Hybrid analytical finite element method for dielectric response of PE/TiO$_2$ nanodielectric materials

Bouchaib Zazoum

Department of Mechanical Engineering, College of Engineering, Prince Mohammad Bin Fahd University, PO Box 1664, Al Khobar 31952, Saudi Arabia

Department of Mechanical Engineering, ETS, University of Quebec, 1100 Notre-Dame Street West, Montreal QC, H3C 1K3, Canada

E-mail: bzazoum@pmu.edu.sa

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Abstract

An accurate three-dimensional (3D) model was developed using hybrid analytical finite element method (H-FEM) for the simulation of frequency-domain dielectric response of low-density polyethylene (PE) filled with titanium dioxide (TiO$_2$) nanoparticles. The input values of dielectric permittivity of nanoparticle and interphase were calculated analytically using the mixture model and adjusted by an optimization procedure. The effective permittivity of PE/TiO$_2$ nanocomposites was then modelled by COMSOL Multiphysics. The model output results agreement with the experimental values indicate that the developed H-FEM 3D model is suitable for use in solving dielectric response problems of different nanodielectric materials in frequency domain. Furthermore, the simulation results also offer further understanding into the effect of the nanoparticle interphase on the final dielectric properties of the nanodielectric materials.

1. Introduction

Polyethylene is extensively used as dielectric in medium and high voltage components. However, the physical properties of this material must be enhanced in order to expand its engineering use. It has been demonstrated that adding sufficiently well dispersed nano size inorganic fillers to the polymer leads not exclusively to a change of its mechanical and thermal properties, but also to improve of its dielectric properties [1–16].

The complex effective permittivity of polymeric nanocomposites is dependant upon its microstructure, the volume fraction, the shapes and type of the components, including the matrix itself, the fillers and a possible third phase known as the interphase. Several studies have shown that this interphase plays a significant role in improving the dielectric performance of nanocomposites [4, 17–22]. This is could be explained by the fact that below 100 nm the volume fraction of the interphase becomes higher as the particle size decreases. However, the exact dielectric properties of the interphase are still not clearly known in most cases.

For some particular geometry, such as periodic or laminated structures, the effective permittivity of a two-phase material can be evaluated from the analytical solution of the field distribution resulting in the various analytical models such as the generally used laws of mixtures and even for some very specific geometries in exact results. However, if the structure is disordered, as it is expected for a real compounded composite, analytical models cannot be directly used for a precise estimation of the material effective permittivity, especially if the information on both real and imaginary parts are needed (which, however, can be determined experimentally). Alternatively, numerical techniques such as finite elements method can be used to calculate the effective dielectric permittivity of composite materials and these techniques have turned to be the most efficient approach to model and predict the physical properties of two or multi-phase materials.

In this paper, numerical simulation of PE/TiO$_2$ nanodielectric were developed to predict their dielectric behavior and to help designing the nanocomposites material with optimum electrical properties for electrotechnique applications. The frequency-domain complex dielectric permittivity of PE/TiO$_2$ composites has been calculated by numerical simulation using a commercial FEM software (Comsol Multiphysics). A new
approach has been proposed to evaluate the input parameters of the FEM model, including dielectric properties of the nanoparticles and their surrounding interphases. A comparison between the numerical results and the experimental ones is also reported.

2. Experimental

Low density polyethylene powder (PE) with a density of 0.922 g cm\(^{-3}\) (Marplex, Melbourne, Australia) was mixed with TiO\(_2\) nanopowder (P25, Sigma-Aldrich, US) to prepare PE/TiO\(_2\) nanodielectric containing 3 wt% of TiO\(_2\) nanoparticles using ball milling and hot-pressing process. More details on ball milling process can be found in the literature \[23\]. For subsequent comparison, neat PE was subjected to the same process. Broadband frequency-domain dielectric spectroscopy (BDS) experiments were performed in the frequency range from 10\(^{6}\) to 10\(^{4}\) Hz at 23°C. The samples with an average thickness of 0.50 mm were sandwiched between two circular brass electrodes of 40 mm in diameter and the complex dielectric permittivity was measured by taking the average value from three points for each frequency. The measurement setup of dielectric permittivity by BDS is shown in Figure 1.

