

ABOUT THE DISTRIBUTION OF RELAXATION TIMES IN DIELECTRICS

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***Abstract:** When studying non – homogenous dielectrics, the distribution of relaxation times is crucial in order to appreciate their behavior in the frequency domain. First, this paper presents an overview of the functions of distribution of relaxation times. Then, a computation method is emphasized, and these functions are graphically represented using MathCAD worksheet. The frequency variation graphics of the two components of the complex permittivity are also outlined, when the influence of the electric conductivity is considered or not.*

***Key Words:** dielectrics, dielectric relaxation, complex permittivity, Cole – Cole diagrams, MathCAD worksheet*

1. INTRODUCTION

A relaxation polarization is, almost always, present in non-homogenous dielectrics. The relaxation phenomenon represents the orientation of dipoles due to an electric field, or the accumulation of real electric charges (free charges) at the boundary between different homogenous parts of non-homogenous dielectrics. This phenomenon causes the change of the two components of the complex relative permittivity and their frequency variation, especially in the range of low or very low frequencies.

Since the dipoles / free charges have a relatively large molecular mass and they are moving in a viscous media, the orientation process is characterized by a relaxation time of the dipole / free charge, τ_d . Non-homogenous dielectrics contain several types of dipoles – with different relaxation times - leading to a distribution of relaxation times that can be expressed by a distribution function. Starting from the empiric formulas of these functions, proposed by different researchers, one can establish the expressions of the two components of the complex

relative permittivity as a function of frequency, when the electric conductivity (σ) is considered or not.

2. DISTRIBUTION OF THE RELAXATION TIMES

The well-known study models for the dielectric relaxation (Debye, Maxwell – Wagner) consider a unique relaxation time. But for non-homogenous dielectrics, experimental data indicate several inconsistencies with Debye's theoretical approach:

- transient absorption / resorption currents do not vary exponentially in time;
- the function $\varepsilon'' = f(\varepsilon')$ isn't a half circle centered on the ε' axe (this would show an exponential rise of the response function of the dielectric), but an arc of a circle, centered below the ε' axe, or an other close form;
- not all the dipoles have the same relaxation time τ_d . Furthermore, its value is between two extremes τ_{dmin} and τ_{dmax} , according to some distribution laws.

If one considers that each type of dipole has a Debye relaxation, the large distribution of relaxation times (figure 1) can be expressed by an empiric function $G(\tau)$ that satisfies the unit normalization condition. The response function of the dielectric in the frequency domain becomes:

$$f(t) = (\varepsilon_s - \varepsilon_\infty) \int_0^\infty G(\tau) \cdot \exp\left(-\frac{t}{\tau}\right) d\tau \quad (1)$$

and the expressions of the two components of the complex permittivity are:

$$\varepsilon'(\omega) = \varepsilon_\infty + (\varepsilon_s - \varepsilon_\infty) \int_0^\infty \frac{G(\tau)}{1 + \omega^2 \tau^2} d\tau; \quad \varepsilon''(\omega) = (\varepsilon_s - \varepsilon_\infty) \int_0^\infty \frac{\omega \tau G(\tau)}{1 + \omega^2 \tau^2} d\tau \quad (2)$$

with $\varepsilon_s, \varepsilon_\infty$ - the permittivities for 0 [Hz] and $f \rightarrow \infty$.

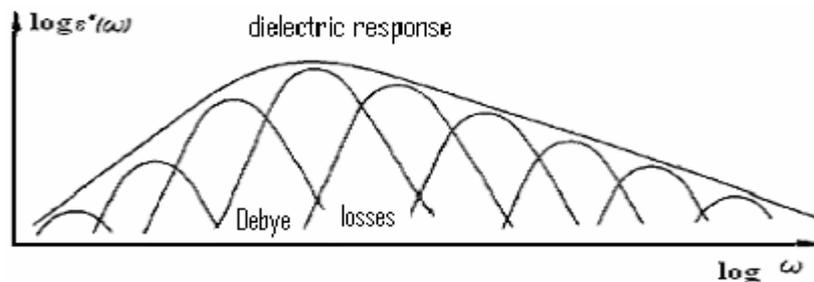


Fig. 1 The distribution of relaxation times

In order to obtain $G(\tau)$ from the experimental data, some easier methods have been proposed: considering a finite number of terms and converting the integrals (2) in finite sums

[2, 3, 5] or using statistical methods, such as Monte Carlo. The most general method is, however, the de-convolution of experimental data.

