ASSESSMENT OF CAVITIES AND WATER CONTENTS THROUGH THE COMPLEX DIELECTRIC PERMITTIVITY COMPUTED BY THE BOUNDARY INTEGRAL EQUATION METHOD

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Abstract. This paper presents a method for characterising the of heterostructures consisting of discrete inhomogeneities embedded in a three-dimensional homogeneous matrix. This method is based on the measurement of the effective complex permittivity. It enables to characterise different media and to determine the presence of inclusions and their concentration in a given heterostructure. To illustrate this technique, measurements are carried out in samples composed of identical aligned inclusions, in the form of circular cylinders, embedded in a polymer matrix and which are filled either by water or air. The experimental results are compared with the results predicted by a numerical approach based on the boundary integral equation method (BIEM) and the PHI3D package. The numerical simulations are found to capture reasonably well the observed trend in the experimental data over a wide range of volume fraction of inclusions.

Keywords: heterostructures, modelling, effective complex dielectric permittivity, voids and water content.

1. Introduction

The knowledge of the cavities (porosity and fissures) and water contents of a given medium or heterostructure is of a great interest. It enables to evaluate the ability of the medium to absorb water (or other fluids), or to place in a prominent position the presence of cavities and/or fluids. Thus, depending on the application, one can evidence the presence of water on a given site (geophysics, agriculture …) or appreciate the penetration degree of moisture and thence the quality and/or the ageing of heterostructure, such as the containers of clays for nuclear waste (diagnostic application) or structural concrete. The knowledge of the presence of fissures, their size and concentration could also inform us about the fragility of heterostructures (building, floor-tiles, beams, pillars …) resulting from different stresses such as an excessive heating or earthquakes, or due to hostile chemical environments or to a bad proportioning of construction materials.

Among different methods used to evaluate the water content in a given medium, one can cite the thermal method [1], the nuclear methods [2], the method based on the measurement of the resistivity called “electrical method” [3–5] and the methods based on the measurement of the dielectric constant called “dielectric methods” (the dielectric constant is the ratio between the absolute permittivity ε and the permittivity of vacuum ε0) [6–8]. These latter consist either to measure the capacitance of a given sample of the considered material or heterostructure (capacitive method) or to analyse the propagation velocity of an electromagnetic wave in the medium (reflectometry method known as T.D.R. method); this propagation velocity varies with the permittivity of the medium. And the knowledge of permittivity enables to go up to the water content. These methods have been successfully applied, for instance, to measure the moisture content variation in unsaturated, compacted clays [1].

During the last decade, non-destructive methods using ultra wideband radar have been developed to perform high resolution imaging of concrete structures to visualise variations of cracks, voids, and rebars inside the concrete and to determine the structural properties [9, 10]. These methods enable, through the measurement of conducting dielectric property, to evaluate the performance of cement concrete or new construction materials, such as polymer concrete developed to reduce the high costs of rehabilitation of structural concrete [11]; polymers and polymer concrete have increasingly been used in many applications, such as in construction and the repair of highways, the manufacture of precast articles, corrosion-resistant concrete in desalination plants, and sewer pipes. These methods can be applied in laboratory as well as in the field.

In this paper, we present a method enabling to evaluate the presence of inclusions and their concentration in a given heterostructure; the inclusions can be voids or cavities filled with water or another material. This method is based on the determination of the complex dielectric permittivity ε of a given heterostructure with inclusions periodically distributed starting from the first principle of electrostatics, namely Laplace’s equation.
The complex dielectric permittivity (CDP) is a parameter that characterises the storage and absorption of electromagnetic fields of a given material. The real part $\varepsilon'$ of CDP expresses the material ability to store the electric field energy and the imaginary part $\varepsilon''$ the absorption phenomena. These two components of the complex permittivity $\varepsilon$ are related as follows: $\varepsilon = \varepsilon' - i\varepsilon''$, and the $\varepsilon''/\varepsilon'$ ratio represents the dielectric loss, $\tan \delta$, of the material. This method includes two already known methods: the electrical method based on the measurement of the resistivity (or conductivity) which intervenes in the calculation of the loss factor and the dielectric method.

The remainder of this paper is organised as follows. In Section 2, we describe the general formulation of the numerical problem used to calculate the effective complex permittivity of dielectric heterostructures. The boundary integral equation method will be reviewed in this section. The experimental technique and measurement are introduced in Section 3. Then, Section 4 discusses the results of the experiments and compares them to the predictions of the numerical model. Finally conclusions of the paper will be presented in Section 5.

