Dielectric dispersion of biological matter: Model combining Debye-type and ''universal'' responses

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The remarkably broad dielectric dispersions exhibited by solid dielectrics are well-known examples of the failure of Debye's relaxation theory; such dispersions are much better represented by a ''fractional power law'' described by Jonscher [A. K. Jonscher, Nature **267**, 673 (1977)] as the ''universal dielectric response.'' As it happens, however, recent experimental advances in this field suggest that neither of the two approaches is general enough to cope with the dielectric response of biological tissues, which combines striking features from *both* types of behavior. A phenomenological function is therefore proposed, which not only reproduces observations on biological tissues but also includes all of Jonscher's ''universal response,'' the Debye, Cole-Cole, and Davidson-Cole functions, as its special cases. [S1063-651X(99)15810-0]

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The Debye theory [1] of dispersion and absorption in dielectrics predicts frequency-dependent permittivity $\varepsilon(f)$ and conductivity $\sigma(f)$ both varying between their low- and high-frequency plateaus that correspond, respectively, to the polarized and relaxed states of the system. Deviations from Debye's ideal characteristics result in broadening of the dispersion, which has classically been correlated with some distribution of relaxation times quantifiable in terms of the Cole-Cole α [2] or the Davidson-Cole β [3] parameters. This type of behavior, collectively termed as the *Debye-type* here to include the Debye [1], the Cole-Cole [2], and the Davidson-Cole [3] responses, is characteristic of some liquids [3–6], colloidal dispersions [7–9], and biological cell suspensions [5,7,10–13].

Jonscher [14,15] has pointed out that, subject to strong interactions among their constituents, a variety of materials—mostly solids [15]—exhibit a ''universal'' dispersion pattern that departs markedly from the Debye-type response. Accordingly, the complex permittivity $\varepsilon^*=\varepsilon-j\sigma/\omega$ [with $j=(-1)^{1/2}$ and $\omega=2\pi f$, the angular frequency] can be more aptly represented by the *constant-phase-angle* (CPA) function having the form $(j\omega)^{n-1}$ (with $0<n<1$) or by the superposition of two such functions. Ngai, Jonscher, and White [16] further developed this idea to propose that solid dielectrics can be classified according to the specific values of *n*, which depend upon the relaxation mechanism(s) underlying the observed dielectric dispersion.

However, when it comes to the interpretation of the audio/radio-frequency dielectric behavior of biological tissues, which is dominated by interfacial polarization across the cell membrane [7,12], a mere application of either the Debye or the Jonscher treatment [12,17] is of limited assistance [18]. In fact, with a few exceptions (notably, the lung), experimental curves [12,19,20] combine features from *both types* of dispersions (i.e., the Debye-type and the CPA-type). More specifically, the high-frequency tails of the observed dispersions may be properly simulated by the classical (say, the Cole-Cole) functions, but their prediction for the low-frequency plateaus of ε and σ does not usually come true [18]; instead, due to electrical interactions between tissue cells [18], a CPA-type function seems to be a better description of the data in the low-frequency region of the plots. I was therefore fully motivated to seek for a more general equation that includes the classical dispersion functions as its special cases, thereby enabling a better agreement with observed curves.

Several empirical functions were examined and the most general I have arrived at is

$$\varepsilon^*=\varepsilon_h+\frac{\Delta}{[(j\omega\tau)^{\alpha}+(j\omega\tau)^{1-\beta}]^{\gamma}}+\frac{\sigma_l}{j\omega}, \tag{1}$$

where subscripts *l* and *h* refer to the low- and high-frequency limiting values; α, β, and γ are real constants taking over the interval [0,1]; τ is the characteristic relaxation time; and Δ is a dimensional constant which in some cases (see below) is called the dielectric increment $(=\varepsilon_l-\varepsilon_h)$.

Equation (1), which is symmetrical with respect to α and β, reduces to the above-mentioned particular functions upon choosing proper values for the α, β, and γ parameters. For $\gamma=1$ and $\alpha+\beta<1$ (or alternatively, $\alpha+\beta>1$), it has the asymptotic behavior

$$\varepsilon-\varepsilon_h\sim(\sigma-\sigma_l)/\omega\sim(\omega)^{-\alpha} \quad \text{for}$$
$$\omega<2\pi/\tau \ \ (\text{or} \ \ \omega>2\pi/\tau) \tag{2}$$

and

$$\varepsilon-\varepsilon_h\sim(\sigma-\sigma_l)/\omega\sim(\omega)^{\beta-1} \quad \text{for}$$
$$\omega>2\pi/\tau \quad (\text{or} \ \ \omega<2\pi/\tau), \tag{3}$$

which is apparently Jonscher's ''universal'' response [14,21,22]. Equation (1) also reduces to the Debye dispersion function for $\alpha=\beta=0$ and $\gamma=1$, to the Cole-Cole function for $\alpha=0$, $0<\beta<1$, and $\gamma=1$, and to the Davidson-Cole function for $\alpha=\beta=0$ and $0<\gamma<1$. In all of the Debye-type

