

Mathematical approach to the relaxation phenomena

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Abstract. The aim of this work is to describe a general method for correlate phenomenological and state coefficients to quantities experimentally measurable both for mechanical and dielectric relaxation phenomena.

M.S.C. 2000: 00A69, 74A15, 74A20, 74D05.

Key words: non-equilibrium thermodynamics, relaxation phenomena, constitutive equations, linear response theory.

1 Introduction

As it is well known any material system which is subjected to a external perturbation shows a change of the set of variables which describes the state. Since a rigorous description of phenomena involves a great number of variables it is useful to introduce some modelization, in agreement with experience, which reduces the number of variables. In the following we refer to continuum scheme in a linear approximation. In such context we will study relaxation phenomena characterized by choosing extensive variables as causa and the relative intensive variables as effect. The differential relation between this kind of variables represents the theoretical approach to describe the phenomenon. In this paper we will consider a continuous medium of order two in the sense that two relaxation phenomena predominate and consequently two relaxation times are introduced.

One method to investigate a system by laboratory measurement is to subject it to a perturbation or input and to analyze the relative response or output. This allows to introduce the so-called response function $h(t)$ which is related to specific physical characteristic of the medium and is unknown quantity. On the contrary to this approach, the theoretical one will determine the mentioned output by knowing input and response function. It is important to specify that we will refer to "linear shift invariant systems" which meaning will be cleared in the next section [1].

2 Approach to linear response theory

As already stated, if signals $f_1(t)$ and $f_2(t)$ give outputs $g_1(t)$ and $g_2(t)$ respectively, then an input $f_1(t) + f_2(t)$ gives an output $g_1(t) + g_2(t)$. A system is shift invariant, [1], if delaying an input has no effect other than to delay the output by the same amount.

For instance if a harmonic signal is applied to a linear shift invariant system then the output consists of a harmonic function at the same frequency.

Let be the impulse response $h(t)$ as the output for an input $f(t)$, it is possible to define the transfer function $H(\omega)$ as a Fourier transform of $h(t)$:

$$(2.1) \quad FT^{-}\{h(t)\} = H(\omega) = \int_{-\infty}^{+\infty} e^{-i\omega t} h(t) dt$$



Fig.1. Schematic response experiment.

It is important to notice that the input $f(t)$, output $g(t)$ and the response $h(t)$ for a linear shift invariant system are related by the convolution relation [1]:

$$(2.2) \quad g(t) = f(t) \otimes h(t)$$

where

$$(2.3) \quad f(t) \otimes h(t) = \int_{-\infty}^{+\infty} f(t_1) h(t - t_1) dt_1$$

From equation (2.2) and taking into account convolution theorem it follows:

$$(2.4) \quad G(\omega) = F(\omega)H(\omega)$$

where

$$(2.5) \quad FT^{-}\{g(t)\} = G(\omega) = \int_{-\infty}^{+\infty} e^{-i\omega t} g(t) dt$$

$$(2.6) \quad FT^{-}\{f(t)\} = F(\omega) = \int_{-\infty}^{+\infty} e^{-i\omega t} f(t) dt$$

This discussion is limited to systems for which input is harmonic. In (2.5) and (2.6) the functions $F(\omega)$ and $G(\omega)$ are interpreted as the complex amplitudes related to harmonic input and output respectively, and $H(\omega)$ as a complex transfer function which take into account the change in amplitude and phase. Obviously it is important to determine the function $h(t)$ which can be obtained easily from relation (2.2) as follows:

$$(2.7) \quad H(\omega) = \frac{G(\omega)}{F(\omega)}$$

from which

$$(2.8) \quad h(t) = FT^{+}\{H(\omega)\} = FT^{+}\left\{\frac{G(\omega)}{F(\omega)}\right\}$$

where $FT^{+}\{\dots\}$ is the inverse Fourier Transform.

