AC Breakdown Performance of Non-isothermally Crystallized Polyethylene/Silicon Nitride Nanocomposites

S.N.H. Kamarudin[1,*], K.Y. Lau[1,2], N.H.Rahim[1], C.W. Tan[2], W.A.W.A. Rahman[3]

[1]Institute of High Voltage and High Current, School of Electrical Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Malaysia

[2]Department of Electrical Power Engineering, School of Electrical Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Malaysia

[3]Biopolymer Research Group, School of Chemical Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Malaysia

*E-mail: sitinoorhazirah.kamarudin@yahoo.com

Abstract— **Polymer nanocomposites has a huge potential to serve as dielectric materials. These materials were found to have a great capability in withstanding high voltage levels. In nanocomposites development, the breakdown strength of the materials was often reported to be higher, lower or similar, in comparison with the unfilled polymer. Because of this, the breakdown performance of nanocomposites is said to be dependent not only on the polymer/nanofiller combination, but also on the sample preparation techniques. Unfortunately, factors leading to increased or decreased the breakdown strength of nanocomposites are less understood. In this paper, investigation into polyethylene blend systems that contain different amounts of silicon nitrite nanofiller with different non-isothermal crystallization processes was reported. The non-isothermal crystallization techniques were determined by fast, medium and slow cooling rate conditions while preparing the samples. Differential scanning calorimetry (DSC) was used to characterize the thermal behavior of the polymer nanocomposites. The AC breakdown data showed that the use of different non-isothermal crystallization techniques affected the AC breakdown strength of each material type.**

Keywords—polymer nanocomposites, polyethylene, silicon nitride, AC breakdown, crystallization

I. INTRODUCTION

The tremendous increase in the use of polymers and polymer-based materials can be seen in various applications. To address future needs of insulation materials, dielectric research has been widely carried out, focusing on improving the dielectric properties of the materials. Due to this situation, polymer nanocomposites have become popular among dielectric researchers.

Polymer nanocomposites are defined as composites with a small amount of nanometer-sized filler dispersed homogeneously in the polymer. According to previous research [1], [2], the study reported that these materials can markedly improve properties, such as mechanical strength, barrier performance, solvent and heat resistance, and optical transparency. In comparison with conventional composites,or known as microcomposite these improvements are achieved with low amounts of nanofillers added to the polymer, normally less than 10 wt%. The use of the nanometer-sized particles, when combined with polymers, have been reported to be capable of enhancing the dielectric behaviors of the materials [3]–[7].

To date, dielectrics researchers have reported that polymer nanocomposites have resulted in many positive impacts and are promising as future dielectric materials. Among many polymers, polyethylene is one of the most widely used polymers in electrical applications. Furthermore, polyethylene blend with the composition of LDPE and HDPE in the ratio 80:20 have been shown to provide promising dielectric results [8], [9]. In this paper, polyethylene blend with different amounts (0 wt%, 1 wt%, and 5 wt%) of silicon nitrate (Si_3N_4) nanofiller was prepared for the investigation. The AC breakdown properties of the nanocomposites was tested by considering the effect of different non-isothermal crystallization techniques imposed on the samples.

II. EXPERIMENT

A. Material Preparation

A polyethylene blend with LDPE and HDPE in the ratio of 80:20 was used in this research. The low density polyethylene (LDPE) grade TITANLENE LDF200YZ and high density polyethylene (HDPE) grade TITANZEX HI2000 from Lotte Chemical Titan were used. The nanofiller used was silicon nitride (Si_3N_4) nanopowder with an average particle size of 15-30 nm (obtained from Nanostructured & Amorphous Materials, Inc). To prepare the polymer nanocomposites, the Brabender melt mixer was used for melt blending the polyethylene (PE), with 1 wt% and 5 wt% of Si_3N_4 for 10 minutes with a rotational speed of 45 rpm and a temperature of 160 °C. Then, the disc-shaped samples were prepared using a hydraulic laboratory hot press at a temperature of ~160 °C and a load of 2.5 ton. The melt pressed samples were left to cool using three different non-isothermal crystallization techniques of fast, medium and slow cooling conditions, as detailed in Table I. For convenience, the investigated materials were denoted as in Table II.

