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Breakdown characteristics of polyethylene/silicon nitride nanocomposites

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Abstract

Silicon nitride (Si_3N_4) has been utilized as a nanofiller in polymeric insulation due to its good characteristics in both electrical insulation and thermal conduction properties. In this work, a comparative study was performed between unfilled polyethylene and polyethylene containing different amounts of Si_3N_4 nanofiller. The study showed that the low density polyethylene (LDPE) added with 15 wt% of Si_3N_4nanofiller could have higher breakdown strength compared to equivalent LDPE with 10 wt% of Si_3N_4nanofiller. Morphological characterizations of the nanocomposite samples were performed using field emission electron microscopy (FESEM) and the results showed that the breakdown performance of the investigated materials were affected by the agglomeration of Si_3N_4 nanoparticles.

Keywords*: aggromeration, breakdown strength, nanofiller, polyethylene*

1. Introduction

Polymer nanocomposites are defined as polymers in which a little amount of nanometer-sized fillers are homogeneously dispersed throughout the polymer matrix by only some weight percentage (wt%). Nanocomposites have attracted a great interest in both the industry and academia, since they frequently demonstrate significant improvements in material properties. The addition of a small weight percentage of nanofillers will give impact on the physical [1, 2], thermal [3, 4], chemical [5] and electrical properties [6-12] of the polymers. Generally, nanocomposites have three main phases, namely the matrix, the filler and the interaction zone; this concept has been introduced since 1994 in the seminal paper by Lewis [13]. Previous research on the breakdown strength of nanocomposites stated that the amount of nanofiller had to be less than 10 wt% in order for the material to achieve good breakdown properties. In this regard, many different results were reported by researchers on the performances of breakdown strength of nanocomposites containing nanofillers less than 10 wt% [14-22]. Nevertheless, the potential improvements in the breakdown strength of nanocomposites are not clear and the mechanisms underlying the breakdown behavior of nanocomposites are not fully understood.

Lately, the presence of water in nanocomposites have been regarded as one of the main factors jeopardizing the breakdown properties of nanocomposites. For oxide-based nanofillers, such as silica (SiO_2) nanofiller, it contains hydroxyl groups around its surface and is therefore prone to water adsorption [23]. This will negatively affect the breakdown performance since water may be present in the interaction zone between the nanofiller and the polymer. Therefore, the use of non-oxide based nanofillers, in the absence of the surface hydroxyl groups, has been proposed as a better alternative to silica nanofiller for use in nanocomposites for breakdown improvements. This paper discusses the breakdown performance of polyethylene (PE) nanocomposites containing non-oxide based, silicon nitride (Si_3N_4) nanofiller. The use of Si_3N_4 as an alternative inorganic filler will contribute to further understanding on the importance of water absorption effect since the surface of Si_3N_4 nanofiller does not have hydroxyl groups as opposed to SiO_2 nanofiller.

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2. Methodology

This section presents the sample preparation and the methods to perform breakdown strength test and morphological characterization.

2.1. Sample Preparation

Low density polyethylene (LDPE) was chosen as the polymeric matrix since PE has been generally used in manufacturing high voltage underground cables. Si_3N_4, with a manufacturer-quoted size of 15-30 nm, was chosen as the nanofiller due to its non-oxide surface properties. The nanocomposites were prepared using a mechanical blending method. In order to prepare the nanocomposites, the LDPE and silicon nitride nanofiller were first mixed together in a plastic bag. Then, they were melted at 140°C for ~25 min in the two-roll mill machine to obtain nanocomposite lumps. Test specimens for breakdown testing were subsequently prepared by melt pressing the nanocomposite lumps using a hydraulic laboratory press at 160°C temperature and 3 ton load. Each test specimen had a thickness of ~100 µm. Table 1 shows the composition of LDPE with Si_3N_4 nanofiller at loadings of 0 wt%, 5 wt%, 10 wt% and 15 wt%.

