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AN APPROXIMATE EVALUATION OF THE PHENOMENOLOGICAL AND STATE COEFFICIENTS FOR VISCO-ANELASTIC MEDIA WITH MEMORY

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In a previous paper we have studied some properties of the phenomenological and state coefficients for a viscoanelastic medium of order one with memory. In that case we have used sometimes numerical approximation. In this paper we study the same coefficients by mean of approximate dynamic moduli expression for low and high frequency and we obtain some properties for viscous coefficients which are connected only on the choice of maximum value of the region of low frequency. In particular we show that the frequency corresponding to minimum value of the viscous coefficient is function of the maximum frequency value in the region of low frequency. Analogous studies have been performed in the region of high frequency. It is shown that the physical implications of the results are in agreement with experiments.

Keywords: Relaxation phenomena. Dynamic moduli. Viscoanelastic media.

1. Introduction

In the last 20 years of the last century a thermodynamic theory of non-equilibrium was proposed and developed through the introduction of tensorial internal variables characterizing the entropy production.

Various irreversible phenomena (viscoanelasticty and plasticity) have been analyzed and rheological equations (stress-strain relations) are obtained in different materials in order to establish differential equations whih describe the mechanical properties of continua under investigations [12]-[21].

Recently [1]-[11], the thermomechanical model, proposed in that theory, has been analyzed by the applications of the linear response theory in which a harmonic shear deformation is assumed as cause and the relative stress as effect and, in particular, numerical values of the phenomenological coefficients which are involved in the production of entropy are obtained and the results are compared with experimental data.

In this paper we consider shear phenomena in viscoanelasticity media and using the linear response theory the coefficients, which occur in the rheological equation of the theory, are calculated as functions of the frequency.

The trend of these coefficients is described both for the high that for the low frequency and the conformity of the results with the related experimental data obtained in the case of a polymeric material (poly-isobutilene) confirm the validity of the mathematical model.

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2. Thermodynamical model for viscoanelastic media with memory

I several papers [17]-[21] it was shown that, by introducing the internal variables which caratacterize theentropy producition and using the general method of non-equilibrium thermodynamics, the total strain $\varepsilon_{ik}\ \left(i,k=1,2,3\right)$ can be splitted in two parts:

$$\varepsilon_{ik}=\varepsilon_{ik}^{(el)}+\varepsilon_{ik}^{(in)} \tag{1}$$

where $\varepsilon_{ik}^{(el)}$ can be considered as the elastic strain tensor and $\varepsilon_{ik}^{(in)}$ is the sum of an arbitrary number, say *n*, of partial inelastic tensors.

In [20],[21] viscoanelastic media of order one ($n = 1$) were considered and by introducing the assumption that the specific entropy *s* is a functio of the specific energy *u*, the tensor of the total strain ε_{ik} and the partial inelastic strain $\varepsilon_{ik}^{(1)}$, i.e.

$$s\left(u,\varepsilon_{ik},\varepsilon_{ik}^{(1)}\right) \quad , \tag{2}$$

the following Gibbs relation is obtained [2]

$$\rho T\,ds=\rho\,du-\tau_{ik}^{(eq)}\,d\varepsilon_{ik}+\tau_{ik}^{(1)}\,d\varepsilon_{ik}^{(1)} \qquad , \tag{3}$$

where

$$T^{-1}=\frac{\partial}{\partial u}s\left(u,\varepsilon_{ik},\varepsilon_{ik}^{(1)}\right) \quad , \tag{4}$$

$$\tau_{ik}^{(eq)}=-\rho T\frac{\partial}{\partial\varepsilon_{ik}}s\left(u,\varepsilon_{ik},\varepsilon_{ik}^{(1)}\right) \ , \tag{5}$$

and

$$\tau_{ik}^{(1)}=\rho T\frac{\partial}{\partial\varepsilon_{ik}^{(1)}}s\left(u,\varepsilon_{ik},\varepsilon_{ik}^{(1)}\right) \ . \tag{6}$$

In (4)-(6) *T* is the temperature, $\tau_{ik}^{(eq)}$ is the equilibrium-stress tensor (which is of a thermoelastic nature) and $\tau_{ik}^{(1)}$ is the affinity stress tensor conjugated to $\varepsilon_{ik}^{(1)}$.