TABLE I. NON-ISOTHERMAL CRYSTALLIZATION TECHNIQUES

Non-isothermal crystallization technique	Rate of cooling (^{0}C/min)
Fast cooling	~25
Medium cooling	~10
Slow cooling	~1

TABLE II. INVESTIGATED MATERIALS

Designation	Description
PE/0/F	Unfilled polyethylene crystallized under the fast cooling condition
PE/0/M	Unfilled polyethylene crystallized under the medium cooling condition
PE/0/S	Unfilled polyethylene crystallized under the slow cooling condition
PE/Si$_3$N$_4$-1/F	Polyethylene with 1wt% Si$_3$N$_4$ crystallized under the fast cooling condition
PE/Si$_3$N$_4$-1/M	Polyethylene with 1wt% Si$_3$N$_4$ crystallized under the medium cooling condition
PE/Si$_3$N$_4$-1/S	Polyethylene with 1wt% Si$_3$N$_4$crystallized under the slow cooling condition
PE/Si$_3$N$_4$-5/F	Polyethylene with 5wt% Si$_3$N$_4$crystallized under the fast cooling condition
PE/Si$_3$N$_4$-5/M	Polyethylene with 5wt% Si$_3$N$_4$ crystallized under the medium cooling condition
PE/Si$_3$N$_4$-5/S	Polyethylene with 5wt% Si$_3$N$_4$ crystallized under the slow cooling condition

B. Characterization

The Perkin Elmer differential scanning calorimetry (DSC7) with Perkin Elmer Pyris software was used to characterize the thermal behavior of the investigated samples. For each sample, the weight of the samples was about 5 mg, placed in a sealed aluminum pan. The DSC was performed under nitrogen atmosphere at a temperature range of 30°C to 160°C and a scan rate of 10°C/min.

AC breakdown testing procedures were conducted by placing the sample between two 6.3 mm diameter spherical electrodes immersed in mineral oil. An alternating current (AC) voltage with a ramp rate voltage of 1 kV for every 20 s was applied until the breakdown occurred. 15 breakdown points were measured on each type of samples with the thickness of ~100μm for each sample.

III. RESULTS AND DISCUSSION

A. Thermal Analysis

Fig. 1, Fig. 2 and Fig. 3 show the melting behaviors of unfilled polyethylene and nanocomposites containing 1wt% of Si$_3$N$_4$ and 5wt% of Si$_3$N$_4$ nanofillers when subjected to different non-isothermal crystallization processes, respectively. Two main peaks were observed, the lower peak indicates the thermal transitions of LDPE and HDPE as the upper peaks. Commonly, the peak temperature of LDPE was ~107 C while the peak temperature of HDPE was higher, ~128 C. By comparing the trends of DSC traces in Fig. 1, Fig. 2 and Fig. 3, for either fast, medium or slow cooling conditions, the results show similar trends for unfilled polyethylene, nanocomposites with 1 wt% of silicon nitride and nanocomposites with 5 wt% of silicon nitride, when crystallized under the respective cooling conditions. It indicated that there were no thermal changes caused by the

inclusion of silicon nitride [3]. Meanwhile, the changes for the melting traces for each material type was affected by different non-isothermal crystallization conditions imposed on the samples.
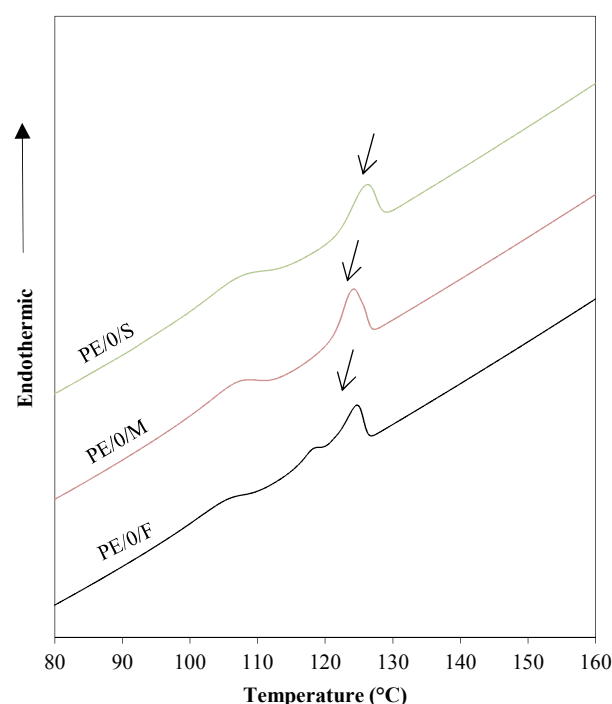
UNFILLED PE