Note that if $f(t)$, $g(t)$ and $h(t)$ are real quantities the transfer function $H(\omega)$ is an essentially complex quantity. It is useful for the study of many physical phenomena to obtain the response function applying a harmonic input (extensive variable) which can be represented by the real (or imaginary) part of a complex function $f(t)$ [1]:

$$(2.9) \quad f(t) = Ae^{i\omega_0 t}$$

and to analyze the output which is the real (or imaginary) part of the function $g(t)$:

$$(2.10) \quad g(t) = B(\omega_0)e^{i(\omega_0 t + \Phi(\omega_0))}$$

where A , B is the amplitudes of the oscillations and ω_0 is angular frequency. This approach can be utilized when relaxation phenomena will be studied. By calculating the Fourier Transform of input (2.9) and output (2.10) it is easy to obtain the transfer function given by the relation (2.7):

$$(2.11) \quad F(\omega) = FT^{-}\{Ae^{i\omega_0 t}\} = 2\pi\delta(\omega - \omega_0)$$

$$(2.12) \quad G(\omega) = FT^{-}\{B(\omega_0)e^{i(\omega_0 t + \Phi(\omega_0))}\} = 2\pi B(\omega_0)e^{i\Phi(\omega_0)}\delta(\omega - \omega_0)$$

and therefore

$$(2.13) \quad H(\omega) = \frac{G(\omega)}{F(\omega)} = \frac{B(\omega)}{A}e^{i\Phi(\omega)}$$

This transfer function allows us to introduce a complex quantity

$$(2.14) \quad \Omega(\omega) = \Omega_1(\omega) + i\Omega_2(\omega) = H(\omega)$$

with real and imaginary part given by:

$$(2.15) \quad \Omega_1 = \frac{B(\omega)}{A} \cos \Phi(\omega)$$

$$(2.16) \quad \Omega_2 = \frac{B(\omega)}{A} \sin \phi(\omega)$$

In a physical context these quantities are called **storage modulus** and **loss modulus** respectively. Their physical meaning depend on the amplitude A and $B(\omega)$ and are directly experimental measurable as function of angular frequency.

In agreement with the pattern "force×increase in path=intensive entity×change in extensive entity" we will show that the quantity Ω_1 is related to non dissipative phenomena and the quantity Ω_2 is related to dissipative one. In fact we can calculate the total energy W dissipated per cycle by considering (without loss of generality) the coefficient of imaginary part of (2.9) and (2.10); we have:

$$(2.17) \quad W = \int_0^T AB\omega \sin(\omega t + \phi) \cos \omega t dt$$

where $T = \frac{2\pi}{\omega}$ is the period of the harmonic input. By equation (2.17) it follows:

$$(2.18) \quad W = \frac{AB}{2} \left[\cos \phi \sin^2 \omega + \frac{\sin \phi \sin 2\omega t}{2} \right]_0^T + \frac{AB}{2} [\omega t]_0^T \sin \phi$$

which is the sum of two terms, one of which is periodic while the other increases linearly with time. Over a complete cycle the first term of (2.18) has zero value while the second is

$$(2.19) \quad W_l = \frac{AB}{2} \omega t \sin \phi$$

and represents the dissipative energy per cycle. The first term (2.18) computed over a quarter of cycle, in which $\frac{T}{4} = \frac{\pi}{2\omega}$ and the strain increase from zero to a maximum, becomes

$$(2.20) \quad W_s = \frac{AB}{2} \cos \phi$$

which is the maximum amount of energy stored in a quarter of cycle. The equations (2.19) and (2.20) can be rewritten taking into account equations (2.15) and (2.16), one obtains:

$$(2.21) \quad W_s = \frac{A^2}{2} \left(\frac{B}{A} \right) \cos \phi = \frac{A^2}{2} \Omega_1$$

$$(2.22) \quad W_l = \frac{A^2}{2} \left(\frac{B}{A} \sin \phi \right) \omega T = \frac{A^2}{2} \Omega_2 \omega T$$

This means that Ω_1 and Ω_2 are related to stored and dissipative phenomena respectively.

Our considerations are restricted to media with

$$(2.23) \quad \begin{cases} 10^5 Pa < \Omega_1(\omega) < 10^{11} Pa \\ 10^3 Pa < \Omega_2(\omega) < 10^9 Pa. \end{cases}$$

We will remember that for materials with one relaxation time the general behaviour of Ω_1 and Ω_2 is shown in Fig.2.

