Mechanical model for relaxation phenomena in viscoanelastic media of order one

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ABSTRACT

In this paper a model for viscoanelastic isotropic material in the formalism of Kluitenberg's theory on mechanical relaxation phenomena is considered. Moreover an expression of the complex modulus is obtained to compare the results obtained with experimental data. In particular, by considering the results obtained from some authors of us in a previous paper, we shall use a different condition that allows to get the phenomenological and state coefficients both low and high frequencies. Finally, the results are applied to a supercooled m-toluidine in order to compare the experimental data and theoretical functions of frequency G_1 and G_2 so obtained.

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1. Introduction

It is well known that rheology is defined as the science of flowing materials or better it studies the relations expressing the stress as function of deformation in a given material [1]. The analysis of the mechanical properties (elasticity, anelasticity, viscosity, plasticity) in different materials is performed in the context of the continuum mechanics theories and numerous are the models introduced from several authors to describe them [2–6] by mean of representations including massless Hookean springs and Newtonian dashpots and these are used in order to establish differential equations which describe the mechanical properties of the continua under investigation [1,7].

It is well known that for a linear viscoelastic solid medium to an strain the initial stress will be proportional to the applied strain and decrease with time at a rate characterized by the relaxation time σ. This behavior is called stress relaxation. For instance one viscoelastic model, called the Maxwell model [2] predicts behavior of a spring (elastic element) being in series with a dashpot (viscous element), while the Voigt model [3] places these elements in parallel. By considering different combinations of a single spring and a single dashpot (in series or in parallel) it is possible to obtained more sophisticated models as viscoanelastic materials, etc. It is not difficult to build generalization of the single models. Very common in rheology is the so called "standard linear solid model" attributed to Zener [8]. An alternative procedure to introduce the relaxation phenomena is to subject the specimen to an harmonic strain and simultaneously measure the stress [9]. For linear viscoelastic behavior the stress and strain will both vary sinusoidally, but the strain lags behind the stress. Indeed, when a material is subjected a shear strain by imposing a sinusoidal deformation $\varepsilon = \varepsilon\sin(\omega t)$ it is possible to assume that the resulting stress τ is also a sinusoidal function $\tau = \sin(\omega t + \varphi)$. This gives rise to a modulus G_1, called storage modulus, which is $\pi/2$ out of phase with the deformation, an another part G_2, called loss modulus, in phase with the rate of deformation. The latter is related to viscous dissipation in the sample tested. This information is important because it allows to compare the storage (e.g. elasticity) and the loss (e.g. viscosity) phenomena. The angle φ represents this information through the ratio of the two moduli: $\tan(\varphi) = G_2/G_1$. These expressions are represented in the complex domain and $G = G_1 + iG_2$ is defined as the complex modulus.

The theory of the thermodynamics of irreversible processes is a good staring point for the derivation of the constitutive equations of materials with mechanical relaxation. Indeed phenomena with mechanical relaxation have been studied extensively with the methods of non-equilibrium thermodynamics [1,10]. In the context of the thermodynamics with internal variable [11] Kluitenberg proposed a theoretical model describing the behavior of an isotropic viscoanelastic medium with memory that generalizes the classical model of the continuum mechanics after suitable positions on the coefficients in the relaxation differential equation.

More recently, on the basis of Kluitenberg theory of mechanical phenomena, a method to measure experimentally the phenomenological coefficients as function of frequency and to

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verify some inequalities which occur in a thermodynamical model for viscoanelastic media with memory when it is subjected to an harmonic shear strain, was proposed [12]. The purpose of the present paper is to suggest a model for the dynamic moduli G_1 and G_2, in case of low and high frequencies, by starting from Kluitenberg's theory and to compare the results obtained with experimental data. Moreover, by taking into account the results obtained in paper [12] we shall modify one condition which leads to a form of the phenomenological and state coefficients in such a way they will be positive for any value of frequency. We shall recall briefly in the following section the fundamental aspects of Kluitenberg's theory [13–16].

2. Extended thermodynamic model for mechanical relaxations

In several papers [11] it was shown that in order to discuss same mechanical relaxation phenomena the total strain tensor ε is influenced by a "hidden" internal variable [1,13] and it has been shown that it can be splits into two parts as follows:

$$\varepsilon_{ik} = \varepsilon_{ik}^{(0)} + \varepsilon_{ik}^{(1)} \tag{2.1}$$

where $\varepsilon_{ik}^{(1)}$ is a function of an internal variable while $\varepsilon_{ik}^{(0)}$ represents the thermoelastic part of the strain if viscous stress does not occur. By taking into account this decomposition and neglecting the cross-effect between viscous and inelastic flows, the following rheological equation for viscoanelastic media of order one with memory can be written [11]

$$\frac{d\tilde{\tau}_{ik}}{dt} + R_0^{(\tau)}\tilde{\tau}_{ik} = R_0^{(\varepsilon)}\tilde{\varepsilon}_{ik} + R_1^{(\varepsilon)}\frac{d\tilde{\varepsilon}_{ik}}{dt} + R_2^{(\varepsilon)}\frac{d^2\tilde{\varepsilon}_{ik}}{dt^2} \tag{2.2}$$

where $\tilde{\tau}_{ik}$ and $\tilde{\varepsilon}_{ik}$ are the deviators of the stress and strain tensors, respectively. The coefficients of the last differential equation assume the following expressions [11]:

$$\begin{cases} R_0^{(\tau)} = a^{(1,1)}\eta_s^{(1,1)} \\ R_0^{(\varepsilon)} = a^{(0,0)}(a^{(1,1)} - a^{(0,0)})\eta_s^{(1,1)} \\ R_1^{(\varepsilon)} = a^{(0,0)} + a^{(1,1)}\eta_s^{(1,1)}\eta_s^{(0,0)} \\ R_2^{(\varepsilon)} = \eta_s^{(0,0)} \end{cases} \tag{2.3}$$

in which $a^{(0,0)}$ and $a^{(1,1)}$ are state coefficients related to elasticity and inelasticity phenomena, respectively, while $\eta_s^{(0,0)}$ and $\eta_s^{(1,1)}$ are phenomenological coefficients related to shear viscosity and fluidity, respectively. The stress–strain relation for ordinary viscous fluid, for elastic media and for Maxwell and Kelvin bodies may be regarded as special case of Eq. (2.2). We will propose to relate the phenomenological and state coefficients (2.3) to quantities that can be experimentally measurable and then to verify the following inequalities, that follows from the principle of entropy production:

$$a^{(0,0)} > 0$$

$$a^{(1,1)} > 0$$

$$\eta_s^{(1,1)} > 0$$

$$\eta_s^{(0,0)} > 0 \tag{2.4}$$

In some previous papers [12], the model proposed by Kluitenberg was used to show a method to measure experimentally the phenomenological coefficients and in particular a condition for applicability of Kluitenberg theory was determined. The mechanical measurements on polymeric material(poly-isobutilene) at different frequencies and fixed temperature was carried out and they allow to give the phenomenological coefficients as functions

of frequencies [12]. Analogously, following the procedure developed in these papers, we will study the mechanical phenomena with relaxation by imposing to the medium a mechanical perturbation considered as extensive variable (causa) and it will analyze the relative response as intensive variable (effect) as Maxwell's representation of the relaxation phenomena [1].

In the following we will consider the case in which just one component of the strain and stress is different from zero and, for sake of simplicity, we will replace the component of the strain $\tilde{\varepsilon}$ with ε and the component of the stress $\tilde{\tau}$ with τ. Let us suppose that a generic material is subjected to a sinusoidal shear strain [17]:

$$\varepsilon = \varepsilon_0 \sin\omega t \tag{2.5}$$

where $\varepsilon_0 = constant$ and $\omega = 2\pi\nu$ are, respectively, the amplitude and the angular frequency of the deformation. As consequence, the shear stress [7,18] (intensive variable=effect) will also vary sinusoidally and it will present a phase lag φ with respect to strain. This will be function of the frequency of the deformation because it results from the time necessary for molecular rearrangement and from dissipative phenomena [18]. The stress will assume the following form:

$$\tau = \tau_0(\omega)\sin[\omega t + \varphi(\omega)] \tag{2.6}$$

or

$$\tau = G_1\varepsilon_0\sin(\omega t) + G_2\varepsilon_0\cos(\omega t) \tag{2.7}$$

where the following functions

$$G_1(\omega) = \frac{\tau_0}{\varepsilon_0}\cos\varphi(\omega) \tag{2.8}$$

$$G_2(\omega) = \frac{\tau_0}{\varepsilon_0}\sin\varphi(\omega) \tag{2.9}$$

are storage and loss moduli. It is possible to show that they are directly proportional to the peak energy stored per cycle and to the energy dissipated as heat per cycle, respectively [17]. The mathematical treatment can be simplified by using a complex representation of stress and strain. Writing applied shear strain as

$$\varepsilon^* = \varepsilon_0 e^{i\omega t} \tag{2.10}$$

then the shear stress will vary according to

$$\tau^* = \tau_0 e^{(i\omega t + \varphi)} \tag{2.11}$$

The complex shear modulus is given by

$$G^* = \frac{\tau^*}{\varepsilon^*} = \frac{\tau_0}{\varepsilon_0}e^{i\varphi} \tag{2.12}$$

By introducing the expressions (2.10) and (2.11) into Eq. (2.2) and taking into account that the coefficient $R_0^{(\tau)} = 1/\sigma$, where σ is the relaxation time, and the expression (2.12), the following relation holds:

$$G^* = \frac{R_0^{(\varepsilon)} + R_1^{(\varepsilon)}i\omega - R_2^{(\varepsilon)}\omega^2}{i\omega + \frac{1}{\sigma}} \tag{2.13}$$

Separating the real and imaginary component of $G^* = G_1 + iG_2$ one obtains:

$$G_1 = \frac{\sigma\left(R_0^{(\varepsilon)} - R_2^{(\varepsilon)}\omega^2\right) + \sigma^2\omega^2 R_1^{(\varepsilon)}}{1 + \omega^2\sigma^2} \tag{2.14}$$

$$G_2 = \frac{\sigma R_1^{(\varepsilon)}\omega - \sigma^2\left(R_0(\varepsilon)\omega - \omega^3 R_2^{(\varepsilon)}\right)}{1 + \omega^2\sigma^2} \tag{2.15}$$

Following the paper [12], since the solution of the relaxation equation of stress and the expression (2.7) represents the same phenomenon, or better limiting the application to those materials