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Abstract: Nanodielectrics have been regarded as a class of material system that can provide significantly improved chemical, mechanical and dielectric properties over conventional microcomposites. This is due to the presence of a high volume fraction of the interphase between nanoparticles and polymers. However, precise effects of nanodielectrics are not well understood, leading to difficulties in interpreting the dielectric behaviours of nanodielectrics. In the current work, effects of nanoparticle distributions, interparticle distances, nanoparticle sizes, interphase permittivities and interphase thicknesses on the possible electric field variations within a nanodielectric model have been simulated using Finite Element Method Magnetics (FEMM) 4.2. The results demonstrate that different nanoparticle and interphase configurations lead to different effects on the electric field intensity within the nanodielectric model. Mechanisms leading to changes in dielectric properties based on the observed electric field variations are discussed.

1 Introduction

Polymer nanocomposites first emerged as engineering materials when polyamide 6/clay nanocomposites were successfully manufactured as engineering plastics on a commercial basis in 1990s. This achievement initiated more research and development efforts to explore the possibility of combining various polymers with various inorganic nanofillers. While nanotechnology has been successfully utilised in semiconductor industries, sensor fields and biological sectors, the utilisation of nanotechnology in dielectrics and electrical insulation has been slow. Nevertheless, the year 1994 marked an important turning point for inspiring the use of nanotechnology in dielectrics and electrical insulation, when Lewis [1] published a revolutionary theoretical paper entitled 'Nanometric Dielectrics' (now widely known as nanodielectrics) highlighting potential design benefits of new dielectric materials utilising nanotechnology.

The addition of nanoparticles to polymers has been regarded as an effective approach to improve dielectric properties of polymers. Recent studies showed that the use of nanoparticles in polymeric insulating materials could enhance dielectric properties of the materials compared to microfilled and unfilled materials. The addition of nanoparticles to polymers has been demonstrated to be capable of reducing space charge accumulation, enhancing resistance to surface discharges and treeing, improving thermal conductivity and endurance, and increasing electrical breakdown strength [2–5]. Significantly, these improvements have been attributed to the presence of high volume fraction of the interphase between the nanoparticles and the polymers, possessing different structures and properties from the nanoparticles and the polymers.

Various models have been proposed to explain the role of the interphase in nanodielectrics [6–10]. According to Raetzke and Kindersberger [6], an interphase could have thickness of <1 nm or >10 nm, depending on chemical and physical bonds affecting the interphase. For example, the interphase structure could be influenced by the type of polymers, the type of nanoparticles and the use of chemical functionalisation. These would lead to different interphase structures with distinct properties – properties that belong to neither that of the polymers nor that of the nanoparticles. Lewis [7] described this through the Intensity Model, where the interphase was regarded as a transitional area between two different materials (i.e. the polymers and the nanoparticles). The interphase, also known as an electric double layer, typically contains a Stern layer surrounded by the Gouy-Chapman diffuse double layer having a Debye shielding length of more than 10 nm. Meanwhile, Tanaka *et al.* [8] proposed a multi-core model as a general hypothesis to further understand various properties and phenomena associated with nanodielectrics. Tanaka *et al.* [8] anticipated that an interphase was multi-layered and consisted of several tens nanometre of a bonded layer, a bound layer and a loose layer, with the Gouy-Chapman diffuse double layer superimposing the interphase, thus causing a far-field effect.

Elsewhere, modelling of nanodielectrics continues to draw interest among dielectric researchers in an attempt to better understand the mechanisms within nanodielectrics. For example, Kuhn and Kliem [11] performed numerical simulations based on a microscopic local field method and a dynamic Monte Carlo algorithm for calculating the properties of nanodielectrics. Daily *et al.* [12], on the other hand, proposed a three-phase theoretical model for describing the effective permittivity of nanodielectrics containing spherical nanoparticles. The model accounted for the presence of an interphase region, which surrounded each nanosphere, where its permittivity was allowed to be different from that of the polymer. This led to the successful analysis of different geometrical regimes of interphase within nanodielectrics, i.e. non-overlapping interphase, interphase overlap through cell faces, interphase overlap through cell faces and edges and matrix displaced entirely by interphase.

Kavitha *et al.* [13] carried out an experimental investigation on the effect of filler permittivity, size, shape, concentration and the interparticle distance on electrical properties of epoxy nanodielectrics containing titania and alumina. The authors then used COMSOL Multiphysics software to study the electric field distribution within the nanodielectrics. Maximum electric stress was found to occur at the interphase due to changes in permittivity between the filler and the polymer. The authors concluded that the volume of stressed region and enhancement in field mainly depended on the filler permittivity, and that the enhancement in field at the interphase would reduce the breakdown strength of nanodielectrics. Meanwhile, Cai *et al.* [14] simulated the nanoparticle distribution effect of barium titanate in five different random distribution cases generated through Matrix Laboratory

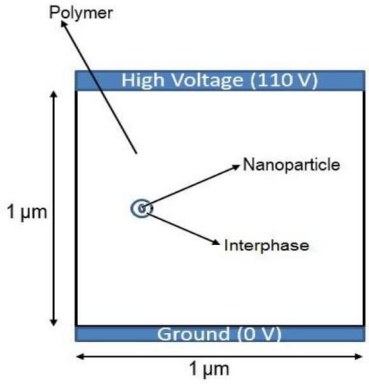

Fig. 1 *Two-dimensional nanocomposite slab with 1 μm × 1 μm dimensions*

Table 1 Properties of materials

Material	Size	Permittivity
polymer (polyethylene)	slab (1 μm × 1 μm)	fixed value (2.3)
nanoparticles (silica)	varying sizes (4, 6 and 8 nm)	fixed value (3.9)
interphase	varying thicknesses (2, 3 and 4 nm)	varying values (1.5, 3.0 and 9.0)

(MATLAB) and COMSOL Multiphysics software. Their results indicated that non-uniform distributions of barium titanate nanoparticles would aggravate the concentration of local electric field, resulting in slightly enhanced dielectric response but drastically reduced breakdown strength of nanodielectrics.