2. Background and numerical approach

The complex permittivity has been regarded as one of the most significant description of the dielectric properties of heterostructures. The major factors affecting the effective permittivity include size, shape and concentration of inclusions and their dielectric properties, geometry, and physico-chemical interactions between the inclusions and the host matrix. Numerous analytical tools with varying degrees of sophistication, such as the effective medium approximation (EMA), percolation, and optimal bounds methods have been used in the past several decades to investigate the macroscopic properties of a heterostructure containing inclusions with different physical properties and to quantify the complex effective permittivity [12]. These analytical tools present some limitations and especially for the analysis of heterostructures with inclusions of high concentrations and complex geometries. That is why, to quantify the effective permittivity of heterostructures, numerical methods have received a great deal of attention for the past two decades. These numerical methods consist in solving the Laplace's equation for the local electric field, including continuity conditions at the interfaces between the different constituents that comprise the media [13–20]. Two approaches have recently been put forward for simulating the complex effective permittivity of dielectric heterostructures. First, there are finite-difference-time-domain (FDTD) methods which involve approximating the partial derivatives in the Maxwell's equations by finite differences. Second, in the numerical approach we used, the effective permittivity is evaluated with the resolution of boundary integral equations (BIE) using the PHI3D field calculation package [16]. Because of its simplicity and low timing cost, this numerical approach is a computational electromagnetics methodology that is making important contributions. In the following, we give the general features of the algorithm which has been described at length elsewhere [13–16].

Consider that the heterostructure consisting of a periodic array (the dispersion being considered as null) of identical anisotropic inclusions (type 1, say) in an isotropic matrix of type 2 and that the volume concentration of inclusions is $c$. We assume first that the heterostructure can be divided into cells. In principle, this method can be applied to any specified unit cell of a given geometry, e.g., cubic, parallelepiped. Note that this problem is scale invariant, i.e., if the entire system is shrunk or dilated uniformly, the effective permittivity does not change. In this simple picture of a composite, all constituents are confined to cells centred at lattice sites which are treated at an appropriate computational level. A major advantage of this numerical approach is the simplicity of the formalism and numerical implementation.

Note that in practice, the inclusions are randomly distributed. However, the assumption of the structure as periodic one made it possible to obtain useful information concerning the dielectric constants since the driving parameter in such structure is the volume fraction rather than the internal morphology. This first approach which consists in considering the dispersion as null enables to get averaged properties of materials. On the other hand, the heterostructure can consists of more than two phases as it is the case of concrete. Indeed, it is known that concrete is considered as a three-phase composite material consisting of hardened cement paste, aggregate and the interfacial region between cement paste and aggregate [21]. The concrete material has also pores which constitute the fourth phase and whose sizes range from around 0.1 mm (capillary voids) to several mm (air voids). The method we present here can be applied to that material to get its averaged properties. For that we make the assumption that the dispersion is null and the inclusions are identical. Thus the simulation is executed in three steps. First, one determines the effective permittivity $\varepsilon_{\text{cp}}$ of the two-phase heterostructure consisting of cement paste of permittivity $\varepsilon_{\text{cp}}$ and the interfacial region of permittivity $\varepsilon_{\text{ip}}$. This two-phase heterostructure will be considered as a homogeneous constituent with its effective permittivity $\varepsilon_{\text{ip}}$. Then, we create another unitary model in which this constituent is embedded in the third one (i.e, the aggregate) of permittivity $\varepsilon_{\text{ag}}$, and we compute the effective permittivity of concrete $\varepsilon_{\text{c}}$. After that, the concrete will be considered at its turn as a homogeneous material the permittivity of which being $\varepsilon_{\text{c}}$. Finally, we create a unitary cell in which the pores of permittivity $\varepsilon_{\text{p}}$ are embedded in the concrete of permittivity $\varepsilon_{\text{c}}$. So, each step, we reduce the problem to a composite consisting of two phases. To take into account the dispersion aspect, one has to use other methods as Monte Carlo method. Moreover if one wants also to consider the fact that the inclusions are not identical then the modeling will be more complicated.

