STUDY OF THE FACTORS THAT INFLUENCE THE EFECTIVE PERMITTIVITY OF THE DIELECTRIC MIXTURES

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The effective permittivity of dielectric mixtures is influenced by important factors as: the shape and geometrical size of inclusions, their orientation with respect to the applied electric field, their space distribution and concentration in the mixture. This paper analysis these influences. One of the methods that can be success fully used in order to compute this permittivity is the numerical modeling of the mixture, using the finite element method (FEM). This method also allows us to consider the above mentioned influence factors. The numerical results obtained are compared to those computed analytically and those determined experimentally.

1. INTRODUCTION

Many of today's electrical insulators are mixtures of two or several materials. One can notice the increased use of composite materials with outstanding mechanical properties (a unique combination of low density but high mechanical resistance). Extensive studies were conducted regarding mechanical properties and manufacturing technologies for these materials, but less importance was given to their electrical properties.

Nowadays, polymer based composites (cheaper and less polluting) replace traditional insulating materials (oil, paper, glass, ceramics, etc.) in insulation systems. Many researchers [1, 2, 3, 4, 9, 10] focused on finding new materials, more reliable for electrical stresses but less expensive. An important goal is to increase the voltage of electrical power lines, and therefore decreasing power loss, by using these new materials.

The main advantage of composites consists of the possibility of designing them for special purposes. Since the classical approach of experimental trail requires time and money, making the improvement of the initial model quite difficult, the use of the computer in the design phase represents a true step forward

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(because an accurate estimation and correction of the required properties is easy to achieve).

Estimating the effective (real) permittivity ε_{ef} of dielectric mixture, knowing the permittivities of the components and their concentration in the mixture, is very useful in designing non-homogenous dielectrics that need to have the characteristics required by a certain application. The computation of this permittivity becomes quite difficult if it's also necessary to estimate how this parameter is influenced by various factors.

The numerical modeling, based on the finite element method (FEM), is one of the methods that can be used in order to determine this permittivity, allowing us to also consider some of the influence factors mentioned in the literature [5, 6, 12, 14].

This paper presents models of some binary mixtures made from a host media with relative permittivity ε_1 and inclusions with relative permittivity ε_2. These systems are described by the volume fraction of the inclusions, q (the ratio between the volume of the inclusions and the entire volume of the mixture). The inclusions are considered to be infinitely long cylinders or prisms with different shapes of the cross-section (ellipse, square, hexagon, etc.). Therefore, a 2D modeling of the material is quite suitable for numerical computation of the effective relative permittivity of binary mixtures.

We performed a systematic study on the influence of concentration, shape, geometrical size, space distribution and orientation of the inclusions with respect to the direction of the applied electric field. This study is conducted first on simple models with a single inclusion of different shapes, at the same concentration, and then on models of statistics or matrix mixtures.

For the experimental study of the glass/polymer mixture we poured 5 samples of each mixture of epoxy resin ($\varepsilon_1 = 4$) and glass balls ($\varepsilon_2 = 6.52$) with the same volume fraction of the glass inclusions. The samples, shaped as discs with 100 mm diameter and depths between 2 and 2.5 mm, were polished and silver was deposited on the surfaces to ensure a better electrical contact [5, 8] with the electrodes of the measurement capacitor. The permittivity measurements were performed with a low voltage Schering bridge of type TR 9701 with a precision of 3.5% for a frequency of 100 Hz.

The numerical results are compared with those computed analytically and those experimentally measured on glass / polymer probes.

2. THEORETICAL BACKGROUND

For the analytical computation of the effective permittivity of statistical or matrix mixtures, one can use formulas proposed by various researchers [6, 7, 8, 11, 12, 14]. The numerical computation can be performed using methods based on the finite element (FEM).

