Calculation of dielectric constant of two-phase disordered composites by using FEM

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Abstract- A simulation for dielectric constant of two-phase disordered composites based on a three-dimensional disordered model is presented. In this model, the inclusions can be arranged at random positions in a composite material that is simulated by 10×10×10 cubic cells. Under the quasi-static approximation, numerical calculations are performed using the finite element method (FEM). In order to get the effective dielectric properties of isotropic composites, an averaging method is adopted because each topological structure of a given disordered model is anisotropic. Then the effect of the permittivity contrast between the inclusions and the matrix phase of composites is studied. The numerical results obtained by the disordered model are also compared with those obtained by the classical mixing rules and the conventional periodic models.

I. INTRODUCTION

Composite materials have been widely used for electrical, mechanical, optical applications due to their excellent properties [1]. Thus, it is of great importance to calculate their dielectric properties in good precision before production. The dielectric properties of mixtures have attracted many attentions in several decades, and a number of researches have been carried out on the calculation of dielectric properties of composite materials. L.C.Shen *et al.* [2] investigated the static conductivity and dielectric constant of two-component periodic composite material by the Fourier expansion technique. B.Sareni *et al.* presented a periodic simulation model [3] and a random model [4] to calculate the effective dielectric constant of composite materials based on the boundary integral equation method. In recent years, E.Tuncer *et al.* [5-7] have published a series of articles on the modeling of the complex dielectric constant based on the finite element method (FEM). Most of the previous calculations are based on the model of regular distribution of inclusions in a composite (periodic model), which is not realistic. Although, the information obtained from calculations based on the periodic model provides an interesting study and is helpful to predict the dielectric properties of composites, random distributions should be more realistic since the distribution within real composites is normally highly irregular. Several researchers have investigated the calculation of the disordered composites. There are several studies considering the random positions of inclusions.

The aim of this paper is to report a numerical three-dimensional calculation for dielectric constant of two-phase disordered composites, in which the inclusions and matrix phase with cubic shapes occupy positions randomly in a big cube consisting of 10×10×10 cubic cells. The dielectric constant of inclusions is ε_1 and the dielectric constant of host material is ε_2. Both materials are homogenous, lossless, and isotropic. The interaction of inclusions plays an important role when the concentration of inclusions is high. When the concentration of inclusions is increased, there appear overlapping inclusions. In this model, the cubic cells of inclusions can be overlapped irregularly, thus there will be many types of interconnect inclusions to simulate the realistic composites at a given volume ratio, especially at high concentration of inclusions. The FEM is applied to calculate the electric filed distribution in the composite material and the effective dielectric constant of composites is derived. The effective permittivity as a function of the volume fraction of inclusions is also calculated by this method. The dependence of the orientation of disordered materials and the effect of permittivity contrast are investigated in our simulation. The calculated dielectric properties are also compared with the classic mixture rules and previous periodic models.

II. CALCULATION METHOD

A. Preconditions for calculation

In our physical model, there are two preconditions for the composite.

(1) The constituent of permittivity ε_1 occupying the volume V_1 is embedded in the region V_2 with the permittivity ε_2. We assume that the net charge density equals to zero in the total spatial domain of V_1 and V_2.

(2) We suppose that the scales of inclusions and matrix phase are much smaller than the wavelength. In this case, the composite can be regarded as a macroscopically isotropic material, in a quasi-static approximation.

B. Physical model

The three dimensional structure presented in this article is composed of 10×10×10 cubic cells for simulating the inclusion and matrix elements. The inclusion elements are placed randomly in the matrix material by means of generating a series of pseudorandom numbers which note the coordinates of the inclusion elements. Fig.1(a) shows a three-dimensional structure sample with 30% volume fraction of inclusions. The presented structure is used to calculated the effective permittivity of two-phase disordered composites with the concentration of inclusions (f) from f =0 to f =0.5. Above the concentration of f =0.5, the inclusion and matrix elements can be interchanged each other. Thus, the concentration of f >0.5 will be changed to f <0.5.