Fig. 1. DSC melting behavior of unfilled polyethylene when subjected to different non-isothermal crystallization processes
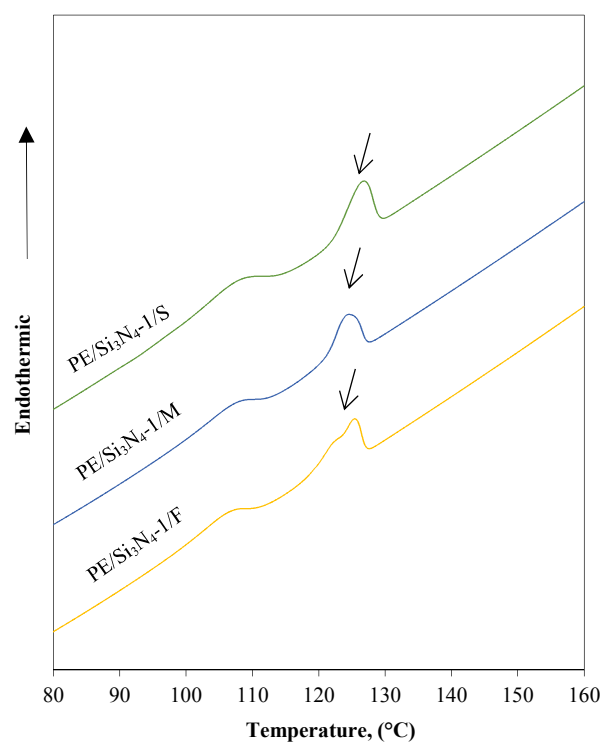
PE with 1 wt% Si$_3$N$_4$

Fig. 2. DSC melting behavior of nanocomposites with 1 wt% of SiO$_2$ when subjected to different non-isothermal crystallization processes
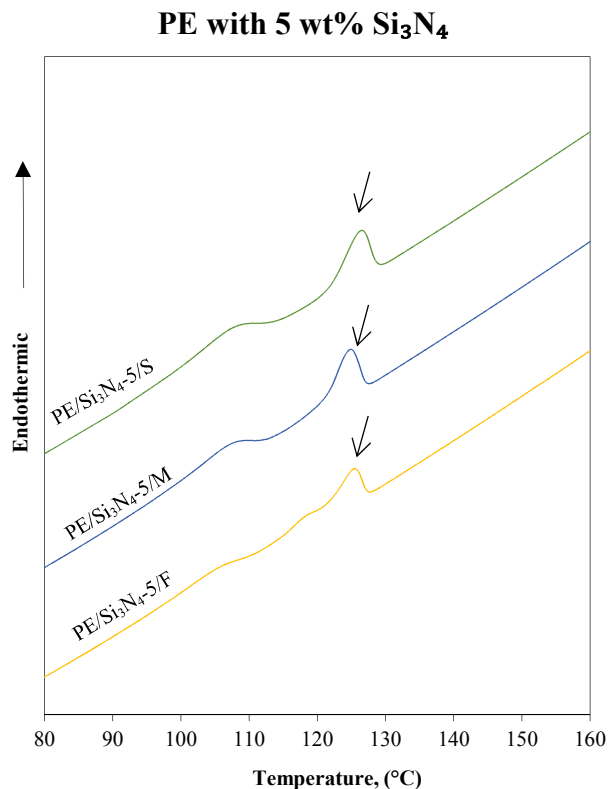
Fig. 3. DSC melting behavior of nanocomposites with 5 wt% of SiO_2 when subjected to different non-isothermal crystallization processes

B. Breakdown Strength Analysis

Fig. 4 shows the Weibull plot of AC breakdown strength of the unfilled polyethylene with different types of cooling conditions; the breakdown parameters are summarized in Table III. The highest AC breakdown strength of 162 kV/mm was recorded for the unfilled polyethylene non-isothermally crystallized under the medium cooling condition (PE/0/M).