Table 1. The Composition of LDPE and Silicon Nitride Nanofiller

Sample	Composition
LDPE	LDPE + 0wt% of Si3N4
LDPE-5	LDPE + 5 wt% of Si3N4
LDPE-10	LDPE + 10 wt% of Si3N4
LDPE-15	LDPE + 15 wt% of Si3N4

2.2. Breakdown Testing

The breakdown strength test was conducted by injecting the step voltage with 2 kV voltage for every 20 s for DC breakdown, and 1 kV voltage for every 20 s for AC breakdown. A digital multimeter was used to display the breakdown voltage after the specimen experienced breakdown. Then, the breakdown voltage readings were recorded and analyzed by using the two-parameter Weibull distribution analysis. Fifteen breakdown values were collected for each specimen type. The breakdown strength value was determined based on (1). The probability of dielectric breakdown strength is determined by using Weibull distribution analysis Bernard's approximation as in (2).

$$\text{BreakdownStrength}\left(\frac{\text{kV}}{\text{mm}}\right) = \frac{\text{BreakdownVoltage(kV)}}{\text{SpecimenThickness (mm)}} \quad (1)$$

$$F(\text{i},\text{N}) = \frac{\text{i}-0.3}{\text{N}+0.4} \quad (2)$$

In (2), *i* represents the rank of dielectric breakdown strength, E_{bd} of specimen from low to high, while *N* is the assigned number of measured values from the breakdown tests. The probability of breakdown strength was estimated by using the cumulative distribution function (CDF) shown in equation (3). The probability of dielectric breakdown strength of the specimen is represented as $F(E_{bd}; \alpha, \beta)$ while α is the scale parameter and β is the shape parameter of the Weibull graph.

$$\text{F}(\text{E}_{\text{bd}};\alpha,\beta) = 1 - \exp\left[-\left(\frac{\text{E}_{\text{bd}}}{\alpha}\right)^{\beta}\right] \quad (3)$$

2.3. Morphological Characterization

The LDPE, LDPE-5 and LDPE-15 samples were characterized using field emission scanning electron microscope (FESEM). This allowed the dispersion state of Si_3N_4 nanoparticles in the polymeric matrix to be determined. Prior to FESEM characterization, each sample was coated with platinum using an automated platinum sputter coater. The FESEM machine was set at high magnifications with a spectrum voltage of 10 kV.

3. Results and Analysis

This section discusses on breakdown and morphological characterization results.

3.1. Breakdown Results

The breakdown results are divided into two sections. Section 1 will discuss on the DC breakdown results and the comparison with the previous studies. Section 2 will discuss on the AC breakdown strength test results and the comparison of results obtained with the previous studies.

3.1.1. DC breakdowns results

The Weibull plots of DC breakdown strength are shown in Figure 1 while the Weibull parameters are shown in Table 2. The scale parameter α represents the breakdown strength while the shape parameter β represents the width of the Weibull distribution. Based on Figure 1, the DC breakdown strength for polyethylene nanocomposites of LDPE-5 sample was the closest to pure polyethylene. The lowest breakdown strength result was LDPE-10 sample with a scale parameter of 117 kV/mm. The breakdown result of LDPE-15 sample showed an improvement of 12% with a scale parameter of 131 kV/mm compared to the LDPE-10 sample.

3.1.2. AC Breakdowns Strength Test

The Weibull distribution analysis was carried out and the Weibull plots for AC breakdown strength are shown in Figure 2. The Weibull parameters are shown in Table 3. From Figure 2 and Table 3, the highest AC breakdown results was possessed by LDPE with the shape parameter β=10.04. The breakdown results for the LDPE-5 sample is slightly lower, which was 4.2% lowered than LDPE, with scale parameter 136 kV/mm, followed by the LDPE-10 sample and the LDPE-15 sample, with 106 kV/mm and 90 kV/mm respectively.

