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LETTER TO THE EDITOR

Analogy between dielectric relaxation and dielectric mixtures: application of the spectral density representation

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Abstract

The spectral representation is an efficient tool to explore electrical properties of material mixtures. It separates the contributions of geometrical topology and intrinsic properties of the constituents in the system. The aim of this letter is to derive an expression for the spectral density representation, which favours dielectric relaxation phenomenon. This unfamiliar form is distinct in that the existing dielectric relaxation models and data analysis tools can be employed for extracting the spectral density function of a given system.

Electrical properties of material mixtures have attracted researchers in academia and industry to seek a relation between overall composite properties and intrinsic properties of the constituents and their spatial arrangement inside the mixture [1]. Bergman [2] has proposed a mathematical way of representing the effective dielectric permittivity ε_e of a binary mixture as a function of permittivities of its constituents, ε_m and ε_i , and an integral equation, which includes the geometrical contributions. This theory is called *the spectral density representation*. Milton [3] gave the corrections, whereas Golden and Papanicolaou [4] presented the rigorous derivation for the spectral representation. The permittivity ε_e of a mixture is expressed as follows in the spectral representation [5]:

$$\varepsilon_e = \varepsilon_m \left\{ 1 + qA(\varepsilon_i \varepsilon_m^{-1} - 1) + \int_{0^+}^1 q \mathbf{G}(x) [(\varepsilon_i \varepsilon_m^{-1} - 1)^{-1} + x]^{-1} dx \right\} \quad (1)$$

where q and x are the concentration of inclusions and spectral parameter, respectively, and $\mathbf{G}(x)$ is the spectral density function. The constant A is related to the percolation strength, which includes the contribution of $\mathbf{G}(0)$. Equation (1) can be expanded by substituting $\varepsilon_i - \varepsilon_j \equiv \Delta_{ij}$, then equation (1) becomes

$$\Delta_{em} \varepsilon_m^{-1} = qA \Delta_{im} \varepsilon_m^{-1} + \int_{0^+}^1 q \mathbf{G}(x) \Delta_{im} (\varepsilon_m + \Delta_{im} x)^{-1} dx. \quad (2)$$

Now, multiplying both sides with ε_m and letting $\Delta_{em}/\Delta_{im} \equiv \xi$ and $qA \equiv \xi_s$, we obtain

$$\xi = \xi_s + q \int_{0^+}^1 \mathbf{G}(x)(1 + \varepsilon_m^{-1} \Delta_{im} x)^{-1} dx. \quad (3)$$

Here, we call ξ the ‘scaled permittivity’ and ξ_s the percolation strength as defined earlier. The mathematical properties of \mathbf{G} , q and ξ_s are expressed in the literature [5]; here $\xi_s + q \int_{0^+}^1 \mathbf{G}(x) dx = 1$ and $\int_{0^+}^1 x \mathbf{G}(x) dx = q(1 - q)d^{-1}$, where d is the dimensionality of the system. In view of equation (3), the dielectric relaxation expression for a process with single relaxation time τ is expressed as [6–8]

$$\varepsilon(i\omega) = \varepsilon_\infty + \Delta\varepsilon(1 + i\omega\tau)^{-1} \quad (4)$$

where ε_∞ , $\Delta\varepsilon$ and ω are the permittivity at optical frequencies ($\omega \rightarrow \infty$), dielectric strength and angular frequency respectively. If there exists a continuous relaxation with a distribution, equation (4) becomes

$$\varepsilon(i\omega) = \varepsilon_\infty + \Delta\varepsilon \int_0^\infty \mathbf{G}(\tau)(1 + i\omega\tau)^{-1} d\tau \quad (5)$$

where $\mathbf{G}(\tau)$ is the distribution function of the relaxation times, and the static dielectric permittivity ε_s is defined as $\varepsilon_\infty + \Delta\varepsilon \int_0^\infty \mathbf{G}(\tau) d\tau = \varepsilon_s$. Observe the similarities between equations (3) and (5). Although the resemblance between the two equations appears superficial, when the Clausius–Mossotti [6, 8] expression is taken into consideration, then the frequency dependent properties of dielectrics can in fact be written as a dynamic response of dipole units embedded in a background medium ε_m , concluding a dielectric mixture approach with inclusions as dipole units. Consequently, interfacial polarization observed in dielectric mixtures can be used to understand the structure–property relationship in physics of dielectrics [9].