2.1. ANALYTICAL RELATIONS FOR COMPUTING THE MIXTURE'S EFFECTIVE PERMITTIVITY

In order to determine the effective permittivity of dielectric mixtures having highly different permittivities, a few calculus relationships are indicated because they yield values that are closer to those experimentally determined [1, 3, 5, 11]. We present here those that we have used for a comparative analysis with the numerical results.

For statistical mixtures the following relationships were used:

$$\log \varepsilon_{ef} = \sum_i q_i \log \varepsilon_i \text{ – Lichtenecker – Rother – logarithmical law of mixtures;} \quad (1)$$

$$\varepsilon_{ef} = A + \sqrt{A^2 + \frac{\varepsilon_1 \varepsilon_2}{2}} \text{ with } A = \tfrac{1}{4}\left[(3q_1 - 1)\varepsilon_1 + (3q_2 - 1)\varepsilon_2\right] \text{ – Odelevsky;} \quad (2)$$

$$\varepsilon_{ef} = \varepsilon_1 \frac{2\varepsilon_1 + \varepsilon_2 + 2q(\varepsilon_2 - \varepsilon_1)}{2\varepsilon_1 + \varepsilon_2 - q(\varepsilon_2 - \varepsilon_1)} \text{ – Maxwell-Wagner;} \quad (3)$$

$$\sqrt{\varepsilon_{ef}} = \sum_i q_i \sqrt{\varepsilon_i} \text{ – Beer;} \quad (4)$$

and for the matrix mixtures:

$$\frac{\varepsilon_2 - \varepsilon_{ef}}{\varepsilon_2 - \varepsilon_1} = (1 - q)\sqrt{\frac{\varepsilon_{ef}}{\varepsilon_1}} \text{ – Bruggerman;} \quad (5)$$

$$\varepsilon_{ef} = \varepsilon_1 \left[1 + \frac{mq(\varepsilon_2 - \varepsilon_1)}{m\varepsilon_1 + (\varepsilon_2 - \varepsilon_1)(1 - q)}\right] \text{ – Sillars.} \quad (6)$$

In the formulas above, q, q_i represent the volume fraction of the inclusions, respectively of the i component of the mixture, ε_{ef} – the effective relative permittivity of the mixture, ε_1, ε_2 – the relative permittivity of the host media, respectively of the inclusions, and m is a shape parameter equal to 3 or 6 for spherical and ellipsoidal inclusions, respectively.

2.2. NUMERICAL APPROACH

The effective permittivity was computed for an electrostatic regime, using a FEM based software developed by the ANSOFT Corporation [5, 6, 7]. Considering the geometrical properties of our model (described in the introduction) a 2D simulation of the composite is quite suitable. The key parameter, which should be obtained for calculation of the dielectric properties of a mixture, is the electric field distribution $E(x,y)$ within a domain. The equation solved by the software program for all electrostatic problems is known as Gauss' law [5, 14]:

$$\nabla \cdot \left(\varepsilon_r \varepsilon_0 E(x,y)\right) = \rho(x,y), \tag{7}$$

where ε_r, ε_0 represent the relative permittivity and the permittivity of vacuum and $\rho(x,y)$ represents the charge density. In our case, the free charge is located only on the plates of the capacitor that represents the computation domain. Therefore, considering there is no free charge inside the domain, the equation solved by the computer in this case is Laplace equation with both Newton and Dirichlet conditions (Fig. 1).

The average permittivity of a composite can be computed using one of the following two methods [13]:

a) The first method (M1) requires the calculation of the average electric field $\widetilde{E}$ and electric flux density $\widetilde{D}$ over the entire computational domain, with:

$$\widetilde{E} = \frac{1}{\Omega}\int_{\Omega} E \,\mathrm{d}\,\Omega \text{ and } \widetilde{D} = \frac{1}{\Omega}\int_{\Omega} D \,\mathrm{d}\,\Omega \,. \tag{8}$$