3. EMPIRIC DISTRIBUTION FUNCTIONS

Since the real distribution of relaxation times is very difficult to find, some empiric formulas have been proposed in order to express the variation of complex permittivity with frequency, formulas based on non Debye relaxation [1, 2, 4]. From these formulas, for $\underline{\varepsilon}$ one can establish the expressions of the empiric function for the distribution of relaxation times $G(\tau)$.

K. S. Cole and R. H. Cole noticed that the locus diagram, determined from experimental data, is a circle centered below the $\varepsilon'' = 0$ axe and they suggested that experimental data would be described by a modified Debye empiric equation:

$$\underline{\varepsilon}(\omega) = \varepsilon_{\infty} + \frac{\varepsilon_s - \varepsilon_{\infty}}{1 + (j\omega \tau_{CC})^{\beta_{CC}}}, \quad (3)$$

where τ_{CC} is the average value of relaxation times and the coefficient β_{CC} is larger than 0 and smaller than 1.

From (3) one can express the components of the complex permittivity, and the functional form of the empiric function $G(\tau)$ is defined as below:

$$\begin{cases} \varepsilon'_{CC} = \varepsilon_{\infty} + \Delta \varepsilon \frac{1 + p^{\beta_{CC}} \cos(\beta_{CC} \pi / 2)}{1 + 2 p^{\beta_{CC}} \cos(\beta_{CC} \pi / 2) + p^{2\beta_{CC}}} \\ \varepsilon''_{CC} = \Delta \varepsilon \frac{p^{\beta_{CC}} \sin(\beta_{CC} \pi / 2)}{1 + 2 p^{\beta_{CC}} \cos(\beta_{CC} \pi / 2) + p^{2\beta_{CC}}} \end{cases} \quad (4)$$

$$G[\tau / \tau_{CC}] = \frac{1}{2\pi} \cdot \frac{\sin[\pi(1 - \beta_{CC})]}{\text{ch}[\beta_{CC} \ln(\tau / \tau_{CC})] - \cos[\pi(1 - \beta_{CC})]}, \quad (5)$$

with $p = \omega \tau_{CC}$ and $\Delta \varepsilon = \varepsilon_s - \varepsilon_{\infty}$.

The distribution function defined in (5) is symmetrical and centered in τ_{CC} .

Cole and Davidson established another empiric formula for $\underline{\varepsilon}$:

$$\underline{\varepsilon}_{CD} = \varepsilon_{\infty} - \frac{\Delta \varepsilon}{(1 + j\omega \tau_{CD})^{\beta_{CD}}}, \quad (6)$$

that leads to:

$$\varepsilon'_{CD} = \varepsilon_{\infty} + \frac{\Delta \varepsilon}{\left(\sqrt{1+p^2}\right)^{\beta_{CD}}} \cos(\beta_{CD} \varphi); \quad \varepsilon''_{CD} = \frac{\Delta \varepsilon}{\left(\sqrt{1+p^2}\right)^{\beta_{CD}}} \sin(\beta_{CD} \varphi) \quad (7)$$

$$G(\tau / \tau_{CD}) = \begin{cases} \frac{\sin \pi \beta_{CD}}{\pi} \left(\frac{\tau}{\tau_{CD} - \tau}\right)^{\beta_{CD}} & \text{for } \tau < \tau_{CD} \\ 0 & \text{for } \tau \geq \tau_{CD} \end{cases} \quad (8)$$

From formula (8) one can realize that there is no larger relaxation time than τ_{CD} , and the maximum loss do not occur for $f=1/2\pi\tau_{CD}$ but for $f_{CD}=(1/2\pi\tau_{CD}) \operatorname{tg} [\pi/2(\beta_{CD}+1)]$.

Williams and Watts first established the most accurate empiric distribution function. This function fits a large number of relaxation data measured for glass and polymer. They considered that the variation in time of polarization can be expressed by an extended exponential function $P_{WW}(t) = e^{-(t/\tau_{WW})^{\beta_{WW}}}$, where the values of β_{WW} are between 0 and 1. Applying the extended exponential Fourier transform for decomposing the above function, they established the formulas:

$$\underline{\varepsilon}_{WW}(\omega) = \varepsilon_{\infty} + \Delta \varepsilon \sum_{n=1}^{\infty} \frac{(-1)^{n-1} \Gamma(n\beta_{WW} + 1)}{(\omega \tau_{WW})^{n\beta_{WW}} \Gamma(n+1)} \left[\cos\left(\beta_{WW} \frac{n\pi}{2}\right) - j \sin\left(\beta_{WW} \frac{n\pi}{2}\right) \right] \quad (9)$$

$$G(\tau / \tau_{WW}) = -\frac{\tau_{WW}}{n\tau} \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \sin(\pi\beta_{WW} n) \Gamma(\beta_{WW} \cdot n + 1) \left(\frac{\tau}{\tau_{WW}}\right)^{\beta_{WW} \cdot n + 1} \quad (10)$$