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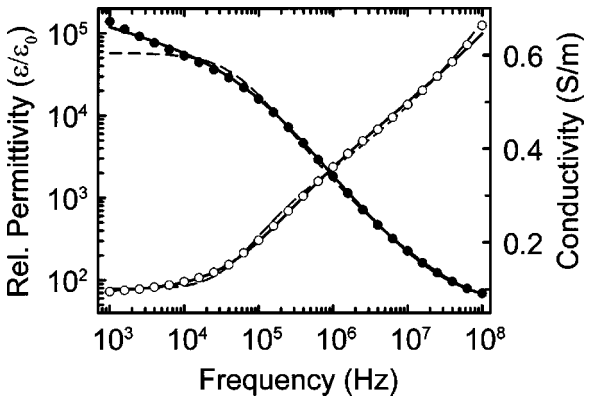

FIG. 1. Experimental data of relative permittivity (●) $\varepsilon/\varepsilon_0$ ($\varepsilon_0=8.854\times10^{-12}$ F/m) and conductivity (○) σ vs frequency for liver tissue *in vivo* (from Ref. [26]), and their comparative simulation by the function [Eq. (1)] (solid line) and the Cole-Cole function (dashed line). Parameters employed are $\Delta/\varepsilon_0=4.28\times10^4$, f_c $(=2\pi/\tau)=104$ kHz, $\alpha=0.248$, $\beta=0.120$, $\gamma=1$, $\sigma_l=0.0956$ S/m, $\varepsilon_h=48$ for Eq. (1), and $\Delta\varepsilon/\varepsilon_0=5.80\times10^4$, $f_c(=2\pi/\tau)$ $=54.1$ kHz, $\alpha=0$, $\beta=0.162$, $\gamma=1$, $\sigma_l=0.101$ S/m, $\varepsilon_h=40$ for the Cole-Cole function.

cases, the dispersion functions have finite limits at low frequencies, for which reason Δ becomes identical with the well-known dielectric increment $\Delta\varepsilon=\varepsilon_l-\varepsilon_h$.

Another special case, still more general than the above, can be made for $\gamma=1$ and for both α and β taken arbitrarily over [0, 1], in which situation one readily obtains

$$\varepsilon^*=\varepsilon_h+\frac{\Delta}{(j\omega\tau)^{\alpha}+(j\omega\tau)^{1-\beta}}+\frac{\sigma_l}{j\omega}. \tag{4}$$

If, further, $\alpha=1-\beta$, we get again the CPA law

$$\varepsilon^*=\varepsilon_h+\left(j\frac{\omega}{s}\right)^{\beta-1}+\frac{\sigma_l}{j\omega}, \tag{5}$$

where s is a scaling factor given by $s=(\Delta/2)^{1/(1-\beta)}\tau^{-1}$. The CPA behavior is believed to reflect [17,21–24], though not exclusively [22], hierarchically organized (fractal) structures, and it follows mathematically from various models of infinite networks of resistors and capacitors or from more general considerations of transport of charge in disordered systems [21,22,25]. Some biological materials (such as the lung [12,19] and plant leaves [17]) apparently fall into this same category.

To illustrate the potential of the new function for predicting experimental data in the context of comparison between the above two approaches, literature data on liver tissue [26] may serve as a typical example. Upon inspection of data in Fig. 1, we notice that the simulation by the Cole-Cole function [i.e., Eq. (1) with $\alpha=0$, $\beta=0.162$, and $\gamma=1$] can adequately reproduce the liver's dielectric response in the high-frequency range; this is an expected outcome of the interfacial polarization occurring in systems of membrane-bounded particles (in this case, cells and organelles) subjected to variable electrical fields [10]. Nevertheless, the low-frequency plateau in ε predictable from the Cole-Cole function is clearly at variance with the experimental data, which, in the log-log plot, suggest a linear behavior at low frequencies. A much better simulation of the data could be achieved when nonzero values were allowed for both α $(=0.248)$ and β $(=0.120)$ parameters in Eq. (1), whose asymptotic behavior at low frequencies is of the CPA-type (see above). According to our previous account of the dielectric relaxation in liver tissue, the marked departure of data at low frequencies from the Debye-type response most probably reflects interactions between neighboring induced di-(multi)poles [18]. Contributions originating from the polarization of electrodes—a common artifact in dielectric measurements on conductive samples [27]—should be ruled out as a possible explanation for the observed behavior, since the data plotted in Fig. 1 have already been corrected for electrode polarization [26]. In any case, the linear portion of the log-log plot shows a slope of -0.41 instead of some -1.5, a value characteristic of the electrode polarization phenomena [28].

A dispersion pattern similar to the one presented in Fig. 1 can be identified in most of the biological tissues [12,20–22] and also in nonbiological systems, either natural (e.g., rocks [23] and sand [29]) or artificial [15,30]. Common to all of these materials is the high volume fraction of their constituent particles and/or the presence of particle aggregates, conditions that are highly supportive to the above-mentioned hypothesis of interparticle interactions.