The effective permittivity is computed with the formula:

$$\varepsilon_{ef} = \widetilde{D} / \varepsilon_0 \widetilde{E} \,. \tag{9}$$

b) The second method (M2) requires the calculation of the electrostatic energy over the computational domain, and the permittivity is obtained from:

$$\frac{1}{2}\varepsilon_{ef}\varepsilon_0 \frac{1}{\Omega}\int_{\Omega} E^2 \,\mathrm{d}\,\Omega = \frac{1}{\Omega}\int_{\Omega}\frac{1}{2} E \cdot D \cdot \mathrm{d}\,\Omega \,. \tag{10}$$

The two methods mentioned above led to the results that will be presented in the following section of the paper.

3. **GEOMETRICAL MODEL**

The boundary conditions assigned to the models are presented in Fig. 1 (the material was considered to form the dielectric of a plane capacitor, having a constant and uniform internal electric field). The computation domain was considered to be a square of 20 μm side.

For a systematic study of the influence of the shape and size of the inclusions on the effective permittivity ε_{ef} of a binary mixture, we first imagined simple models with a single polygonal inclusion, with a different number of sides (n = 3, 4, … ∝), inside a host media (square) – (Fig. 2). For ellipsoid inclusions, we presented the influence of the orientation of the ellipse with respect to the applied electric field over the effective permittivity (β is the angle between the long semi-axis of the ellipse and the direction of the field – Fig. 1).

For the 2D modeling of matrix mixtures, ordered structures were considered, having the same matrix of the host material (16 x 16 elements in a square with 20μm side) but with different shapes (cylindrical and ellipsoidal) of the inclusions. Fragments of the modeled structrures are given in Fig. 3.

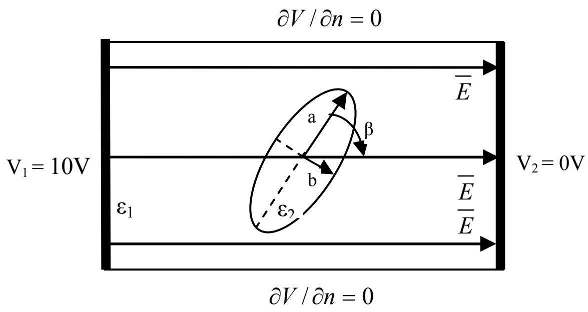


Fig. 1 – Boundary conditions assigned to the model.

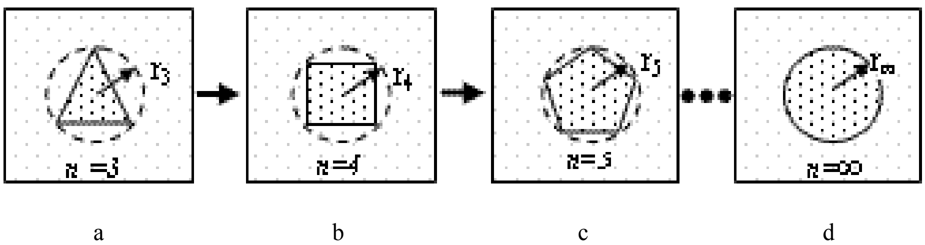
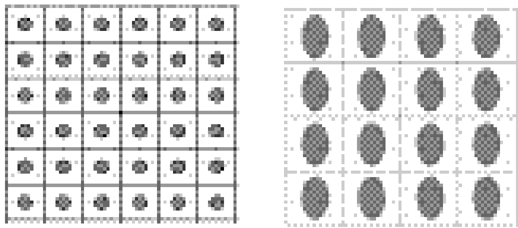


Fig. 2 – Dielectric models with polygonal inclusions, with a different number of sides: a) $n = 3$; b) $n = 4$; c) $n = 5$; d) $n = \infty$.

The models considered for statistic structures with different volume fractions q of the cylindrical inclusions are represented in Fig. 4. For the same volume fraction of the inclusions, q = 0.098, we considered square (Fig. 5) or triangular (Fig. 6) inclusions of different sizes and with a random space distribution.



a b

Fig. 3 – Fragments of modeled matrix mixtures with different shapes of the inclusions: a – cylinders; b – ellipsoids.