Plotting $G(\tau/\tau_{WW}) = f(\log(\tau/\tau_{WW}))$ for different values of β_{WW} (fig. 2), one can see that the graphic is symmetrical with respect to $\alpha = \tau / \tau_{WW} = 1$ for low values of β_{WW} , but tends asymptotically to a delta function at $\beta_{WW} \rightarrow 1$.

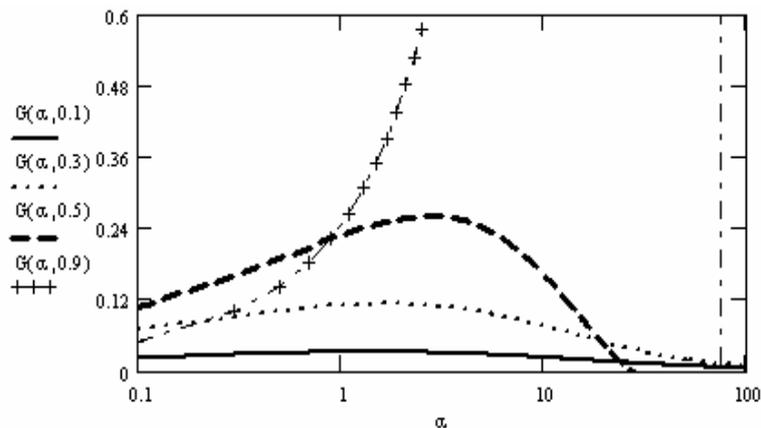


Fig. 2. MathCAD worksheet calculating and displaying the William – Watts relaxation time distribution function for several values of β_{WW}

4. MATCHAD FILES FOR COMPUTATION AND PLOTTING

Based on formulas (4), (7) and (9) we created some MathCAD files for computing the two components of the complex permittivity, for different frequencies, and plotting the Cole – Cole diagrams. Figure 3 emphasizes these diagrams for the same average relaxation time and for two different values of β (0,2 and 0,7).

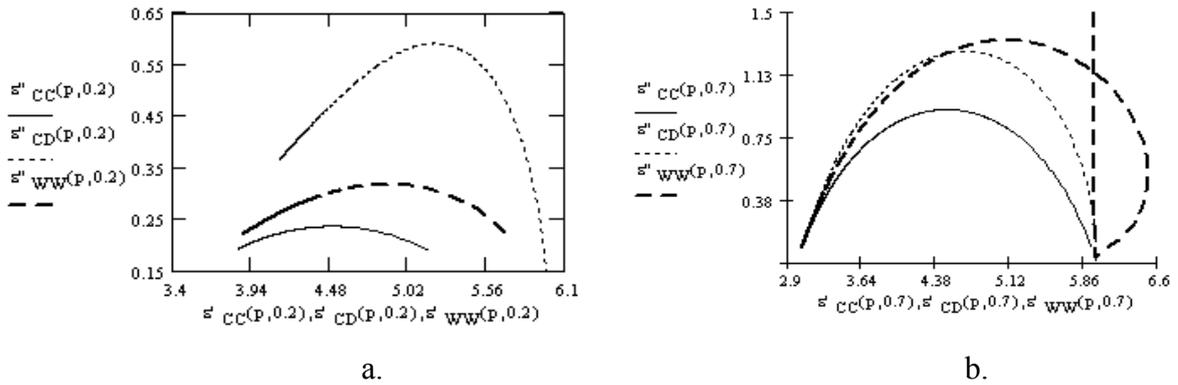


Fig. 3. MathCAD worksheet for computing and plotting the Cole – Cole diagrams according to the relaxation models proposed by Cole-Cole, Davidson – Cole, William – Watts for: a. $\beta = 0,2$; b. $\beta = 0,7$.