Modelling of nanodielectrics based on polyethylene (polymer) and nanosilica (nanoparticle) has also been carried out previously by Lau *et al.* [15, 16] using Finite Element Method Magnetics (FEMM) 4.2 software. From the analysis, the presence of nanoparticle with higher permittivity than that of a polymer was found to distort the electric field intensity surrounding the nanoparticle. With the presence of interphase, the variation in electric field intensity across nanoparticle region and the polymer region became less drastic if the interphase permittivity value lied between that of the nanoparticle and the polymer. Besides that, when the separation distance between adjacent nanoparticles became greater (representing reduced concentration of nanoparticles), the electric field became less distorted. The current work seeks to extend the previous simulation by first comparing the electric field distribution in nanodielectrics containing homogenously and non-homogeneously distributed nanoparticles. Effects of electric field distribution in relation to different nanoparticle sizes and interphase thicknesses are later analysed. Finally, mechanisms leading to changes in dielectric properties based on the proposed nanodielectric model are discussed.

2 Modelling and simulation

The electrostatics module in FEMM 4.2 was used for nanodielectric modelling and electric field analysis purposes. To solve the problem precisely, the mesh size parameter was chosen to be 0.001, which led to more than 1,000,000 nodes and 3,000,000 elements generated. This allowed FEMM 4.2 to fill the mesh region with nearly equilateral triangles where the sides were approximately the same length as the specified mesh size parameter. Of note, the boundaries for different materials with different values of permittivity could not be competently resolved using FEMM 4.2, thus resulting in discontinuities along the boundaries [16]. Due to the limitation of FEMM 4.2, space charge effects were not considered in the current work.

A nanodielectric model consisted of a polymer (polyethylene), a nanoparticle (nanosilica) and an interphase was modelled with a two-dimensional polymer slab (with 1 μm × 1 μm dimensions) placed between a high voltage electrode (+110 V) and a ground electrode, as illustrated in Fig. 1. The properties of the materials were assumed as in Table 1 [16–18]. The model was varied with different nanoparticle sizes, interphase thicknesses and permittivity values of the interphase. To simulate effects of nanoparticles distributions within the nanodielectric model, the position and distance between 110 nanoparticles were varied randomly.

3 Results

Fig. 2 shows the electric field distribution within the proposed nanodielectric model containing 110 nanoparticles distributed homogeneously and non-homogenously. Of note, the diameter of the nanoparticles and interphase thickness were assumed at 4 and 3 nm, respectively, while the permittivity value of the polymer, nanoparticles and interphase were assumed at 2.3, 3.9 and 3.0, respectively.

From Fig. 2*a*, the highest electric field intensity (∼127 kV/mm) occurred at the boundary between the interphase and the polymer. When nanoparticles distributions became slightly non-homogeneous (see Fig. 2*b*), the electric field intensity at the boundary between the interphase and the polymer increased slightly to ∼127.4 kV/mm. As nanoparticles distributions worsened, the electric field intensity further increased to ∼129.7 kV/mm (see Fig. 2*c*) and ∼143.7 kV/mm (see Fig. 2*d*). In addition, the range of electric field intensity within the nanodielectric model widened as nanoparticles distributions worsened. These are mainly attributed to reduced distances among nanoparticles.

Since achieving homogeneously distributed nanoparticles is difficult in nanodielectrics, five different interparticle distances (estimated from the centre of nanoparticles) from the proposed nanodielectric model were chosen for subsequent analyses, and the electric field intensities are shown in Fig. 3. From Fig. 3*a*, the highest electric field intensity (∼129.8 kV/mm) was recorded at the boundary between the interphase and the polymer when two nanoparticles (interparticle) were separated at 100 nm. With the interparticle distance reduced to 50 nm (see Fig. 3*b*), variations in electric field intensity became slightly higher (∼130.6 kV/mm). With touching interphase (see Fig. 3*c*), overlapping interphase (see Fig. 3*d*) and touching nanoparticle (see Fig. 3*e*), variations in electric field intensity became much pronounced. Of note, the red vertical line in Fig. 3 indicates where the electric field intensity data were taken from each case for subsequent analyses.

Based on the aforementioned five different interparticle distances, Fig. 4 depicts in detail how the electric field distribution varied with different interphase permittivities of 1.5, 3.0 and 9.0. The size of nanoparticles and interphase thickness were assumed at 4 and 3 nm, respectively. From Fig. 4*a* (100 nm interparticle distance), the distortion of electric field happened at the boundary between the nanoparticle and the interphase as well as the boundary between the interphase and the polymer. These were mainly caused by changes in permittivities of the nanoparticle, interphase and polymer. When the interphase permittivity was lower ($\varepsilon_r = 1.5$) than the nanoparticle ($\varepsilon_r = 3.9$) and the polymer ($\varepsilon_r = 2.3$), high electric field intensity was observed at the boundary immediately between the nanoparticle and the interphase. The intensity then gradually reduced along the interphase region towards the polymer, before experiencing a change in the field at the boundary between the interphase and the polymer. The intensity later plateau at 100 kV/mm towards the polymer. Meanwhile, when the interphase permittivity was higher ($\varepsilon_r = 9.0$) than the nanoparticles ($\varepsilon_r = 3.9$) and the polymer ($\varepsilon_r = 2.3$), entirely opposite electric field distribution effects to the case with interphase permittivity of 1.5 were observed. Variations in electric field intensity across the nanoparticle-interphase-polymer regions appeared less drastic when the interphase permittivity value ($\varepsilon_r = 3.0$) was between the nanoparticle ($\varepsilon_r = 3.9$) and the polymer ($\varepsilon_r = 2.3$).