The viscous stress tensor $\tau_{ik}^{(\mathrm{vi})}$ is defined by

$$\tau_{ik}^{(\mathrm{vi})}=\tau_{ik}-\tau_{ik}^{(1)} \qquad , \tag{7}$$

where τ_{ik} is the mechanical stress tensor which occurs in the first law of thermodynamics:

$$\rho\frac{d\,u}{d\,t}=-\mathrm{div}\,\mathbf{j}^{(q)}+\tau_{ik}\frac{d\varepsilon_{ik}}{d\,t} \qquad , \tag{8}$$

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[2] we use the Einstein's convection for the indices.

where $\mathbf{j}^{(q)}$ is the heat flux.

By utilizing this last equation and the Gibbs relation (3), the entropy production $\sigma^{(s)}$ can be written in the following form:

$$\sigma^{(s)} = T^{-1}\left(-T^{-1}\mathbf{j}^{(q)} \cdot \operatorname{grad} T + \tau_{ik}^{(\mathrm{vi})}\frac{d\varepsilon_{ik}}{dt} + \tau_{ik}^{(1)}\frac{d\varepsilon_{ik}^{(1)}}{dt}\right) \quad . \tag{9}$$

Neglecting cross effects among viscous and anelastic flows in [20]-[21] it was shown that the deviator of the viscous stress tensor, $\tilde{\tau}_{ik}^{(\mathrm{vi})}$, satisfies the following phenomenological equation:

$$\tilde{\tau}_{ik}^{(\mathrm{vi})} = \eta_s^{(0,0)}\frac{d\tilde{\varepsilon}_{ik}}{dt} \quad . \tag{10}$$

In equation (10) $\eta_s^{(0,0)}$ is the shear viscosity and $\tilde{\varepsilon}_{ik}$ is the deviator of the total strain.

From (9) and applying the general method of non-equilibrium thermodynamic, the following rheological equation for viscoanelastic media of order one is determined:

$$\frac{d\,\tilde{\tau}_{ik}}{dt} + R_0^{(\tau)}\tilde{\tau}_{ik} = R_0^{(\varepsilon)}\tilde{\varepsilon}_{ik} + R_1^{(\varepsilon)}\frac{d\,\tilde{\varepsilon}_{ik}}{dt} + R_2^{(\varepsilon)}\frac{d^2\,\tilde{\varepsilon}_{ik}}{dt^2} \tag{11}$$

where

$$\begin{cases} R_0^{(\tau)} = a^{(1,1)}\eta_s^{(1,1)} \\ R_0^{(\varepsilon)} = a^{(0,0)}\left(a^{(1,1)} - a^{(0,0)}\right)\eta_s^{(1,1)} \\ R_1^{(\varepsilon)} = a^{(0,0)} + a^{(1,1)}\eta_s^{(0,0)}\eta_s^{(1,1)} \\ R_2^{(\varepsilon)} = \eta_s^{(0,0)} \end{cases} \tag{12}$$

in which $a^{(0,0)}$ and $a^{(1,1)}$ are state coefficients related to the elastic and anelastic properties of the medium, while $\eta_s^{(1,1)}$ is the coefficient related to irreversible anelastic shear phenomena. In [20]-[21], from stability considerations, it was shown that the following inequalities hold:

$$a^{(0,0)} \geq a^{(1,1)} \quad \text{and} \quad \eta_s^{(0,0)} \geq 0 \ , \ \eta_s^{(1,1)} \geq 0 \quad . \tag{13}$$

In the following sections it will be seen that these inequalities are verified from experimental data.

