DISTRIBUTION OF RELAXATION TIMES IN VISCOELASTIC INHOMOGENEOUS MEDIA

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Applying Ter Haar, Alfrey, and Gross approximations, we have calculate distributions of viscoelastic relaxation times for relaxation functions given by Schiessel et al. Fractional derivatives calculus has been applied to Maxwell model of viscoelasticity and it has been proved by means of the Gross formula that fractional derivatives brought about appearance of continuous spectrum of visoelastic relaxation frequencies. The width of this spectrum depends on the value of the fractional parameter α .

Keywords : viscoelasticity, relaxation times, fractional calculus.

1. INTRODUCTION

In dynamics of complex materials, relaxation processes deviating from the classical exponential Debye behaviour are often encountered [1-4] . Such situation is in the case of stress relaxation in viscoelastic materials, polymers, in critical gels, in the charge carrier transport in amorphous semiconductors, in dielectric relaxation, in attenuation of seismic waves, in transient photoconductivity etc. Viscoelastic phenomena are experimentally studied by two methods. In the first one stress changes of materials caused by applied harmonic changes of strain is studied , and one obtains as a result the complex elastic modulus of investigated material. In the second one time changes of stress in material , produced by unit jump of strain in the form of Heaviside functions, are investigated , and as a result the so called relaxation function is obtained. According to the work [1, 2, 4] , experimentally observed decay of the relaxation function with increase in time that elapsed from the moment of unit jump of strain, may often be described by the following formulae:

$$\Phi_1(t) \propto \exp(-(t/\mu)^{\alpha}) \qquad (1)$$

or

$$\Phi_2(t) \propto (t/\mu)^{-\beta} \tag{2}$$

where α and β are fractional numbers from the range: $0<\alpha<1$, $0<\beta<1$. More complex behaviours of viscoelastic materials may also be exhibited [2], typified by changes from one form of power law relaxation function decay to another , for instance if motion of polymer segments are restricted by finite extensions of the investigated system.

2. THE RELAXATION FUNCTION AS A LAPLACE INTEGRAL

The relaxation function is a continuous, decreasing function which for $t \to \infty$ diminishes to 0 .Thus it can be represented in integral form as [4] :

$$\Phi(t) = \Phi(0) \int_0^\infty N(\tau) \exp(-t/\tau) d\tau \tag{3}$$

where $N(\tau)$ denotes the distribution function of relaxation times. Introducing the relaxation frequency $s = 1/\tau$, and the frequency function:

$$M(s) = \Phi(0) N(1/s)/s^2 \tag{4}$$

eq. (3) is transformed into the form of Laplace integral:

$$\Phi(t) = \int_0^\infty M(s) \exp(-ts) ds \tag{5}$$

3. APPROXIMATION METHODS FOR THE DETERMINATION OF THE RELAXATION SPECTUM

The relaxation spectrum can be determined rigorously by means of the inverse Laplace transform of the above formula only if the relaxation function is known in the form of analytical expression over the entire time scale from 0 to ∞ . But not all values contribute to the integral to the same extent. The weighing factor $\exp(-ts)$ reduces the influence of the far-off end of the spectrum upon a given $\Phi(t)$. Measurements of relaxation function provide however only a set of experimental data, plots in a graph, which extend over a limited interval of time only. For this reason, many attempts were made to obtain an approximation methods for determination the relaxation spectrum [4] . In this work we have considered three of them. In the Alfrey approximation, it is assumed that $\exp(-ts)$ in the formula (5) may be approximated by:

$$\exp(-ts) = 1 \quad \text{for} \quad s \le 1/t \qquad (6)$$

$$\exp(-ts) = 0 \quad \text{for} \quad s > 1/t$$

In this approximation the eq. (5) takes the form:

$$\Phi(t) = \int_0^{1/t} M(s)ds \qquad (7)$$

and after a simple calculation, one obtains from eq. (4) and eq. (7) the following formula:

$$\Phi(0)N(\tau) = -\frac{d\Phi(\tau)}{d\tau} \qquad (8)$$

Ter Haar has assumed the following approximation:

$$s\exp(-ts) = \delta(s - 1/t)/t^2 \qquad (9)$$

which applied to eq. (5) changes it to the form:

$$\Phi(t) = \int_0^{\infty} M(s)(\delta(s - 1/t)/t)ds = M(1/t)/t \qquad (10)$$

from which it results the following formula:

$$\Phi(0)N(\tau) = \frac{\Phi(\tau)}{\tau} \qquad (11)$$

According to Gross, the following assumption may be made:

$$\exp(-ts) = 1 - ts \quad \text{if} \quad s \le 1/t \qquad (12)$$

$$\exp(-ts) = 0 \quad \text{if} \quad s > 1/t$$

under which the eq. (5) takes the form :

$$\Phi(t) = \int_0^{1/t} M(s)(1 - ts)ds \qquad (13)$$

and after simple calculations it results from eq. (4) and eq. (13):

$$\Phi(0)N(\tau) = \tau\frac{d^2\Phi(\tau)}{d\tau^2} \qquad (14)$$

Using the above mentioned approximations , distributions of relaxation times $N(\tau)$ have been estimated for media in which relaxation functions are given by formulae (1) and (2) .In Fig. 1 distributions of relaxation times for relaxation function given by the formula (1) have been plotted for $\alpha = 0.7$ and for the three approximations of Alfrey , Ter Haar and Gross .

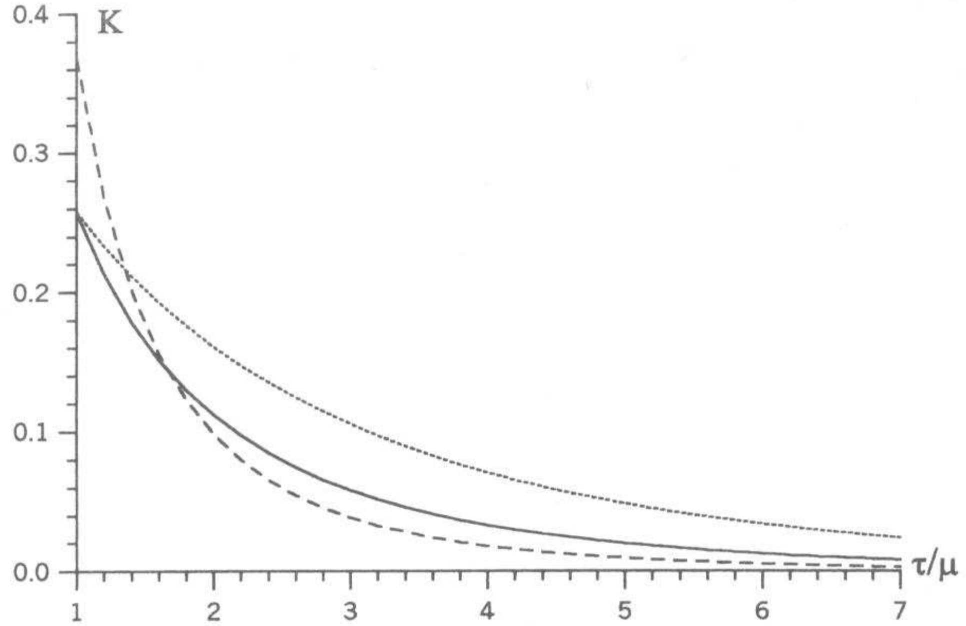

Fig. 1. Distribution of relaxation times $K = \Phi(0)N(\tau)\mu$ for relaxation function given by formula (1) and $\alpha = 0.7$. The heavy line – Alfrey's approximation , the dashed line – approximation of Ter Haar , the pointed line – approximation of Gross.

