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Phenomenological approach on wave propagation in dielectric media with two relaxation times

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ABSTRACT

In this paper dielectric phenomena with two relaxation times are discussed. By assuming a sinusoidal form for induction vector **D** a sinusoidal electric field is generated and it depends on unknown phenomenological coefficients whose expressions together to their numerical values as functions of frequency are obtained. Moreover, electromagnetic wave propagation is analysed obtaining wave vector as function of the aforementioned coefficients. The results are applied to a Vinylidene Chloride-Vinyl Chloride (VDC–VC) to test the applicability of the model.

1. Introduction

In a previous paper [1] the connection between phenomenological coefficients and quantities experimentally measurable, e.g. real and imaginary parts of complex dielectric constant, has been obtained for media with dielectric relaxation phenomena and in the case in which just one relaxation time is considered.

In Refs. [2–4] a phenomenological equation was proposed in which two relaxation times occur and this is connected to physical behaviour of materials. In fact the instantaneous increasing or decreasing in the polarization is impossible because any change of the polarization is related to the motion of any kind of microscopic particles which cannot be infinitely fast. The phenomenological equation will be discussed in Section 2 and we will refer to the case in which two relaxation times are taken into account and in such a context we will study electromagnetic wave propagation obtaining wave vector as function of the aforementioned quantities experimentally measurable.

Our aim is to study the system under a sinusoidal perturbation represented by the induction vector D, which is an extensive variable (cause), and to analyse the relative electric field as an intensive variable (effect) inside the medium [5]. In particular, if we consider a generic dielectric medium placed between the plain plate of a capacitor where a sinusoidal voltage has been applied, then on the plates a sinusoidal surface charge arises whose density is characterized by the normal component of induction vector $D = \mathbf{D}\cdot\mathbf{n}$ ($\mathbf{n}$ is the unit normal to the plates) generating a sinusoidal electric field inside the capacitor [6].

The linear-response theory establishes that if D (cause) evolves harmonically [5], i.e.

$$D^* = D_0 e^{i\omega t} \quad (1.1)$$

where D_0 is the displacement amplitude and ω the angular frequency, then the normal component ($E = \mathbf{E}\cdot\mathbf{n}$) of the electric field inside the capacitor is also harmonic and characterized by the same frequency but different phase and amplitude:

$$E^* = E_0 e^{i(\omega t + \phi(\omega))} \quad (1.2)$$

where E_0 is the field amplitude and ϕ the phase lag. Furthermore we have

$$D^* = \varepsilon^* E^* = (\varepsilon' - i\varepsilon'')E \quad (1.3)$$

where ε^* is the complex dielectric constant and

$$\varepsilon' = |\varepsilon^*|\cos\phi, \quad \varepsilon'' = |\varepsilon^*|\sin\phi, \quad \frac{\varepsilon''}{\varepsilon'} = \tan\phi \quad (1.4)$$

the quantities ε' and ε'' are the real and imaginary components of the complex dielectric constant ε^* [7].

The quantities ε' and ε'' are related to the relative dielectric constants ε_1, ε_2 by the following expressions:

$$\varepsilon' = \varepsilon_0\varepsilon_1, \quad \varepsilon'' = \varepsilon_0\varepsilon_2 \quad (1.5)$$

where ε_0 is the dielectric constant in vacuum. These are usually called the dielectric storage factor and dielectric loss factor, respectively, and $\tan\phi$ is termed the loss tangent. Let us remark that in a relaxation region ε' is decreasing with frequency from a value of ε_R to ε_U. This decrease represents the dispersion of the

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dielectric constant [6], the difference $(\varepsilon_R - \varepsilon_U)$ is known as the magnitude of the relaxation and it expresses a measure of the orientation polarization [8]. In relaxation region ε'' passes through a maximum at a frequency $\omega_{\varepsilon''}$. By computing the real part of Eq. (1.2) one has

$$E = D_0 s_1 \sin(\omega t) + D_0 s_2 \cos(\omega t) \tag{1.6}$$

where

$$s_1 = \frac{E_0(\omega)}{D_0}\cos\phi(\omega) \tag{1.7}$$

$$s_2 = \frac{E_0(\omega)}{D_0}\sin\phi(\omega) \tag{1.8}$$

If electric charge density on the plates (extensive quantity) is viewed as the cause determining the electric field inside capacitor (intensive variable), it allows us to study dielectric relaxation phenomena. By defining the reciprocal complex dielectric constant $s^* = E^*/D^* = s_1 + is_2$ the complex dielectric constant is related to it as

$$\varepsilon^* = \frac{1}{s^*} = \varepsilon' - i\varepsilon'' \tag{1.9}$$

where

$$s_1 = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2}, \quad s_2 = \frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} \tag{1.10}$$