When the concentration of inclusions is increased, there will be overlapping inclusions. In this case, the mutual interaction of particles is important for the calculation of effective permittivity of composites. According to the calculation

structure shown in Fig.1(a), the elements of inclusions can be overlapped irregularly, thus there will be many types of overlapping inclusions to simulate the realistic composites at a given volume fraction. Fig.1(b) shows the inclusion cells according to the same topology shown in Fig.1(a). Moreover, the overlapping inclusions can also be simulated by this calculation model even in the case of low concentrations. When the volume fraction of inclusions is low, the inclusions in the real composites may be overlapped in a little probability. However, the overlapping inclusions in the case of low concentrations can not be fulfilled by the conventional periodic models, in which the overlap occurs only in the case of high concentrations and the size of the inclusion needs to exceed a critical value.

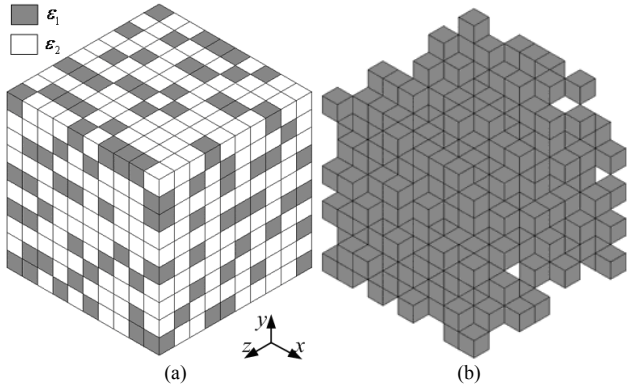

Fig.1. A three-dimensional cubic structure with 30% volume fraction of inclusions. It is composed of 10×10×10 cube cells in which the inclusions occupy the positions randomly in the matrix material: (a) 10×10×10 cube cells structure composed of both constituent materials, (b) inclusion elements corresponding to the same topology shown in (a).

C. Finite-element method

Considering absence of charge density in the composites, numerical solutions of electrostatic problems in a nonconducting material are based on the solutions of Laplace's equation

$$\mathrm{div}(\varepsilon \cdot \mathrm{grad}\varphi) = 0 , \tag{1}$$

where φ is a potential distribution in the calculated three-dimensional domain.

FEM is used to solve the equation and obtain the electric filed distribution in the material. Fig.2 shows the boundary conditions of the three-dimensional structure.

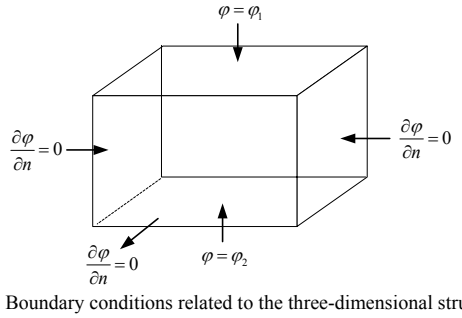

Fig.2. Boundary conditions related to the three-dimensional structure

The material is exposed to a static electric field, which is generated by a voltage across the opposite faces of the cube, and the other faces of the cube meet the requirement of $\partial\varphi/\partial n$=0. By dividing the domain into finite elements, the calculation of the potential distribution for each element is carried out by interpolation of the potential φ and its normal derivative $\partial\varphi/\partial n$ with the corresponding nodes [8, 9]:

$$\begin{cases} \varphi = \sum_i \lambda_i \varphi_i \\ \dfrac{\partial \varphi}{\partial n} = \sum_i \lambda_i \left(\dfrac{\partial \varphi}{\partial n} \right)_i \end{cases} \tag{2}$$

where λ_i represents the interpolation functions.