The shape of the distribution functions proposed by Cole-Cole, Cole-Davidson or Williams-Watts, with the ratio $\alpha = \tau_i/\tau_{CC}; \tau_i/\tau_{CD}; \tau_i/\tau_{WW}$ and for $\beta_{CC} = \beta_{CD} = \beta_{WW} = 0,45$ is shown in figure 4.

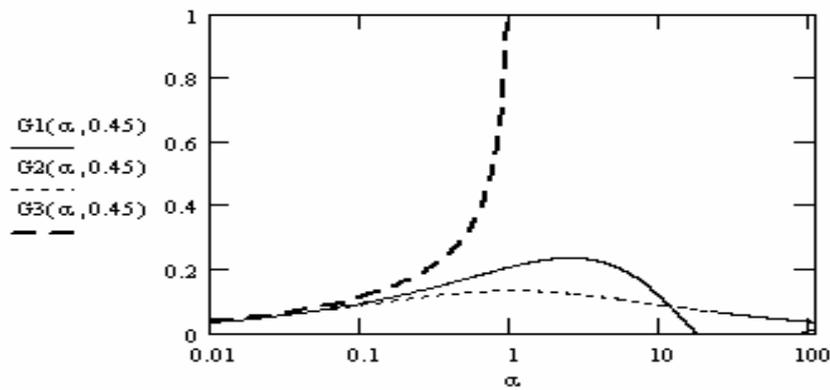


Fig. 4. MathCAD worksheet for computing and plotting the function of relaxation times according to Cole- Cole, Davidson – Cole and Williams – Watts models for $\beta = 0,45$.

If we also consider the influence of electric conductivity, the loss component ϵ'' changes by addition of a new term due to conduction. The diagrams $\epsilon'' = f(\epsilon')$ and $\epsilon'' = f(\log f)$ are plotted in figure 5 for several values of the electric conductivity ($\sigma = 0; 10; 100; 1000$).

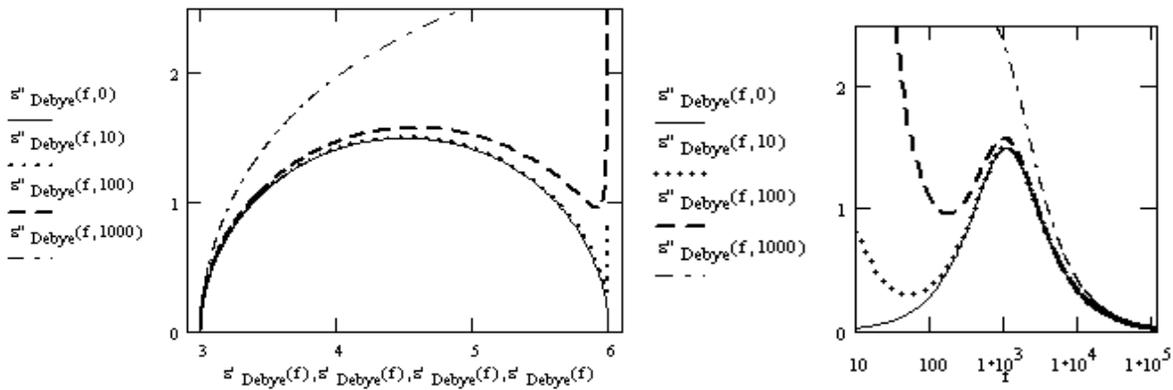


Fig. 5 MathCAD worksheet for computing and plotting the Cole – Cole diagram ($\epsilon'' = f(\epsilon')$) and losses ($\epsilon'' = f(\log f)$) of a dielectric with Debye relaxation time, for $\sigma = 0; 10; 100; 1000$.

5. DISCUSSIONS AND CONCLUSIONS

The MathCAD files created permit a fast computation of the complex permittivity frequency variation for a wide range of values of the β coefficient, from the most accurate empiric formulas. The graphical plots of the Cole – Cole diagrams indicate important changes of the shape, even for the same value of β : starting from a half – circle centered below the ϵ' axe (Cole – Cole model), to a pear shape (Cole – Davidson model) or a deformed bell (Williams – Watts model). For different values of β , the shapes are quite changed.

Comparing the Cole – Cole diagrams plotted from experimental data with those computed through the indicated method, one can choose the empiric formula most adequate to show the frequency behavior of a certain non-homogenous dielectric. After establishing the distribution function of relaxation times in a composite material, one can start simulating the behavior of the composite using equivalent electric circuits in order to study the relaxation phenomenon in complex situations, or to analyze the aging coefficient of insulating systems.

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