Correlating the breakdown strength with electric field analysis for polyethylene/silica nanocomposites

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Abstract

The experimental findings on the DC and AC breakdown strength of polyethylene/silica nanocomposites are reported and correlated with simulation results on the electric field distribution of possible nanocomposite models. Specifically, the effects of interphase permittivity and interparticle distances on the electric field intensity and the breakdown strength are discussed with the aid of the Finite Element Method Magnetics (FEMM) 4.2 software. The results showed that the presence of the interphase, when assigned a unique interphase permittivity value, led to variations in electric field distributions. The electric field also changed as adjacent nanoparticles separated from each other with different interparticle distances.

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1. Introduction

Polymer nanocomposites concern polymers with nanometer-sized fillers homogeneously dispersed at just a few weight percentage (wt%). In the dielectrics community, the term “nanometric dielectrics” [1], “nanodielectrics” [2-3], or “dielectric polymer nanocomposites” [4] has been introduced to refer to nanocomposites of specific interest in connection with their dielectric characteristics. These materials have been envisioned to be the future of high voltage electrical insulation, and are capable of enhancing the dielectric performance of conventional insulation systems which commonly suffer from reduced breakdown strength [5-7]. This is attributed to the presence of the much smaller size of the filler (nanometer-sized filler), which subsequently leads to the presence of an extensive interphase—an interaction zone between the nanofiller and the polymer [8-12].

For anticipating the presence of the interphase in nanodielectrics, two prominent interphase models have been proposed by Lewis [8] and Tanaka et al. [9] in the early 2000s. Based on Lewis’ diffuse electrical double layer model [1,8,10], a Stern layer and a Gouy-Chapman diffuse layer were assumed to exist at the interface between the filler and the polymer. The formation of these layers was thought to change the internal charge activity and the resulting electrical potential distribution at the interphase, which could then influence the dielectric properties of the resulting nanodielectrics. The multi-core model proposed by Tanaka et al. [9,11], on the other hand, assumed the presence of four layers, i.e., the bonded layer, the bound layer, the loose layer, and the Gouy-Chapman diffuse layer, within the interphase. Through the multi-core model, the interphase of a nanoparticle was permitted to overlap with the interphase of its neighboring nanoparticle, thus causing a far-field effect which results in a cooperative effect between the nanoparticles.

In addition, many other interphase models have been proposed in an attempt to explain the phenomena at the interphase that influence the dielectric behaviors of nanocomposites. For example, the multi-region structure model, the dual layer model, and the polymer chain alignment model were suggested by Li et al. [12], Singha and Thomas [13], and Andritsch [14], respectively, by assuming that nanoparticle/polymer interactions led to the formation of two interphase layers around the nanoparticle. The presence of a single interphase layer was also proposed by Raetzke et al. [15] and Daily et al. [16] in an attempt to clarify the electrical properties of nanocomposites based on the distinct features at the interphase.

Although various nanocomposite systems have been
investigated and positive experimental results on the breakdown strength of nanocomposites have been reported [17–20], there are many fundamental challenges yet to be addressed for the use of nanocomposites as electrical insulating materials [21,22]. For example, the experimental work of Huang et al. [23] revealed that the breakdown performance of polyethylene/aluminum nanocomposites was not as good as the base polymer, even though surface-treated nanoparticles were introduced. Also, the electrical breakdown performance of polymers was found to be adversely affected by the presence of large aggregates of nanofillers [24]. Although chemical means such as silane coupling agents and compatibilizers can be used to improve the interfacial adhesion between the nanofiller and the polymer [23,25], the use of different silane coupling agents may result in entirely different dielectric behaviors, a consequence which is closely associated with processes at the interphase. This shows that the incorporation of nanofillers does not always improve the breakdown strength and that there might be subtle unidentified factors that jeopardize the breakdown performance due to nanoinclusions.

Nevertheless, breakdown performances of polymer nanocomposites have been reported to be much better than their equivalent microcomposites [12,17]. In this regard, the size of the filler — or the presence of the interphase — was found to critically influence the breakdown strength, where microcomposites tended to fail at far lower applied fields as compared with nanocomposites or unfilled polymers [26]. Consequently, the relative improvement of electrical breakdown properties of nanocomposites could be resulted from a more uniform electric field distribution within thin films [8].