As already mentioned in the introductory paragraphs, deviations of dielectric dispersion curves from the simple Debye characteristics are traditionally quantified [2,3,6,22] in terms of the distribution of relaxation times, $F(y)$, defined by

$$\varepsilon^*=\varepsilon_h+\delta\varepsilon\int_{-\infty}^{\infty}\frac{F(y)}{1+j\omega\tau}d\ln(y)+\frac{\sigma_l}{j\omega}, \tag{6}$$

where $y=\tau/\tau_p$ with τ_p the most probable relaxation time. Although its physical meaning seems quite a bit difficult to fully grasp, I will derive a distribution function for the relaxation times involved in Eq. (1) in order to make a comparison with previous studies. To this end, we shall make use of the relationship [6]

$$F(y)=\frac{1}{2\pi\delta\varepsilon}\left|\varepsilon^*[1/(ye^{j\pi})]-\varepsilon^*[1/(ye^{-j\pi})]\right|,$$

where $\varepsilon^*[1/(ye^{\pm j\pi})]$ can be obtained by substituting $1/(ye^{\pm j\pi})$ for $j\omega\tau$ in Eq. (1) which, after some algebraic manipulation, gives

$$F(y)=\frac{y^{(1+\alpha-\beta)\gamma/2}}{2^{\gamma/2}\pi}\times\frac{\left|\sin(\gamma\theta)\right|}{\{\cosh[(1-\alpha-\beta)\ln y]+\cos[\pi(1-\alpha-\beta)]\}^{\gamma/2}} \tag{7}$$

with

$$\theta=\arctan\frac{y^{\alpha}\sin[\pi(1-\beta)]+y^{1-\beta}\sin[\pi\alpha]}{y^{\alpha}\cos[\pi(1-\beta)]+y^{1-\beta}\cos[\pi\alpha]}.$$

This distribution reduces to

$$F(y)=\frac{1}{2\pi}\frac{y^{\alpha}\sin[\pi(1-\beta)]+y^{1-\beta}\sin[\pi\alpha]}{\cosh[(1-\alpha-\beta)\ln y]+\cos[\pi(1-\alpha-\beta)]} \tag{8}$$

in the particular case of Eq. (4), to the Cole-Cole and Davidson-Cole distributions [2,3,6] for $\alpha=0$, and to the distribution function associated with the universal response (i.e., the Pareto distribution [22]) for $\alpha=1-\beta$ and $\gamma=1$.

The distribution functions for relaxation times corresponding to the theoretical simulations in Fig. 1 are plotted against y in Fig. 2, together with the Pareto distribution. It is worth noting that the distribution implied by the best simulation of the experimental data (see *solid* lines in Figs. 1 and 2) appears as an intermediate step toward the ''degeneration'' of the Cole-Cole distribution into the Pareto distribution as α increases from 0 to $1-\beta$. This fact comes to bridge once again the apparent gap between the Jonscher and the Debye (or Cole-Cole) approaches, at the basic level of their distribution functions for relaxation times.

In summary, I suggest that the Debye-type (i.e., the Debye, the Cole-Cole, and the Davidson-Cole) dispersion functions and the universal response, which are otherwise useful in emphasizing the specific differences between classes of polarizable materials, are often too particular to completely describe the dielectric behavior of such complex systems as, but not restricted to, biological tissues. By contrast, a dispersion function proposed herein reproduces typical experimental data with excellent accuracy over a broad range of frequencies, and appears to unify the two different types of dispersion functions by including them as its limiting cases. These features taken together may shed light on the manner in which we presently classify electrically polarizable materials.

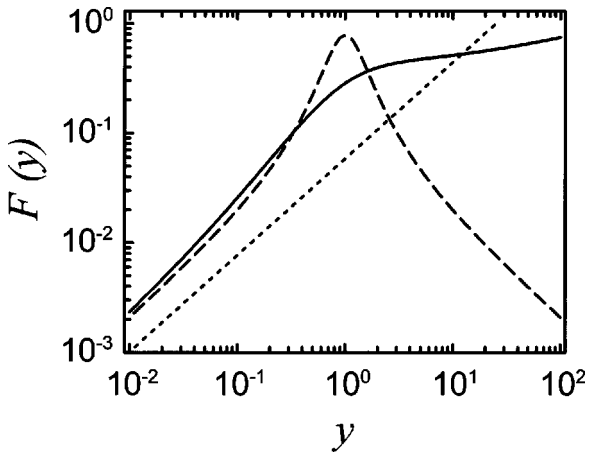

FIG. 2. Distribution functions for relaxation times [Eq. (7)] corresponding to the theoretical curves in Fig. 1. The α, β, and γ parameters as well as the type of the lines are exactly as in Fig. 1. Also added, for comparison, is the Pareto distribution (dotted line, $\alpha=1-\beta=0.880$, $\gamma=1$).

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