To compute the permittivity we start from the first principles in electrostatics, namely Laplace's equation, i.e., $\nabla \cdot (\varepsilon \nabla V) = 0$, where $V$ is a potential distribution inside a spatial domain $\Omega$ with a density of charge equals to zero in the whole space. The local potential $V(M \in \Omega)$ can be
written using Green’s theorem [22] in terms of \( V(P) \) and of the normal derivative \( \frac{\partial V(P)}{\partial n} \) with \( P \) being any point on the boundary \( S \) (with no overhangs) of \( \Omega \):

\[
V(M) = -\frac{4\pi}{A} \int_S \left( V(P) \frac{\partial G}{\partial n} - G \frac{\partial V(P)}{\partial n} \right) ds,
\]

where \( A \) stands for the solid angle under which the point \( M \) sees the oriented surface \( S \), \( n \) is the normal unit vector oriented outward to \( S \), \( ds \) is a surface element of \( S \) and \( G \) denotes the Green’s function. Figure 1, represents an arbitrarily shaped homogeneous inclusion occupying a volume \( \Omega_1 \) and permittivity \( \varepsilon_1 \), which is embedded in a homogeneous matrix of volume \( \Omega_2 \) and permittivity \( \varepsilon_2 \). Absence of charge density is tacitly assumed through the analysis. Given these assumptions, Eq. (1) leads to

\[
V = -\frac{4\pi}{A} \int_{S_1} \left( V(P) \frac{\partial G}{\partial n} - G \frac{\partial V(P)}{\partial n} \right) ds
\]

for domain 1, and

\[
V = -\frac{4\pi}{A} \int_{S_2} \left( V(P) \frac{\partial G}{\partial n} - G \frac{\partial V(P)}{\partial n} \right) ds
\]

for domain 2.

Moreover, the normal component of the electric displacement vector at the interface is conserved

\[
\varepsilon_1 \left( \frac{\partial V}{\partial n} \right)_1 = \varepsilon_2 \left( \frac{\partial V}{\partial n} \right)_2
\]

To evaluate numerically the electrostatic potential distribution, we have to solve the above two integral equations (2) and (3). For that purpose, we implement the boundary integral element method. It consists in dividing the boundaries into finite element and carrying out calculations for each finite element by interpolating \( V \) and \( \frac{\partial V}{\partial n} \) with the corresponding nodal values \( V = \sum \lambda_j V_j \) and \( \frac{\partial V}{\partial n} = \sum \lambda_j \left( \frac{\partial V}{\partial n} \right)_j \), where \( \lambda_j \) denote interpolating functions [22]. Thus one has to mesh only the surface bounding the volume. Following this way, integral equations are transformed in a matrix equation which is numerically solved using the boundary conditions on each side of the unit cell (Fig 1). It is worth emphasizing that the entire procedure outlined above makes use of the field calculation package PH13D.

Once the potential distribution and its normal derivative are calculated, the permittivity can be obtained. Two cases have to be distinguished:

- the inclusion is isolated (low concentration): the medium of permittivity \( \varepsilon_1 \) doesn’t intercept the sides of the unit cell (Fig 1). In that case the effective permittivity, in the direction corresponding to the applied field, is calculated using the equation

\[
\left\{ \frac{\varepsilon_2}{S_2} \left( \frac{\partial V}{\partial n} \right)_2 \right\} + \left\{ \frac{\varepsilon_1}{S_1} \left( \frac{\partial V}{\partial n} \right)_1 \right\} = \frac{V_2 - V_1}{e} (S_1 + S_2).
\]

where \( V_2 - V_1 \) denotes the difference of potential imposed in the z-direction, \( e \) is the composite thickness in the same direction and \( S \) is the surface of the unit cell perpendicular to the applied field;

- the inclusions are allowed to fuse each other (high concentration): the medium of permittivity \( \varepsilon_1 \) can intercept the sides of the unit cell (Fig 2). In that case, we must take into account the electric displacement flux through the area \( S_1 \) to calculate the effective permittivity in the direction of the applied field [13–15]. Then, Eq (5a) turns into

\[
\left\{ \frac{\varepsilon_2}{S_2} \left( \frac{\partial V}{\partial n} \right)_2 \right\} + \left\{ \frac{\varepsilon_1}{S_1} \left( \frac{\partial V}{\partial n} \right)_1 \right\} = \frac{V_2 - V_1}{e} \varepsilon (S_1 + S_2).
\]

Note that the BIE method gives an accurate description of the electric potential by taking into account edge and proximity effects even for a high concentration of