According to Wang et al. [27], finite element simulations can provide a statistical view of the local electric field distribution within nanocomposites. When high dielectric constant fillers are used, the polymer matrix may sustain most of the electrical stress — the locally enhanced field can be linked to the electrical breakdown strength for a short term breakdown process. Significantly, the overall permittivity of a composite material is often related to the compatibility of its constituents, hence electrical performance of the composite [28]. In oil-pressboard insulation, for example, permittivity mismatches between the two materials usually assists flashover phenomenon at the interface [29]. This is due to the non-uniform electric field distribution results from the permittivity mismatches. This raises the electrical stress and may act as a critical flaw in dielectric breakdown [30]. By reducing the mismatch in dielectric constant at the matrix-filler interface, the buildup of field concentrations responsible for the premature breakdown events could be mitigated.

According to conventional mixing rules, the addition of fillers into polymers should result in an overall permittivity according to the constituents (fillers and polymers) with a value between the constituents [31–33]. Nevertheless, the presence of interphases in nanocomposites has resulted in a seemingly new concept of permittivity that defies conventional mixing rules. According to Frechet et al. [21] and Andritsch et al. [34], the permittivity of nanocomposites can increase or decrease remarkably, out of the permittivity range of the constituents, depending on the nanofiller/polymer combinations and processing methods. Considering the effect of the interphase, the mixing rules may be generalized to have an additional term representing the interphase [33–37]. Consequently, if the volume fraction and the permittivity of the interphase dominate, the overall permittivity will reflect the permittivity of the interphase rather than a simple binary combination of permittivity of the filler and the polymer, as in conventional microcomposites. This observation is not unusual and has been anticipated by Lau et al. [38], where water molecules may reside within the interphase of a nanocomposite, resulting in higher overall permittivity of the nanocomposite due to the presence of more polar dipoles (i.e. hydroxyl groups) from the water that can readily respond to the applied field.

Since the breakdown characteristics of an insulating material depends on its resistance against voltage or electric field [39], the analysis of the electric field intensity within the insulating material is important to understand the material’s breakdown performance [40]. In this paper, we seek to correlate the experimental findings on the breakdown strength of polyethylene/silica nanocomposites with the simulation results on the electric field distribution of our proposed nanocomposite models. Specifically, the effects of interphase permittivity and interparticle distances on the electric field intensity and the possible breakdown strength are discussed with the aid of the Finite Element Method Magnetics (FEMM) 4.2 software.

2. Experimental

2.1. Materials and sample preparation

The polymers used in the experimental work were the low density polyethylene (LDPE) grade Titanlene LDF200Y2Z and the high density polyethylene (HDPE) grade Titanx H2000, obtained from Lotte Chemical Titan. The nanofiller used was silicon dioxide ($\text{SiO}_2$) nanopowder (nanosilica) obtained from NanoAmor, with an average particle size of 80 nm.

Nanocomposite samples were prepared using a mechanical mixing method. The desired amount of nanosilica (1 wt%, 3 wt%, and 5 wt%) was mixed with polyethylene (LDPE or HDPE) in a laboratory two-roll mill at a temperature of ~140 °C for ~30 min. Samples for breakdown measurements were then prepared using a hydraulic press at a temperature of ~160 °C. The thickness of the prepared samples was ~100 μm.

2.2. Characterization

Breakdown testing was conducted by placing a test sample between two opposing 6.35 mm diameter steel ball-bearing electrodes immersed in Hyrax Hypertrans transformer oil to prevent surface flashover. A DC voltage with a step voltage of 2 kV every 20 s or an AC voltage with a step voltage of 1 kV every 20 s was applied until the sample failed. Six breakdown tests were performed on each type of material. The voltage obtained from each measurement was divided by the specimen thickness at the breakdown point in order to obtain the breakdown field. The resulting dielectric breakdown data were statistically analyzed assuming two-parameter Weibull statistics [41]; the cumulative probability of failure was approximated using the median rank method [42].