Fig. 4*b* shows the electric field intensity as the interparticle distance reduced to 50 nm. The electric field intensity demonstrated the same behaviour as seen in Fig. 4*a*, but the intensity towards the polymer was slightly distorted further away from 100 kV/mm for interphase permittivities of 1.5 and 9.0. Fig. 4*c* illustrates that, as the interphases touched each other, the distortion of electric field intensity became much noticeable. The

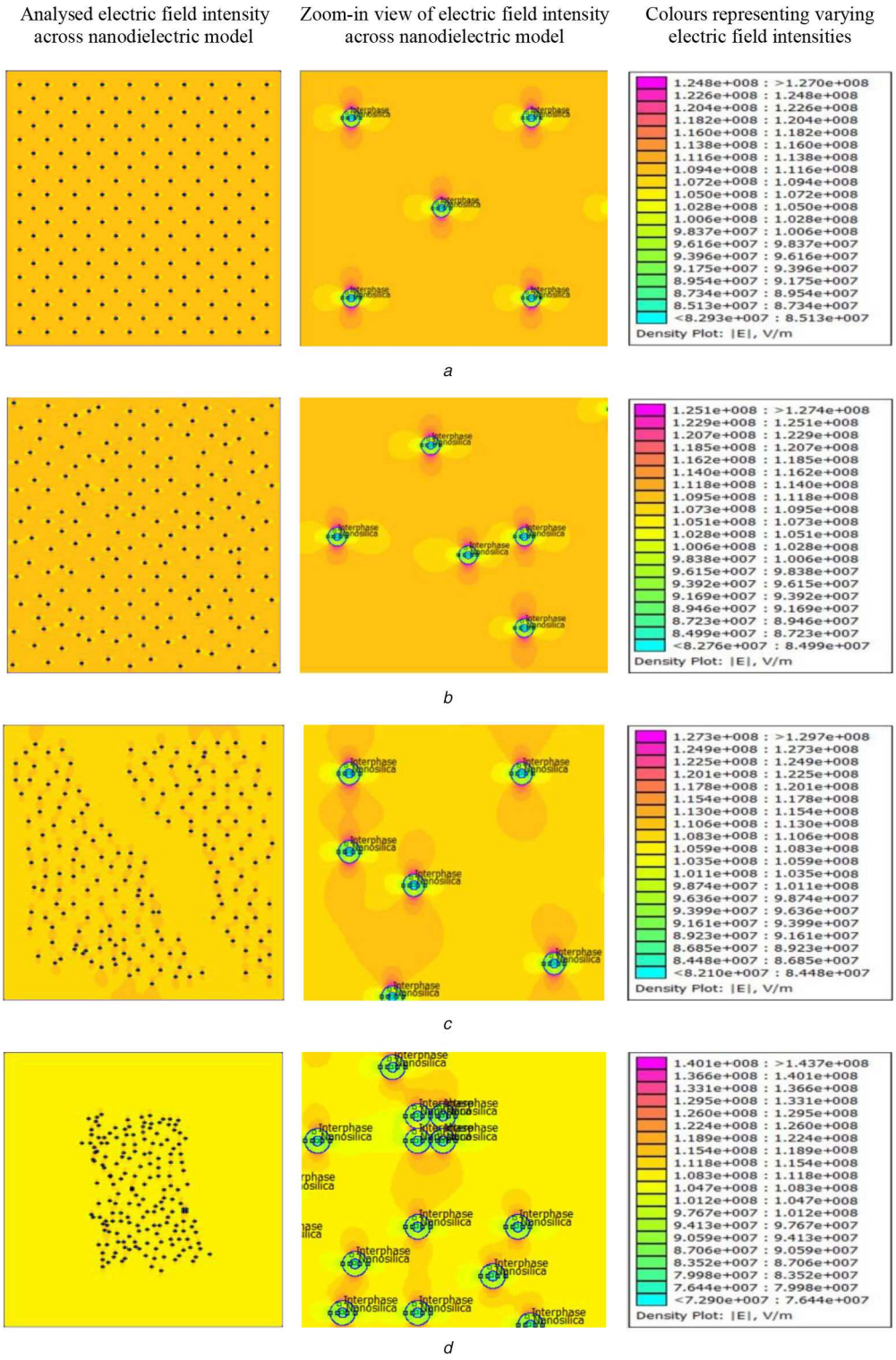

Fig. 2 *Four cases of nanoparticles distributions analysed for nanodielectric model*
(a) Homogenous nanoparticles distribution, ***(b)*** Slightly non-homogenous nanoparticles distribution, ***(c)*** Non-homogenous nanoparticles distribution, ***(d)*** Concentrated nanoparticles distribution

advantage of having an interphase with the permittivity between those of the constituents ($\varepsilon_r = 3.0$) became more apparent. The electric field intensity became less distorted compared to the case of lower ($\varepsilon_r = 1.5$) and higher ($\varepsilon_r = 9.0$) interphase permittivities. These effects became more pronounced with overlapping interphases (see Fig. 4*d*) and touching nanoparticles (see Fig. 4*e*). Significantly, for the case of nanocomposites having interphase permittivity between ($\varepsilon_r = 3.0$) those of the constituents, the electric field distribution within the interphase region of the nanocomposite model was much less distorted.

Since the interphase permittivity of 3.0 was favourable for the polymer permittivity of 2.3 and the nanoparticle permittivity of 3.9, these values were maintained for subsequent analyses. Again based on the aforementioned five different interparticle distances, Fig. 5 shows how the electric field distribution varied with different nanoparticle sizes (4, 6 and 8 nm). Of note, the interphase thickness was fixed at 3 nm.