**Fig 1.** Diagram of the unit cell of a two-component periodic heterostructure (three-dimensional) investigated in the numerical computation. The isolated inclusion (low concentration) of volume \( \Omega_1 \) with dielectric constant \( \varepsilon_1 \) is periodically arranged in a three-dimensional cubic lattice structure. The dielectric constant of the remaining space is \( \varepsilon_2 \). In the following, \( e \) is taken as unit length

**Fig 2.** Same as in Fig 1 for fused inclusion (high concentration)
inhomogeneities. Therefore, this numerical value does not suffer from the disadvantages of the traditional boundary-value approach. One salient feature of BIEM is that only the boundaries of the geometry need be discretised which has for effect to reduce the memory space required for manipulation of data, but the matrix equation to solve is asymmetric and full.

3. Experimental technique

The heterostructure we consider is a “periodic” anisotropic two-component composite material. To provide the simplest geometry for subsequent analysis of the system, we have manufactured material samples consisting of a cube of dimensions 40 mm × 40 mm × 40 mm in which 9 parallel non-overlapping identical cavities in the form of circular cylinders have been periodically spaced in the plane transverse to the cylinders. Fig 3 a presents the axially symmetric configuration investigated in the present study in which the orientation of the cylinders’ axis is parallel to the direction of the electric field vector. The host matrix is made of poly(vinyl chloride) (PVC) the permittivity of which is \(\varepsilon = 6.5 - 0.08i\) in the range of frequency we investigate (ie 20 Hz – 1 MHz). The inclusions contain either air \((\varepsilon = 1 - 0i)\) or de-ionized water \((\varepsilon = 90 - 15i)\). To vary the volume fraction of inclusion, \(c\), we modify either the radius of the circular cylinder \((0.5 \leq r \leq 5.5 \text{ mm})\), or we add spacers of PVC of 2 and 4 mm thickness (Fig 3 b).

\[\varepsilon' = C_p / C_0 \]
\[\varepsilon'' = 1/(C_0 R_p \omega)\]

\[\text{with } C_0 = \varepsilon_0 S_a / l \text{ and } \omega = 2\pi f\]

\(S_a\) and \(l\) are respectively the active section of the sample and its thickness; \(f\) is the frequency.

The loss factor is given by

\[\tan \delta = \varepsilon'' / \varepsilon' = 1/(R_p C_p \omega).\]

The choice of this frequency (100 kHz) is justified by the fact that the measurements of the resistance \(R_p\) and the capacitance \(C_p\) from which we compute respectively \(\varepsilon'\) and \(\varepsilon''\) are more stable at this frequency than at a lower frequency. Note that the conductivity and dielectric constant of each constituent of heterostructure are generally independent of frequency at least below 1 GHz, but
the composite system shows a strong dispersion at low frequencies [Maxwell-Wagner-Sillars (MWS) effect] [23]. Above 10 GHZ, the permittivity is frequency-dependent; for instance, the permittivity of water decreases for frequencies higher than 10 GHz.

4. Results and discussion

This section is dedicated to the confrontation of the experimental measurements of the complex permittivity versus the volume fraction of inclusions to the simulated results.

Fig 6 shows the meshing of the heterostructure unitary model, that is a single cylinder embedded in the PVC matrix, which we modelled in the PHI3D package. In the simulations for the given model, we introduced the known dielectric characteristics of both PVC and cylinder constituents. This way, through the simulations we could obtain the effective complex permittivity of the model as a function of the volume fraction of inclusions and then compare it with the experimental results.

Figs 7 a–b and 8 a–b give the measured and computed real part \(\varepsilon'\) and imaginary part \(\varepsilon''\) of the effective permittivity versus volume fraction of inclusions containing voided materials (inclusions with air) and de-ionised water, respectively. We observe a good qualitative agreement between the experimental values and the predictions of the numerical data. The noteworthy feature is that there are significant differences between the results obtained with the two types of materials filling the inclusions. Indeed, for the volume fraction we investigate, the calculated values of \(\varepsilon'\) and \(\varepsilon''\) for inclusions containing water are always larger than the corresponding measured values, even if the two variations vs volume fraction show the same trend; the calculated value and measured value are the same only when \(c = 0\). In contrast, \(\varepsilon'\) and \(\varepsilon''\) decrease gradually as the void volume fraction increases with numerical values being smaller than the experimental values over the explored range of volume fraction.