3. Modeling in FEMM 4.2

The electrostatics module in FEMM 4.2 was used for electric field analysis purposes; the use of electrostatic analysis is common in simulating electric field distribution such as those related to high voltage research [43,44], electromagnetics [45], materials science [46], experimental and particle physics [47,48], and medicine [49]. The mesh size parameter, which defines a constraint on the largest possible elements size allowed in the associated section, was chosen to be 0.001: this led to more than 1,000,000 nodes and 2,000,000 elements generated. This allowed the mesh generator in FEMM 4.2 to fill the region with nearly equilateral triangles in which the sides were approximately the same length as the specified mesh size parameter. Meanwhile, the solver precision used to determine the single precision accuracy was set at $10^{-8}$. Nevertheless, as with many other finite element methods, the boundaries
for different materials with different values for permittivity could not be competently resolved using FEMM 4.2, thus resulting in discontinuities along the boundaries; solving these boundary conditions was not of our main interest here, but it can be pursued elsewhere in the literature [50–54].

Several assumptions were made for simulation purposes, such as: i) the particle was spherical in size and it was enclosed in an interphase of a constant thickness, ii) the particle distribution within the polyethylene was homogeneous, iii) the particle interacted strongly with the polymer, iv) the electric field intensity was mainly affected by the variation in permittivity, and v) the electric field intensity was not significantly affected by temperature or space charge effects.

3.1. Nanocomposite models

In the current work, we propose a two-dimensional unit cell model based upon a polymer and a spherical nanoparticle with a surrounding interphase layer of discrete thickness that was permitted to overlap with neighboring interphase layers. For this, three unique phases were assumed to exist within the nanocomposites: the polymer, the nanoparticle, and the single-layer interphase. The model (1 μm × 1 μm) was placed between a high voltage electrode (+200 V) and a ground electrode, as shown in Fig. 1.

The nanocomposite models were based upon polyethylene as the polymer and silicon dioxide as the nanoparticle (nanosilica), with assumed permittivity values of 2.3 and 3.9, respectively [38,55,56]. In all models, the size of each nanoparticle was assumed 80 nm in diameter; this would somehow match the nanosilica’s [38,55,56]. In all models, the size of each nanoparticle was assumed with assumed permittivity values of 2.3 and 3.9, respectively

\[
\varepsilon_2 = 3.9; \quad \varepsilon_3 = 3.0, \quad \varepsilon_4 = 2.3.
\]

The thickness of the interphase was again considered for representing the non-overlapping interphase effects. For Nano Model II and Nano Model III, adjacent nanoparticles with 0 nm (see Fig. 2b) and 40 nm (see Fig. 2c) interparticle distances were considered for representing the overlapping interphase effects. For Nano Model IV, V, and VI, adjacent nanoparticles with 30 nm (see Fig. 2d), 120 nm (see Fig. 2e), and 240 nm (see Fig. 2f) interparticle distances were considered for representing the non-overlapping interphase effects. Of note, the nanoparticle and the interphase for Nano Model II, III, IV, V, and VI were assumed similar with Nano Model I.

Lastly, Particle Size Model was introduced to determine the effect of particle size distribution on the electric field distribution. For this, about 10% and 90% particle under-sizing was considered — the electric field distribution for nanoparticles with 70 nm and 10 nm diameters was analyzed and compared with that of the 80 nm diameter. In these, the thickness of the interphase was, again, assumed half the diameter of the nanoparticle. The permittivity values for the polymer, nanoparticle, and interphase were fixed at 2.3, 3.9, and 3.0, respectively.

Fig. 3a shows an illustrating example of the analyzed electric field intensity in Nano Model I. The field distribution of the analyzed area represents the case where the permittivity of the interphase became different from the permittivity of the polymer and the nanoparticle. The y-line and the x-line in Fig. 3a show where the electric field intensity data were taken from each case for comparison purposes. Meanwhile, Fig. 3b illustrates how the interparticle distance between adjacent nanoparticles was modeled, with analyzed electric field intensity. The colors representing varying electric field intensities across a sample are shown in Fig. 3c.