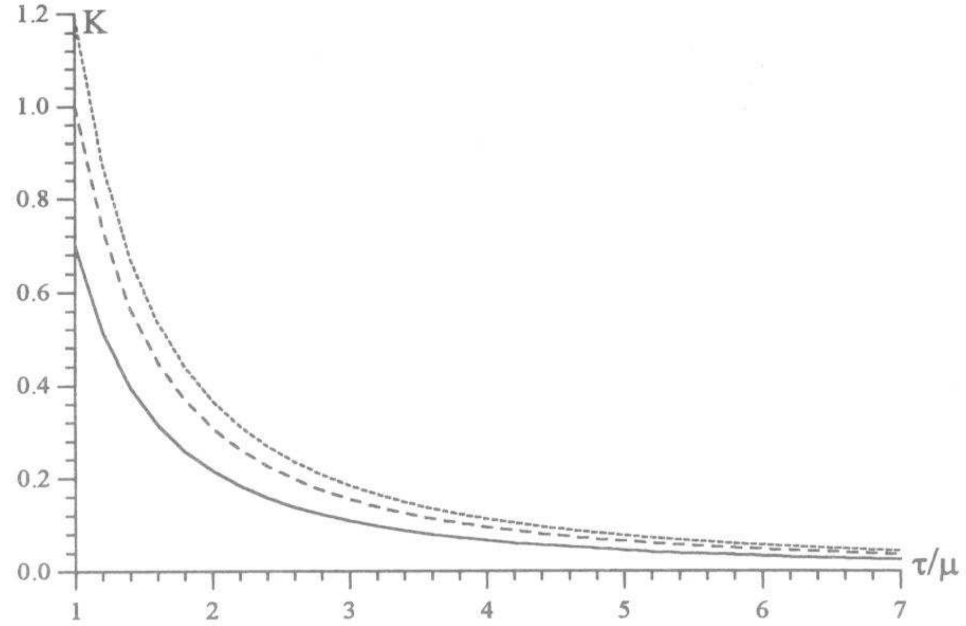

Fig. 2. Distribution of relaxation times $K = \Phi(0)N(\tau)\mu$ for relaxation function given by formula (2) and $\alpha = 0.7$. The heavy line – Alfrey's approximation , the dashed line – approximation of Ter Haar , the pointed line – approximation of Gross.

4. RELATIONS BETWEEN COMPLEX MODULUS FUNCTION, RELAXATION FUNCTION AND RELAXATION SPECTRUM

The complex modulus function is defined as a stress developed under a sinusoidally varying strain, applied for a very long time so that the transient has disappeared [4]:

$$E^*(i\omega) = i\omega \int_0^\infty \Phi(\tau)\exp(-i\omega\tau)d\tau + Eo \tag{15}$$

where ω denotes the cyclic frequency of the varying strain, $i^2 = -1$, and E_0 is the static elastic modulus. After introducing the formula (5) for the relaxation function, and integrating over τ , eq. (15) takes the form:

$$E^*(i\omega) = i\omega \int_0^\infty M(s)/(s+i\omega)ds + Eo \tag{16}$$

The following formulae for the real and imaginary components of the dynamical elastic modulus result from eq. (16) :

$$E_1(\omega) = \int_0^\infty M(s)\omega^2/(\omega^2+s^2)ds \tag{17}$$

$$E_2(\omega) = \int_0^\infty M(s)\omega s/(\omega^2+s^2)ds \tag{18}$$

Using eq.(16) Gross [4] has derived the following formula connecting the complex modulus function and distribution function of relaxation frequencies:

$$M(\omega) = \pm\frac{1}{\pi\omega E_\infty}\operatorname{Im} E^*(\omega\exp(\pm i\pi)) \tag{19}$$

where Im denotes the imaginary part of the complex modulus. It may be demonstrated [4] that the frequency function $M(\omega)$ may also be calculated from the real and imaginary components of the dynamical elastic modulus, by means of the following formulae:

$$M(\omega) = \pm\frac{2}{\pi\omega E_\infty}\operatorname{Im} E_1(\omega\exp(\pm i\pi/2)) \tag{20}$$

$$M(\omega) = \frac{2}{\pi\omega E_\infty}\operatorname{Re} E_2(\omega\exp(\pm i\pi/2)) \tag{21}$$

