Modelling the effect of nanofiller’s shape and alignment on the dielectric strength and permittivity of polyethylene nanocomposite insulation

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Abstract

Solid insulation defects are a major cause of power failures. Incorporation of micro and nanoparticles in the solid insulation materials has proven its advantage. It has been reported that nanocomposites are far superior in terms of insulation to microcomposites because of larger interphase area between nanofillers and polymer in nanocomposites. In addition to that, size, shape, concentration and alignment of nanofillers affect the properties of resulting nanocomposite insulation. A lot of work has reported the effect of nanofiller’s nature, size and concentration but very little is reported on the effect of filler’s shape and alignment on the insulation and dielectric properties. In this paper nanocomposite dielectrics with nanofillers of various shapes and alignment are modelled by varying their permittivity using finite element method. To characterize the effect of various shapes and orientation electric field intensity and energy density models are computed. Simulations are performed using AC/DC module of COMSOL Multiphysics. The results revealed that shape and alignment of nanofillers are the two key factors in tuning the breakdown strength and permittivity of nanocomposite dielectrics for their applications in insulation as well as high voltage capacitors.

1. Introduction

Polymer composite materials are being used in high voltage industry over the past few decades due to their superior electrical and mechanical properties as compared to conventional ceramic materials [1]. Fillers are added in a polymer to enhance its electrical and mechanical characteristics. Addition of microfillers results in improved mechanical properties with a little degradation in electrical breakdown strength. However, addition of nanofillers into polymer results in improvement of both electrical and mechanical properties at the same time [2–4]. Nanocomposite dielectric is a composite having nanofillers with dimensions of at most 100 nm [5]. Interphase is the region around nanofiller in which it interacts with a polymer [6]. Nanocomposite dielectric has a larger interphase area between polymer and nanofiller as compared to the microcomposite dielectric.

Different models have been developed in order to investigate the properties of interphase and its size [6–10]. Two famous classical interphase models were developed by Lewis and Tanaka. Lewis modelled the interphase area around nanofiller by splitting it into two layers, named as, Stern and Diffuse layer. Layer attached to the nanofiller with high charge concentration having a width of about 1 nm is named as Stern layer. Outer layer which is attached to the polymer having low concentration of charges is Diffuse layer. Width of this layer depends on the nature of nanofiller. Thickness of this layer is few nm for high permittivity fillers and tens of nm for low permittivity fillers [6, 10]. Tanaka modelled the interphase using multi core model. This model consists of three layers named as Transition, Bound and Loose layers along with a double layer over these three layers. Transition is the innermost layer with least thickness and highest charge concentrations. Layer next to Transition is Bound layer having thickness up to 10 nm with moderate charge concentration. The outermost layer is the Loose layer, having a thickness of tens of nm and possesses low charge concentrations [8]. A two dimensional interphase model demonstrating the effect of properties of interphase on other dielectric
characteristics was developed by Z. Hashim et al. through finite element method [11]. Interphase with varying thickness and permittivity were investigated in this modeling and it was found that improved dielectric properties could be achieved with an interphase thickness of half the diameter of nanofiller as well as interphase permittivity close to the mean permittivity of nanofiller and polymer. Statistical analysis of electric field strength of nanocomposite dielectrics was performed by using finite element method [12]. Finite element simulations are useful in predicting the breakdown strength of nanocomposite dielectrics.

Polyethylene/silica nanocomposites were modelled by K Y Lau et al. [13] using finite element method. In addition to interphase permittivity and width, filler’s permittivity, concentration and size were considered to compute their effect on electric field distribution. A 3D model was developed by A N Asokan et al. [14] to investigate the mean electric field and the coefficient of volume fraction. Input parameters were nanofiller’s size, concentration, conductivity, permittivity and interphase thickness.

Shapes of nanofillers and their alignment through external field result in tuned dielectric properties. X. Huang et al. analyzed the effect of nanofiller’s shape on the suppression of space charge [15]. In this study, spheres and sheets of Boron Nitride nanofiller were blended with low density polyethylene and respective analysis revealed that spherical nanofillers were more effective regarding space charge suppression. However, the impact of nanofiller’s shape upon other dielectric characteristics such as electric field intensity and energy density were not reported in this study. Moreover, elliptical/tube shapes and their orientation were also not involved in the analysis.