4. Results

4.1. Experimental breakdown strength

4.1.1. DC breakdown strength

Fig. 4a compares Weibull plots of DC breakdown strength for LDPE containing 0 wt%, 1 wt%, 3 wt%, and 5 wt% of nanosilica. The DC breakdown strength of the unfilled LDPE was 240 kV/mm. The DC breakdown strength of LDPE containing 1 wt% of nanosilica reduced to 131 kV/mm. As the amount of nanosilica increased from 1 wt% to 3 wt%, the DC breakdown strength increased slightly to 150 kV/mm. Nevertheless, considering the uncertainties in Weibull analysis, the slight variation of the DC breakdown strength for LDPE containing 1 wt% and 3 wt% of nanosilica is negligible. Meanwhile, the DC breakdown strength of LDPE containing 5 wt% of nanosilica was the lowest among all, i.e., 103 kV/mm. To confirm the behavior of polyethylene on the addition of nanosilica, another batch of
material systems was investigated, where HDPE was used in place of LDPE, and a similar breakdown trend was observed (see Fig. 4b).

### 4.1.2. AC breakdown strength

Fig. 5a compares Weibull plots of AC breakdown strength for LDPE containing 0 wt%, 1 wt%, 3 wt%, and 5 wt% of nanosilica. The AC breakdown strength of the unfilled LDPE was 148 kV/mm. The AC breakdown strength of LDPE containing 1 wt% of nanosilica was 129 kV/mm. As the amount of nanosilica increased from 1 wt% to 3 wt%, the DC breakdown strength decreased slightly to 125 kV/mm. Nevertheless, considering the uncertainties in Weibull analysis, the variation of the AC breakdown strength for LDPE containing 0 wt%, 1 wt%, and 3 wt% of nanosilica is negligibly small. Meanwhile, the AC breakdown strength of LDPE containing 5 wt% of nanosilica was the lowest among all, i.e., 105 kV/mm. Again, HDPE was used in place of LDPE to confirm the AC breakdown behavior of polyethylene on the addition of nanosilica, and a similar breakdown trend was observed (see Fig. 5b).

### 4.2. Electric field analysis

Fig. 6 shows plots of the electric field intensity along the y-line, from the center of origin of the nanoparticle, extending towards the polymer region. The electric field was found to be homogeneously distributed in the unfilled polymer (Polymer Model). The electric field intensity of the unfilled polymer was found to remain constant at 200 kV/mm irrespective of permittivity values (see the plot for “Unfilled” in Fig. 6).

With the addition of a nanoparticle without interphase having a permittivity value of \( \varepsilon_2 = 3.9 \), higher than that of the polymer, the electric field intensity within the nanoparticle became lower (149 kV/mm) than that in the polymer region (see the plot for “No Interphase” in Fig. 6b). However, the electric field intensity immediately adjacent to the surface of the nanoparticle (along the y-line) increased significantly up to 251 kV/mm. Further away from the nanoparticle’s surface, the electric field intensity resumed slowly to that of the polymer. Meanwhile, the electric field intensity for the regions along the x-line showed an opposite effect (see Fig. 6c) — in a dielectric, an increase in electric field intensity at a location is compensated by a decrease in electric field intensity elsewhere. Therefore, the electric field distribution was distorted.

When an interphase was assumed to surround a nanoparticle (Nano Model I), the electric field intensity within the interphase and at regions immediately adjacent to the interphase along the y-line became every different from that without an interphase. For the case where the interphase permittivity was assumed to be lowered (\( \varepsilon_3 = 1.5 \)) than that of the nanoparticle (\( \varepsilon_2 = 3.9 \)) and the polymer (\( \varepsilon_1 = 2.3 \)), high electric field intensity was recorded within the interphase (see the plot for “Interphase 1.5” in Fig. 6b). The intensity reduced significantly at regions immediately adjacent to the interphase, before resuming to the value exhibited by the unfilled polymer. Meanwhile, a higher permittivity value of the interphase (\( \varepsilon_3 = 9.0 \)) compared to that of the nanoparticle (\( \varepsilon_2 = 3.9 \)) and the polymer (\( \varepsilon_1 = 2.3 \)) resulted in an entirely opposite field effect within the interphase and at regions immediately adjacent to the interphase (compare the plots between “Interphase 9.0” and “Interphase 1.5” in Fig. 6b). If the interphase permittivity value lies between (\( \varepsilon_3 = 3.0 \)) that of the nanoparticle (\( \varepsilon_2 = 3.9 \)) and the polymer (\( \varepsilon_1 = 2.3 \)), the variation in electric field intensity is within the nanoparticle region, the interphase region and the polymer region appears to be less drastic (compare the plots for “Interphase 3.0”, “Interphase 9.0”, “Interphase 1.5” and “No Interphase” in Fig. 6b). Along the x-line, the effects of permittivity on the electric field intensity immediately adjacent to the surface of the nanoparticle are entirely opposite to that along the y-line (compare Fig. 6c with Fig. 6b). For brevity, we will focus on the electric field intensity along the y-line from this point onwards.