3. Linear response theory

In this section we consider the case in which only one component of the deviator of stress is different from zero. Let be τ this component and ε the corrispondent component of the deviator of the strain, so from (11) we have

$$\frac{d\tau}{dt}+R_0^{(\tau)}\tau=R_0^{(\varepsilon)}\varepsilon+R_1^{(\varepsilon)}\frac{d\varepsilon}{dt}+R_2^{(\varepsilon)}\frac{d^2\varepsilon}{dt^2} \tag{14}$$

In the theory of linear response [10] one assume that in a generic continuum medium subjected (at constant temperature) to an harmonic shear deformation (causa), i.e.

$$\varepsilon=\varepsilon^{(0)}\sin(\omega t) \tag{15}$$

with amplitude $\varepsilon^{(0)}$= constant and angular frequency ω, the effect is a stress with the same frequency but different amplitude $\tau^{(0)}$ and phase lag φ which depend on ω, i.e.

$$\tau(\omega)=\tau^{(0)}(\omega)\sin(\omega t+\varphi(\omega)) \tag{16}$$

From (15) and (16) we have

$$\tau(\omega)=G_1(\omega)\varepsilon^{(0)}\sin(\omega t)+G_2(\omega)\varepsilon^{(0)}\cos(\omega t) \tag{17}$$

where

$$\begin{cases} G_1(\omega)=\dfrac{\tau^{(0)}(\omega)}{\varepsilon^{(0)}}\cos(\varphi(\omega)) & \text{(storage modulus),} \\ G_2(\omega)=\dfrac{\tau^{(0)}(\omega)}{\varepsilon^{(0)}}\sin(\varphi(\omega)) & \text{(loss modulus).} \end{cases} \tag{18}$$

The quantities $G_1(\omega)$ and $G_2(\omega)$ are, respectively, connected to non dissipative and dissipative phenomena and their experimental curves are plotted in Fig.1 .

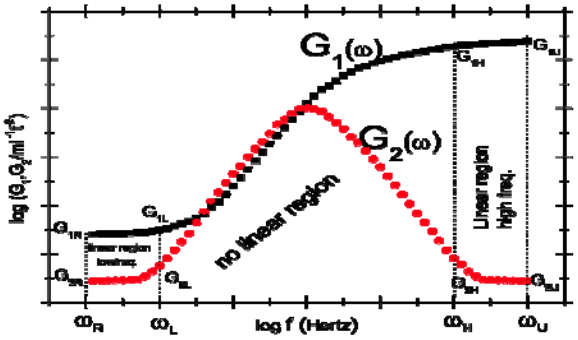

Fig. 1. Generic storage and loss moduli.

We considere two ranges: $\omega_R\le\omega\le\omega_L$ (low frequency) and $\omega_H\le\omega\le\omega_U$ (high frequency) in which we have:

$$G_{1R}=G_1(\omega_R),\quad G_{2R}=G_2(\omega_R),\quad G_{1H}=G_1(\omega_H),\quad G_{2H}=G_2(\omega_H) \tag{19}$$

and we put [9]-[11]:

- low frequency: $\omega_R\le\omega\le\omega_L$

$$\begin{cases} G_1(\omega) = G_{1R}(1.001)^r \\ G_2(\omega) = G_{2R}(1.001)^r \end{cases} \quad \text{where} \quad r = 263\frac{\omega - \omega_R}{\omega_L - \omega_R} \quad , \tag{20}$$

- high frequency: $\omega_H \le \omega \le \omega_U$

$$\begin{cases} G_1(\omega) = G_{1H}(1.001)^s \\ G_2(\omega) = G_{2H}(1.001)^s \end{cases} \quad \text{where} \quad \mathrm{s} = 357\frac{\omega - \omega_H}{\omega_U - \omega_H} \quad . \tag{21}$$

From (20) and (21) we obtain:

- low frequency: $\omega_R \le \omega \le \omega_L$

$$\begin{cases} G_{1L} = G_1(\omega_L) = 1.3G_{1R} \quad , \\ G_{2L} = G_2(\omega_L) = 1.3G_{2R} \quad , \end{cases} \tag{22}$$

- high frequency: $\omega_H \le \omega \le \omega_U$

$$\begin{cases} G_{1U} = G_1(\omega_U) = 1.43G_{1H} \\ G_{2U} = G_2(\omega_U) = 0.7\,G_{2H} \end{cases} \quad \text{or} \quad \begin{cases} G_{1H} = 0.7G_{1U} \\ G_{2H} = 1.43G_{2U} \end{cases} \quad . \tag{23}$$