Epoxy/Sodium Titanate-nanowire nanocomposites were prepared by S Chen et al. [16]. Nanowires were aligned perpendicularly and parallel to the applied field. It was reported that perpendicularly aligned nanowires gave better dielectric breakdown strength as compared to parallel aligned nanowires. However, author did not

![COMSOL flow chart for nanocomposite dielectric modelling.](image-url)
analyze the impact of nanofiller’s shapes on tuning the dielectric properties. Additionally, the effect of alignment regarding different nanofiller’s shapes, other than nanowires, was not reported. Moreover, impact of alignment on energy density was not investigated.

Dielectric properties of nanocomposite dielectrics are influenced by nanofiller’s size, shape, permittivity, concentration and its alignment. It’s always beneficial to determine the dielectric behavior of nanocomposite dielectrics through modelling before their actual preparation. In this paper the effect of nanofiller’s shape, alignment and permittivity on the electric field intensity and energy density of various formulations of nanocomposite dielectric materials was investigated using finite element analysis modelled in AC/DC module of COMSOL Multiphysics.

2. Methodology

A two dimensional nanocomposite dielectric model with dimensions of 1000 nm by 1000 nm is selected. Polyethylene with permittivity of 2.3 is selected as the base polymer. To simulate stress of 40 kV mm\(^{-1}\) which is comparable to breakdown strength of polyethylene, a voltage of 40 V is set at anode at the bottom while the upper anode is set at ground potential. Four nanofillers; Clay, SiO\(_2\), Al\(_2\)O\(_3\) and TiO\(_2\) with permittivity values of 3.2, 3.7, 9.8 and 100 respectively were selected. Weight percentage of all these nanofillers is kept constant at 1 wt%. Nanofillers with three different shapes i.e. spherical, elliptical and tubes were embedded in the developed

| Material | Permittivity | Permittivity of Interphase |
|----------|-------------|---------------------------|
| PE       | 2.3         | —                         |
| Clay     | 3.2         | 2.7                       |
| SiO\(_2\) | 3.7         | 3.0                       |
| Al\(_2\)O\(_3\) | 9.8     | 6.05                      |
| TiO\(_2\) | 100         | 51.15                     |

Figure 2. Electric field intensity distributions of nano-clay fillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.
In case of elliptical and tube shape nanofillers the effect of horizontal and vertical alignment was also investigated separately. Spherical nanofillers having diameter of 60 nm and tube nanofillers with diameter of 10 nm and length of 60 nm were used in all cases. Elliptical nanofillers used have a diameter of 90 nm on the major axis and 60 nm on the minor axis. Interphase permittivity was chosen as mean of the permittivity values of polymer and nanofiller. Width of interphase for spherical nanofillers was selected as half of the diameter of

Figure 3. Electric field intensity of clay nanofillers.

Figure 4. Electric field intensity distributions of SiO$_2$ nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.

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nanofiller (30 nm) while for elliptical nanofillers, 30 nm for minor axis and 45 nm for major axis were selected. Interphase width of 5 nm was selected for tube shape nanofillers. Inter-particle distance was kept constant at 360 nm for all simulations cases. Information regarding the materials and their permittivity and the permittivity of interphase is tabulated in table 1.

Simulations based on finite element method were performed using COMSOL Multiphysics. Flow chart describing various steps of modelling is shown in figure 1.

3. Results

Two parameters; electric field intensity and energy density were computed for four nanofillers of different permittivity. Each of nanofiller was classified regarding its shape and alignment into five cases of spherical shape, horizontal aligned elliptical and tube shape and vertical aligned elliptical and tube shape. A total of 40 simulation cases were investigated.

3.1. Electric field intensity of spherical, elliptical and tube shape nanofillers

3.1.1. Electric field intensity of nano-clay fillers

Electric field intensity distributions of nano-clay fillers with various shape and alignments are shown in figure 2. Electric field intensity has its maximum value at the interphase of nanofiller and polymer and the least value inside the nanofiller as can be seen in figure 3. Highest electric field intensity of 45 kV mm$^{-1}$ is shown by vertical aligned elliptical shape nanofillers and the lowest of 42 kV mm$^{-1}$ by horizontal aligned tube shape nanofillers. No significant difference is observed in electric field intensities of spherical shape, horizontal aligned elliptical shape and vertical aligned tube shape for low permittivity nano-clay fillers.