5. DISTRIBUTION OF RELAXATION FREQUENCIES IN GENERALIZED FRACTIONAL MODELS OF VISCOELASTICITY

We have calculated the complex elastic modulus, using the generalized Maxwell model, which in fractional derivatives [5, 6] is described by the following formula:

$$\sigma + \tau^{\alpha} D^{\alpha}\sigma = E_{\infty}\tau^{\alpha} D^{\alpha}\varepsilon \tag{22}$$

where σ is the stress, ε is the strain, $\tau = const$ is the relaxation time of the simple Maxwell model, E_{∞} - instantaneous modulus of elasticity, and $D^{\alpha}\sigma$, $D^{\alpha}\varepsilon$ denote the Riemann-Liouville derivatives of order α , where $0 < \alpha \leq 1$ [6-10]. Applying the Fourier transformation to eq. (22), we have obtained the following expression for the complex modulus:

$$E^{*}(i\omega) = E_{\infty}\frac{\tau^{\alpha}(i\omega)^{\alpha}}{1+\tau^{\alpha}(i\omega)^{\alpha}} \tag{23}$$

which real and imaginary parts are:

$$E_1(\omega) = E_{\infty}\frac{\cos(\alpha\pi/2) + (\omega\tau)^{\alpha}}{(\omega\tau)^{-\alpha} + (\omega\tau)^{\alpha} + 2\cos(\alpha\pi/2)} \tag{24}$$

$$E_2(\omega) = E_{\infty}\frac{\sin(\alpha\pi/2)}{(\omega\tau)^{-\alpha} + (\omega\tau)^{\alpha} + 2\cos(\alpha\pi/2)} \tag{25}$$

Introducing $\omega \to \omega\exp(i\pi/2)$ in the formula (25), according to the Gross formula (21), we have obtained the following expression:

$$E_2(\omega\exp(i\pi/2)) = E_{\infty}\frac{[(\omega\tau)^{\alpha} + (\omega\tau)^{-\alpha} + 2]\sin(\alpha\pi/2)\cos(\alpha\pi/2) - i[(\omega\tau)^{\alpha} - (\omega\tau)^{-\alpha}]\sin^2(\alpha\pi/2)}{[(\omega\tau)^{\alpha} + (\omega\tau)^{-\alpha} + 2]^2\cos^2(\alpha\pi/2) + [(\omega\tau)^{\alpha} - (\omega\tau)^{-\alpha}]^2\sin^2(\alpha\pi/2)}$$

from which it follows according to eq. (21):

$$M(\omega) = \frac{\tau[(\omega\tau)^{\alpha} + (\omega\tau)^{-\alpha} + 2]\sin(\alpha\pi)}{\pi(\omega\tau)\{[(\omega\tau)^{\alpha} + (\omega\tau)^{-\alpha} + 2]^2\cos^2(\alpha\pi/2) + [(\omega\tau)^{\alpha} - (\omega\tau)^{-\alpha}]^2\sin^2(\alpha\pi/2)\}} \tag{26}$$

As one can see, fractional derivatives in Maxwell model generate distribution of relaxation frequencies. We have proved that the width of this spectrum depends on the value of the fractional parameter α , describing order of the fractional derivatives. Graphs of the distribution function $M(\omega)$ have been presented in Fig. 3 for various values of the parameter α . If $\alpha \to 1$, the distribution of relaxation frequencies changes to a single component.