The results from Fig. 5 show that PE/Si_3N_4-1/M also had the highest value of AC breakdown strength. Meanwhile, the AC breakdown strength for PE/Si_3N_4-1/F was comparable with PE/Si_3N_4-1/S. To confirm the AC breakdown behavior of polyethylene blends with the addition of Si_3N_4, 5 wt% of Si_3N_4 was used with respect to different non-isothermal crystallization conditions. From Fig. 6, the AC breakdown strength for PE/Si_3N_4-5/M is the highest in comparison with PE/Si_3N_4-5/F and PE/Si_3N_4-5/S.

From the Weibull plots, it seems that the samples non-isothermally crystallized under the medium cooling condition had the highest breakdown strength compared to other cooling conditions. Coincidently, the observation of the current breakdown trend based on different non-isothermal crystallization conditions is in line with the authors' previously published results based upon polyethylene/silica nanocomposites [10].
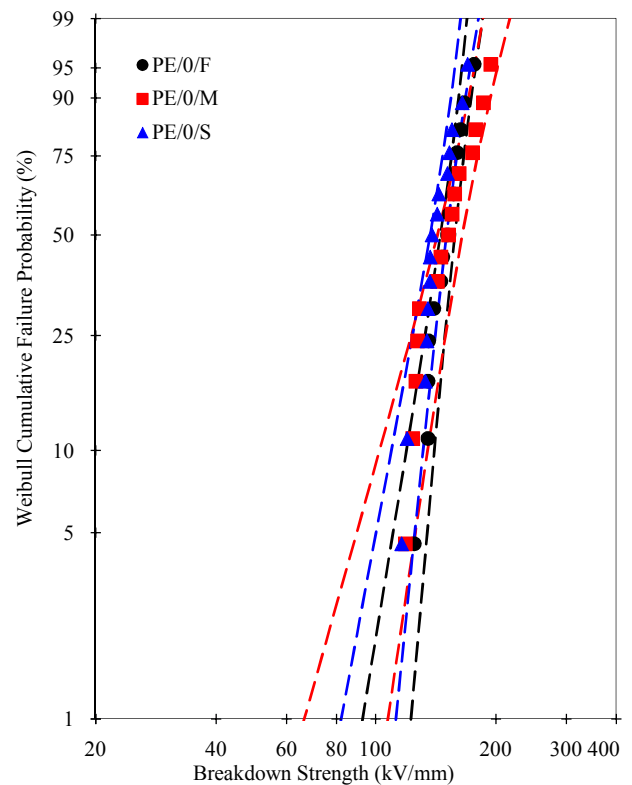

Fig. 4. Weibull plots of unfilled polyethylene when subjected to different non-isothermal crystallization conditions
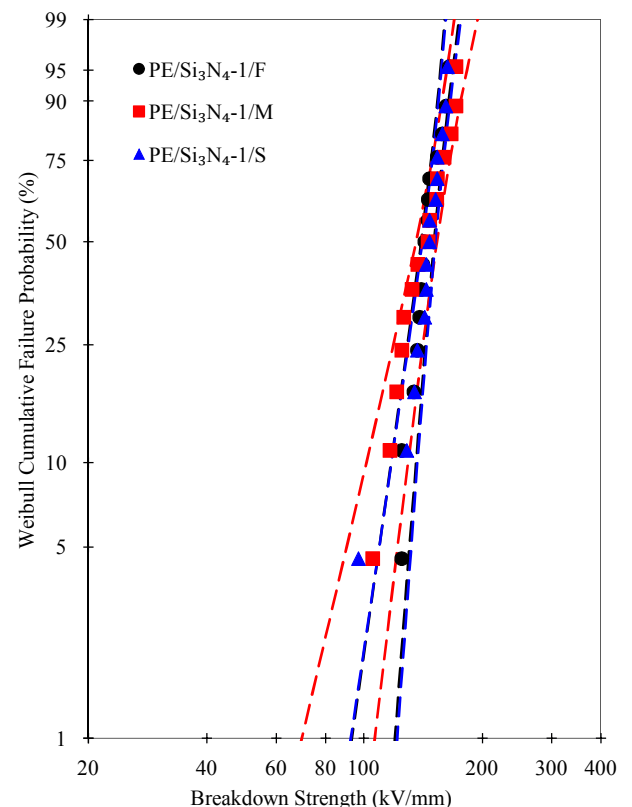