3.1.2. Electric field intensity of SiO$_2$ nanofillers

Electric field intensity distributions of SiO$_2$ nanofillers with different shapes and alignments are shown in figure 4. As per studies performed by different authors, high permittivity materials have higher electric field intensities as compared to the low permittivity materials $^{[17, 18]}$. It is vivid from figure 5 that electric field intensity values of SiO$_2$ nanofillers are slightly higher than nano-clay fillers for various shapes and alignments. This rise in electric field intensity is due to higher permittivity of SiO$_2$ nanofillers (3.7) as compared to nano-clay fillers (3.2). Incorporation of high permittivity nanofiller into base polymer material resulted in higher permittivity of nanocomposite dielectric material which ultimately caused higher electric field intensity at the interphase between nanofiller and polymer. It is interesting to note that for SiO$_2$ nanofillers, approximately 5% rise in electric field intensity values of spherical shape, horizontal aligned elliptical shape and vertical aligned tube is observed as compared to nano-clay fillers. Maximum rise of approximately 6.66% is observed for vertical aligned elliptical shape and minimum of 1% is observed for horizontal aligned tube shape nanofillers. For small
Figure 6. Electric field intensity distributions of Al$_2$O$_3$ nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.

Figure 7. Electric field intensity of Al$_2$O$_3$ nanofillers.
rise in permittivity, vertical aligned elliptical shape nanofillers show significant rise while horizontal aligned tube shape nanofillers show negligible rise in electric field intensity.

Figure 8. Electric field intensity distributions of TiO$_2$ nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.

Figure 9. Electric field intensity of TiO$_2$ nanofillers.
3.1.3. Electric field intensity of Al₂O₃ nanofillers

Electric field intensity distributions of moderate permittivity Al₂O₃ nanofiller for five different cases of shapes and alignments are shown in figure 6. Higher values of electric field intensity are observed for vertical aligned nanofillers and lower values for horizontal aligned nanofillers. Moderate values of electric field intensity are shown by spherical shape nanofillers. It is obvious from figure 7 that the highest electric field intensity of 70 kV mm⁻¹ is observed for vertical aligned elliptical shape nanofillers and lowest value of 45 kV mm⁻¹ is observed for horizontal aligned tube shape nanofillers. For moderate permittivity Al₂O₃ nanofillers, 55% rise in electric field intensity of vertical aligned elliptical shape and only 7% rise of horizontal aligned tube shape nanofillers is observed in comparison to low permittivity nano-clay nanofillers.

![Figure 6. Energy density distributions of Clay nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.](image)

### Table 2. Electric Field Intensity (kV mm⁻¹) values at the interphase.

| Nanofiller | Spherical | Horizontal | Vertical | Horizontal | Vertical |
|------------|-----------|------------|----------|------------|----------|
| Clay       | 44        | 44         | 45       | 42         | 44       |
| SiO₂       | 46        | 46         | 48       | 42         | 46       |
| Al₂O₃      | 60        | 55         | 70       | 45         | 60       |
| TiO₂       | 80        | 70         | 120      | 60         | 100      |

3.1.4. Electric field intensity of TiO₂ nanofillers

Electric field intensity distributions of high permittivity TiO₂ nanofillers for five cases of different shapes and alignments are shown in figure 8. Analysis revealed a significant difference in electric field intensity for all five cases with high permittivity nanofillers, as can be seen in figure 9. In electrical insulation materials, the permittivity of nanofiller is higher than that of base polymer. Moreover variation of permittivity occurs at the interphase as compared to the nanofiller and base polymer which ultimately leads to a higher electric field stress at the interphase. Greater the difference of permittivities between nanofiller and base polymer, higher will be the electric field intensity at the interphase [11, 13, 19]. It is evident from table 2 that horizontal aligned tube shape
nano fillers exhibit least values of electric field intensity. For high permittivity TiO$_2$ nano fillers, rise in electric field intensity of 43% is observed for horizontal aligned tube shape and 166% for vertical aligned elliptical shape nano fillers in comparison to low permittivity nano-clay fillers. In general, horizontal aligned nano fillers offer less electric field intensity than vertical counter parts in all kinds of nano fillers and base polymers.

Figure 11. Energy density of Clay nanofillers.