With 100 nm interparticle distance (see Fig. 5*a*), smaller nanoparticles sizes resulted in slightly lower electric field intensities at the boundary between the nanoparticle and the interphase as well as the boundary between the interphase and the polymer. For example, the intensity value between the interphase

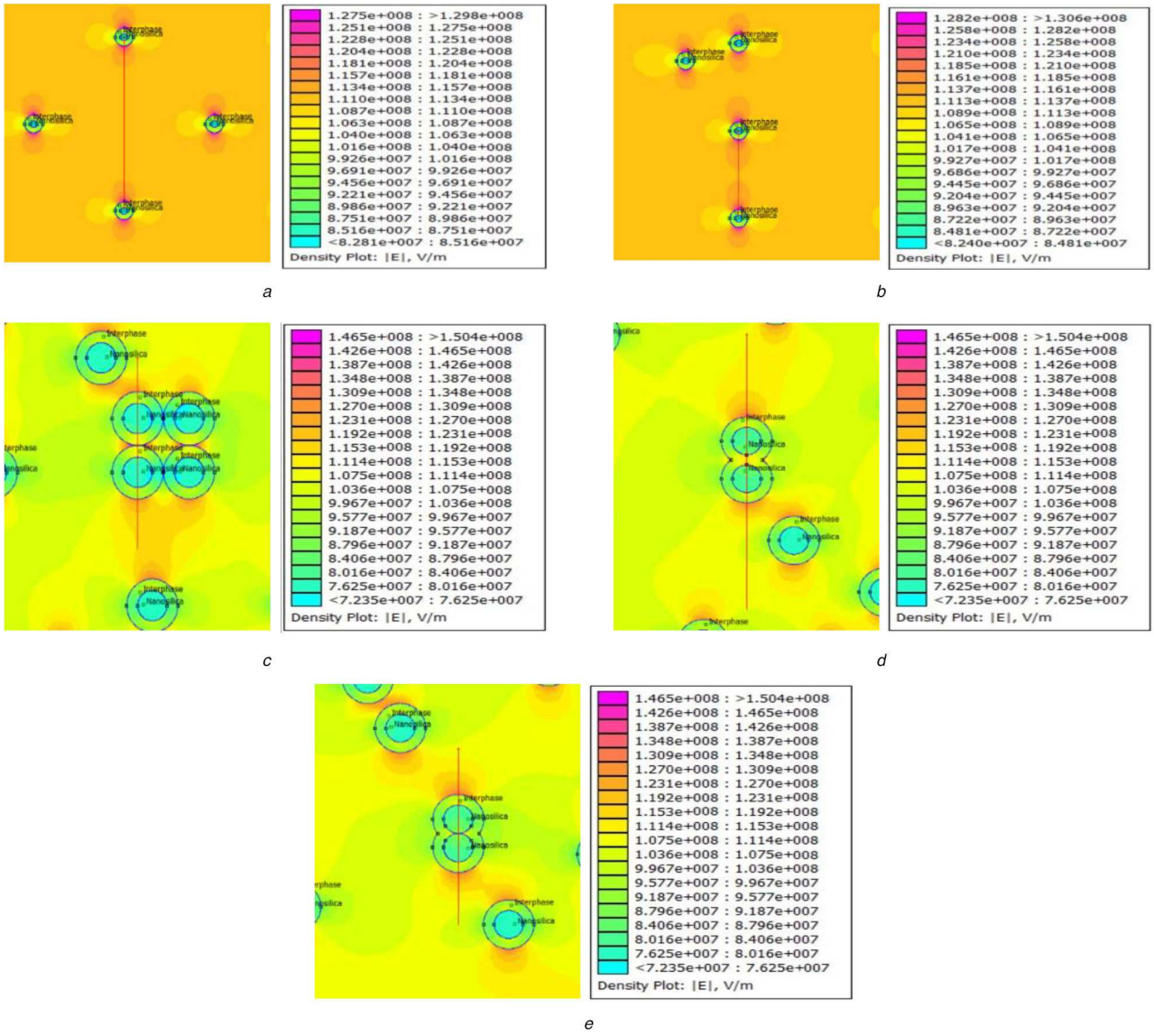

Fig. 3 *Nanodielectric model analysed with different interparticle distances (estimated from the centre of nanoparticles). The red vertical line indicates where the electric field intensity data were taken from each case for analysis purposes*
(a) 100 nm interparticle distance, ***(b)*** 50 nm interparticle distance, ***(c)*** Touching interphases, ***(d)*** Overlapping interphases, ***(e)*** Touching nanoparticles

and the polymer was lower for 4 nm nanoparticle (126 kV/mm) compared to 6 nm nanoparticle (127 kV/mm) and 8 nm nanoparticle (128 kV/mm). The electric field intensity plateau at 100 kV/mm towards the polymer. As the interparticle distance reduced to 50 nm (see Fig. 5*b*), the field intensities for all three cases remained similar, but the electric field intensities towards the polymer were slightly higher than 100 kV/mm for 6 and 8 nm nanoparticles. As the interparticle distance reduced such that the interphases of the nanoparticles touched each other, the variation of electric field intensity within the nanodielectric model became greater (see Fig. 5*c*). The distortion of electric field was significant at the touching interphase. Again, the electric field distortion was slightly less pronounced for smaller nanoparticles. Similar observations were found for interparticle distances with overlapping interphases (see Fig. 5*d*) and touching nanoparticles (see Fig. 5*e*).