Fig. 7 shows the effects of electric field distribution within a model nanocomposite containing two adjacent nanoparticles separated 0 nm apart (with overlapping interphases) (Nano Model II). If the interphase permittivity became lower (1.5) than the constituents’ permittivity (\( \varepsilon_1 = 2.3 \) for the polymer and \( \varepsilon_2 = 3.9 \) for the nanoparticle), the electric field intensity adjacent to the nanoparticles’ touching surface (see line \( A_1 \) in Fig. 7b) and at the interphase (between lines \( B_1 \) and \( C_1 \) in Fig. 7b) increased significantly compared to that of the unfilled polymer. Outwards from the interphase (see line \( C_1 \) onwards in Fig. 7b), the electric field intensity was slightly lower than the unfilled polymer case, but resumed to that of the polymer at distances further away from the interphase. An entirely opposite trend to the situation discussed above can be observed for the case of nanocomposites having an interphase permittivity higher (\( \varepsilon_3 = 9.0 \)) than the constituents’ permittivity. Meanwhile, the electric field distribution for the case of nanocomposites having interphase permittivity between those of the constituents (\( \varepsilon_3 = 3.0 \)) seems to be less distorted when compared with the aforementioned two cases. The trend seems comparable to the case of nanocomposites having no interphase, albeit that the variation of electric field intensity at regions immediately adjacent to the nanoparticle surface is greater for the case of nanocomposites having no interphase.

When two adjacent nanoparticles became 40 nm separated apart (with overlapping interphases), as shown in Fig. 8 (Nano Model III), the electric field distortion within the overlapped interphase (between lines \( A_2 \) and \( B_2 \) in Fig. 8b) became noticeable.

### Table 1

Models and parameters.

<table>
<thead>
<tr>
<th>Model</th>
<th>Permittivity</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polymer Model</td>
<td>Fixed: 2.3</td>
<td>No nanoparticle</td>
</tr>
<tr>
<td>Nano Model I</td>
<td>Fixed: 2.3</td>
<td>Interparticle distance: 0 nm</td>
</tr>
<tr>
<td>Nano Model II</td>
<td>Fixed: 2.3</td>
<td>Interparticle distance: 40 nm</td>
</tr>
<tr>
<td>Nano Model III</td>
<td>Fixed: 2.3</td>
<td>Interparticle distance: 40 nm</td>
</tr>
<tr>
<td>Nano Model IV</td>
<td>Fixed: 2.3</td>
<td>Interparticle distance: 0 nm</td>
</tr>
<tr>
<td>Nano Model V</td>
<td>Fixed: 2.3</td>
<td>Interparticle distance: 120 nm</td>
</tr>
<tr>
<td>Nano Model VI</td>
<td>Fixed: 2.3</td>
<td>Interparticle distance: 240 nm</td>
</tr>
<tr>
<td>Particle Size Model</td>
<td>Fixed: 2.3</td>
<td>Single nanoparticle with varied sizes: 80 nm, 70 nm, or 10 nm</td>
</tr>
</tbody>
</table>
Again, the lower ($\varepsilon_3 = 1.5$) or higher ($\varepsilon_3 = 9.0$) interphase permittivity as compared with the constituents’ permittivity resulted in highly distorted electric field intensity along the analyzed regions. Meanwhile, for the case of nanocomposites having interphase permittivity between ($\varepsilon_3 = 3.0$) those of the constituents, the electric field intensity seems to be less distorted when compared with the case of nanocomposites having no interphase, and the electric field distribution within the interphase region (between lines A2 and B2 and between lines C2 and D2 in Fig. 8b) of the nanocomposite was somehow comparable with the unfilled polymer.