In ref. [1] it was discussed that the following relations hold:

$$\begin{cases} \eta_s^{(0,0)} = \dfrac{G_{2R}}{\omega} \quad \text{(low frequency) ,} \\ \eta_s^{(0,0)} = \dfrac{G_{2R}}{\omega} \quad \text{(high frequency) .} \end{cases} \tag{24}$$

In (24) G_{2R} and G_{2U} represent the minimum values of G_2, respectively, in the range of low and high frequencies. Finally, from (14) it can observe that is $R_0^{(\tau)}$ the inverse of relaxation time for the stress and by using $(12)_1$ we put:

$$R_0^{(\tau)} = a^{(1,1)}\,\eta_s^{(1,1)} = \frac{1}{\sigma}\,, \tag{25}$$

where σ is the relaxation time.

4. Rheological coefficients

By virtue of (15) and (25), from (14) we obtain:

$$\frac{d\tau}{dt} + \frac{1}{\sigma}\tau = \alpha\sin(\omega t) + \beta\cos(\omega t)\,, \tag{26}$$

where

$$\begin{cases} \alpha = \varepsilon^{(0)}\left(R_0^{(\varepsilon)} - \omega^2 R_2^{(\varepsilon)}\right) \\ \beta = \varepsilon^{(0)}\omega\,R_1^{(\varepsilon)} \end{cases} \tag{27}$$

The solution of (26) is

$$\tau\left(t\right)=c_1\,e^{-\frac{t}{\sigma}}+\frac{\sigma\left(\alpha+\beta\,\sigma\,\omega\right)}{1+\sigma^2\,\omega^2}\sin\left(\omega t\right)+\frac{\sigma\left(\beta-\alpha\,\sigma\,\omega\right)}{1+\sigma^2\,\omega^2}\cos\left(\omega t\right), \tag{28}$$

with c_1 an arbitrary constant. Neglecting the term of $e^{-\frac{t}{\sigma}}$ (σ is very small, for instance $\sigma=10^{-7}$ s. for polyisobutilene) compared (17) and (28) one has:

$$\begin{cases}\varepsilon^{(0)}G_1=\dfrac{\sigma\left(\alpha+\beta\,\sigma\,\omega\right)}{1+\sigma^2\omega^2}\,,\\[2ex] \varepsilon^{(0)}G_2=\dfrac{\sigma\left(\beta-\alpha\,\sigma\,\omega\right)}{1+\sigma^2\omega^2}\,.\end{cases} \tag{29}$$

from which we obtain:

$$\begin{cases}\alpha=\varepsilon^{(0)}\,\dfrac{G_1-G_2\,\sigma\,\omega}{\sigma}\,,\\[2ex] \beta=\varepsilon^{(0)}\,\dfrac{G_2+G_1\,\sigma\,\omega}{\sigma}\,.\end{cases} \tag{30}$$

By virtue of $(12)_4$, (27) and (30) we have

$$\begin{cases}R_0^{(\varepsilon)}=\dfrac{G_1+\omega\,\sigma\left(\eta_s^{(0,0)}\omega-G_2\right)}{\sigma}\,,\\[2ex] R_1^{(\varepsilon)}=\dfrac{G_1\,\omega\,\sigma+G_2}{\sigma\,\omega}\,.\end{cases} \tag{31}$$

Finally, from (12), (23), (24),(25) and (31) one has

$$\begin{cases}a^{(0,0)}=\dfrac{G_1\,\sigma\,\omega\;+G_2-G_{2\,R/U}}{\sigma\,\omega}\,,\\[2ex] a^{(1,1)}=\dfrac{\left(G_2-G_{2\,R/U}+G_1\,\sigma\,\omega\right)^2}{\sigma\,\omega\left(G_2-G_{2\,R/U}\right)\left(1+\sigma^2\,\omega^2\right)}\,,\\[2ex] \eta_s^{(0,0)}=\dfrac{G_{2\,R/U}}{\omega}\,,\\[2ex] \eta_s^{(1,1)}=\dfrac{\omega\left(G_2\;-G_{2\,R/U}\right)\left(1+\sigma^2\,\omega^2\right)}{\left(G_2\;-G_{2\,R/U}+G_1\,\sigma\,\omega\right)^2}\,.\end{cases} \tag{32}$$

where $G_{2\,R/U}$ means $G_{2\,R}$ for low frequency and $G_{2\,U}$ for high frequency.