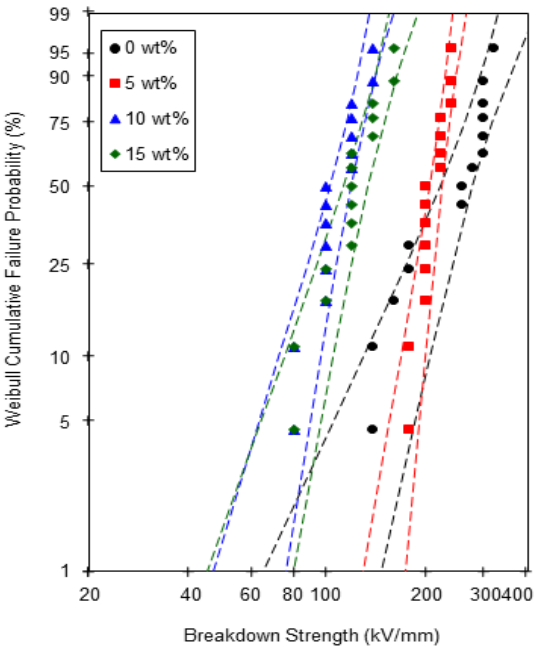

Figure 1. Weibull distribution analysis of DC breakdown strength

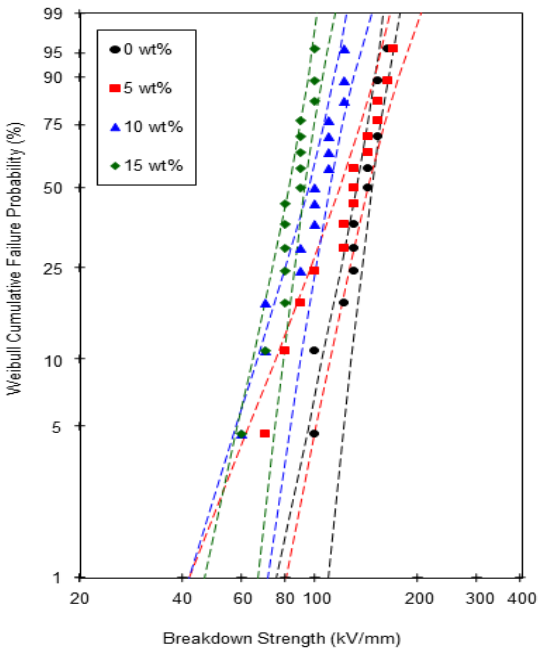

Figure 2. Weibull distribution analysis of AC breakdown strength test

Table 2. Weibull Distribution Parameters

Sample	α (kV/mm)	β
0 wt%	266 ± 27	5 ± 2
5 wt%	220 ± 9	12 ± 5
10 wt%	117 ± 8	7 ± 2
15 wt%	131 ± 10	6 ± 2

Table 3. Weibull Distribution Parameters

Sample	α (kV/mm)	β
0 wt%	142 ± 6	10 ± 4
5 wt%	136 ± 12	5 ± 3
10 wt%	106 ± 7	7 ± 3
15 wt%	90 ± 4	10 ± 3

3.2. Comparison of AC and DC Breakdown Strength

Figure 3 shows the difference between AC and DC breakdown results. Clearly, these nanocomposites samples behaved differently under AC and DC applied fields. It can be noticed that the detrimental effect of Si_3N_4 on LDPE is less significant in AC than in DC. These DC and AC breakdown trends are not unusual and have also been reported elsewhere for SiO_2-filled LDPE [24, 25]. With increasing amounts of Si_3N_4, the AC breakdown strength of the nanocomposites reduced accordingly. Meanwhile, the DC breakdown strength for nanocomposites with 15 wt% of Si_3N_4 increased slightly compared to those with 10 wt% of Si_3N_4. In this case, the addition of a high amount of Si_3N_4 resulted in better breakdown performance, albeit that the value was not comparable to that of the unfilled LDPE sample.