Fig 6. The unitary model executed in the PHI3D package

The quantitative differences likely originates from several limitations of the experimental model. The first is that our experimental model is not rigorously periodic, since it is of finite size. Thus it is important that the number of inclusions should be sufficiently large. Only in this case we can assume that this experimental model is quasi-periodic. The second limitation is the actual sensitivity of the measured capacitance to air bubbles due to ill-controlled filling of the cylindrical inclusions. All experimental data presented here were obtained out using a reasonable number N of cylinder inclusions, but it should be noted that extending the range of the experimental model to a higher number is in principle possible even if the range of N that can be studied is limited by the model.

An important feature that has also to be underlined is that the shape anisotropy and spatial orientation of inclusions affect significantly the effective complex permittivity as shown elsewhere [24]. For example, if the electric field is perpendicular to the cylinder axis, the value of the real and imaginary parts of CDP will be quite different (Fig 9 a–b).

Fig 7. The real part \(\varepsilon'\) of the effective permittivity as a function of the hole volume fraction, for z-aligned samples: numerical simulation (♦), experimental (□). (b) Same as in (a) for the imaginary part \(\varepsilon''\) of the effective permittivity

Despite these limitations, this work shows that knowing the real and imaginary parts of the complex effective permittivity, one can deduce a global concentration of cavities or water in a given medium. Such an information is a of major interest for numerous applications.
Fig 8. (a) The real part $\varepsilon'$ of the effective permittivity as a function of the volume fraction of de-ionised water: numerical simulation (♦), experimental (□). (b) Same as in (a) for the imaginary part $\varepsilon''$ of the effective permittivity.

Fig 9. (a) The simulated real part $\varepsilon'$ of the effective permittivity as a function of the volume fraction of de-ionised water, when the electric field is perpendicular to the cylinder axis. (b) Same as in (a) for the simulated imaginary part $\varepsilon''$ of the effective permittivity.

The experimental design can be also used to characterise concrete/mortar/paste matrix material, in the range of frequencies we considered. The specimens of the considered material are cut in cylinders or cubes or either parallelepipeds, and inserted between the plane-plane electrode arrangement; the electrode planes can be circular, square or rectangular respectively.

5. Conclusions

In this paper, we have investigated the complex effective permittivity of circular cylinders arranged periodically in a regular simple cubic lattice, in the quasistatic limit as a function of the volume fraction of the different components that comprise the media.

Despite the fact that it is a simple experimental model of an anisotropic and periodic composite material, and despite its limitations, the model has a rich and complex dielectric behaviour.

A good agreement observed between the experimental and numerical results indicates that the boundary integral equations method using the PHI3D package to solve Laplace's equation is a valid descriptive tool for the effective permittivity of dielectric heterostructures. Thus, the simulated and experimental measurements of the complex effective permittivity and BIEM can be used to characterise different media and to determine the presence of inclusions and their concentration in a given heterostructure or to predict the properties of a heterostructure depending on its initial constituents.

References


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**TUŠTUMŲ IR VANDENS KIEKIO ĮVERTINIMAS PAGAL KOMPLEKSINĘ DIELEKTRINĮ SKVARBĄ, APSKAICIUOTĄ KRAŠTINIŲ INTEGRALINIŲ LYGČIŲ METODU**

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**Santrauka**

Pateikiamas nevienalytės struktūros, susidedančios iš diskrečiųjų intervų, esančių trimateje vienalytėje matricijoje, apibūdinimo metodas. Šis metodas pagrįstas komplexinės skvarbos matavimu. Jis leidžia apibūdinti įvairias terpes ir prognozuoti intervų kiekį bei jų koncentraciją nagrinėjamoje nevienalytėje struktūroje. Metodą patikrai atlikti eksperimentinių matavimų. Į polimerinę matricią įdėtos apvalaus cilindro formos, pripildytos vandens arba oro. Kiekviename bandinėje intervų išdėstymas buvo vienodas. Eksperimentinių tyrimų rezultatai palyginti su teoriniai rezultatai, gautais taikant skaičių kūrinių integruotų lygčių metodą (KILM) bei programą PH13D. Rezultatų palyginimas parodė gerą teorinių skaičiavimų atitikimą eksperimentiniams matavimams.

**Reikšminiai žodžiai:** nevienalytė struktūra, modeliavimas, efektyvioji kompleksinė dielektrinė skvarba, tuštumų ir vandens kiekis.

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