Fig. 6 shows the electric field distributions when different interphase thicknesses of 2, 3 and 4 nm were analysed. The size of the nanoparticle was fixed at 4 nm and permittivities of polymer, nanoparticles and interphase remained at 2.3, 3.9 and 3.0, respectively. From Fig. 6*a*, the interphase thickness with 2 nm recorded slightly higher electric field intensity compared to the interphase thickness of 3 and 4 nm. For example, at the boundary between the interphase and the polymer, the intensity was 128 kV/mm for the interphase thickness of 2 nm. The intensity reduced slightly to 127 and 126 kV/mm for interphase thicknesses of 3 and 4 nm, respectively. With interparticle distance reduced to 50 nm (see Fig. 6*b*), the electric field distribution remained similar. Again, the electric field distribution became highly distorted especially when interphases touched each other (see Fig. 6*c*), where the highest field intensity appeared between the touching interphases. With the interphase thickness of 2 nm, the field intensity became to 145 kV/mm. For interphase thicknesses of 3 and 4 nm, the intensities were 144 and 143 kV/mm, respectively. Figs. 6*d* and *e* show the distortion of intensity across the nanodielectric model for overlapping interphases and touching nanoparticles. The field intensity reduced at the overlapped interphases but increased at the boundary between the interphase and the polymer. Again, slightly lowered field intensities were recorded with increased interphase thicknesses.

Finally, Fig. 7 compares the electric field intensity of an unfilled polymer (polyethylene) and a polymer with the inclusion of a microparticle (0.5 µm silica), and a nanoparticle (4 nm silica without and with an interphase). For the unfilled polymer, the field intensity remained constant at 100 kV/mm. For the microcomposite, the electric field intensity within the microparticle lowered to 82.9 kV/mm, but increased significantly up to 147 kV/mm immediately adjacent to the surface of the particle. For the nanodielectric, the addition of nanoparticle did not largely affect the field intensity within the nanodielectric (compared to the microcomposite) albeit that the field intensity became higher at

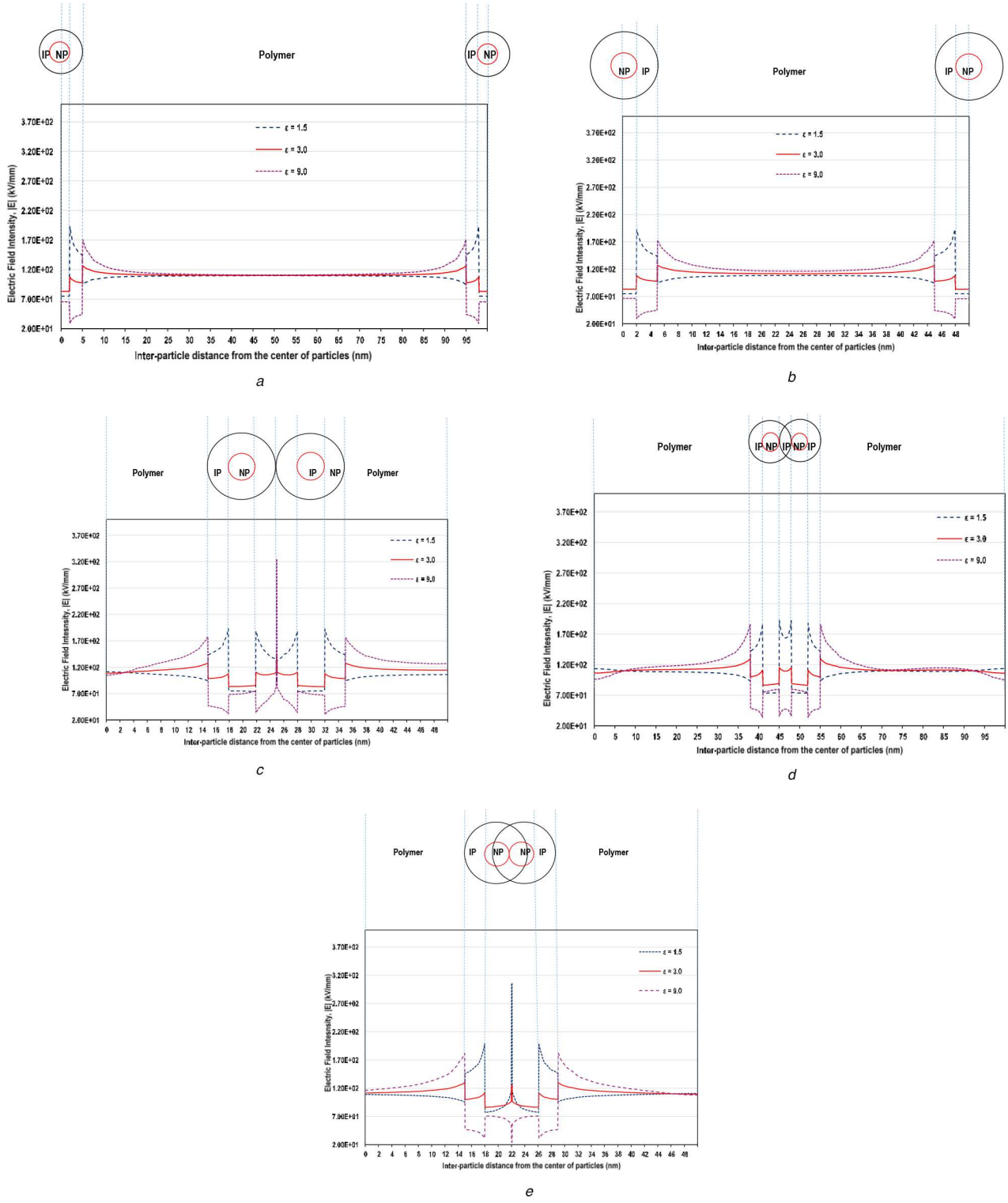

Fig. 4 *Electric field distribution versus interphase permittivity*
(a) 100 nm interparticle distance, ***(b)*** 50 nm interparticle distance, ***(c)*** Touching interphases, ***(d)*** Overlapping interphases, ***(e)*** Touching nanoparticles

region surrounding the nanoparticle. The presence of the interphase would mitigate the effect.