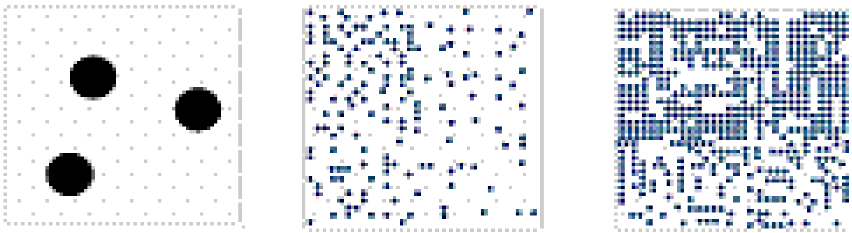


Fig. 4 – Fragment from modeled statistical structures with different values of the volume fraction of the inclusions.

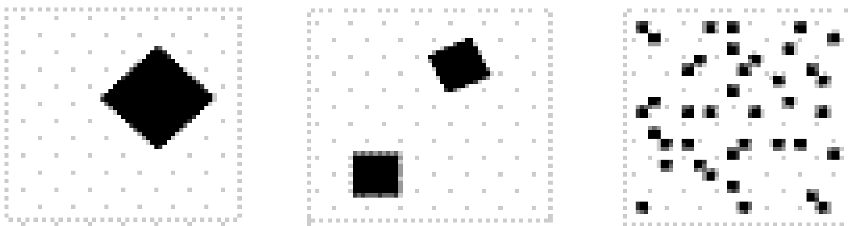


Fig. 5 – Fragment from modeled statistical structures with different values of the square's side (that represents the inclusion), for the same volume fraction $q = 0{,}098$.

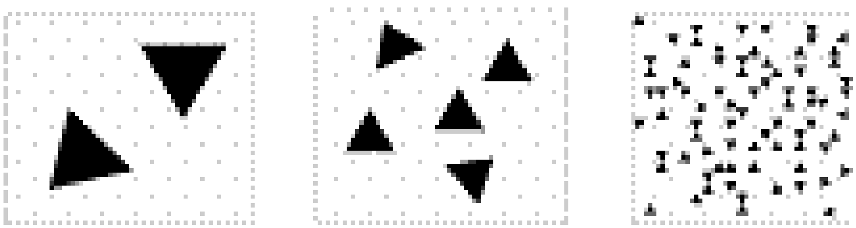


Fig. 6 – Fragment from modeled statistical structures with triangular inclusions of different sizes, for the same volume fraction $q = 0{,}098$.

4. **RESULTS AND DISCUSSIONS**

The results obtained by numerical modeling of simple structures, with a single polygonal inclusion (with a different number of sides and relative permittivity $\varepsilon_2 = 6.5$) placed inside a square (host media) with permittivity $\varepsilon_1 = 4$, for a volume fraction $q = 0.2$, are given in Table 1.

Table 1

Effective permittivity of mixtures with simple polygonal inclusions, $q = 0.2$

No. of sides of the inclusion	3	4	6	10	20	70	99	∞
M1	4.39884	4.39697	4.39457	4.39428	4.3943	4.39513	4.39510	4.39510
M2	4.30995	4.30891	4.30603	4.30525	4.3048	4.30569	4.30567	4.30564

One can see that the permittivity decreases by increasing the number of sides of the polygonal inclusion, and for a sufficiently large number of sides this one remains practically constant. The differences between the values of ε_{ef} for $n = 3$ and $n \to \infty$ for this volume fraction ($q = 0.2$) are small (0.937 % for M1 and 1.39 % for M2). Another ordered structure studied is the one with ellipse inclusions.