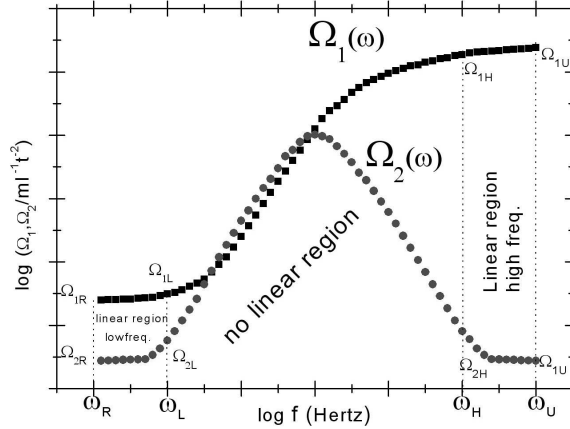


Fig.2. Generic $\Omega_1(\omega)$ and $\Omega_2(\omega)$.

Moreover, phenomena suggest to admit two range of frequencies for which a fixed medium shows a linear behaviour. Denoting by σ constant relaxation time of the medium we introduce (see Fig.2) :

- a region of low frequencies ($\omega\sigma \ll 1$):

$$(2.24) \quad \begin{aligned} \Omega_1(\omega) &\leq \Omega_{1R} + p(\omega)\Omega_{1R} \\ \Omega_2(\omega) &\leq \Omega_{2R} + r(\omega)\Omega_{2R} \\ 10^{-3} &\leq p(\omega) \leq 0.3 \\ 10^{-3} &\leq r(\omega) \leq 0.3 \end{aligned}$$

- a region of high frequencies ($\omega\sigma \gg 1$):

$$(2.25) \quad \begin{aligned} \Omega_1(\omega) &\geq \Omega_{1U} - q(\omega)\Omega_{1U} \\ \Omega_2(\omega) &\leq \Omega_{2U} + s(\omega)\Omega_{2U} \\ 10^{-3} &< q(\omega) \leq 0.3 \\ 10^{-3} &< s(\omega) \leq 0.3 \end{aligned}$$

- two values of ω : ω_L and ω_H for which both $\Omega_1(\omega)$ and $\Omega_2(\omega)$ assume the values

$$(2.26) \quad \begin{aligned} \Omega_1(\omega_L) &= \Omega_{1L} = \Omega_{1R} + 0.3\Omega_{1R} = 1.3\Omega_{1R}, \\ \Omega_2(\omega_L) &= \Omega_{2L} = \Omega_{2R} + 0.3\Omega_{2R} = 1.3\Omega_{2R}, \end{aligned}$$

and

$$(2.27) \quad \begin{aligned} \Omega_1(\omega_H) &= \Omega_{1H} = \Omega_{1U} - 0.3\Omega_{1U} = 0.7\Omega_{1U}, \\ \Omega_2(\omega_H) &= \Omega_{2H} = \Omega_{2U} + 0.3\Omega_{2U} = 1.3\Omega_{2U}. \end{aligned}$$

These values identify the regions that we call *linear regions* (see fig.1).

In particular when we write $\omega\sigma \ll 1$ or $\omega\sigma \gg 1$ we mean that it must be $\omega\sigma < 10^{-2}$ and $\omega\sigma > 10^2$, respectively.

3 Determinations of Ω_1 and Ω_2

In this paper we suppose relaxation phenomena of order two [8], [15]. A general linear relation between extensive and intensive phenomena can be expressed by the following differential equation:

$$(3.1) \quad \ddot{g} + \alpha\dot{g} + \beta g = \mu f + \nu \dot{f} + \pi \ddot{f}$$

where the dot means the derivative with respect to time and the coefficients α , β , μ , ν , π are algebraic functions of phenomenological and state coefficients which appear in the theory that one considers [11], [12]; these coefficients in the following will be expressed as functions of the moduli Ω_1 and Ω_2 which are experimentally determinable. Taking into account (2.9), (2.10), (2.13) and (2.14) one obtains the following expressions:

$$(3.2) \quad \Omega_1(\omega) = \frac{B(\omega)}{A} \cos \phi(\omega),$$

$$(3.3) \quad \Omega_2(\omega) = \frac{B(\omega)}{A} \sin \phi(\omega)$$

It is possible to express the physical meaning of (2.9) and (2.10) also by specifying A and $B(\omega)$; it is useful consider without loss generality the coefficients of imaginary part which we indicate with

$$(3.4) \quad f(t) = A \sin \omega t$$

$$(3.5) \quad g(t) = B(\omega) \sin(\omega t + \phi(\omega))$$

It is easy to get [16]:

$$(3.6) \quad g(t) = A \left[\left(\frac{B(\omega)}{A} \cos \phi(\omega) \right) \sin \omega t + \left(\frac{B(\omega)}{A} \sin \phi(\omega) \right) \cos \omega t \right]$$