5. The coefficient of state $a^{(0,0)}$.

By virtue of (19)-(22), the relation $(32)_1$ becomes:

1) low frequency:

$$a^{(0,0)}=G_{1R}\left(1.001\right)^{r}+\frac{G_{2R}\left[\left(1.001\right)^{r}-1\right]}{\sigma\left[\dfrac{r\left(\omega_{L}-\omega_{R}\right)}{263}+\omega_{R}\right]},\quad\left(0\leq r\leq 263\right),\tag{33}$$

2) high frequency:

$$a^{(0,0)}=0.7\,G_{1U}\left(1.001\right)^{s}+\frac{G_{2U}\left[1.43\cdot\left(1.001\right)^{-s}-1\right]}{\sigma\left[\dfrac{s\left(\omega_{U}-\omega_{H}\right)}{357}+\omega_{H}\right]},\left(0\leq s\leq 357\right),\tag{34}$$

For polyisobutilene we have [15] the following characteristic values:

$$\begin{gathered}\sigma=10^{-7}\,\mathrm{sec.},\ \omega_{R}=10^{-3}\,Hz,\ \omega_{L}=1.3Hz,\ \omega_{U}=6\cdot 10^{14}\,Hz,\\ \omega_{H}=3.2\cdot 10^{14}\,Hz,\ G_{1R}=2.5\cdot 10^{5}\,Pa,\ G_{1U}=2.4\cdot 10^{9}\,Pa,\\ G_{2R}=G_{2U}=2.75\cdot 10^{4}\,Pa.\end{gathered}\tag{35}$$

and so the graphics in Fig.2

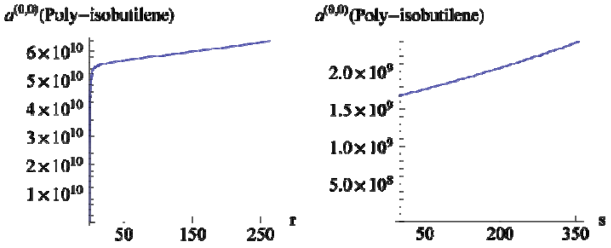

Fig. 2. The theoretical curves of coefficient $a^{(0,0)}$ for low (left) and high (right) frequencies.

The experimental curves (see [15]) for $a^{(0,0)}$ are plotted in Fig.3.

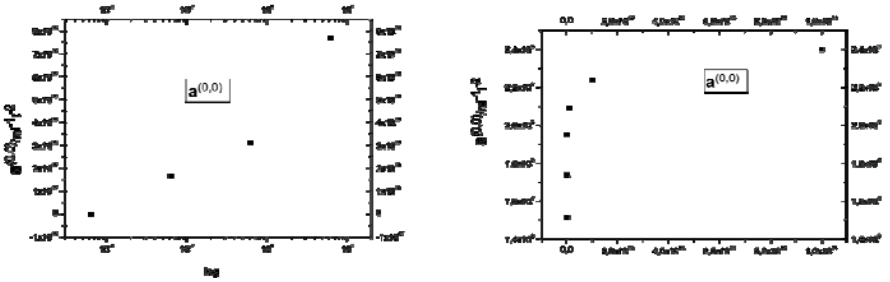

Fig. 3. Poly-isobutilene: the experimental curves of coefficient $a^{(0,0)}$ for low (left) and high (right) frequencies (M.w. = 10^{6} *g/mol*; T_0 = 273 *K*).