Taking into account Eqs. (1.7) and (1.8) the following expressions are obtained:

$$\varepsilon' = \frac{D_0}{E_0(\omega)}\cos(\omega) \tag{1.11}$$

$$\varepsilon'' = \frac{D_0}{E_0(\omega)}\sin\phi(\omega) \tag{1.12}$$

Since the phase difference ϕ depends on frequency, it follows that for values of ω sufficiently small ϕ approaches to zero obtaining from Eqs. (1.12) and (1.12):

$$\varepsilon' \cong \frac{D_0}{E_{0R}} = \varepsilon_R, \quad \varepsilon'' \cong 0 \tag{1.13}$$

where E_{0R} is the value of $E_0(\omega)$ for sufficiently small values of ω and ε_R is the relaxed value of ε'. Analogously, for sufficiently large values of ω the phase ϕ goes to zero and one has

$$\varepsilon' \cong \frac{D_0}{E_{0U}} = \varepsilon_U, \quad \varepsilon'' \cong 0 \tag{1.14}$$

where E_{0U} is the value of $E_0(\omega)$ for large values of ω, and ε_U is the un-relaxed value of ε'.

2. Phenomenological coefficients

Following Refs. [9–15], a general hypothesis concerning the entropy allows us to decompose the polarization vector **P** as

$$\mathbf{P} = \mathbf{P}^{(0)} + \mathbf{P}^{(1)} \tag{2.1}$$

where $\mathbf{P}^{(0)}$ and $\mathbf{P}^{(1)}$ are the reversible (elastic) and irreversible parts of **P**, respectively. In the linear approximation and by neglecting cross effects as the influence of electric conduction, heat conduction and (mechanical) viscosity on electric relaxation, the following relaxation equation may be derived [9]:

$$\chi^{(0)}_{EP}\mathbf{E} + \frac{d\mathbf{E}}{dt} = \chi^{(0)}_{PE}\mathbf{P} + \chi^{(1)}_{PE}\frac{d\mathbf{P}}{dt} + \chi^{(2)}_{(PE)}\frac{d^2\mathbf{P}}{dt^2} \tag{2.2}$$

where **E** is the electric field and $\chi^{(0)}_{(EP)}, \chi^{(i)}_{(PE)}$ $(i = 0, 1, 2)$ are algebraic functions of the coefficients occurring in the phenomenological equations (describing the irreversible processes) and in the equations of state.

By considering the expression for the polarization vector:

$$\mathbf{P} = \mathbf{D} - \varepsilon_0\mathbf{E} \tag{2.3}$$

where ε_0 is the dielectric constant in vacuum; by substituting it in Eq. (1.3) the equation for dielectric relaxation reduces to

$$\chi^{(0)}_{(ED)}\mathbf{E} + \chi^{(1)}_{(ED)}\frac{d\mathbf{E}}{dt} + \chi^{(2)}_{(ED)}\frac{d^2\mathbf{E}}{dt^2} = \chi^{(0)}_{(DE)}\mathbf{D} + \chi^{(1)}_{(DE)}\frac{d\mathbf{D}}{dt} + \chi^{(2)}_{(DE)}\frac{d^2\mathbf{D}}{dt^2} \tag{2.4}$$

where we are setting

$$\chi^{(0)}_{(ED)} = \chi^{(0)}_{(EP)} + \varepsilon_0\chi^{(0)}_{(PE)} \tag{2.5}$$

$$\chi^{(1)}_{(ED)} = 1 + \chi^{(1)}_{(PE)}\varepsilon_0 \tag{2.6}$$

$$\chi^{(2)}_{(ED)} = \chi^{(2)}_{(PE)}\varepsilon_0 \tag{2.7}$$

$$\chi^{(0)}_{(DE)} = \chi^{(0)}_{(PE)} \tag{2.8}$$

$$\chi^{(1)}_{(DE)} = \chi^{(1)}_{(PE)} \tag{2.9}$$

$$\chi^{(2)}_{(DE)} = \chi^{(2)}_{(PE)} \tag{2.10}$$

By dividing $\chi^{(2)}_{(ED)} \neq 0$ Eq. (2.4) one obtains the following normal form:

$$\frac{\chi^{(0)}_{(ED)}}{\chi^{(2)}_{(ED)}}\mathbf{E} + \frac{\chi^{(1)}_{(ED)}}{\chi^{(2)}_{(ED)}}\frac{d\mathbf{E}}{dt} + \frac{d^2\mathbf{E}}{dt^2} = \frac{\chi^{(0)}_{(DE)}}{\chi^{(2)}_{(ED)}}\mathbf{D} + \frac{\chi^{(1)}_{(DE)}}{\chi^{(2)}_{(ED)}}\frac{d\mathbf{D}}{dt} + \frac{1}{\varepsilon_0}\frac{d^2\mathbf{D}}{dt^2} \tag{2.11}$$

By computing the time derivative of D expressed by the real part of Eq. (1.1) having used the normal component of electric field $E = \mathbf{E}\cdot\mathbf{n}$, the differential equation (2.11) can be written as

$$\frac{d^2E}{dt^2} + \frac{\chi^{(1)}_{(ED)}}{\chi^{(2)}_{(ED)}}\frac{dE}{dt} + \frac{\chi^{(0)}_{(ED)}}{\chi^{(2)}_{(ED)}}E = \alpha\sin(\omega t) + \beta\cos(\omega t) \tag{2.12}$$

where

$$\alpha = D_0\left(\frac{\chi^{(0)}_{(DE)}}{\chi^{(2)}_{(ED)}} - \frac{\omega^2}{\varepsilon_0}\right), \quad \beta = D_0\frac{\chi^{(1)}_{(DE)}\omega}{\chi^{(2)}_{(EwD)}}. \tag{2.13}$$

The integration of differential equation (2.12) gives the following general solution that gives the electric field:

$$E(t) = c_1c_1e^{\lambda_1 t} + c_2e^{\lambda_2 t} + \left[\frac{\alpha(\lambda_1\lambda_2 - \omega^2) - \beta\omega(\lambda_1 + \lambda_2)}{(\lambda_1^2 + \omega^2)(\lambda_2^2 + \omega^2)}\right]\sin(\omega t)$$

$$+ \left[\frac{\beta(\lambda_1\lambda_2 - \omega^2) + \alpha\omega(\lambda_1 + \lambda_2)}{(\lambda_1^2 + \omega^2)(\lambda_2^2 + \omega^2)}\right]\cos(\omega t) \tag{2.14}$$

where c_1 and c_2 are two arbitrary integration constants, λ_1 and λ_2 are solutions of the homogeneous equation associated to (2.12)

$$\lambda^2 + \frac{\chi^{(1)}_{(ED)}}{\chi^{(2)}_{(ED)}}\lambda + \frac{\chi^{(0)}_{(ED)}}{\chi^{(2)}_{(ED)}} = 0 \tag{2.15}$$

moreover the quantities $-\lambda_1^{-1}$ and $-\lambda_2^{-1}$ represent the two relaxation times. Since Eqs. (1.6) and (2.14) describe mathematically the same phenomenon, by neglecting any transitory effect, the identification of two equations leads to the following:

$$D_0s_1 = \frac{\alpha(\lambda_1\lambda_2 - \omega^2) - \beta\omega(\lambda_1 + \lambda_2)}{(\lambda_1^2 + \omega^2)(\lambda_2^2 + \omega^2)} \tag{2.16}$$

$$D_0s_2 = \frac{\beta(\lambda_1\lambda_2 - \omega^2) + \alpha\omega(\lambda_1 + \lambda_2)}{(\lambda_1^2 + \omega^2)(\lambda_2^2 + \omega^2)} \tag{2.17}$$

By using Eqs. (1.10) and (2.13), from Eqs. (2.16) and (2.17) we obtain

$$\chi^{(1)}_{(PE)} = \frac{\chi^{(0)}_{(EP)}\varepsilon'' + \omega(\varepsilon' - \varepsilon_0)}{\omega[(\varepsilon' - \varepsilon_0)^2 + \varepsilon''^2]} \tag{2.18}$$

$$\chi^{(2)}_{(PE)} = \frac{\chi^{(0)}_{(PE)}}{\omega^2} + \frac{\chi^{(0)}_{(EP)}(\varepsilon_0 - \varepsilon') + \varepsilon''\omega}{\omega^2[(\varepsilon' - \varepsilon_0)^2 + \varepsilon''^2]} \tag{2.19}$$