By using the expression (3.4)₁ the last differential equation becomes:

$$(3.7) \quad \ddot{g} + \alpha \dot{g} + \beta g = \bar{A} \sin \omega t + \bar{B} \cos \omega t$$

where

$$(3.8) \quad \bar{A} = A(\mu - \pi \omega^2)$$

$$(3.9) \quad \bar{B} = \nu A \omega$$

The integration of differential equation (3.7) gives the following general solution that represents the intensive function:

$$(3.10) \quad g(t) = c_1 e^{r_1 t} + c_2 e^{r_2 t} + \left[\frac{\bar{A} \omega (r_2 + r_1) + \bar{B} (r_1 r_2 - \omega^2)}{(\omega^2 + r_1^2)(\omega^2 + r_2^2)} \right] \cos \omega t +$$

$$(3.11) \quad + \left[\frac{\bar{A} (r_1 r_2 - \omega^2) - \bar{B} \omega (r_1 + r_2)}{(\omega^2 + r_1^2)(\omega^2 + r_2^2)} \right] \sin \omega t$$

where c_1 and c_2 are two arbitrary integration constants, $-r_1^{-1}$ and $-r_2^{-1}$ are two relaxation times [15]. Obviously r_1 and r_2 are solution of the following characteristic equation associated to homogeneous differential equation:

$$(3.12) \quad r^2 + \alpha r + \beta = 0$$

from which one obtains the following relations:

$$(3.13) \quad r_1 + r_2 = -\beta$$

$$(3.14) \quad r_1 r_2 = \alpha$$

Since the aim is to find a functional relation between the coefficients of differential equation and the quantities expressed by equations (2.15) and (2.16) which are experimentally measurable, it is reasonable to neglect any transitory phenomenon, so that the solution (3.10) can be written as follows:

$$(3.15) \quad g(t) = g_0(\omega) \sin(\omega t + \phi)$$

in which

$$(3.16) \quad \cos \phi = \frac{\bar{A} (r_1 r_2 - \omega^2) - \bar{B} \omega (r_1 + r_2)}{\sqrt{(A^2 + B^2)(\omega^2 - r_1 r_2)^2}}$$

$$(3.17) \quad \sin \phi = \frac{\bar{A} \omega (r_2 + r_1) + \bar{B} (r_1 r_2 - \omega^2)}{\sqrt{(A^2 + B^2)(\omega^2 - r_1 r_2)^2}}$$

and

$$(3.18) \quad g_0 = \frac{\sqrt{(A^2 + B^2)[(\omega^2 - r_1 r_2)^2 + \omega^2(r_1 + r_2)^2]}}{(r_1^2 + \omega^2)(r_2^2 + \omega^2)}$$

Since equations (3.6) and (3.15) are two mathematical representations of the same phenomenon by identifying these equations one has:

$$(3.19) \quad A\Omega_1 = \frac{\bar{A}(r_1 r_2 - \omega^2) - \bar{B}\omega(r_1 + r_2)}{(\omega^2 + r_1^2)(\omega^2 + r_2^2)}$$

$$(3.20) \quad A\Omega_2 = \frac{\bar{A}\omega(r_2 + r_1) + \bar{B}(r_1 r_2 - \omega^2)}{(\omega^2 + r_1^2)(\omega^2 + r_2^2)}$$

The quantities r_1 and r_2 are experimentally known because represent the inverse of relaxation times changed in sign, therefore by solving the equations (3.19) and (3.20) the unknown coefficients \bar{A} and \bar{B} can be obtained as follows:

$$(3.21) \quad \bar{A} = A[\Omega_1(r_1 r_2 - \omega^2) + \Omega_2 \omega(r_1 + r_2)]$$

$$(3.22) \quad \bar{B} = A[\Omega_2(r_1 r_2 - \omega^2) - \Omega_1 \omega(r_1 + r_2)]$$