Fig. 9a illustrates two adjacent nanoparticles that were separated 80 nm apart such that the interphases of the nanoparticles did not overlap, but touched each other (Nano Model IV). In this...
case, the advantage of having an interphase with the permittivity between \( \varepsilon_3 = 3.0 \) those of the constituents became more apparent. Overall, the distortion of the electric field intensity for the case of nanocomposites having interphase permittivity between \( \varepsilon_3 = 3.0 \) those of the constituents was less than the case of nanocomposites having no interphase and having interphase permittivity lower \( \varepsilon_3 = 1.5 \) and higher \( \varepsilon_3 = 9.0 \) than those of the constituents (see Fig. 9b).

Fig. 10 shows the effects of interphase permittivity as the adjacent nanoparticles were separated further apart (120 nm), such that the interphases of the adjacent nanoparticles did not touch each other (Nano Model V). For all investigated cases with interphases, the distortion of the electric field intensity in the polymer region adjacent to the nanoparticle surfaces (between lines B4 and C4 in Fig. 10b) became great. The nanocomposite having interphase permittivity between \( \varepsilon_3 = 3.0 \) those of the constituents showed a sudden increase of electric field intensity within the region encompassing lines B4 and C4. Nevertheless, the electric field intensity was, again, somehow comparable with the case of nanocomposite having no interphase.

Similar effects as discussed in Fig. 10 can be observed in Fig. 11 for the case of adjacent nanoparticles separated further apart (240 nm; Nano Model VI). However, the distortion effects of the electric field intensity in the polymer region adjacent to the
nanoparticle surfaces (between lines B$_3$ and C$_3$) became mitigated as the separation distance between the adjacent particles increased. Again, the electric field distortion effects for the cases of having no interphase and interphase permittivity between ($\varepsilon_3 = 3.0$) those of the constituents seem to be comparable, but the maximum value of the electric field intensity was higher for the case of having no interphase.

Fig. 12 shows that the electric field distribution is dependent upon the size — and the interphase thickness — of the nanoparticle. The much smaller-sized nanoparticle (10 nm) showed quick, momentarily changes in electric field intensity when compared with those with larger diameter nanoparticles (70 nm and 80 nm).

5. Discussion

From the experimental results, the DC breakdown strength of the unfilled LDPE and HDPE was the highest in comparison with their nanofilled counterparts containing 1 wt%, 3 wt%, and 5 wt% of nanosilica. For this, the addition of nanosilica into polyethylene reduced the DC breakdown strength of the polyethylene. Meanwhile, the variation of AC breakdown strength for both the LDPE and HDPE containing 1 wt% and 3 wt% of nanosilica was found to be insignificant in comparison with their unfilled counterparts. These DC and AC breakdown trends are not unusual and have also been reported elsewhere for polyethylene blend samples added with 0–10 wt% of nanosilica (10–20 nm size range) [58].

The simplest explanation for the reduced DC breakdown strength can be attributed to the effect to nanoparticles agglomeration, which seems to be more pronounced in DC than in AC, and more sensitive to the nanofiller loading level. Nevertheless, another possible explanation for the reduced DC breakdown strength is that the addition of nanosilica introduces permittivity mismatches that could enhance the electric field intensity — an effect that as well seems to be more critical in DC field than in AC field. We will now discuss the analysis of DC electric field in detail.

If the permittivity of a nanoparticle is higher than that of a polymer, the electric field intensity surrounding the nanoparticle will be distorted. This resembles the electric field distribution in commonly used polymers and nanoparticles as nanocomposites — in our case, polyethylene ($\varepsilon_1 = 2.3$) and nanosilica ($\varepsilon_2 = 3.9$). Clearly, adding nanoparticles into polymers results in distorted electric field intensity within the material.

By assuming that an interphase is present around a nanoparticle, and the interphase has a unique permittivity itself, the electric field distribution within the interphase region would be very different from that of the nanoparticle and the polymer. Significantly, our analysis suggests that the permittivity of the interphase needs to lie between the permittivity of the constituents for mitigating electric field distortion effects. Although the electric field distribution in these cases is nowhere as homogeneous as the case of unfilled polymer, the electric field distortion effects can be mitigated as adjacent nanoparticles separated further away from each other. This may help in explaining improved DC breakdown performance of nanocomposites having low nanofiller contents in comparison with nanocomposites having high nanofiller contents as noticed in our breakdown experiments — as the nanofiller amount is reduced, the separation distance between adjacent nanoparticles becomes greater, and the electric field becomes less distorted, so the breakdown strength increases.

