Communications to SIMAI Congress,
ISSN 1827-9015, Vol. 2 (2007)
DOI: 10.1685/CSC06157

RHEOLOGICAL COEFFICIENTS FOR MEDIA WITH MECHANICAL RELAXATION PHENOMENA

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Abstract.

In this paper the phenomenological coefficients, which occur in the rheological equations of K.C.'s theory[1]-[8] for viscoanelastic media of order one with memory, are determined as functions of an harmonic shear deformation and it is shown that these coefficients verify some inequalities which follow from principle of entropy production. Experimental confirmations are obtained for a polimeric material as the poly-isobutilene.

Keywords: (MSC 2000): rheological coefficients (35E20); mechanical relaxtion phenomena (82C21).

1. Experimental approach.

Let a generic continuum medium be subject to one-dimensional harmonic shear deformation (extensive variable = cause) of the form:

$$\varepsilon = \varepsilon_0 \sin \omega t \,, \tag{1}$$

where ε_0 and $\omega = 2\pi\nu$ are respectively the amplitude and the angular frequency of the deformation.
Of course the medium will oppose by a stress (intensive variable=effect) of the same frequency as the deformation but of different amplitude τ_0 and with a phase lag δ. These will be functions of the frequency of deformation because they result from the time necessary for molecular rearrangement and from dissipative phenomena; so we have $\tau_0 = \tau_0(\omega)$ and $\delta = \delta(\omega)$.
The form of this stress will be:

$$\tau(t) = \tau_0(\omega) \sin[\omega t + \delta(\omega)] \,, \tag{2}$$

or

$$\tau(t) = (\varepsilon_0 G_1) \sin \omega t + (\varepsilon_0 G_2) \cos \omega t \,, \tag{3}$$

where

$$G_1 = \frac{\tau_0(\omega)}{\varepsilon_0}\cos\delta(\omega)\ , \tag{4}$$

$$G_2 = \frac{\tau_0(\omega)}{\varepsilon_0}\sin\delta(\omega)\ . \tag{5}$$

The quantity $G_1(\omega)$ and $G_2(\omega)$ are called dynamic storage and loss moduli respectively and are related to **non dissipative** phenomena and to **dissipative** ones[10]-.[11]
If we don't take in account transition phenomena in which the linear response theory cannot be applied, we can observe that for sufficiently small ω the phase lag $\delta = \delta(\omega)$ vanish. The same is obtained for ω sufficiently large.
More exactly, we assume (by experimental observations) that there exist two values ω_R and ω_U such that :

$$\begin{cases} \delta(\omega) = \delta_R \cong 0 & \text{for} \quad \omega \leq \omega_R\ , \\ \delta(\omega) = \delta_U \cong 0 & \text{for} \quad \omega \geq \omega_U\ , \end{cases} \tag{6}$$

and

$$\begin{cases} G_1(\omega) = G_{1R} \cong const. & for \quad \omega \leq \omega_R\ , \\ G_1(\omega) = G_{1U} \cong const. & for \quad \omega \geq \omega_U\ , \end{cases} \tag{7}$$

Consequently, from (5), we have

$$\begin{cases} G_2(\omega) = G_{2R} \cong 0 & for \quad \omega \leq \omega_R \\ G_2(\omega) = G_{2U} \cong 0 & for \quad \omega \geq \omega_U \end{cases} \tag{8}$$

From (4) and (5) we have:

$$G_1(\omega_R) = \frac{\tau_0(\omega_R)}{\varepsilon_0}\cos\delta(\omega_R) = \frac{\tau_{0R}}{\varepsilon_0} = G_{1R}\,, \tag{9}$$

$$G_1(\omega_U) = \frac{\tau_0(\omega_U)}{\varepsilon_0}\cos\delta(\omega_U) = \frac{\tau_{0U}}{\varepsilon_0} = G_{1U}\,, \tag{10}$$

with

$$G_{1R} < G_{1U}\ .$$

In (9) and (10) G_{1R} represents the value assumed by $G_1(\omega)$ when the deformation is so slow that the medium remain near the mechanical equilibrium state (low frequency) and G_{1U} represents the value assumed by $G_1(\omega)$ when the deformation changes so rapidly that no relaxation has time to occur (high frequency). This mean that for low frequencies the medium has a meanly viscous behaviour (small G_1), while for high frequencies the medium shows an elastic behaviour (large G_1).