Since for low and high frequencies (as suggested by phenomena) the coefficient Ω_1 will assume values remaining constant which we indicate as $\Omega_{1R/H}$ where we select Ω_{1R} or Ω_{1H} (see Fig.2) for the symbol $\Omega_{1R/H}$ depending on we refer to low or high frequencies respectively, and in the equation (3.1) we can neglect time derivative, it follows that the coefficients μ and β which appear in (3.1) will assume values such that:

$$(3.23) \quad \mu = \Omega_{1R/H} \beta$$

Therefore it is obtained the unknown coefficient of differential equation (3.7) as functions of quantities which depend on the frequency and experimentally determinable. By substituting the expressions (3.8) and (3.9) in equations (3.21) and (3.22) respectively it is possible to obtain the following explicit form for the unknown quantities π and ν :

$$(3.24) \quad \pi = - \frac{[\Omega_1(r_1 r_2 - \omega^2) + \Omega_2 \omega(r_1 + r_2)] + r_1 r_2 \Omega_{1R}}{\omega^2}$$

$$(3.25) \quad \nu = \frac{\Omega_2(r_1 r_2 - \omega^2) - \Omega_1 \omega(r_1 + r_2)}{\omega}$$

Then, equations (3.23), (3.24), (3.25) together with

$$(3.26) \quad r_1 + r_2 = -\beta$$

$$(3.27) \quad r_1 r_2 = \alpha$$

represent two systems of four equations in unknown coefficients α , β , μ , ν and π of differential equation (3.7) according to consider low or high frequencies selected by $\Omega_{1R/H}$. These systems in general are incomplete, in the next section it is given some cases particular in which it is possible to complete.

3.1 Identifications of phenomenological coefficients in media with dielectric and mechanical relaxations

By comparing these results with other one obtained in [11], [12], it is possible to explicit the coefficients of differential equation (3.7) in case of media with dielectric [10] and mechanical [17] relaxations and to complete the system.

a) Dielectric Case.

Let be denote with \mathbf{D} and \mathbf{E} the induction vector and the electric field respectively; the identification of the coefficients A and B with the amplitudes $D_0 = \mathbf{D}_0 \cdot \mathbf{n}$ and $E_0 = \mathbf{E}_0 \cdot \mathbf{n}$ (\mathbf{n} is the unit normal to an arbitrary surface surrounding the charge) of induction vector \mathbf{D} and the relative electric field \mathbf{E} lead to determinations of algebraic functions of phenomenological coefficients. In this case it is enough in equation (3.7) to make the following positions:

$$(3.28) \quad D = f, \quad E = g$$

where $D = \mathbf{D} \cdot \mathbf{n}$ and $E = \mathbf{E} \cdot \mathbf{n}$, therefore the relations (3.23)-(3.27) lead to expressions of dielectric case if it is identified the real and imaginary parts of reciprocal complex dielectric constant with Ω_1 and Ω_2 as follows:

$$(3.29) \quad \begin{aligned} \Omega_1 &= \frac{\epsilon'}{\epsilon'^2 + \epsilon''^2}, \quad \Omega_2 = \frac{\epsilon''}{\epsilon'^2 + \epsilon''^2} \\ \mu &= \Omega_{1R}/H\beta \\ \pi &= -\frac{[\Omega_1(r_1 r_2 - \omega^2) + \Omega_2 \omega(r_1 + r_2)] + r_1 r_2 \Omega_{1R}}{\omega^2} \\ \nu &= \frac{\Omega_2(r_1 r_2 - \omega^2) - \Omega_1 \omega(r_1 + r_2)}{\omega} \\ \beta &= -(r_1 + r_2) \\ \alpha &= r_1 r_2 \end{aligned}$$

where ϵ' and ϵ'' are real and complex parts of complex dielectric constant. The relations (3.29) lead to that of the reference [4] if we consider $r_1 = r_2$ and identify in equation (3.7) of paper [4]:

$$(3.30) \quad \begin{aligned} b &= \frac{1 + h_1 \epsilon_0}{h_2 \epsilon_0} = \alpha, \quad a = \frac{h_0 \epsilon_0 + k_0}{h_2 \epsilon_0} = \beta \\ \mu &= \frac{h_0}{h_2 \epsilon_0}, \quad \nu = \frac{h_1}{h_2 \epsilon_0}, \quad \frac{1}{\epsilon_0} = \pi \end{aligned}$$

In this case the unknown functions are α , β , μ , ν since $\pi = \frac{1}{\epsilon_0}$ and the system is complete. Therefore these relations express a connection between the phenomenological coefficients and quantities that are directly measurable.