6. CONCLUSIONS

If the relaxation function is given in the form of Laplace integral, the relaxation spectrum may be simply estimated using the approximate methods of Alfrey, Ter Haar and Gross. It has been stated that the Alfrey and Ter Haar methods gave relaxation spectra very close one to another if the relaxation function is given by the formula (1). In the case of the relaxation function given by the expression (2) approximations of Ter Haar and of Gross give close results.

It has been stated that application of fractional derivatives in models of viscoelastic materials generates continuous spectrum of relaxational frequencies. As one can see from the Fig. 3 ,the distribution of this spectrum depends on the fractional parameter α describing the order of derivatives .

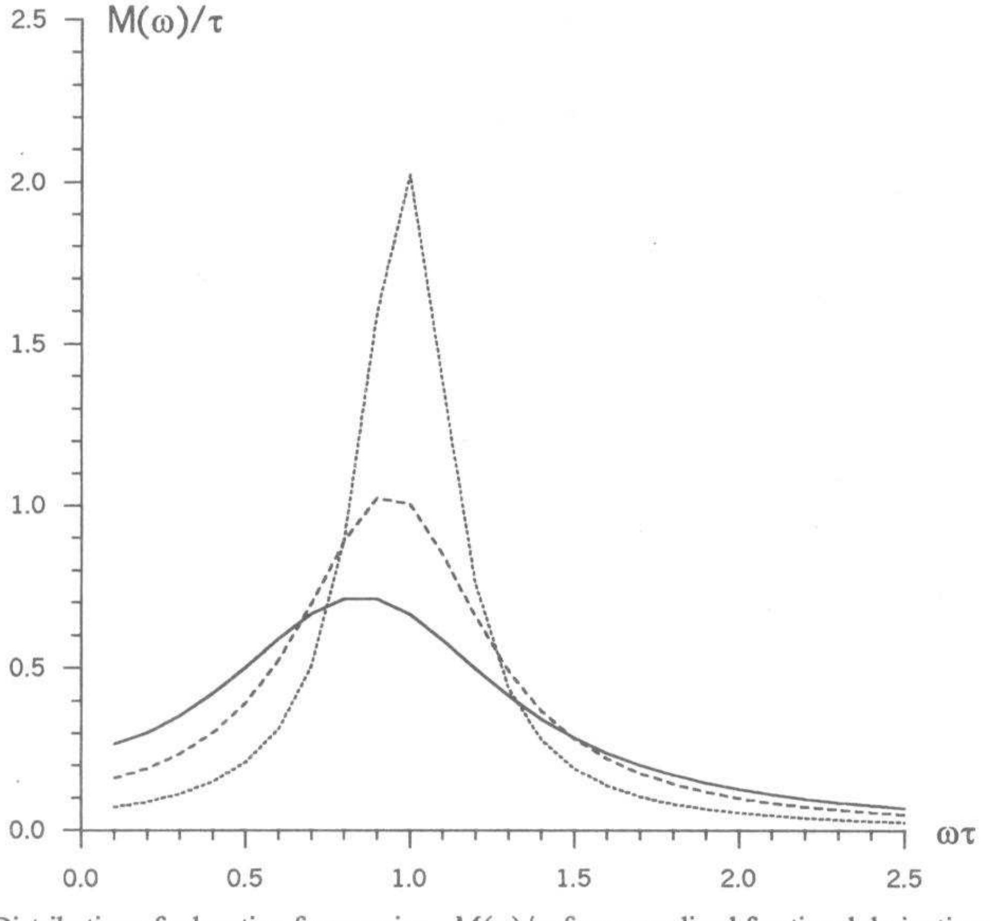

Fig. 3. Distribution of relaxation frequencies $M(\omega)/\tau$ for generalized fractional derivatives Maxwell model for various values of the fractional derivative order α . The heavy line $\alpha = 0.85$, the dashed line $\alpha = 0.90$, the pointed line $\alpha = 0.95$. τ denotes the relaxation time of a simply Maxwell model.

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