The results regarding the influence of the direction of the applied electric field on the relative effective permittivity are presented graphically in Fig. 7, where eps1 represents the permittivity computed with method M1, while eps2 is computed with M2. An ordered matrix mixture with ellipse inclusions was modeled, with a volume fraction of the inclusions of $q = 0.3$.We present the variation of the relative effective permittivity with respect to the orientation angle β (β represents the angle between the direction of the electric field vector and the longest semi-axis of the ellipse, *a*).

In order to study the influence of the geometrical sizes of the inclusions on the effective permittivity, we first considered the simple models of Figs. 2 and 1, with cylindrical inclusions, respectively ellipsoidal ones, with $\varepsilon_2 = 6.5$, of different sizes inside a host media with $\varepsilon_1 = 4$, which is a square of variable side, computed with formula $l = r\sqrt{\pi / q}$ for cylindrical inclusions, and $l = \sqrt{\pi a b / q}$ for ellipsoidal ones.

For statistical mixtures of the same volume fraction of the inclusions ($q = 0.098$) and different shapes (circle, square or triangle) and dimensions, we considered permittivity $\varepsilon_2 = 60$ inside a host media of permittivity $\varepsilon_1 = 2.2$ (mixture polyester/rutilus). The numerical results are given in Table 2 and Table 3.

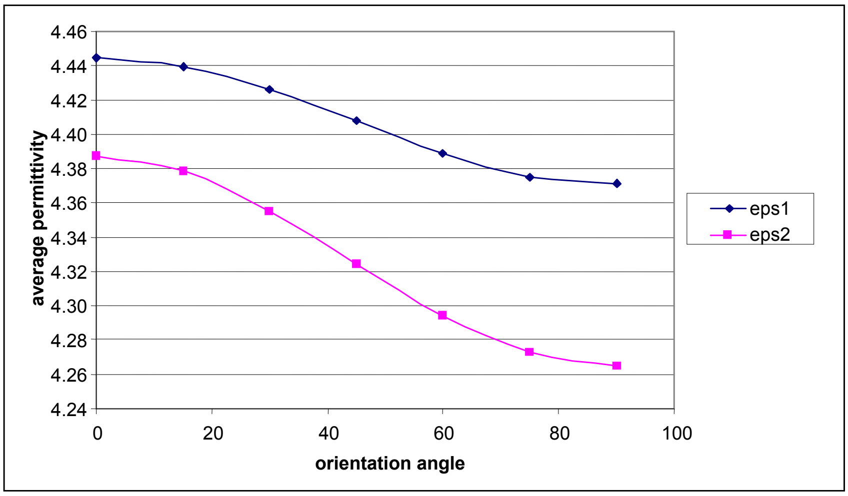


Fig. 7 – Relative effective permittivity of the mixture versus orientation angle of the ellipse inclusions (volume fraction is $q = 0.3$).

Table 2

The effective permittivity of simple models with inclusions of different dimensions, $q = 0.2$

Radius/ long axis µm	1	4	10	100	4	5	6	10	40
M1	4.3947	4.3949	4.3969	4.4041	4.0884	4.0884	4.0865	4.0962	4.1776
M2	4.3047	4.3053	4.3091	4.3098	4.0644	4.0626	4.0593	4.0645	4.1139
	Cylindrical inclusions				Ellipsoidal inclusions with short axis of 3 µm				

Table 3

Variation of the effective permittivity ε_{ef} of statistical mixtures as a function of inclusions size

Shape of inclusion	Circle's radius or square's side / triangle's side [µm]							
	10^{-3}	$2 \cdot 10^{-3}$	$5 \cdot 10^{-3}$	10^{-2}	63	100	400	700
	Effective permittivity of mixtures with different shapes and dimensions of the inclusions							
circle	-	4.033	4.065	-	-	-	4.43977	4.43929
square	4.299	4.350	4.395	4.342	4.44907	4.45888	4.469896	4.51487
triangle	-	4.373	4.403	4.403	4.45180	4.452974	4.46269	4.49806