5. The coefficient of state $a^{(1,1)}$.

By virtue of (19)-(22), the relation $(32)_1$ becomes:

2) low frequency:

$$a^{(1,1)} = \frac{\left(G_{2R}\left[(1.001)^r - 1\right] + G_{1R}(1.001)^r\,\sigma\,\omega\right)^2}{\sigma\,\omega\,G_{2R}\left[(1.001)^r - 1\right]\left(1+\sigma^2\,\omega^2\right)}\quad , \tag{36}$$

$$\text{where}\qquad \omega = \frac{r\left(\omega_L - \omega_R\right)}{263} + \omega_R\ ,\quad \left(0 \le r \le 263\right).$$

2) high frequency:

$$a^{(1,1)} = \frac{\left(G_{2U}\left[1.43(1.001)^{-s} - 1\right] + 0.7G_{1R}(1.001)^s\,\sigma\,\omega\right)^2}{\sigma\,\omega\,G_{2U}\left[1.43(1.001)^{-s} - 1\right]\left(1+\sigma^2\,\omega^2\right)}\quad , \tag{37}$$

$$\text{where}\qquad \omega = \frac{s\left(\omega_U - \omega_H\right)}{357} + \omega_H\ ,\quad \left(0 \le r \le 263\right)$$

By using the characteristic value (35) we have the fig.4

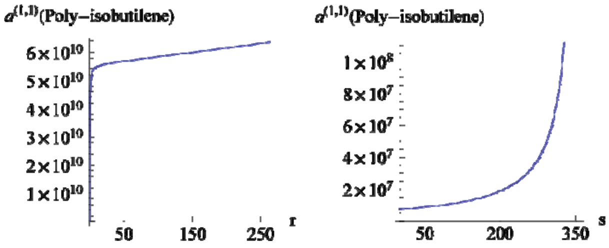

Fig. 4. The theoretical curves of coefficient $a^{(1,1)}$ for low (left) and high (right) frequencies.

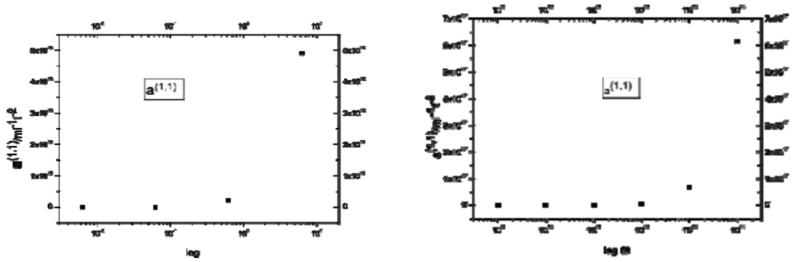

Fig. 5. Poly-isobutilene: the experimental curves of coefficient $a^{(1,1)}$ for low (left) and high (right) frequencies (M.w. = 10^6 *g/mol*; T_0 = 273 *K*).

6. The phenomenological coefficient $\eta_s^{(0,0)}$.

By virtue of (23) and (24), one has:

1. low frequency:

$$\eta_s^{(0,0)} = \frac{263 G_{2R}}{\omega_R \left(263 - r\right) + \omega_L r} , \quad \left(0 \le r \le 263\right), \tag{38}$$

2. high frequency:

$$\eta_s^{(0,0)} = \frac{357 G_{2U}}{\omega_H \left(357 - s\right) + \omega_U\, s} , \quad \left(0 \le s \le 357\right), \tag{39}$$

By putting in (40) and (41) the values (35) we have the fig. 6

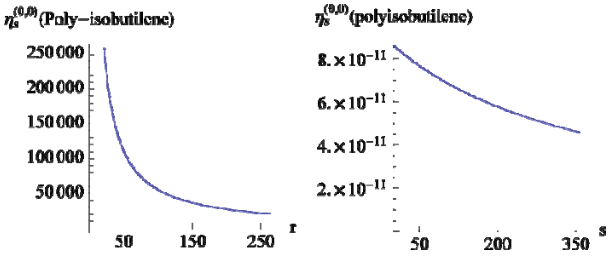

Fig. 6. The theoretical curves of coefficients $\eta_s^{(0,0)}$ for low (left) and high (right) frequencies.