2. Viscoanelastic media of order one.

The Kluiteberg-Ciancio's model[1]-[8] for isotropic viscoanelastic medium of order one with memory lead to a differential equation "*stress-strain*" in which phenomenological and state coefficients appear. In one-dimensional case and neglecting cross-effect between viscous and inelastic flow, this equation is

$$\frac{d\tau}{dt} + R_0^{(\tau)}\tau = R_0^{(\varepsilon)}\varepsilon + R_1^{(\varepsilon)}\frac{d\varepsilon}{dt} + R_2^{(\varepsilon)}\frac{d^2\varepsilon}{dt^2}\,, \tag{1}$$

where

$$\begin{cases} R_0^{(\tau)} = a^{(1,1)}\eta_s^{(1,1)}; \quad R_0^{(\varepsilon)} = a^{(0,0)}\left(a^{(1,1)} - a^{(0,0)}\right)\eta_s^{(1,1)}\,, \\ R_1^{(\varepsilon)} = a^{(0,0)} + a^{(1,1)}\eta_s^{(1,1)}\eta_s^{(0,0)}; \quad R_2^{(\varepsilon)} = \eta_s^{(0,0)}\,, \end{cases} \tag{2}$$

and

$$\left[R_0^{(\tau)}\right] = t^{-1}, \left[R_0^{(\varepsilon)}\right] = ml^{-1}t^{-3}, \left[R_1^{(\varepsilon)}\right] = ml^{-1}t^{-2}, \left[R_2^{(\varepsilon)}\right] = ml^{-1}t^{-1}\,,$$

$$a^{(0,0)} \Rightarrow \text{elasticity}\,; \qquad a^{(1,1)} \Rightarrow \text{inelasticity}\,,$$

$$\eta_s^{(1,1)} \Rightarrow \text{fluidity}\,; \qquad \eta_s^{(0,0)} \Rightarrow \text{viscosity}\,.$$

where $a^{(0,0)}$, $a^{(1,1)}$ are state coefficients and $\eta_s^{(0,0)}$, $\eta_s^{(1,1)}$ are phenomenological coefficients. For entropy production principles the above coefficients must satisfy the following inequalities:

$$\begin{cases} R_0^{(\tau)} > 0\,;\; R_0^{(\varepsilon)} > 0\,;\; R_2^{(\varepsilon)} > 0\,, \\ R_1^{(\varepsilon)}R_0^{(\tau)} - R_0^{(\varepsilon)} > 0\,;\; R_1^{(\varepsilon)} - R_0^{(\tau)}R_2^{(\varepsilon)} > 0\,. \end{cases} \tag{3}$$

By substituting the relation (1) into the equation (1) we have:

$$\frac{d\tau}{dt} + R_0^{(\tau)}\tau = \left(R_0^{(\varepsilon)} - \omega^2 R_2^{(\varepsilon)}\right)\varepsilon_0 \sin\omega t + R_1^{(\varepsilon)}\varepsilon_0\omega\cos\omega t\,. \tag{4}$$

Now, putting

$$\begin{cases} \alpha = \left(R_0^{(\varepsilon)} - \omega^2 R_2^{(\varepsilon)}\right)\varepsilon_0\,, \\ \beta = R_1^{(\varepsilon)}\varepsilon_0\,\omega\,, \\ \sigma = \dfrac{1}{R_0^{(\tau)}}\,. \end{cases} \tag{5}$$

we obtain

$$\frac{d\tau}{dt} + \frac{\tau}{\sigma} = \alpha\sin\omega t + \beta\cos\omega t\,, \tag{6}$$

where σ is the relaxation time, experimentally measurable.
The solution of (6) is:

$$\tau(t) = \frac{\alpha\sigma + \beta\omega\sigma^2}{1+\omega^2\sigma^2}\sin\omega t + \frac{\beta\sigma - \alpha\omega\sigma^2}{1+\omega^2\sigma^2}\cos\omega t\,. \tag{7}$$

Equations (3) and (6) are two mathematical representations of the same phenomena therefore their equality allows to the following system of two equations:

$$(8)\qquad \begin{cases} \dfrac{\alpha\sigma + \beta\omega\sigma^2}{1+\omega^2\sigma^2} = \varepsilon_0 G_1\,, \\ \dfrac{\beta\sigma - \alpha\omega\sigma^2}{1+\omega^2\sigma^2} = \varepsilon_0 G_2\,, \end{cases}$$

from which, taking in account (5) and (3), we obtain:

$$(9)\qquad \begin{cases} R_0^{(\varepsilon)} = \dfrac{R_2^{(\varepsilon)}\omega^2\sigma + G_1 - G_2\omega\sigma}{\sigma}\,, \\ R_1^{(\varepsilon)} = \dfrac{G_2 + G_1\omega\sigma}{\omega\sigma}\,, \\ R_0^{(\tau)} = \dfrac{1}{\sigma}\,. \end{cases}$$