One can observe that the effective permittivity increases along with the increase of circle's radius or the long axis of the ellipse. This rise is much more significant for ellipsoidal inclusions: 2.13 % for a 25 times augmentation of the ellipse area, compared to only 0.21 % rise for a 100 times augmentation of the circle's area. For statistical mixtures too, one care observe an increase of the permittivity for larger inclusions, differences that are due to inclusions (square or triangle) orientation with respect to the direction of the electric field. Numerical results obtained for the effective permittivity of statistical and matrix mixtures of various volume fraction q_i (with the permittivity of the host media ε_1 = 4 and the inclusion ε_2 = 6.52) are compared to those computed using existing analytical formulas and to those measured experimentally – Table 4.

Table 4

Effective permittivity for mixtures with cylindrical inclusions

q	Maxwell Wagner	Lichte-necker Rother	Beer	Odelev-schi	M1	M2	Exp.	q	Error Brugger-man-M1	Sillars	M1	M2
0.039	4.0817	4.0763	4.0867	4.0819	4.0778	4.0624	4.073	0.1	-0.179	4.1859	4.1946	4.1510
0.078	4.1646	4.1548	4.1745	4.1653	4.1555	4.1237	4.141	0.2	-0.154	4.0587	4.3977	4.3127
0.117	4.2487	4.2347	4.2631	4.2502	4.2352	4.1885	4.182	0.3	-0.1297	4.3386	4.6114	4.4892
0.156	4,3339	4.3162	4.3527	4.3366	4.3159	4.2544	4.217	0.4	-0.106	4.6257	4.8367	4.6836
0.234	4.5079	4.4838	4.5347	4.5135	4.4846	4.3979	4,405	0.5	-0.083	4.9205	5.0752	4.9002
0.312	4.6869	4.6580	4.7204	4.6961	4.6659	4.5545	4.581	0.7	-0.0435	55341	5.5999	5.4329
Statististical mixtures								Matrix mixtures				

Analyzing data in Table 4 one observes that for statistical mixtures the experimental values are closer to analytical data computed with Lichtenecker-Rother, and to the numerical ones computed with M1. For matrix mixtures, the best concordance is between Sillars analytical results and numerical values computed with M2.

For statistic mixture, one can also see that the relative error between values computed with M1 and Lichtenecker-Rother formula decreases as the volume fraction increases, up to q = 0.156. The relative error between the numerical methods M1 and M2, but also between M1 and experimental values are rising as the volume fraction increases, but the maximum error remains below 2.5 %.

For matrix mixtures, the relative error between M1 and Silars increases with the volume fraction up to q = 0.2, but then decreases for higher values of q.

For statistical mixtures, the values of the effective permittivity, computed numerically using FEM (with both methods M1 and M2), are quite close to those measured experimentally. The differences that one can notice are explained mainly by the numerical computation errors (due to the mesh of the computation domain or the number of steps taken for solving the system of equation). Some experimental hazards can also be considered, such as a poor accuracy of preparing the mixture samples (including clusters of impurities or air inclusions in the dielectric mixture, etc.).

5. **CONCLUSIONS**

One can conclude that in binary dielectric mixtures, the influence factors considered (shape, size and inclusions orientation in the electric field) do not change significantly the values of the effective permittivity. Among the numerical computation methods, M1 gives results in good agreement with analytical ones. For statistical mixtures, the permittivities computed with Lichtenecker-Rother`s formula are close to those computed with the method M1 and those measured on glass/epoxy resin mixtures. For matrix mixtures, the values of the effective permittivity computed with Sillars formula agree with those computed by method M2.

For statistic mixtures, the relative errors between numerical data (M1) and analytical values computed with Lichtenecker-Rother formula do not exceed 0.2 %. For matrix mixtures, the error between M1 and Silars is larger, reaching a maximum for q = 0.2 (7.708 %).

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