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SCALE-INVARIANCE OF TIME HIERARCHIES IN VISCOELASTIC MEDIA RELAXATION PROCESSES†

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It is shown that the time hierarchy governing shear and bulk relaxation in viscoelastic media has a fractal (scale-invariant) structure. It is noted that temporal fractality eases the modelling of viscoelastic media and leads to simple universal relaxation functions. In particular, in media with scale-invariant distributions of relaxational characteristics an algebraic relaxation law may occur, which leads to rheological models and equations of state containing fractional derivatives.

Relaxation phenomena in rheophysically complex media are associated with the slow development of rearrangement processes amongst structural units of different length-scales. (For polymers these are flexible molecules and separate segments or clusters formed by these molecules.) These processes result in delayed changes in the deformation after changes in stress (hysteresis, elastic after-effects, stress relaxation, etc.) and can be described by models of elastic bodies with internal friction and viscous bodies with elasticity [1–5]. Mechanical models of viscoelastic bodies are useful for understanding the qualitative properties of relaxation phenomena, but their use for the quantitative description of actual materials requires the construction of very complicated systems consisting of a large number of different springs and viscous elements (due to the presence of a hierarchy of structural units of different scales, leading to a hierarchy of broadly distributed relaxation times). It is clear that complicated models cannot be efficient: there are very great difficulties associated with determining the numerous relaxation parameters from experimental data, and also with solving modelling problems for moving media with a wide spectrum of relaxation times.

We shall show below that these difficulties can be circumvented by specifying the structure of the time hierarchies governing relaxation in rheophysically complex media.

1. STRESS RELAXATION IN VISCOELASTIC MEDIA

We consider a generalized Maxwellian body, the mechanical model of which is an array of links connected in parallel and consisting of springs and viscous elements connected in series. The rheology of such a body is governed by the well-known relations

$$\sigma = \sum_{n=1} \sigma_n, \quad \varepsilon = \varepsilon_n^{(1)} + \varepsilon_n^{(2)}$$

where ε is the deformation of the body, σ is the stress, $\sigma_n = E_n\varepsilon_n^{(1)} = \eta_n D\varepsilon_n^{(2)}$ is the stress for the nth element with spring stiffness E_n and coefficient of viscous damping η_n, $\varepsilon_n^{(1)}$ and $\varepsilon_n^{(2)}$ are the

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elongation of the nth spring and displacement of the nth viscous element, and $D=d/dt$ is the differentiation operator.

Suppose that at a time $t=0$ the body experiences a deformation $\varepsilon=h(t)$, where $h(t)$ is the Heaviside function. Stress relaxation is then governed by the function [1–5]

$$\Phi(t)=\sum_n E_n \exp\left(-\frac{t}{\tau_n}\right) \tag{1.1}$$

where $\tau_n=\eta_n/E_n$ is the relaxation of the nth element. The quantity E_n $(n=1, 2, \ldots)$ governs the contribution of the nth element to the total stress.

It has recently been shown that multilevel relaxation processes in many very different systems are characterized by a scale-invariant (fractal) distribution of characteristic times [6, 7]. Based on this, we shall assume that the quantities E_n and τ_n are governed by scale laws of the form

$$E_n=E_0/\lambda_1^n=E_0\exp(-n\lambda),\quad \lambda=\ln\lambda_1 \tag{1.2}$$

$$\tau_n=\tau_0\mu_1^n=\tau_0\exp(n\mu),\quad \mu=\ln\mu_1 \tag{1.3}$$

or instead of (1.3)

$$\tau_n=\tau_0 n^{\nu} \tag{1.4}$$

Thus, when there is temporal scale-invariance, ln E should decrease linearly as n increases.

The existence of such a relation is confirmed by data from [5], which gives values of E_n and τ_n for several hierarchical levels in specimens of monodisperse and polydisperse polystyrenes. According to these data the logarithm of the relaxation time also depends linearly on the level number, which could be a manifestation of the law (1.3). However, it should be noted that for high hierarchical levels (which were those considered in [5]), law (1.4) also leads to an approximately linear relation between n and τ_n, so that on the basis of the above data [5] alone one cannot give reference to any of the above possible relations for the relaxation time.

