An Empirical Dielectric Mixture Expression and its Application to Composites

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Abstract— **In this contribution, we focus not only on predicting the dielectric properties of heterogeneous media but also on extracting the topological description of such materials. We propose a novel empirical expression for binary dielectric mixtures with phase permittivities ε_1 and ε_2. The expression is derived for a scaled permittivity level, denoted with $\xi[=(\varepsilon_e-\varepsilon_1)(\varepsilon_2-\varepsilon_1)^{-1}]$, here subscript 'e' represents the permittivity of the effective medium. The proposed expression yields the volume fraction of constituents and topological information when the permittivities of the constituents are known in advance. We apply the expression to various selected composites to acquire the structure-property relationship in these materials.**

Introduction

Predicting the dielectric properties of composite materials with or without knowing the composition of the mixture is an old problem in science and technology that has large implications in various fields such as physics, electrical engineering, materials science, geophysics, pharmaceutics, biology, *etc.* A deeper understanding would be of great value to be able to calculate the dielectric constant of either a mixture of substances of known dielectric constants or, knowing the dielectric constant of a mixture of two components and that of one of the components, to calculate the dielectric constant of the other [1]. It would also be usedful to estimate the morphology of the mixture knowing the dielectric constants of a mixture of two components and that of two components [2], [3]. Recently, it has been shown [4] that the application of the knowledge in dielectric relaxation phenomena and the theory developed by Bergman [5]–[7] and Milton [8]–[10], can provide significant information from the frequency-dependent dielectric properties of a mixture when the dielectric properties of the constituents are known. Here, we present a new dielectric-mixture formula, based on an dielectric relation term, and utilize it to analyse brine-porous rock (B/PR) [11] and glass-beads-paraffin wax (GB/PW) [12] systems.

Theory and results

We first introduce a 'scaled' dielectric permittivity ξ of the two-component mixture system as,

$$\xi = (\varepsilon_e-\varepsilon_m)(\varepsilon_i-\varepsilon_m)^{-1} \tag{1}$$

where the subscripts 'e', 'm' and 'i' denote the complex dielectric permittivities ε with ohmic loss contributions of the effective, matrix and inclusion medium, respectively. Note that ξ varies between 0 and 1. Second, we propose that the dielectric permittivity of a mixture can in principle be written as the dielectric relaxation with the 'scaled' permittivity replacing the dielectric permittivity of the material [4],

$$\xi = \xi_s + \Delta\xi[1+(\varpi x)^{\alpha}]^{-\beta} \tag{2}$$

Here ξ_s is the fraction of inclusion phase 'i' forming a percolating network in the direction of the applied field—indicating the fraction of infinite cluster(s) which does not contribute to the interfacial polarization. The quantity $\Delta\xi$ is the fraction of inclusion phase that is related to the isolated, deserted, clusters, which contribute to the polarization of the system. The permittivities of the constituents construct the so-called *probing spectral frequency*, $\varpi(\equiv \varepsilon_i/\varepsilon_m-1)$, which is analogous to the frequency of the applied field in impedance measurements in the dielectric relaxation phenomenon [4]. Consequently the frequency-dependent permittivities of the constituents generate a range of spectral frequencies, which are utilized to scan the depolarization contributions that are needed for the description of the composite's topology [2], [4]. The quantity x in Eq. (2) is the spectral parameter, and it contains information on the effective depolarization factors inclusions that could be related to the topology [2]. The parameters α and β depend on the geometrical description of the system. Consequently the parameters x, α and β yield the distribution of depolarization factors for the given system in hand [13]. The concentration q of the inclusions are simply defined with $q=\xi_s+\Delta\xi$; as a result for non-percolating systems $q\equiv\Delta\xi$, and for completely percolating systems $q\equiv\xi_s$. The defined parameters in Eq. (2) are positive and smaller than one, $0<\{\xi_s,\ \Delta\xi,\ x,\ \alpha,\ \alpha\beta\}\leq 1$. Rearranging Eq. (2), we propose a dielectric mixing law as follows

$$\varepsilon_e = \varepsilon_m + (\varepsilon_i-\varepsilon_m)\{\xi_s+\Delta\xi[1+(\varpi x)^{\alpha}]^{-\beta}\} \tag{3}$$