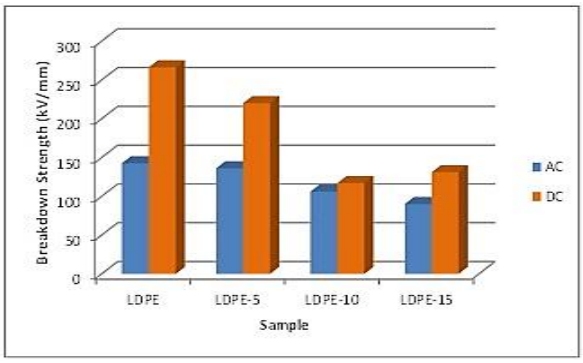

Figure 3. The comparison results AC and DC breakdown strength test

3.3. Morphological Characterization of LDPE Containing Nanofiller

Morphological characterization of the LDPE/Si_3N_4 nanocomposite samples was carried out to investigate the dispersion of Si_3N_4 nanoparticles in the LDPE matrix. The FESEM micrographs for the unfilled LDPE, LDPE-5 and LDPE-15 samples are shown in Figure 4, Figure 5 and Figure 6 respectively.Figure 4 is the micrograph taken by FESEM for the reference, unfilled LDPE sample. There were no nanoparticles present in the LDPE matrix. Meanwhile, the images for the LDPE-5 and LDPE-15 samples shown in Figure 5 and Figure 6, respectively, showed the presence of nanoparticles in the LDPE matrix; these images display the agglomerated state of nanoparticles. Of note, more agglomerations were found in the LDPE-15 sample. While it was expected that increasing nanofiller amounts would lead to reduced breakdown strength, this did not happen for the case of LDPE-15 sample in comparison with the LDPE-10 sample. Nevertheless, an alternative method to fabricate the samples could be explored to improve the dispersion the nanoparticles across the samples in an attempt to improve the breakdown performance.