3. Analytical approaches

In analytical methods for solving single inclusion problems, the effectual properties of heterogeneous media is the way out to such problems where an electric field \(E_0\) is applied away from inclusion in z-direction \[24–34\]. For the case of spherical inclusion with radius \(R\), the solution of the Laplace equation in terms of spherical coordinates is given as:

\[
V(r, \varphi) = -E_0 r \cos \varphi - A E_0 \frac{\cos^2 \varphi}{r^2} \quad r \geq R
\]

\[
V(r, \varphi) = -E_0 r \cos \varphi + B E_0 r \cos \varphi \quad r \leq R
\]

where,

\[
A = -R^3 \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1}
\]

\[
B = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1}
\]

Here \(V(r, \varphi)\) denotes the electrical potential, \(r\) represents the radial coordinate, \(\varphi\) is the angle between the position vector and the z-axis, \(\varepsilon_1\) is permittivity of the matrix and \(\varepsilon_2\) is the permittivity of the inclusion. The electric field along the z-axis inside the inclusion according to (1) is given by

\[
e_1 E_0 = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1}
\]
\[ E_2 = \left( 1 + \frac{1}{3} \frac{\epsilon_2 - \epsilon_1}{\epsilon_1} \right)^{-1} E_o \]  

(2)

For a general case of ellipsoidal inclusion, analogous calculations give

\[ E_2 = \left( 1 + A_1 \frac{\epsilon_2 - \epsilon_1}{\epsilon_1} \right)^{-1} E_o \]  

(3)

Here \( A_1 \) is the depolarization factor in the direction of ellipsoid principal axis and parallel to electrical field \( E_o \). According to definition, for a two-component heterogeneous linear material the effective dielectric permittivity given as

\[ \epsilon_c = \frac{q_1 \epsilon_1 \langle E_1 \rangle + q_2 \epsilon_2 \langle E_2 \rangle}{E} \]  

(4)

where \( \epsilon_c \) denotes effective permittivity, \( q_1 \) is volume fraction of matrix, \( q_2 \) the volume fractions of the inclusion and brackets \( \langle \rangle \) represents an average over phase 1, 2 or over the volume of material. In composite materials the analytical calculation of electric field is possible only when the minority phase exists in regular shape inclusions and in minor concentration. Eventually dealing with several matrix systems and periodic regular inclusion arrangements, the exact solution to these matrixes can be calculated. For instance, by assuming \( \langle E_i \rangle = \langle E_j \rangle = \langle E \rangle \) in (4), the equation gives the mixture model.

\[ \epsilon_c = q_1 \epsilon_1 + q_2 \epsilon_2 \]  

(5)

So, this gives the exact solution for a laminated structure that is applied parallel to the electrical field. The solution of single inclusion problem equation (3) can likely be used for a dilute suspension having a permittivity \( \epsilon_2 \) in a continuum matrix of permittivity \( \epsilon_1 \) and ellipsoidal shape inclusion. While dealing with such cases it is assumed that electrical field \( (E_o) \) and average field in matrix are equivalent, this gives

\[ \epsilon_c = \epsilon_1(1 - q_2)(1 - A_1) + \epsilon_2[q_2 + A_1(1 - q_2)] \]  

(6)

For inclusions that are randomly oriented, the similar method leads to

\[ \epsilon_c = \epsilon_1(1 - q_2) + \frac{\epsilon_2 q_2}{3} \sum_{i=1}^{A_1} \frac{\alpha_i}{(\epsilon_1 + \epsilon_2 - \epsilon_1 A_i)} \]  

(7)

In above equation \( A_i \) denotes the depolarization factor of the ellipsoid for the \( i \)-th-axis. While, for spherical particles since \( A_1 = A_2 = A_3 = 1/3 \) as a result equations (7) and (6) are identical. The analytical expressions for depolarization factor \( A_i \) for spheroids \( (a = b \neq c) \), oblate spheroids \( (a = b > c) \) and prolate spheroids \( (a = b < c) \) can be found in literature.

Additionally, another method known as effective medium approximation also depends on the solution of single inclusion boundary value problem. Generalizing the Maxwell approximation for ellipsoidal inclusion having two-phase composite, comprising of matrix (phase 1) containing a perfectly orientated ellipsoidal inclusion (phase 2) gives:

\[ \sum_{j=1}^{2} q_j (\epsilon_c - \epsilon_j) \left[ 1 + A_1 \frac{\epsilon_j - \epsilon_1}{\epsilon_1} \right]^{-1} = 0 \]  

(8)

It can further be verified that equations (6) and (8) are analogous to each other, in case of randomly orientated ellipsoidal inclusion it would be analogous to (7) [38].

This equation also represents the exact solution of the coated spheres model. Moreover, equation (8) represents the multi component composites having various kinds of ellipsoidal inclusions.