We have applied this method to PMMA (PolyMethylMethaCrylate) and PVC polymers in order to obtain phenomenological coefficients for such materials.

The dielectric measurements were performed by Rheometric Scientific Analyser (DETA). The analysis chamber is purged with nitrogen. The frequencies scanned the range 10 Hz - 10^5 for PMMA and 10^2 Hz - 10^6 Hz for PVC.

The PMMA and PVC samples [9, 7], in the shape of suitable disks, were previously metallised with gold to ensure a good contact with stainless steel blocking electrodes of the DETA; ε' and ε'' were determined at 90°C and 100°C , respectively. The experimental results are shown in the next figures.

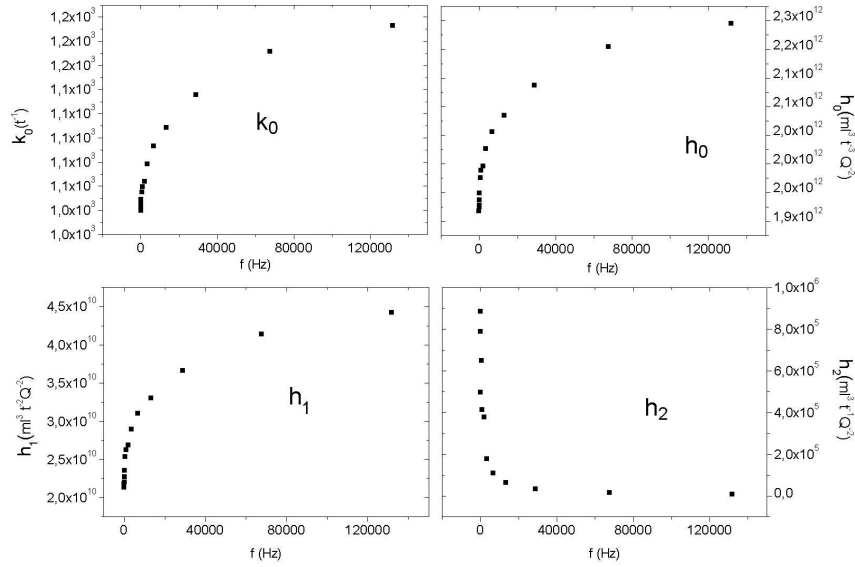


Fig.3. The trend of the phenomenological coefficients h_0, h_1, k_0 and h_2 for PolyMethylMethaCrylate (PMMA) at 90°C in the range $10 - 10^5$ Hz.

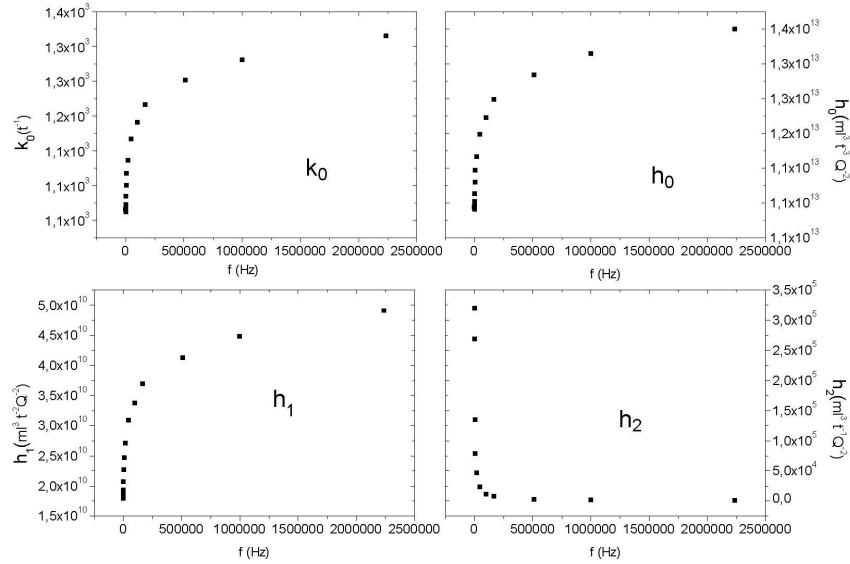


Fig.4. The trend of the phenomenological coefficients h_0, h_1, k_0 and h_2 for PolyVinylChloride (PVC) at 100°C in the range $10^2 - 10^6$ Hz.

b) The mechanical case.