4 Discussion

Analyses of electric field distributions on the inclusion of nanoparticles in polymers can provide an insight into the effects of constituents permittivities, nanoparticles distributions and interphase thicknesses on the dielectric behaviours of nanodielectrics. As demonstrated through the analysed results, the widest range of electric field variation is recorded especially when nanoparticles distributions are non-homogenous (concentrated), and the highest electric field intensity occurred at the boundary between the interphase and the polymer. This is in line with the results presented by Kavitha *et al.* [13], where the maximum electric stress at the interphase was attributed to changes in permittivity between two constituents. Commonly, breakdown will be initiated at high field region, so largely distorted electric field distribution across a dielectric will lead to lowered breakdown strength of the dielectric. In this regard, concentrated nanoparticles distributions – which resemble nanoparticles agglomeration – are likely to happen when the amount of nanoparticles increased. Breakdown can therefore be initiated at the closest points between neighbouring nanoparticles, where the electric field is the highest. This is in agreement to the work of Cai *et al.* [14], where non-uniform distributions of nanoparticles would aggravate the

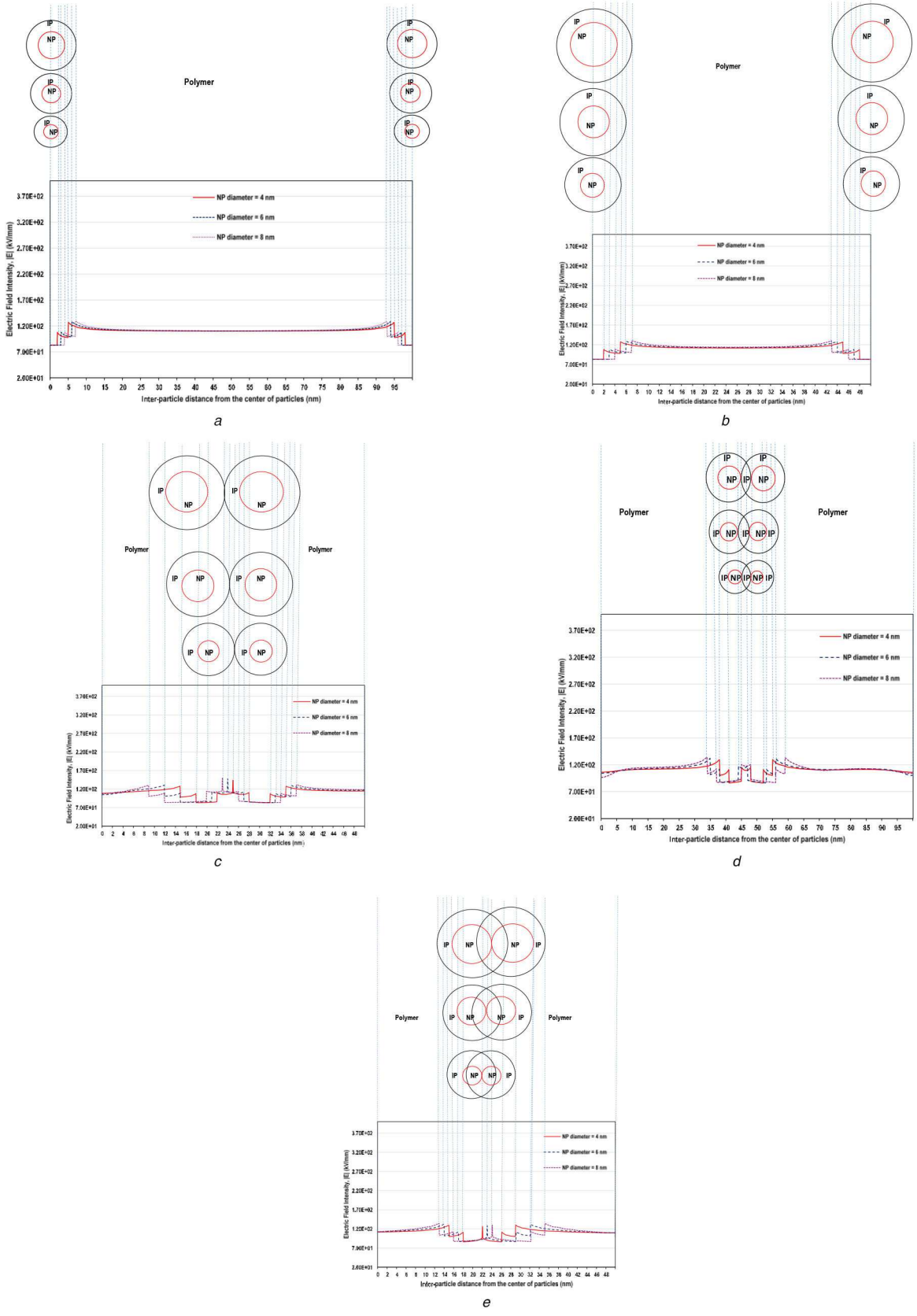

Fig. 5 *Electric field distribution versus nanoparticle size*
(a) 100 nm interparticle distance, ***(b)*** 50 nm interparticle distance, ***(c)*** Touching interphases, ***(d)*** Overlapping interphases, ***(e)*** Touching nanoparticles

concentration of local electric field, thus reducing the breakdown strength of nanodielectrics. In contrast, the electric field distribution is more uniform under homogeneous nanoparticles distributions compared to non-homogeneous nanoparticles. Since the electrical breakdown strength of a dielectric depends on the electric field distribution within the dielectric, homogeneous distributions of nanoparticles are of utmost importance in mitigating adverse electric field distributions, thus increasing breakdown strength.

