conference Series: Materials Science and Engineering*, 2017, vol. 210, no. 1, p. 12034.

[10] H. Illias, Teo Soon Yuan, A. H. A. Bakar, H. Mokhlis, G. Chen, and P. L. Lewin, "Partial discharge patterns in high voltage insulation," in *2012 IEEE International Conference on Power and Energy (PECon)*, 2012, no. May 2014, pp. 750–755.

[11] M. H. Ahmad *et al.*, "Temperature Effect on Electrical Treeing and Partial Discharge Characteristics of Silicone Rubber-Based Nanocomposites," *J. Nanomater.*, vol. 2015, pp. 1–13, 2015.