Choosing laws (1.2) and (1.3) and converting the summation (1.1) into an integral, we obtain

$$\Phi(t)=E_0\int_0^{\infty}\exp(-x\lambda)\exp\left(-\frac{t}{\tau_0}\exp(-x\mu)\right)dx$$

To find the asymptotic behaviour of this integral at long times we change the variable $z=\exp(-x\mu)$ and use Laplace's method to obtain

$$\Phi(t)\approx\frac{E_0}{\mu}\Gamma\left(\frac{\lambda}{\mu}\right)\left(\frac{t}{\tau_0}\right)^{-\lambda/\mu} \tag{1.5}$$

If, however, the relaxation time is given by (1.4), then

$$\Phi(t)\approx\exp\left(-\left(\frac{t}{\tau}\right)^{1/(\nu+1)}\right),\quad \tau=\frac{\tau_0\lambda^{-\nu}}{\nu}\left(1+\frac{1}{\nu}\right)^{-(\nu+1)} \tag{1.6}$$

Hence, the scale-invariance of the relaxation processes considerably simplifies their description and enables one to use fairly simple universal relaxation functions of the type (1.5) and (1.6).

We note that a relaxation function of the type (1.5) with exponent equal to $-1/2$ can be obtained in the Rouse and Bikki molecular theory of viscoelasticity [5]. However, that theory cannot explain the often-observed deviation of the exponent from that magnitude, and moreover, the origin of a relaxation function of the type (1.6).

The scale-invariance of the distribution of relaxation parameters can serve to explain the principle of temperature–time superposition [5] which is expressed by the constraint

$$\Phi(k(T)t) = k_1(T)\Phi_0(t) \tag{1.7}$$

where T_0 is some characteristic temperature, $\Phi(t)$ and $\Phi_0(t)$ are relaxation functions at temperatures T and T_0, and k and k_1 are temperature-dependent coefficients ($k(T_0) = k_1(T_0) = 1$). Indeed, if we assume that the exponents λ and μ do not depend on temperature, then from (1.5) we obtain (1.7) when

$$k = \tau_0(T)/\tau_0(T_0), \quad k_1 = E_0(T)/E_0(T_0)$$

As an example we considered the stress relaxation curve in a sample of monodisperse polystyrene [5]. Calculations showed that this curve was described quite well by Kohlrausch's law (1.6) when $(1/\nu+1) = 0.50$.

2. RHEOLOGICAL MODELS WITH FRACTIONAL DERIVATIVES

We will now consider a viscoelastic body represented by a series of Voight bodies (links consisting of springs and viscous elements connected in parallel). Then the connection between the rate of deformation and the stress is governed by the relations [1, 3]

$$D\varepsilon(t) = \frac{\sigma(t)}{\eta} + \int_0^t \psi(t-\xi)d\sigma(\xi), \quad \psi(t) = \sum_n \frac{1}{\eta_n}\exp\left(-\frac{t}{\tau_n}\right) \tag{2.1}$$

As above, we assume the presence of scale-invariance in the distribution of the relaxation parameters

$$\eta_n = \eta_0 \exp(\lambda' n), \quad \tau_n = \tau_0 \exp(n\mu)$$

Then (see (1.5))

$$\psi(t) \approx Lt^{-\gamma_1}, \quad L = \Gamma(\gamma_1)\tau_0^{\gamma_1}/(\eta_0\mu) \tag{2.2}$$

and (2.1) can be rewritten in the form

$$D\varepsilon(t) = \eta^{-1}\sigma(t) + \alpha D^{-\gamma} D\sigma(t) \tag{2.3}$$

$$\gamma = 1-\gamma_1, \quad \gamma_1 = \lambda'/\mu, \quad \alpha = L\Gamma(\gamma)$$

$$D^{-\gamma}f(t) \equiv \frac{1}{\Gamma(\gamma)}\int_0^t (t-\xi)^{\gamma-1} f(\xi)d\xi$$

(where $D^{-\gamma}f(t)$ is a fractional derivative of order $-\gamma$.)