The (9) is an algebraic system in three equations with four unknown functions : $R_0^{(\tau)}$, $R_0^{(\varepsilon)}$, $R_1^{(\varepsilon)}$, $R_2^{(\varepsilon)}$. The quatities G_1, G_2 and σ can be experimentally measured.
To complete the system (9) we need a fourth equation that can be derived observing that the following relation, obtained by equation (1):

$$(10)\qquad \tau = \frac{R_0^{(\varepsilon)}}{R_0^{(\tau)}}\varepsilon$$

is connected with elastic and inelastic deformation . Since generally for low frequencies a mainly viscous behaviour is shown by viscoanelastic medium, the relation (10) assumes a character of small not dissipative effects and then the coefficient $R_0^{(\varepsilon)}/R_0^{(\tau)}$ can be set equal to G_{1R} which is the minimum value of G_1 in the linear region of low frequencies. This means that the linear increasing of G_1 in low frequency region don't change this ratio.
In the linear high frequencies region the medium shows an elastic behaviour and the ratio $R_0^{(\varepsilon)}/R_0^{(\tau)}$ must assume the minimum value of G_1 in this region. We have seen that this value can be set equal to G_{1H}; this means that the linear increasing of G_1 in high frequency region don't change this ratio.
We put

$$(11)\qquad G_{1R/H} = \frac{R_0^{(\varepsilon)}}{R_0^{(\tau)}}\,,$$

where we select the values G_{1R} or G_{1H} for the symbol $G_{1R/H}$ depending on we refer to low or high frequency respectively.
Using (11) we obtain the following algebraic system:

$$(12)\qquad \begin{cases} R_0^{(\varepsilon)} = \dfrac{R_2^{(\varepsilon)}\omega^2\sigma + G_1 - G_2\omega\sigma}{\sigma} \\ R_1^{(\varepsilon)} = \dfrac{G_2 + G_1\omega\sigma}{\omega\sigma} \\ R_0^{(\tau)} = \dfrac{1}{\sigma} \\ R_0^{(\varepsilon)} = R_0^{(\tau)} G_{1R/H} \end{cases}$$

The solutions of this system are:

$$(13)\qquad \begin{cases} R_0^{(\tau)} = \dfrac{1}{\sigma} \geq 0\,, \\ R_0^{(\varepsilon)} = \dfrac{G_{1R/H}}{\sigma} \geq 0\,, \\ R_1^{(\varepsilon)}\,(\omega) = G_1 + \dfrac{G_2}{\omega\sigma} \geq 0\,. \\ R_2^{(\varepsilon)}\,(\omega) = \dfrac{G_{1R/H} + G_2\omega\sigma - G_1}{\omega^2\sigma}\,. \end{cases}$$

From (13_4) we have:

$$(14)\qquad R_2^{(\varepsilon)}\,(\omega) \geq 0 \qquad \text{if} \qquad G_1 \leq G_{1R/H} \,+\, G_2\,\omega\,\sigma\,.$$

For polimeric materials as PolyIsobutilene [see Fig.1], the inequality (14) is verified only for high frequencies because in this range the dissipative phenomena don't occur.[9]

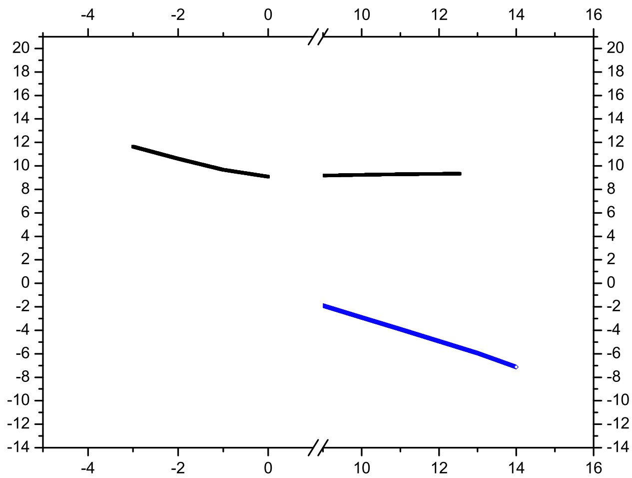

Fig. 1. Poly-isoButylene ($M.w. = 10^{-6} g/mol.$; $T_0 = 273K$; $G_{1R} \approx 10^{5.4} Pa$; $G_{1U} \approx 10^{9.38} Pa$; $\sigma \approx 10^{-5} s.$).

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