We have selected two different systems to test the proposed expression in Eq. (3); (*i*) brine-in-porous-rock [11] and (*ii*) metal-coated glass-beads-in-paraffin wax [12] composite systems. The data for the first systems is digitized from the appropriate reference [14]. The previously obtained results for the system from the mentioned reference [14] are also shown in the figure below to better illustrate the utility of the presented mixture expression. Note that, in the analysis (the representation of results), we present the complex resistivity $\rho[\equiv(\imath\varepsilon_0\varepsilon\omega)^{-1}]$ level [15], [16] of the systems as well to

TABLE I
THE NON-LINEAR COMPLEX LEAST-SQUARES FIT RESULTS USING EQ. (2).

System	ξ_s	$\Delta\xi$	x	α	β
B/PR	0.064	0.123	0.032	1	0.741
GB/PW	0.025	0.269	0.152	1	1

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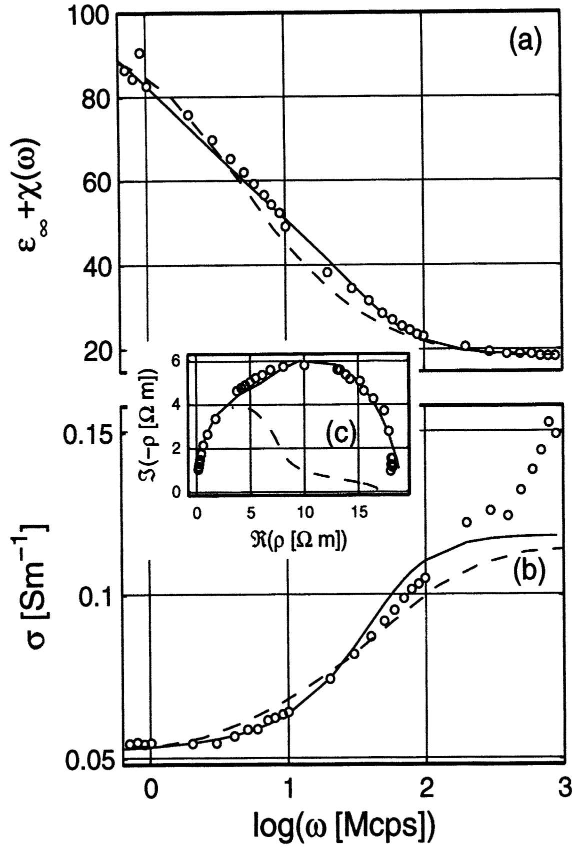

Fig. 1. Frequency-dependent (a) relative dielectric permittivity and (b) conductivity σ of the brine-porous-rock system. The Argand plot of the complex resistivity ρ is shown in (c). The measured and modelled data are presented with the open-symbols (o) and the solid line (——), respectively. The actual fits from Stroud et al. [14] are shown by the dashed lines (- - -). The resistivity level plot of the immittance from Stroud et al. [14] is in the range of the plot. However, it is only successful for low complex resistivity values. Their model generates over- and underestimate values for the complex dielectric permittivity of the system.