Alternatively, if the permittivity of the interphase is higher than the permittivity of the constituents, a large distortion of the electric field would occur, and this would theoretically jeopardize the dielectric performance of the resulting nanocomposites. In reality, this phenomenon resembles the presence of water within the interphase of nanocomposites, since the interphase can be a preferred location for the aggregation of water molecules. As water molecules couple strongly with the applied field, changes in their environment will be reflected in changes in the electric field intensity, particularly under DC fields. Since the nanosilica used in our experiment will inevitably contain surface hydroxyl groups, water tends to create bonding with the hydroxyl groups. Consequently, higher permittivity values may be noticed within the
interphase, thus distorting the electric field distribution and subsequently reducing the breakdown strength of the nanocomposites.

It has been reported that the overall permittivity of a nanocomposite could decrease remarkably, out of the permittivity range of its constituents [21,34]. In this case, the lowered overall permittivity could be attributed to the lowered interphase permittivity than the permittivity of the constituents. While lowered permittivity values of the interphase is thought to enhance the breakdown performance of a nanocomposite, our analysis showed otherwise — a large distortion of the electric field would occur under this condition, and reduced breakdown performance could be expected.

In addition, our analysis also showed that, with the interphase maintained to be half the diameter of the nanoparticle, much smaller sized nanoparticles would theoretically lead to less electric field distortion when compared with larger sized nanoparticles.

Therefore, provided that nanoparticles could be homogenously dispersed in nanocomposites and the interphase within nanocomposites could be properly managed, the use of nanoparticles with small diameters would seem to be preferable over their larger...
It is noteworthy that our discussion thus far is mainly for the material’s breakdown behavior under DC electric fields. This is because, under AC electric fields, the material’s response may not be able to keep up with changes in AC frequencies when taking into account the phase lag between AC and material responses [59]. Consequently, the AC breakdown behaviors are quite different from the DC breakdown behaviors, as noticed in our experiments. To further understand these breakdown behaviors, polarization effects upon nanoinclusions can be pursued elsewhere in the literature [38, 60, 61].

6. Conclusions

The results from our current work show that the polyethylene/silica nanocomposites’ breakdown behaviors are different under DC and AC applied fields. The addition of an increasing amount of nanosilica into the LDPE and the HDPE progressively reduces the DC breakdown strength of the polyethylenes; such an effect is not apparent under AC applied fields. While the simplest explanation is to attribute the effect to nanoparticle agglomeration, which seems to be more pronounced in DC than in AC, we seek to clarify the DC effect based upon the concept of permittivity mismatch between the nanoparticle and the polymer. Our analysis shows that, if the permittivity of a nanoparticle is higher than that of a polymer, the electric field intensity is nowhere as homogeneous as the case of the unfilled polymer, and this may subsequently lead to reduced breakdown strength. Meanwhile, the presence of the interphase, when assigned a unique interphase permittivity value between those of the constituents, results in less distorted electric field distribution in comparison with the cases of nanocomposites having interphase permittivity lower or higher than the constituents’ permittivity. Also, the electric field distortion effects can be mitigated as adjacent nanoparticles separated far away from each other, a phenomenon commonly associated with nanocomposites containing a lower amount of nanoparticles, thus improving the breakdown performance. It is noteworthy that our current analysis only considers the effects of a single particle or adjacent particles in our attempt to relate the electric field distribution of our proposed nanocomposite models with the observed experimental
breakdown strength. As such, our analysis falls short of providing a comprehensive understanding on the electric field effects within a practical bulk nanocomposite system. Of note, breakdown mechanisms in a dielectric material is not solely affected by the permittivity of the material constituents or the electric field distribution within the material, and other factors such as thermal and chemical effects need to be considered altogether. Nevertheless, we believe our current work helps further to understand the experimentally observed DC and AC breakdown performances of the nanocomposites from the perspective of electric field distribution within the nanocomposites, based upon our proposed nanocomposite models.

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References


Fig. 12. Effects of (a) nanoparticle size distribution (with interphase) on the plots of electric field intensity along the (b) y-line, from the center of origin of a nanoparticle, extending towards the polymer region (Nano Model VII).
constants to increase dielectric breakdown strength, in: Proc. 19th International Conference on Composite Materials, Montreal, Canada, July 28–August 2, 1–9, 2013.