The solutions of FEM are carried out by the commercial FEM software Ansys. In the software, the region of interest is meshed using an adaptive meshing technique. Once the potential distribution is known, the effective permittivity of a composite is calculated by the energy balance method which has been used in the earlier simulations. The electrostatic energy for each element can be expressed as

$$\delta W_e(k) = 1/2 \int_{S_k} \varepsilon_k \left[\left(\partial\varphi/\partial x\right)^2 + \left(\partial\varphi/\partial y\right)^2 \right] dxdy , \tag{3}$$

where ε_k and S_k are the dielectric constant and the surface of the kth element, respectively. The total electrostatic energy in the entire composite can be calculated by summation of all the elements, i.e.,

$$W_e = \sum_k \delta W_e(k) . \tag{4}$$

The composite material can be regarded as a capacitor that stores the electrostatic energy when it is exposed to the electric field. The stored electrostatic energy of the capacitor can be calculated macroscopically by

$$W_e = \frac{1}{2} \varepsilon_{eff} \frac{S}{d} (\varphi_2 - \varphi_1)^2 \tag{5}$$

where S is the area of each surface exposed to the electric field and d is distance between them.

D. Statistical mean of a given structure

For a given topological structure exposed to the electric fields of orthogonal orientations, the electric field distributions in the calculated domain are not same and the obtained dielectric constants in directions x, y and z (ε_x, ε_y and ε_z) are different. Thus, an anisotropic disordered composite is simulated, instead of a macroscopically one. The calculated effective permittivity depends on the orientation of the disordered model.

In order to make up the disadvantage of the orientation of the disordered model, the effective permittivity is calculated by a method of statistical mean. The processes of calculating the mean values are comprised of two steps. At first, for a given volume fraction, the effective permittivity of a topological structure i (ε_{eff}^i) is computed by taking a statistical mean of the permittivity in the three directions x, y, and z:

$$\varepsilon_{eff}^i = \frac{1}{3} \left(\varepsilon_x^i + \varepsilon_y^i + \varepsilon_z^i \right) . \tag{6}$$

Then, in order to get a better estimate of the effective permittivity, the arithmetical mean of the calculated statistical means ε_{eff}^{i} for a number of topological structures is computed, i.e.,

$$\varepsilon_{eff} = \frac{1}{N}\sum_{i}^{N}\varepsilon_{eff}^{i} \tag{7}$$

where N is the number of the topological structures for a given volume fraction, equal to 10 in our calculation.

This procedure may be justified by the fact that each ε_{eff}^{i} is close to the mean value of the effective permittivity at a given volume fraction of inclusions, and the relative errors of ε_{eff}^{i} to their mean value are small. Fig.3(a) shows the numerical results of ε_{eff}^{i} and their mean values as a function of volume fraction f in the case of $0 \le f \le 0.5$. Fig.3(b) shows the relative errors of ε_{eff}^{i} to their mean values. It can be seen that the relative errors locate in the range of ±5%, especially they are smaller than 1% in the cases of ε_1=10 and ε_2=1, and ε_1=3 and ε_2=1. So, this procedure for calculating the effective permittivity is valid, and the numerical results reported in the rest subsections are calculated by performing this procedure.

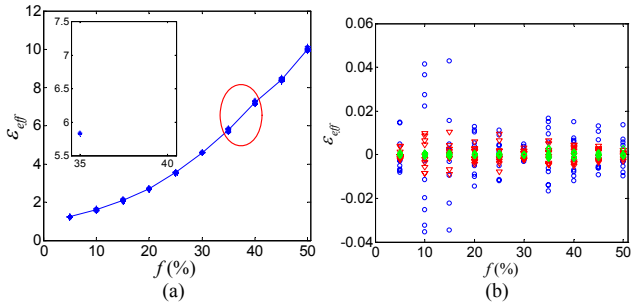

Fig.3. The calculated effective permittivity and the relative errors: (a) the effective permittivity as a function of volume fraction f due to 10 different types of topological structures at each f in the case of ε_1=30 and ε_2=1, (b) the relative errors of the calculated permittivities to their mean value at each topological structure in the cases of ε_1=30 and ε_2=1 (circles), ε_1=10 and ε_2=1 (triangles), and ε_1=3 and ε_2=1 (diamonds).