From nanodielectrics point of view, an interphase is formed as an electric double layer or a Gouy-Chapman diffuse layer between a nanoparticle and a polymer, as evidenced from the electrostatic force microscopy work of Deschler *et al.* [19]. This mesoscopic phase is capable of modifying the molecular chain movement,

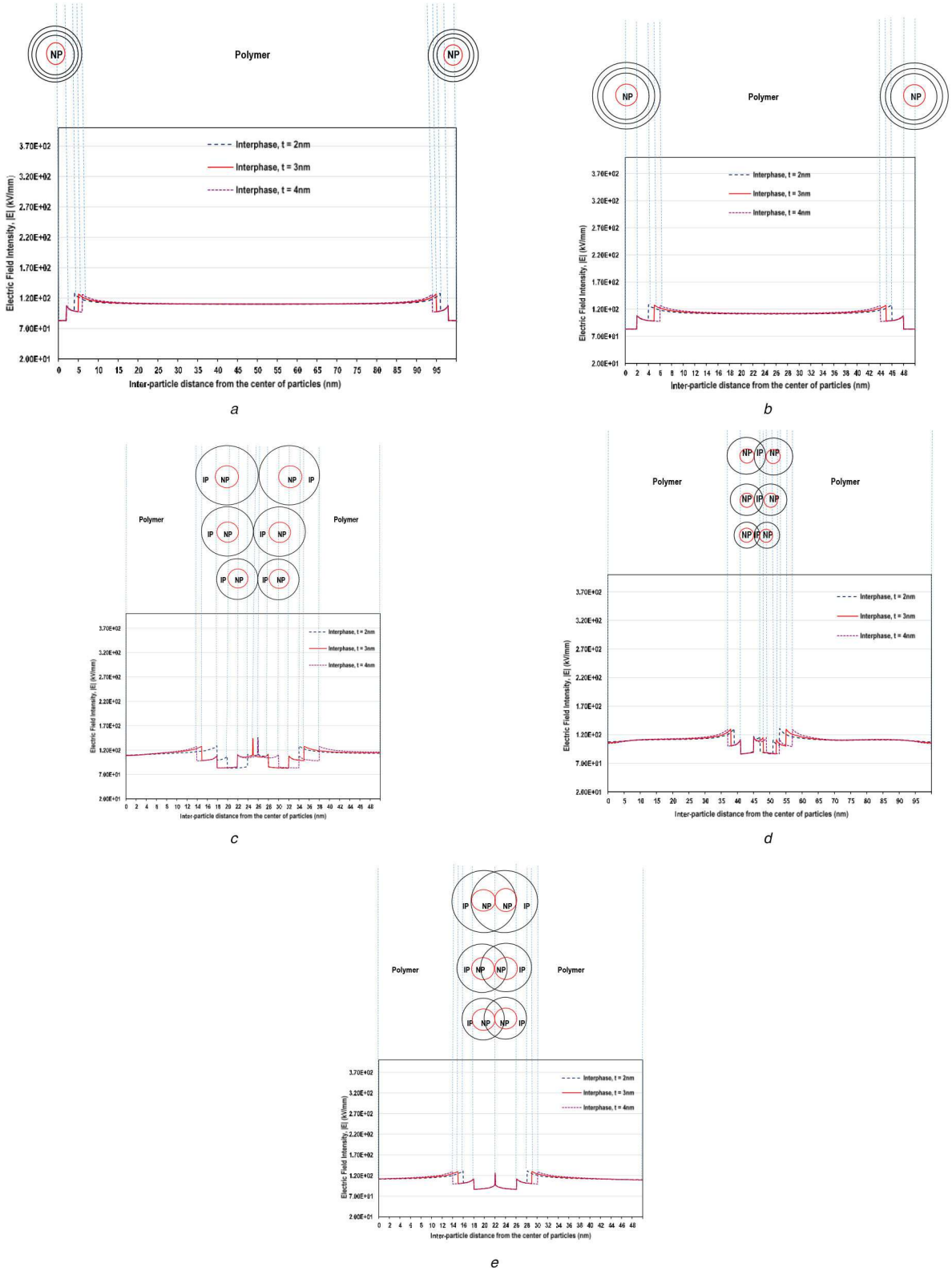

Fig. 6 *Electric field distribution versus interphase thickness*
(a) 100 nm interparticle distance, ***(b)*** 50 nm interparticle distance, ***(c)*** Touching interphases, ***(d)*** Overlapping interphases, ***(e)*** Touching nanoparticles

charge carrier mobility, trap distribution and free volume therein, thus affecting the macroscopic properties of a nanodielectric [4, 7, 8, 20]. Due to the unique bonding between the nanoparticle and polymer chain within the interphase, the interphase can therefore have permittivity values different from that of the nanoparticle and the polymer [6]. For example, the increase in the interphase permittivity can resemble the presence of water within the interphase of nanodielectrics which would otherwise reduce the breakdown strength of nanodielectrics [15]. Meanwhile, the decrease in the interphase permittivity can be as a consequence of nanoparticle surface modification that subsequently restricts polymer molecular chain movement which, if properly engineered, could lead to desirable dielectric properties [4]. Nevertheless, the current work suggests that the interphase permittivity is best to lie between that of the nanoparticle and the polymer to minimise electric field distortions, in accordance with the effective medium theory suggested by Myrochnychenko and Brosseau [21].