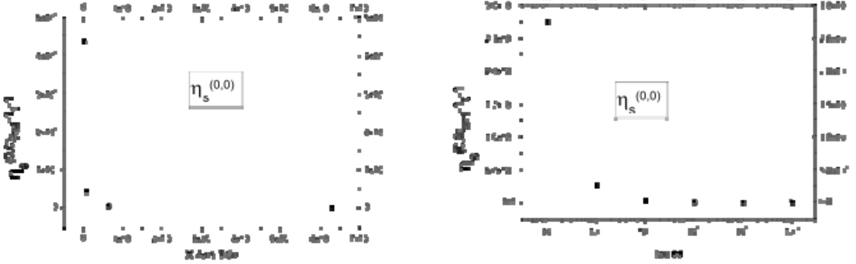

Fig. 7. Poly-isoButilene: the experimental curves of coefficient $\eta_s^{(0,0)}$ for low (left) and high (right) frequencies (M.w. = 10^6 *g/mol*; T_0 = 273 *K*).

7. The phenomenological coefficient $\eta_s^{(1,1)}$.

By virtue of (23) and (24), one has:

1. low frequency:

$$\eta_s^{(1,1)} = \frac{\omega G_{2R}\left[(1.001)^r - 1\right]\left(1+\sigma^2\omega^2\right)}{\left(G_{2R}\left[(1.001)^r - 1\right] + G_{1R}(1.001)^r\sigma\omega\right)^2}\ , \tag{40}$$

where $\omega = \dfrac{r(\omega_L - \omega_R)}{263} + \omega_R\ , \quad (0 \le r \le 263)\ .$

2. high frequency:

$$\eta_s^{(1,1)} = \frac{\omega G_{2U}\left[1.43(1.001)^{-s} - 1\right]\left(1+\sigma^2\omega^2\right)}{\left(G_{2U}\left[1.43(1.001)^{-s} - 1\right] + 0.7G_{1U}(1.001)^s\sigma\omega\right)^2}\ , \tag{41}$$

where $\omega = \dfrac{s(\omega_U - \omega_H)}{357} + \omega_H\ , \quad (0 \le s \le 357)\ .$

By putting in (40) and (41) the characteristic values (35) one obtains the fig. 8

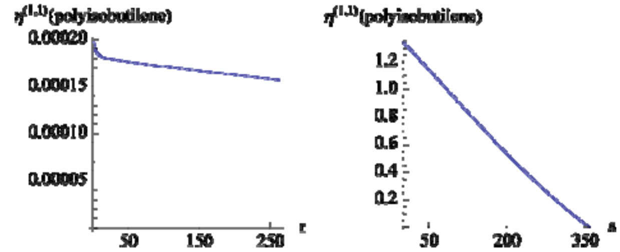

Fig. 8. The theoretical curves of coefficient $\eta_s^{(1,1)}$ for low (left) and high (right) frequencies.

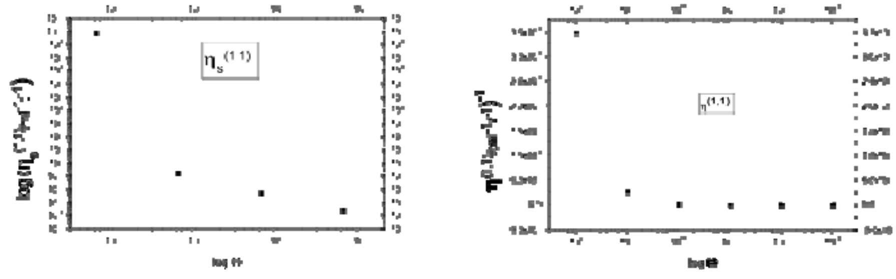

Fig. 9. Poly-isobutilene: the experimental curves of coefficient $\eta_s^{(1,1)}$ for low (left) and high (right) frequencies (M.w. = 10^6 *g/mol*; T_0 = 273 *K*).

8. Conclusions

The phenomenological coefficients which occur in the rheological equation obtained under a thermodynamic theory for viscoanelastic media are determined as functions of the dynamic moduli of linear response theory. It is shown that the theoretical results [see figures 6,4,6,8] are in agreement with experimental data in the case of polymeric material (Poly-isobutilene) [see figures 3,5,7,9]. This comparison confirms the validity of the model in the range of frequencies performed.

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