In the family of effective medium approximation, Bruggeman introduced self-consistent approximation [39] that leads to minor changes in (8)

\[ \sum_{j=1}^{2} (\epsilon_c - \epsilon_j) \left[ 1 + A_1 \frac{\epsilon_j - \epsilon_1}{\epsilon_c} \right]^{-1} = 0 \]  

(9)

and aiding to quadratic equation for the effective permittivity. For randomly oriented ellipsoidal, equation (9) is written as [40].

\[ \sum_{j=1}^{3} q_j (\epsilon_1 - \epsilon_c) + \frac{q_2 (\epsilon_2 - \epsilon_c)}{A_1 (\epsilon_2 - \epsilon_c) + \epsilon_c} = 0 \]  

(10)
Contrary to (8), each phase in self-consistent approximation equations (9), (10) is treated symmetrically and is not affected if indexes are interchanged simultaneously. Mostly minority phases are embedded in majority phase (known as matrix) so practically phases are not symmetrical. Lastly with the implementation of a

Figure 2. Unit cell 3D model of PE/TiO$_2$ nanodielectric, containing 3 wt% of TiO$_2$ nanoparticles.

Figure 3. Measured real (a) and imaginary (b) part of the complex dielectric permittivity versus frequency for the PE and PE/TiO$_2$. 
symmetric integration technique [41] and irrespective of the shape of ellipsoids the Looyenga equation for randomly oriented ellipsoids is given as

$$\varepsilon_c^{1/3} = q_1\varepsilon_1^{1/3} + q_2\varepsilon_2^{1/3}$$  \hspace{1cm} (11)

For steady-state AC conditions, the electric fields and permittivity in the above equations are substituted by their respective phasors and complex representations. Consequently, for the prediction of a composite dielectric response equations (6)–(11) can be implemented.

4. Numerical model description

When the electric field vector present inside the material is well-known, the effective permittivity of nanodielectric can be calculated. For an entirely electrostatic case, it can be calculated by solving the Poisson’s equation given as:

$$\nabla \cdot (\varepsilon_r \varepsilon_0 \nabla V) = -\rho$$  \hspace{1cm} (12)

where $\varepsilon_r$ and $\varepsilon_0$ are relative permittivity and vacuum permittivity respectively, $V$ is electrical potential and $\rho$ presents charge density. When $\rho = 0$ (neutral condition) and possible conductivity $\sigma$ and dielectric losses are considered, (12) can generally be written as [42–44]:

$$\nabla \cdot [j\omega(\varepsilon_r \varepsilon_0 \nabla V)] = 0$$  \hspace{1cm} (13)

Here $\varepsilon_r$ (the complex permittivity) is given by

$$\varepsilon_r = \varepsilon_r' - j \left( \frac{\varepsilon_r''}{\omega\varepsilon_0} \right)$$  \hspace{1cm} (14)

In above equation (14) $\varepsilon_r''$ is the involvement to the imaginary portion because of dielectric relaxation. When the properties of each phase and material microstructure are familiar, we can numerically solve (12) and (13) with the help of finite elements method (FEM). After the numerical evaluation of field distribution, the relative permittivity ($\varepsilon_r'$) and dielectric losses ($\varepsilon_r''$) can be calculated by several equivalent methods. One such method is the calculation of total current and complex impedance.

$$\varepsilon_r' = \left( \frac{-Z''}{2\pi f\varepsilon_0([Z']^2 + [Z'']^2)} \right) \cdot \frac{d}{S}$$  \hspace{1cm} (15)

$$\varepsilon_r'' = \left( \frac{Z'}{2\pi f\varepsilon_0([Z']^2 + [Z'']^2)} \right) \cdot \frac{d}{S}$$  \hspace{1cm} (16)

where $Z'$ and $Z''$ are the real and imaginary part of the complex impedance $Z$, while $d$ represents the distance between the two electrodes, $f$ is the frequency, $S$ is the area of the plate and $\varepsilon_r''$ is the imaginary part of equation (14), including the contribution of direct conductivity.
As a simplified approach, 27 particles with weight fraction of 3 wt% are considered randomly distributed in a block of polyethylene matrix. The inclusions were assumed to have spherical geometry with a diameter of 50 nm and each nanoparticle has an interphase of thickness of 10 nm. A Matlab script was created to generate the random non-overlapping nanoparticles in a unit cubical domain based on dynamic collision algorithm (DCA) as shown in figure 2. The top face of cube was fixed to a constant potential of 1 V and the opposed face was set to ground (V = 0), while the other faces of the cube were set to the periodic conditions.

In the first approximation, the interphase was considered has the same value as the nanoparticles. The numerical results of the real and imaginary part of the effective permittivity obtained with the developed 3D model were compared with experimental measurement from BDS and then, the values for the permittivity of interphase will be adjusted to match well the numerical results with the experimental values.