Fig. 5. Weibull plots of nanocomposites with 1 wt% of Si_3N_4 when subjected to different non-isothermal crystallization conditions.
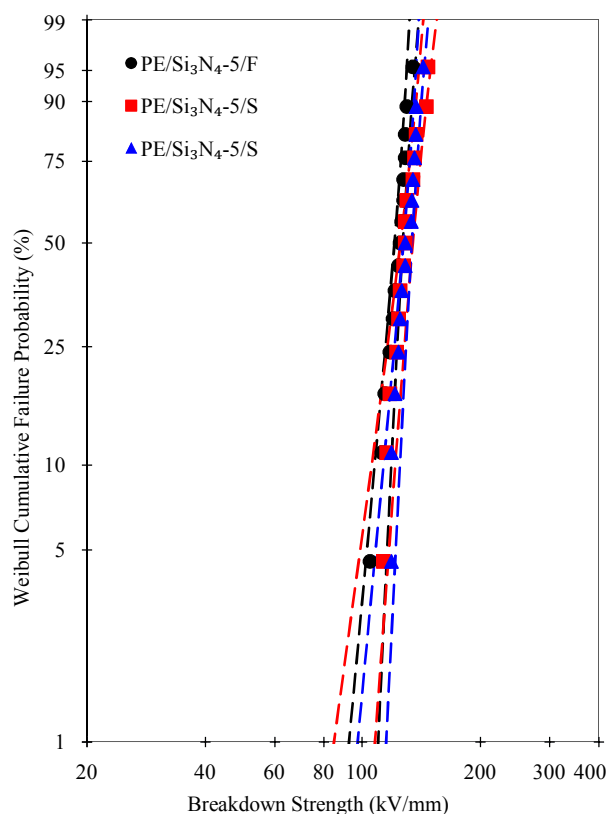
Fig. 6. Weibull plots of nanocomposites with 5 wt% of Si_3N_4 when subjected to different non-isothermal crystallization conditions

TABLE III.   Weibull Parameters

Material	Scale Parameter α (kV/mm)	Shape parameter, β
PE/0/F	156 ± 6	12 ± 5
PE/0/M	162 ± 10	7 ± 3
PE/0/S	149 ± 6	11 ± 3
PE/Si₃N₄-1/F	150 ± 5	13 ± 5
PE/Si₃N₄-1/M	151 ± 8	8 ± 4
PE/Si₃N₄-1/S	151 ± 5	13 ± 6
PE/Si₃N₄-5/F	126 ± 3	21 ± 8
PE/Si₃N₄-5/M	133 ± 5	14 ± 5
PE/Si₃N₄-5/S	133 ± 3	20 ± 8

IV. CONCLUSIONS

The use of different non-isothermal crystallization techniques (slow, medium and fast cooling conditions) in preparing the unfilled polyethylene and nanocomposite samples had subsequent effect on the breakdown strength of the materials. All the samples, when subjected to medium cooling condition, resulted in the highest breakdown values compared to fast and slow cooling conditions. Moreover, different non-isothermal crystallization techniques also affects the thermal traces of the samples. Therefore, the variations in the DSC melting behaviors of the materials are believed to contribute to the changes in the AC breakdown strength of the materials under different non-isothermal crystallization conditions.

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