There are couple of expressions that are extensively used in the dielectric data analysis, e.g. Havriliak–Negami [10], Davidson–Cole [11], and Cole–Cole [12], which have known distribution functions $\mathbf{G}(\tau)$ [7]. In addition, there exists a vast literature on dielectric data analysis [7, 13]. The total polarizability of a material is given as $\varepsilon(\omega \rightarrow 0)$; therefore, $\varepsilon_\infty + \Delta\varepsilon = \varepsilon_s$, which is actually similar to the definitions and properties of $\mathbf{G}(x)$ and ξ_s , which can be obtained by converting polarizations to fractional polarizations, which results by dividing both sides by ε_s . As a consequence, we can complete the derivation and the hypothesis about the relation between the dielectric relaxation and two-component dielectric mixtures as the relationship between various parameters in both representations, $\varepsilon_\infty \varepsilon_s^{-1} \Leftrightarrow \xi_s$, $\Delta\varepsilon \varepsilon_s^{-1} \Leftrightarrow q$ and $\mathbf{G}(x) \Leftrightarrow \mathbf{G}(\tau)$ and $\omega \Leftrightarrow \varpi \equiv \varepsilon_m^{-1} \Delta_{im}$, where ϖ is the scaled frequency of the mixture system.

Due to the similarities between equations (3) and (5), and the commutability of various parameters, one can express a general ‘scaled’ dielectric mixture formula as in the dielectric dispersions, i.e. the Havriliak–Negami expression [10],

$$\xi(\varpi) = \xi_s + q[1 + (\varpi x)^\alpha]^{-\beta}. \quad (6)$$

As a result the dielectric permittivity ε_e of the mixture then becomes

$$\varepsilon_e = \varepsilon_m + \Delta_{im} \{ \xi_s + q[1 + (\varpi x)^\alpha]^{-\beta} \}. \quad (7)$$

Yet the scaled permittivity notation can somewhat be used to calculate the time-domain dielectric mixture relations by using an inverse transform as in the case of dielectric relation theory, i.e. the relationship between the response function and the dielectric susceptibility. As

a note, one more resemblance between the two representations expressed by the Havriliak–Negami expression is that it yields the Maxwell–Garnett equation [14], which formulates the dielectric permittivity of a mixture with spherical inclusions ($d = 3$), while in the dielectric relaxation form it results in the simple Debye relaxation in equation (4), when $\alpha = \beta = 1$, $\xi_s \approx 0$ and $x = (1 - q)d^{-1}$. Similarly for the Clausius–Mossotti expression, $\alpha = \beta = 1$, and the spectral parameter localizes at 3^{-1} ; q becomes the number density of dipoles.

In this letter, it is ‘described how’ the dielectric relaxation phenomenon and expressions developed thereof for analysing dielectric dispersions can be employed to investigate the topological description or spectral density functions of two-component composites. Once the data are transformed to the scaled permittivity notation as shown in equations (3) or (6), the existing dielectric relaxation data analysis tools can in fact be readily applied; there exist many dispersion expressions in the literature. Finally, it can be inferred that even pure one-component materials can also be expressed in terms of scaled permittivity notation, wherein the smallest parts of the material are embedded in vacuum $\epsilon_m = 1$. In such a case the spectral density function would be indicative of the structure of the material and its parts, which in turn can be valuable to calculate the local fields [8] and interaction energies [15].

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