Let be denoted by $\tilde{\tau}_{ik}$ and $\tilde{\epsilon}_{ik}$ the deviator of the stress and strain tensors respectively. If we consider the case for which just one component of the stress and strain is different from zero for example $\tilde{\tau}_{12}$ and $\tilde{\epsilon}_{12}$, the identifications of coefficients A and B with the amplitudes $\tilde{\tau}_{12}^{(0)}$ and $\tilde{\epsilon}_{12}^{(0)}$ of stress and strain respectively, lead to determination of an incomplete system of algebraic functions of phenomenological and state coefficients, [13], [14]. In this case it is enough to put:

$$(3.31) \quad f = \tilde{\epsilon}_{12}, \quad g = \tilde{\tau}_{12}$$

the relations (3.23)-(3.27) lead to expressions of mechanical case if it is identified real and imaginary parts G_1 and G_2 of complex dynamic modulus [19] with Ω_1 and Ω_2 , i.e.:

$$\Omega_1 = G_1, \quad \Omega_2 = G_2$$

then the system assumes the following form:

$$(3.32) \quad \begin{aligned} \mu &= \Omega_{1R/H}\beta \\ \pi &= -\frac{[\Omega_1(r_1r_2 - \omega^2) + \Omega_2\omega(r_1 + r_2)] + r_1r_2\Omega_{1R}}{\omega^2} \\ \nu &= \frac{\Omega_2(r_1r_2 - \omega^2) - \Omega_1\omega(r_1 + r_2)}{\omega} \\ \beta &= -(r_1 + r_2) \\ \alpha &= r_1r_2 \end{aligned}$$

These relations will be the same of (19.9) of reference [11], if we identify

$$\begin{aligned} \mu &= R_{(d)0}^{(\epsilon)}; \quad \nu = R_{(d)1}^{(\epsilon)}, \quad \pi = R_{(d)2}^{(\epsilon)} \\ \beta &= R_{(d)0}^{(\tau)}; \quad \alpha = R_{(d)1}^{(\tau)} \end{aligned}$$

Moreover, if the media is of order one, it follows from (3.32) the equations (3.33)-(3.36) of reference [2, 5], if it results in eq.(3.1) $\ddot{g} = 0$, $\alpha \neq 0$ and

$$\frac{\beta}{\alpha} = R_0^{(\tau)}; \quad \frac{\mu}{\alpha} = R_0^{(\epsilon)}; \quad \frac{\nu}{\alpha} = R_1^{(\epsilon)}; \quad \frac{\pi}{\alpha} = R_2^{(\epsilon)}$$

These last relations represent the expressions of coefficients in the case under consideration.

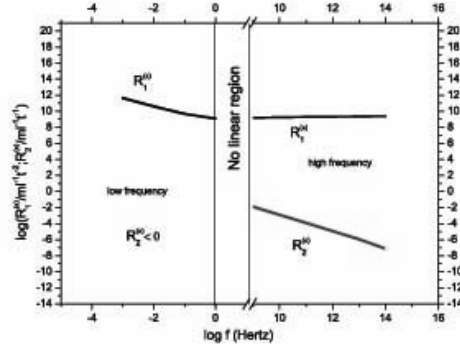


Fig.5. Poly-isoButylene; $M.w. = 10^6 g/mol$; $T_0 = 273K$.

4 Conclusions

In this paper we have unified some mechanical and dielectric aspects of relaxation phenomena, obtaining a mathematical generalization of the method developed in refs [4], [5]. It has been possible, by correlation of linear response theory and a general second order differential equation relating extensive variables (considered as cause) and the corresponding intensive (considered as effect), to show that a same procedure is able to determine, in both mechanical and dielectric cases, phenomenological and state coefficients as function of two frequency dependent quantities experimentally measurable. It is remarked that this procedure is important because it shows a common behaviour in two different fields of physics. Related results can be found in [18, 6, 3].

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