III. CALCULATION RESULTS

A. *Effect of permittivity contrast*

If the mixture is very dilute or very dense, or if the permittivity contrast $\varepsilon_1/\varepsilon_2$ is close to one, the classical mixing rules and the periodical models are valid for calculating the effective permittivity in a good accuracy. But the permittivity contrast between constituents of composites may be very large in realistic materials. This case will result in the failure of all the classical mixing rules and periodical models in obtaining a reliable effective permittivity[14]. Therefore, it is necessary to study the effect of permittivity contrast of constituent components on the calculated effective permittivity.

Based on the disordered model, the effective permittivity of composites is calculated in the cases of three different values of permittivity contrast: ε_1=3 and ε_2=1, ε_1=30 and ε_2=1, and ε_1=30 and ε_2=1. The numerical results are shown in Fig.4(a)-4(c), respectively. It should be pointed out that the distributions of electric field have no significant change due to different permittivity contrast at a given topological structure. However, the orientation of the disordered model is sensitive to the permittivity contrast. The deviation of ε_x, ε_y and ε_z will grow in the case of increasing the permittivity contrast. Moreover, the deviation between ε_{eff}^{i} and ε_{eff} becomes bigger when the permittivity contrast increases. The result can be checked by Fig.3(b). The relative errors are very small (<5‰) in the case of ε_1=3 and ε_2=1, while the biggest of them is near to 1% when ε_1=10 and ε_2=1, is about 5% when ε_1=30 and ε_2=1. So, for a low permittivity contrast of components, the effective permittivity of composites is not affected by the disordered topological structure, while it is greatly dependent on the topological structure for a high permittivity contrast. It indicates that the conventional periodic model cannot obtain a good estimate of the effective permittivity in the case of high permittivity contrast.

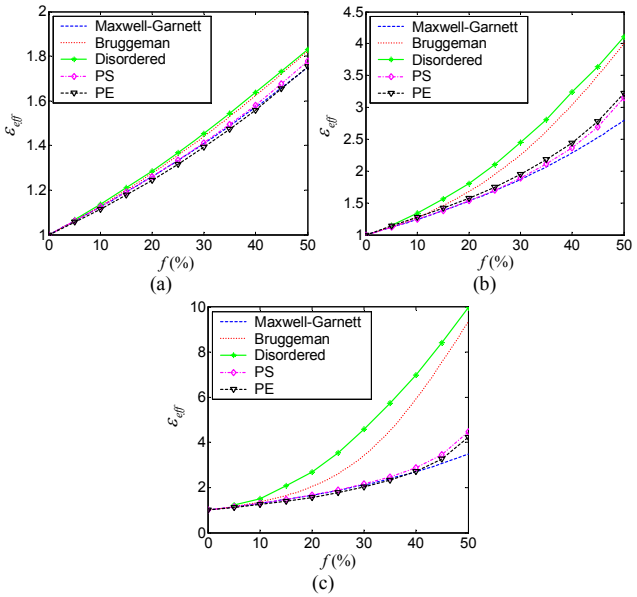

Fig.4. Comparison of the numerical results with the previous approaches: (a) ε_1=3 and ε_2=1, (b) ε_1=10 and ε_2=1, (c) ε_1=30 and ε_2=1. PS represents the results obtained by the periodic sphere model of SC and PE represents the results obtained by the periodic ellipsoid model of SC [3, 4]. In the PE model, an ellipsoid with b=c=a/4 is placed at the center of a cuboid. In this case, the depolarization factors L_x, L_y, L_z are 0.0754, 0.4623 and 0.4623, respectively.