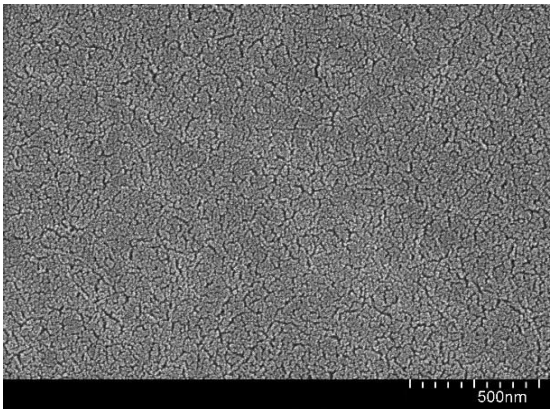

Figure 4. FESEM micrograph for the unfilled LDPE sample/

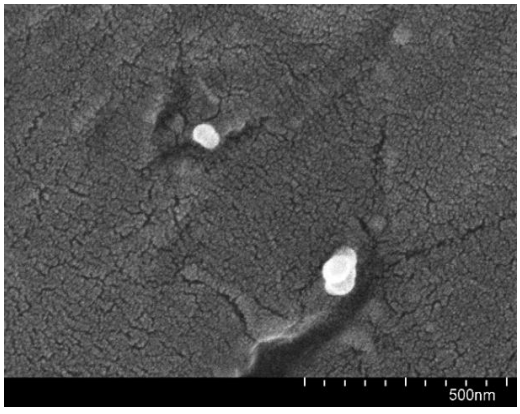

Figure 5. FESEM micrographs for the LDPE-5 sample

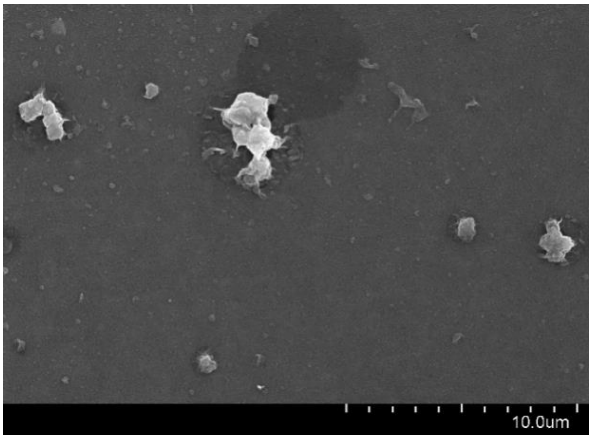

Figure 6. FESEM micrograph for the LDPE-15 sample

4. Conclusion

This study was conducted to investigate the breakdown strength of unfilled polyethylene and polyethylene nanocomposites containing high amounts of silicon nitride nanofillers. It can be concluded that the breakdown strength of polyethylene nanocomposites under AC and DC were lower compared to unfilled polyethylene. However, in comparing the breakdown performance of LDPE-15 and LDPE-10 under the DC field, LDPE-15 sample showed 12% improvement in breakdown strength compared to the LDPE-10 sample. Therefore, the results from this study showed that the presence of a high amount of silicon nitride nanofillers in LDPE did not necessarily result in reduced breakdown strength, but better breakdown performance could be achieved.

References

[1] M Ghasemzadeh-Barvarz, C Duchesne, D Rodrigue. Mechanical, Water Absorption, and Aging Properties of Polypropylene/Flax/Glass Fiber Hybrid Composites. *J. Compos. Mater.* 2015; 49(30): 3781–3798.

[2] CP Sugumaran, I Member. *Experimental Study on Dielectric and Mechanical Properties of PVC Cable Insulation with SiO 2 / CaCO 3 Nanofillers.* in 2015 Annual Report Conference on Electrical Insulation and Dielectric Phenomena. 2015; 503–506.

[3] W Cheewawuttipong, D Fuoka, S Tanoue, H Uematsu, Y Iemoto. Thermal and Mechanical Properties of Polypropylene/Boron Nitride Composites. *Energy Procedia*. 2013; 34: 808–817.

[4] CW Ping Zhu, Jing Chen. Crystallization Behavior and Mechanical Properties of Polypropylene/Modified Carbon Black Composites. *Polym. Compos.* 2014; 116(1): 37–43.

[5] C Xiaohong, W Weiwang, L Shengtao, F Michel, Z Xiaohong. Polymer Nanocomposites in High-Voltage Insulation. *IEEE Nanotechnol. Mag.* 2018; 33–40.

[6] Q Guo, Y Chen, J Zhang, Z Yao. Large impact in electrical properties of polypropylene by improving the filler-matrix interface effect in PP/PS blends. *Polym. Test*. 2017; 63: 587–595.

[7] BX Du, H Xu, J Li, Z Li. Space Charge Behaviors of PP / POE / ZnO Nanocomposites for HVDC Cables. *IEEE Trans. Dielectr. Electr. Insul.* 2016; 23(5): 3165–3174.

[8] AN Ramani, AM Ariffin, G Vijian, ABA Ghani. The Effects of Nano Fillers on Space Charge Distribution in Cross-Linked Polyethylene. *Int. J. Electr. Comput. Eng.* 2017; 7(6): 3147–3152.

[9] JK Nelson, Y Hu, J Thiticharoenpong. *Electrical Properties of TiO2 Nanocomposites.* 2003 Annu. Rep. Conf. Electr. Insul. Dielectr. Phenom. 2003; 719–722.

[10] K Lau, A Vaughan, G Chen, I Hosier, A Holt, K Ching. On the Space Charge and DC Breakdown Behavior of Polyethylene/Silica Nanocomposites. *IEEE Trans. Dielectr. Electr. Insul.* 2014; 21(1): 340–351.