Figure 12. Energy density distributions of SiO$_2$ nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.
3.2. Energy density of spherical, elliptical and tube shape nanofillers

Energy density of nanocomposite dielectric material is computed at two points. Energy density at the interphase of nanofiller and polymer is named as interphase energy and energy at the mid-point between two adjacent nanoparticles is referred to as mid-point energy onwards.

Figure 13. Energy density of SiO$_2$ nanofillers.

Figure 14. Energy density distributions of Al$_2$O$_3$ nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.
3.2.1. Energy density of nano-clay fillers

Energy density distributions of nano-clay fillers with different shapes and alignments are shown in figure 10. Maximum interphase energy is shown by vertical aligned elliptical shape and the minimum by horizontal aligned tube shape nanofiller. Difference in interphase energy values of spherical and vertical aligned tube shape nanofiller.

Figure 15. Energy density of Al₂O₃ nanofillers.

Figure 16. Energy density distributions of TiO₂ nanofillers with (a) Spherical shape (b) Elliptical shape horizontal aligned (c) Elliptical shape vertical aligned (d) Tube shape horizontal aligned (e) Tube shape vertical aligned.

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nano filler is negligible. Maximum mid-point energy is observed for vertical aligned elliptical shape nano filler and minimum for horizontal and vertical aligned tube shape nanofillers as can be seen in figure 11.

3.2.2. Energy density of SiO\textsubscript{2} nanofillers
Energy density distributions of SiO\textsubscript{2} nanofillers with different shapes and alignments are shown in figure 12. Interphase and mid-point energy density of SiO\textsubscript{2} nanofiller with various shapes and alignments are shown in figure 13. Interphase and mid-point energy values of SiO\textsubscript{2} nanofillers are slightly higher than of nano-clay fillers for different shapes and alignments. For SiO\textsubscript{2} nanofillers, rise in interphase energy values is 17% for vertical aligned elliptical shape nanofillers and 3% for vertical aligned tube shape nanofiller compared to nano-clay fillers. Mid-point energy shows 6% rise in case of vertical aligned elliptical shape nanofiller and insignificant rise in case of horizontal aligned tube shape nanofillers.

3.2.3. Energy density of Al\textsubscript{2}O\textsubscript{3} nanofillers
Energy density distributions of moderate permittivity Al\textsubscript{2}O\textsubscript{3} nanofiller with different shapes and alignment is shown in figures 14 and 15. Highest values of interphase and mid-point are observed in case of vertical aligned elliptical shape nanofillers. Interphase and mid-point energy values of spherical shape nanofillers are higher than vertical aligned tube shape nanofiller.

3.2.4. Energy density of TiO\textsubscript{2} nanofillers
Energy density distributions of high permittivity TiO\textsubscript{2} nanofiller are shown in figure 16. Significant rise is seen in interphase and mid-point energy values as evident from figures 16 and 17. For high permittivity TiO\textsubscript{2} nanofillers, 644% rise in interphase energy and 156% rise in mid-point energy is observed for vertical aligned elliptical shape nanofillers compared to nano-clay fillers as evident from table 3. For horizontal aligned tube shape nanofillers 23% rise in interphase energy and 4% rise in mid-point energy is observed compared to nano-clay fillers. Vertical aligned elliptical shape nanofillers exhibit highest values of interphase and mid-point energy while the horizontal aligned tube shape nanofillers show the lowest. Spherical shape nanofillers show moderate values of interphase and mid-point energy values.

4. Discussion
In this research, nanocomposite dielectric model of polyethylene with four different nanofillers having varying permittivities was developed by using COMSOL Multiphysics. In this modeling, clay, SiO\textsubscript{2}, Al\textsubscript{2}O\textsubscript{3} and TiO\textsubscript{2} nanofillers with three different shapes; spherical, elliptical and tubes, along with horizontal and vertical alignment of elliptical and tube shape were investigated. Nano-clay filler was selected as the base nanofiller (control sample) for comparative analysis.
Table 3. Energy density ($J m^{-3}$) values.