Taking $E_n = E_0\exp(-\lambda n)$ we obtain

$$\tau_n = \eta_0 E_0^{-1}\exp((\lambda'+\lambda)n)$$

from which $0<\gamma_1<1$, $0<\gamma<1$.

Hence the presence of temporal scale-invariance leads to the need to use rheological models with fractional derivatives. Note that similar models (arising from other considerations) have been previously introduced (e.g. [3, 4, 8]). The result obtained here also has connections with [9], where it was shown that the temporal self-similarity of the processes leads to equations with fractional derivatives. We emphasize that a rheological law with fractional derivatives has been obtained for models that only include different spring and viscous elements, unlike in [4], where the existence of an independent type of deformation was postulated: a highly-flexible deformation which cannot be reduced to a sum of elasticity and viscous friction.

3. RELAXATION PROCESSES FOR BULK DEFORMATION

We will now consider relaxation processes for bulk deformation. It has been noted in a number of experiments [10, 11] that if a vessel is filled with a structured liquid (for example, petroleum with asphalt-tar impurities), and then pressurized and hermetically sealed, the pressure in the vessel falls slowly to some steady-state value. Relaxation processes of this kind are associated with rearrangements of macromolecules and clusters formed by them. When rapidly compressed, such a system undergoes an instantaneous elastic deformation whose magnitude is governed by the coefficient of bulk elasticity of the medium in its initial state. There is then a slow rearrangement of structural units of differing complexity, with the medium becoming denser and its volume becoming smaller, with, as a consequence, a fall in pressure. Assuming the structural units to be viscoelastic elements, one can obtain

$$-\frac{\delta V(t)}{V_0}=\beta_0\delta p(t)+\int_0^t \psi_1(t-\xi)D\delta p(\xi)d\xi \tag{3.1}$$

$$\psi_1(t)=\beta'\sum_{n=1}^{\infty}\frac{1}{E_n}\left[1-\exp\left(-\frac{t}{\tau_n}\right)\right]$$

where $-\delta V$ is the reduction in the volume of the medium when the pressure has been increased by δp, V_0 is the initial volume, $\psi_1(t)$ is the relaxation function, and β' is a quantity governing the change in volume due to the shifting of structural elements.

Differentiating (3.1) with respect to time we obtain the equation of state of a viscoelastic medium in the form

$$\frac{1}{\rho_0}D\rho=\beta_0 Dp+\int_0^t \psi(t-\xi)Dp(\xi)d\xi$$

$$\left(\frac{1}{\rho_0}D\rho=\frac{1}{V_0}D\delta V\right),\quad \psi(t)=\beta'\sum_{n=1}^{\infty}\frac{1}{\eta_n}\exp\left(-\frac{t}{\tau_n}\right)$$

(where ρ_0 is the density of the medium).

We again take scale laws of the form (1.2) and (1.3) and retaining the previous notation, we obtain, like (2.3)

$$\rho_0^{-1}D\rho(t)=\beta_0(Dp+\beta_1 D^{-\gamma}Dp),\quad \beta_1=\beta'\Gamma(\gamma_1)\tau_0^{\gamma_1}/(\mu\beta_0\eta_0) \tag{3.2}$$

Thus the equation of state of a viscoelastic medium can also contain fractional derivatives. (The order of the derivatives in (2.3) and (3.2) can differ, but for the time being we will keep the same notation.)

As an example, consider data from the following experiment performed by G. M. Panakhov. A high-pressure thermostatically-controlled container was filled with structured petroleum, containing impurities in the form of paraffins and tar. After being filled the container was thoroughly evacuated and the pressure was then abruptly raised by rapidly injecting into the container a small quantity of petroleum from a PVT bomb. After this the container was closed and the pressure drop was measured as a function of time. The results of one such experiment, during which the pressure in the closed container fell from 5 to 4.64 MPa, are shown below:

$t\times10^{-2}$, s	0	1.5	3	6	15	30	60
P, MPa	5.00	4.91	4.85	4.78	4.72	4.68	4.65

We assume that the pressure relaxation in the container is described by Eq. (3.2). To identify this model we use the operational methods of [12, 13]. Since the petroleum density does not change during the relaxation process

$$\delta\rho(t)=\rho_0\beta_0\delta p(0)h(t)$$

Putting

$$\delta p(t)=\delta p(0)[h(t)-1]+\delta p_1(t)$$

and performing a Laplace transformation, we find from (3.2) that

$$\ln\left(\frac{1}{sU}-1\right)=\ln\beta_1-\gamma\ln s,\quad U=\frac{1}{\delta p(0)}\int_0^\infty \exp(-st)\delta p_1(t)dt \tag{3.3}$$

where $p_1(t)$ is the pressure measured in the experiment. Thus, if the bulk relaxation is in fact described by model (3.2), the pressure variation curve should become straight in $Y=\ln(1(sU)-1)$, In s coordinates. To verify this fact various values of s were specified in the interval $[5/T;\ 20/T]$ (T being the period of time over which the experimental curve was measured; in the present case it was $T=6000$ s), and the transform of the function $\delta p_1(t)$ was calculated using the formula

$$U(s)=\frac{\delta p(0)}{s}+\frac{1}{s^2}\sum_i\left[\frac{\delta p_1(t_{i+1})-\delta p_1(t_i)}{t_{i+1}-t_i}\left(e^{-st_i}-e^{-st_{i+1}}\right)\right]$$

The calculations show that the relaxation curve does indeed straighten in these coordinates. The slope of the line was found to be $\gamma=0.78$.

The results obtained can be used to derive equations of motion for relaxing media. We will first consider the motion of a structured relaxing liquid in a tube of radius R. We write the rheological equation of the medium in the form (cf. (2.3))

$$-\frac{\partial U}{\partial r}=\frac{\sigma}{\eta}+\alpha D^{-\gamma}\frac{\partial\sigma}{\partial t} \tag{3.4}$$

where $v(r, t)$ is the component of the velocity along the tube axis, σ is the shear stress, and η is the viscosity of the medium. Averaging Eq. (3.4) over the tube cross-section one can obtain the following equation of motion within the framework of the quasisteady approximation [14]

$$\rho_0\left(\frac{\partial w}{\partial t}+2aw\right)=-\left(\frac{\partial p}{\partial x}+\alpha D^{-\gamma}\frac{\partial^2 p}{\partial x\partial t}\right),\quad 2a=\frac{8\eta}{\rho_0 R^2} \tag{3.5}$$

where w is the velocity averaged over the cross-section, and $\partial p/\partial x$ is the pressure gradient along the tube axis.

Using (3.2), the equation of continuity

$$\partial p / \partial t = -\rho_0 \partial w / \partial x$$

can be written in the form

$$\frac{\partial p}{\partial t} + \beta_1 D^{-\gamma'} \frac{\partial p}{\partial t} = -\rho_0 c_0^2 \frac{\partial w}{\partial x}, \quad c_0 = (\beta_0 \rho_0)^{-1/2} \tag{3.6}$$

where c_0 is the "instantaneous" velocity of sound in the medium. Eliminating the velocity from (3.5) and (3.6), we obtain the equation of motion of a relaxing fluid in the form

$$(D+2a)(Dp + \beta_1 D^{-\gamma'} Dp) = c_0^2 (1 + \alpha D^{-\gamma} D) \frac{\partial^2 p}{\partial x^2} \tag{3.7}$$

The filtration equation can be obtained by ignoring the inertial term $\partial w/\partial t$ in (3.7) and putting $1/(2a) = k/\eta$, where w is now the filtration velocity and k is the permeability of a porous medium. Following the usual methods (for example, of [15]), we obtain in this case the following analogue of (3.7)

$$Dp + \beta_1 D^{-\gamma} Dp = \kappa(1 + \alpha D^{-\gamma} D)\,\mathrm{div}(\mathrm{grad}\, p), \quad \kappa = k/(\eta m \beta_0)$$

where κ is the coefficient of piezoconductivity and m is the porosity.

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