[11] Y Zhou, J Hu, B Dang, J He. Effect of Different Nanoparticles on Tuning Electrical Properties of Polypropylene Nanocomposites. *IEEE Trans. Dielectr. Electr. Insul.* 2017; 24(3): 1380–1389.

[12] NAM. Jamail, MAM Piah, NA. Muhammad, QE Kamarudin. PDC Analysis of LLDPE-NR Nanocomposite for Effect of Moisture Absorption. *Int. J. Electr. Comput. Eng.*2017; 7(6):. 3133–3139.
[13] TJ Lewis. Nanometric Dielectrics. *J. Phys. D. Appl. Phys*. 2005; 38(2): 202–212.
[14] T Tanaka. Polymer Nanocomposites as Dielectrics and Electrical Insulation-perspectives for Processing Technologies, Material Characterization and Future Applications. *IEEE Trans. Dielectr. Electr. Insul.* 2004; 11(5): 763–784.
[15] T Imai, F Sawa, T Ozaki, Y Inoue, T Shimizu, T Tanaka. Comparison of Insulation Breakdown Properties of Epoxy Nanocomposites Under Homogeneous and Divergent Electric Fields. in *Electrical Insulation and Dielectric Phenomena (CEIDP).* 2006; 306–309.
[16] P Preetha, MJ Thomas. AC Breakdown Characteristics of Epoxy Nanocomposites. *IEEE Trans. Dielectr. Electr. Insul*. 2011; 18(5): 1526–1534.
[17] GC Montanari, D Fabiani, F Palmieri, D Kaempfer, R Thomann, R Mülhaupt. Modification of Electrical Properties and Performance of EVA and PP insulation through Nanostructure by Organophilic Silicates. *IEEE Trans. Dielectr. Electr. Insul.* 2004; 11(5): 754–762.
[18] C Sunada, MN Dinesh, N Vasudev. *Performance Evalution of Silicon Rubber Insulating Material with MgO And ZnO Nanofillers.* in IEEE International Conference on High Voltage Engineering and Application (ICHVE). 2016; 1–5.
[19] S Virtanen, H Ranta, S Ahonen, M Karttunen, J Pelto, K Kannus, M Pettersson. Structure and dielectric breakdown strength of nano calcium carbonate/polypropylene composites. *J. Appl. Polym. Sci.* 2014; 131(1): 39504–39504.
[20] W Lei, K Wu, Y Wang, Y Cheng, X Zheng, LA Dlssado, SJ Dodd, NM Chalashkanov, JC Fothergill, C Zhang, W Li. Are nano-composites really better DC insulators? A study using silica nanoparticles in XLPE. *IEEE Trans. Dielectr. Electr. Insul.* 2017; 24(4): 2268–2270.
[21] KY Lau. Structure and Electrical Properties of Silica-Based Polyethylene Nanocomposites. University of Southampton, 2013.
[22] SNH Kamarudin, KY Lau, WAW A Rahman. *The Effect of Non-Isothermal Crystallization on the AC Breakdown Performance of Polyethylene/Silicon Dioxide Nanocomposites*. in IEEE Conference on Energy Conversion (CENCON). 2017; 7–10.
[23] KY Lau, AS Vaughan, G Chen, IL Hosier, AF Holt. On the Dielectric Response of Silica-Based Polyethylene Nanocomposites. *J. Phys. D. Appl. Phys.* 2013; 46(9).
[24] KFTA Bakar, N Eusoff, KY Lau, MAM Piah, KY Ching. *DC and AC Breakdown Behaviors of Polyethylene/Silica Nanocomposites*. in International Conference on Power and Energy (PECON). 2017; 591–594.
[25] KY Lau, MAM Piah, KY Ching. Correlating the Breakdown Strength with Electric Field Analysis for Polyethylene/Silica Nanocomposites. *J. Electrostat.* 2017; 86: 1–11.