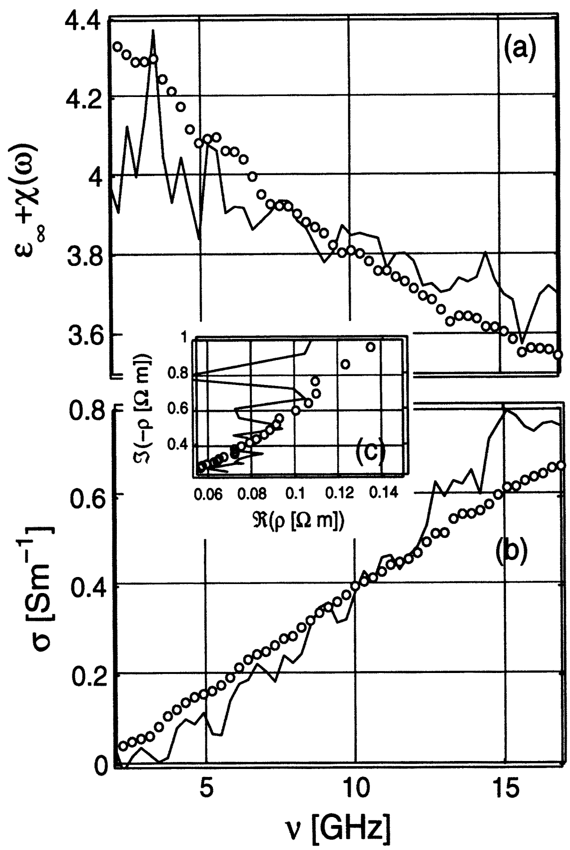

Fig. 2. Frequency-dependent (a) relative dielectric permittivity and (b) conductivity σ of the glass-bead-paraffin wax system. The Argand plot of the complex resistivity ρ is shown in (c). The measured and modelled data are presented with the open-symbols (o) and the solid line (——), respectively.

show the significance of this representation in lossy dielectric composites.

Kenyon [11] presented the dielectric data on Whitestone saturated with salt water, the system being a brine-porous-rock mixture. Here we adapted the complex permittivities of the water and the rock taken from Stroud et al. [14]. The fitting results are presented in Fig. 1, and the fit parameters are listed in Table I. It was stated that the porosity of the stone was 80%$[= (1-q)\times 100]$ in [11], a value in accord with our finding $\approx 0.813(= 1-q \equiv 1-\xi_s-\Delta\xi)$. In addition, even though the morphology of the rock was complex, the proposed expression gives evidence that some part of the brine was located in infinite pore clusters ($\xi_s > 0$).

Youngs et al. [12] have recently presented the dielectric properties of paraffin wax filled with metal-coated hollow glass-beads at microwave frequencies. In the present investigation, we adopt the complex permittivity of paraffin wax from the measurements , and estimate the inclusion permittivity from a dilute mixture measurement by utilizing an effective-medium approximation [17]. Except for the data at low frequencies ($\nu < 5$ GHz), Eq. (3) yields results in accordance with the measurements, cf. Fig. 2. It is striking that the predicted concentration $q(= 0.294)$ is in good agreement with the nominal volume fraction given by Youngs et al. [12]. The fit results listed in Table I demonstrate that most of the particles are arranged in clusters ($\xi_s < \Delta\xi$).

Conclusions

The application of the proposed expression to several data sets has illustrated its significance and potential in predicting and in comprehending the dielectric properties and structure of composites. We demonstrated that valuable information regarding the nature of mixtures can be obtained by the proposed expression. It can even be extended to systems exhibiting several depolarization processes,

$$\xi = \xi_s + \sum_n \Delta\xi_n[1+(\varpi x_n)^{\alpha_n}]^{-\beta_n}, \tag{4}$$

which is analogous to theoretical representations of dielectric relaxation. Last but not least, it is suggested that the complex resistivity level is used while analysing dielectric properties of composites [15].

Acknowledgment

We would like to express our thanks to Dr. Ian J. Youngs (DSTL, UK) for supplying the dielectric data on the GB/PW system and for fruitful comments. One of the authors' (ET) research is sponsored by the U.S. Department of Energy, Office of Electricity Delivery and Energy Reliability, Superconductivity Program for Electric Power Systems, under contract No. DE-AC05-00OR22725 with UT-Battelle, LLC.

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