5. Results and discussion

5.1. Dielectric spectroscopy

Figure 3 presents the dielectric response of PE and PE/TiO₂ nanocomposites over the 10³ to 10⁴ Hz frequency range at room temperature (23 °C). From figure 3(a), it can be observed that over the 10³ to 10⁴ Hz frequency range, the relative dielectric permittivity (ε’) of PE/TiO₂ nanocomposites is higher as compared to that of PE. At high frequencies (higher than the relaxation frequency shown in figure 3(b)), this can be attributed to the fact, that the ε’ of TiO₂ is much higher than the ε’ of PE. The additional increase in the permittivity at lower frequencies is related to the relaxation mechanism shown in figure 3(b). In the case of dielectric losses (ε’’), it can

![Figure 3](image-url)
be seen from figure 3(b) that for non-polar PE no relaxation peak can be observed. However, for PE/TiO2, one relaxation peak was observed in the vicinity of 10 Hz. This behavior is known as the Maxwell-Wagner-Sillars polarization, which is mostly related to a difference in conductivity between the two phases [45].

5.2. Model validation
In this 3D model, the dielectric permittivity of polyethylene matrix and that of the TiO2 nanoparticles were frequency dependent, while the conductivities were assumed constants. The electrical conductivity of PE was fixed to $\sigma_1 = 10^{-15}$ $\Omega^{-1}$m$^{-1}$, as measured by Keithley 6517 electrometer and those of the nanoparticle, $\sigma_2$ and interphase, $\sigma_3$ were chosen to be between $10^{-8}$ and $10^{-10}$ $\Omega^{-1}$m$^{-1}$, as often found in the literature. The real, $\varepsilon'_i$ and imaginary, $\varepsilon''_i$ parts of the complex relative permittivity, $\varepsilon_i$ assumed to PE matrix are obtained from BDS measurement. However, the real, $\varepsilon'_2$ and imaginary, $\varepsilon''_2$ parts of the complex dielectric permittivity of the nanoparticles, $\varepsilon_2$ were calculated separately by using the above mixture’s formula (equation (5)).

In the first estimation, the complex relative permittivity of the interphase $\varepsilon_3$ was considered has the same value as the nanoparticles,

$$\varepsilon_3 = \varepsilon'_3 - j\varepsilon''_3 = \varepsilon'_2 - j\varepsilon''_2$$ (17)

where $\varepsilon'_3$ and $\varepsilon''_3$ are the real and imaginary parts of the complex relative dielectric permittivity of the nanoparticle.
The surface plots of the electric field distribution in the 3D model were obtained by FEM simulations and are shown in figure 4. As it can be seen, there is a field enhancement at the matrix–nanoparticle interface in the z-direction, and the field is almost constant and smaller within the inclusions. This is due the fact that the two inhomogeneous phases (polymer and nanoparticle) have different permittivities.

Figure 5 shows simulation and experimental results about the effective real and imaginary part of PE/TiO₂ nanodielectric. It appears that at low frequency the real relative dielectric permittivity predicted by the developed H-FEM model agree well with the experimental data obtained by BDS. However, at high frequency once noticed that the simulated values are lower than those experimentally measured. In the case of dielectric loss permittivity, a good agreement can be observed between the experimental and simulated results in terms of profile shape, but the experimental values are higher than that the numerical ones.

To reduce errors and optimise the validity of this H-FEM model, the imaginary part of the dielectric permittivity of the interphase that was previously calculated by mixture model, is scaled with a factor of 2.3.

Figure 6 shows a comparison of experimental results and the numerical values predicted by the corrected H-FEM model with the adjusted complex dielectric permittivity of the interphase which can be written in this case as

\[ \tilde{\varepsilon}_3 = \varepsilon'_3 - 2.3j\varepsilon''_3 \]  

It can be noticed that the numerical values for both real and imaginary parts of the effective relative dielectric permittivity of the nanodielectric now agree well with those obtained experimentally by BDS. These results indicate that the corrected model is reliable and confirm the validity of our simulation procedure.

6. Conclusion

This paper presents H-FEM 3D model for predicting the dielectric properties such as electrical field distribution and dielectric permittivity of PE/TiO₂ nanodielectric materials in frequency domain. The model was built based on FEM combined with the experimental BDS measurement and mixture model. The numerical results supported by experimental data obtained by BDS indicate the validity of the developed 3D model and reliability of the proposed strategy to calculate the dielectric permittivity of nanoparticle and interphase by using the adjusted mixture model. Finally, this numerical model can be extended to design nanocomposites materials with optimum dielectric properties for electrotechnical applications.

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ORCID iDs

Bouchaib Zazoum https://orcid.org/0000-0001-7641-5809

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