Meanwhile, interparticle distances are closely related to nanoparticles distributions within a nanodielectric. This is because non-homogenously distributed nanoparticles will commonly lead

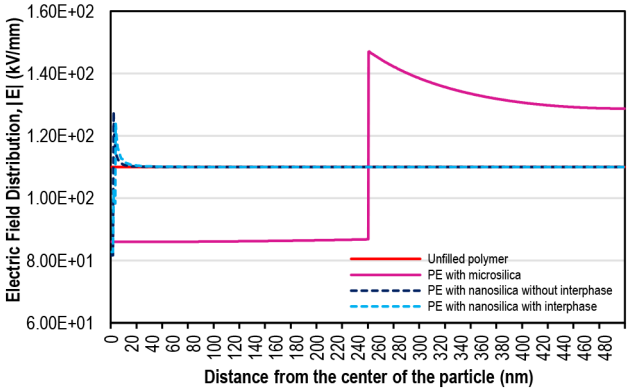

Fig. 7 *Electric field distribution within an unfilled polymer and polymer with microparticle and nanoparticle (without and with interphase)*

to reduced interparticle distances. Reduced interparticle distances, on the other hand, increases electric field intensity within nanodielectrics, as demonstrated through the current work. Li *et al.* [22] observed that, with increasing nanoparticles concentration, the interparticle distance decreases drastically while the deep trap charge density increases, resulting in greatly distorted electric field. Of note, interparticle interactions can also result in collaborative effects that affect the Coulombic barrier height with respect to electron affinity, Fermi level (electrochemical potential) and charge transport mechanisms [1]. The formation of percolation networks through the interphase will lead to lowered breakdown voltages and reduced maximum energy density of nanodielectrics [23–25]. According to Nelson [23], percolation that occurs especially when interphases touch but do not overlap each other will also result in a marked increase in conductivity. The current results demonstrating high electric field variations under touching interphases or touching nanoparticles somehow agree with the concept of percolation in affecting breakdown properties.

The current work demonstrates that larger sizes nanoparticles results in higher electric field intensities within the nanodielectric model. High electric field intensity can then lead to breakdown at an electric field far below the intrinsic strength of a dielectric. Therefore, the use of small size nanoparticles seems to be preferable over larger sizes nanoparticles. Meanwhile, increasing interphase thicknesses results in reduced electric field distortion within the nanodielectric model. This agrees with the findings of Smith *et al.* [26], where large interphase area is expected to create opportunities for increased charge scattering, a mechanism crucial for increasing the breakdown strength of nanodielectrics. Lastly, the inclusion of a microparticle and a nanoparticle in a polymer affects the electric field distribution compared to the unfilled polymer. However, the nanodielectric model shows much less distorted electric field compared to the microcomposite model. This indicates that nanodielectrics can possess enhanced breakdown strength compared to microcomposites. It is noteworthy, however, that electrical percolation network, as explained previously, can be much more influential on the properties of nanocomposites, as demonstrated elsewhere [27–29]. Although the distorted electric field within the nanodielectric model indicate potentially lowered breakdown strength of nanodielectrics compared to unfilled polymers, this largely depends upon the configurations of the nanoparticles and the interphase within nanodielectrics in creating nanodielectrics' properties distinct from that of unfilled polymers.

Of note, the electric field analysed in the current work serves as a theoretical reference to the electric field of the material and by no means represent the actual breakdown field of the material. This is because the experimental breakdown strength of a dielectric depends on many factors, including the condition of the material, the thickness of the material, the configuration of the breakdown test and other surrounding factors affecting the breakdown test [30]. It should also be noted that breakdown mechanism in a dielectric is not solely affected by the permittivity of the material constituents or the electric field distribution within the material. Other factors such as thermal and chemical effects as well as the percolation network effects need to be considered altogether. As such, the analysis may not provide a comprehensive understanding on the electric field effects within a practical nanodielectric system.

Although silica nanoparticles should ideally be spherical in size, nanoparticles can have irregular shapes in practice. However, analytical solutions for non-ideal nanoparticles are complex in three-dimensional modelling. Therefore, as a model concept, the current work considered two-dimensional modelling that allowed non-ideal, cylindrical nanoparticles to be analysed. The advantages of this two-dimensional modelling included its capability to simplify the model and reduce the calculation time. These, however, came with a trade-off, where the length of the nanodielectric and its nanoparticles had to be assumed infinite, with the nanoparticles having small radius to depth ratios. Although this would not resemble a practical nanodielectric system, available literature [31, 32] demonstrated that two-dimensional models with infinite, cylindrical constituents could provide reasonable approximations of analytical results compared to equivalent three-dimensional models with ideally spherical constituents.

5 Conclusions

In the current work, a polyethylene nanodielectric model embedded with 110 silica particles, dispersed homogeneously and non-homogeneously, was successfully modelled using FEMM 4.2 software. The results demonstrated that the distribution of nanoparticles affected the electric field intensity of the nanodielectric model. Homogeneously distributed nanoparticles resulted in less electric field distortion compared to non-homogenously distributed nanoparticles. As the interparticle distance increased, the electric field distortion reduced slightly. When the permittivity of the interphase was between that of the nanoparticles and the polymers, the electric field distribution across the nanodielectric model was less distorted. Meanwhile, the electric field distribution was also dependent upon the nanoparticle size, where smaller nanoparticles resulted in the less electric field distortions. The thickness of interphase also affected the electric field distribution, where increased interphase thicknesses reduced electric field variations. Although the current analysis is a two-dimensional representation of the electric field distribution within polyethylene/silica nanodielectrics, it has great potential to model the electric field distribution within other nanodielectric systems with reasonable approximations of analysed results. Significantly, the analysed results can be helpful towards understanding how electric field distributions within nanodielectrics affect breakdown performances of the materials.

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