| Nano filler | Spherical | Elliptical | Vertical | Tubes |
|-------------|-----------|------------|----------|-------|
|             | Mid point | Horizontal | Vertical | Horizontal | Vertical |
| Clay        | 17 300    | 17 710     | 18 000   | 16 420   |
| SiO$_2$     | 17 880    | 18 520     | 19 050   | 16 480   |
| Al$_2$O$_3$ | 21 720    | 23 550     | 20 500   | 16 800   |
| TiO$_2$     | 27 680    | 30 560     | 46 150   | 17 100   |
|             | 76 930    | 53 240     | 156 300  | 21 720   |
|             |           |            |          | 18 850   |
|             |           |            |          | 67 750   |
Permittivity of nanocomposite dielectric material depends on the permittivity values of nanofiller, polymer and interphase [20–22]. Analysis revealed that permittivity difference at the interphase of nanofiller and polymer resulted in a higher electric field intensity at the interphase, as shown in figures 3, 5, 7 and 9. Accordingly, a maximum rise of 2.7 times in electric field intensity was observed for high permittivity TiO₂ nanofiller as compared to the control sample. Irregular electric field intensity of higher magnitudes results in poor field distribution which ultimately degrades the breakdown strength of a dielectric material [23, 24]. Hence, it is essential to use low permittivity nanofillers to produce nanocomposite dielectric materials with high breakdown strength.

Shape and alignment of nanofillers could also greatly alter the electrical properties of resulting nanocomposite dielectric material [25–27]. Analysis revealed that the least electric field intensity was observed for the tube shape nanofillers as compared to the spherical and elliptical shape nanofillers, as shown in table 2. Further table 2 shows that the electric field intensities of horizontal aligned nanofillers are lower than the vertical aligned nanofillers. For tube shape nanofillers, a rise in electric field intensity of 1.42 times was observed for horizontal aligned TiO₂ nanofiller as compared to the control sample.

Energy density of a dielectric material reflects its charge storage capability [28]. Energy density of a dielectric material can be computed by using equation (1) [29].

\[ U = \frac{1}{2} \kappa \varepsilon_0 E^2 \]  

In equation (1), \( U \) is the energy density of dielectric material, \( \kappa \) is the dielectric constant, \( \varepsilon_0 \) is the permittivity of free space and \( E \) is the electric field intensity. Energy density of a dielectric material is linearly proportional to its permittivity and quadratically proportional to the electric field intensity.

Increase in the energy density of a nanocomposite dielectric material is directly related with the permittivity of nanofiller [30]. Analysis revealed that the use of high permittivity nanofiller resulted in improved energy density at the interphase as well as at the mid-point between two nanofiller particles. Accordingly, a maximum rise of 7.4 times and 2.6 times in energy density at the interphase and mid-point was observed respectively for TiO₂ nanofiller as compared to the control sample.

Investigations revealed that the elliptical shape nanofillers resulted in greater energy density than the spherical and tube shape nanofillers. Further, energy density of vertical aligned nanofillers was observed to be greater than the horizontal aligned nanofillers. Moreover, results demonstrated that the interphase energy density of vertical aligned elliptical shape TiO₂ nanofiller was 2.3 times the vertical aligned tube shape nanofiller. Mid-point energy density of elliptical shape nanofillers in both horizontal and vertical alignment had the highest values in comparison to spherical and tube shape nanofillers. Mid-point energy density of vertical aligned elliptical shape TiO₂ nanofiller was 2.4 times the energy density of vertical aligned tube shape and 1.7 times the spherical shape nanofiller.
5. Conclusions

This research reports on the effect of nanofiller’s permittivity, shape and alignment on tuning the electric field intensity and energy density of nanocomposite dielectric material. Modeling and analysis revealed that higher permittivity of nanofiller resulted in increased electric field intensity at the interphase. Further, the increased electric field intensity degraded the electric field distribution of resulting nanocomposite dielectric, decreasing its breakdown strength. Additionally, increased permittivity of nanofiller enhanced the charge storage capability of nanocomposite dielectric material. It’s worth mentioning that significant difference in electric field intensity and energy density of spherical, elliptical and tube shape nanofillers was observed while using nanofiller with higher permittivity values. Small difference was observed in the case of low permittivity nanofillers.

Modeling and analysis also revealed that the horizontal alignment of nanofillers resulted in improved electric field distribution along with a reduction in charge storage capability of nanocomposite dielectric material. Least distortion in electric field distribution was observed for horizontal aligned tube shape nanofiller. Vertical alignment of nanofillers resulted in increased energy density of the resulting nanocomposite dielectric material. Moreover, highest energy density was observed for vertical aligned elliptical shape nanofiller. A comparison of charge storage capability and dielectric breakdown strength of nanocomposite dielectric with different shape and alignments of nanofillers is shown in figure 18.

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