B. *Comparison with previous approaches*

Fig.4(a)-4(c) show the effective permittivity as a function of volume fraction of inclusions obtained by the disordered model, the classical rules and the conventional periodic models for three different values of permittivity contrast. In Fig.4, it can be seen that the predictions from different models are very close when the volume fraction of inclusions is less than 10% which can be regarded as the dilute limit. In the case of low permittivity contrast, the effective permittivity values obtained by the disordered model are in good agreement with the values predicted from the Maxwell-Garnett rule, the Bruggeman rule, and the conventional periodic models. As the permittivity contrast increases, the numerical results obtained by the

disordered model and the Bruggeman rule are much bigger than those obtained by the Maxwell-Garnett rule and the conventional models. The disordered model and Bruggeman rule get similar results of effective permittivity, which can be seen in Fig.4(b)-4(c). Therefore, the validity of the numerical results obtained by different models depends on the value of the permittivity contrast and the volume fraction of inclusions. The Maxwell-Garnett rule is based on the assumption that the inclusions are spherical and the distance between neighboring spheres is much larger than their radius. Thus, for high permittivity contrast, it can only be used at low volume fraction ($f \leq 10\%$).

By comparing the effective permittivities obtained by the disordered model with those obtained by the periodic models, we can find that the arranged type of inclusions has an effect on the dielectric properties of the composites. For a low permittivity contrast, the effective permittivity is not affected by the arranged type of inclusions, and the disordered model and the periodic models get similar results. Increasing the contrast value shows its influence, i.e., in this case the deviation between the disordered model and the periodic models becomes greater. B.U.Felderhof has found that in the case of high permittivity contrast, the response of composites to a potential is coupled by multipole modes that are not contained in simple dipole mixture [10]. The periodic models can not succeed in simulating the multipole response, while the disordered model can fulfill this simulation. This is supposed to be the reason of the deviation of numerical results between the disordered model and the conventional models in the case of high permittivity contrast. Of the previous approaches, the Bruggeman rule appears to be closest to the numerical results obtained by the disordered model, even in the case of high permittivity contrast. The inclusions occupy random positions in the matrix phase of composites in the presented disordered model, thus the clustering effects will be allowed for the inclusion cells. The identical result has been found that the Bruggeman prediction is close to the simulations due to the clustering effects, which has been reported in [11].

IV. CONCLUSIONS

In summary, we have presented a simulation method for the effective permittivity of two-phase composites based on a three-dimensional disordered model. In this model, the inclusions can occupy random positions in a composite material, which is simulated by 10×10×10 cubic cells. In our calculation, the FEM is used to calculate the electric field distributions in the developed model, and then the effective permittivity is derived on the basis of the energy balance method. Three different values of permittivity contrast of components have been considered. The dependence of the effective permittivity on the orientation of the disordered model is studied. According to the orientation, an averaging procedure is adopted to get the macroscopical effective permittivity of composites in a good accuracy. The effect of permittivity contrast on the obtained effective permittivity has also been studied. And the numerical results are compared with the classical mixing rules and the conventional periodic models.

Our calculations confirm that the disordered model can be applied to high permittivity contrast and large volume fractions of inclusions, while the Maxwell-Garnett rule and the conventional periodic models fail to predict the effective permittivity, because they are not able to consider the overlapping of inclusions. The numerical results obtained by the disordered model are close to those obtained by the Bruggeman rule due to the clustering effects. Although the presented disordered model is relative close to realistic two-phase composites, there is a long way to describing the dielectric properties of realistic composites. Developing a similar topological structure to the realistic mixtures is a great challenge. The interface between the inclusions and the matrix phase has a great effect on the dielectric properties of composite material. How to introduce them to simulations will be our next research subject.

ACKNOWLEDGMENT

This work is financially supported by the National Natural Science Foundation of China (No. 50577048) and Youth Research Foundation of State Key Laboratory of Electrical Insulation and Power Equipment, Xi'an Jiaotong University, China.

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