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On fractional modelling of viscoelastic foams.

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Abstract

Empiric models have been introduced to describe frequency dependence of dielectric permittivity. Simple exponential models are often not satisfactory, while advanced non-exponential models (usually referred as "anomalous relaxation") are commonly required to better explain experimental observations of complex systems. For viscoelastic materials, the so-called fractional derivatives models are powerful for both dynamic and loss moduli prediction. In this paper, the analysis of the main models used in the characterization of dielectric and viscoelastic materials such as five-parameter fractional Zener model and empiric Havriliak-Negami model are analysed. The fractional shape parameters describing the symmetric and asymmetric broadening of the complex modulus don't have the same influence in low and high frequencies. In contrast to the five-parameter Zener model, the empiric model asymmetry parameter has an influence on complex modulus at low frequencies comparing to the loss modulus peak frequency. A no resonance technique based on a forced vibrations procedure is used to investigate the frequency dependent complex shear modulus of a polyurethane foam, not influenced by its fluid phase, in the range 0.1-500 Hz. It is shown that the Havriliak-Negami model can predict the frequency dependence for a wide frequency range.

Keywords: polyurethane foams, fractional models, dynamic modulus, loss modulus, wide frequency range

1. Introduction

Porous materials like polymer foam and glass wool are widely used for noise control in several engineering activities such as aeronautics and automotive industries. Their properties are two-fold: sound absorption and damping of the nearby structure [1].

Viscoelastic behavior was analogically modelled by ideal springs and dashpots representing respectively the elasticity and the dissipation phenomenon during material deformation. The most popular models (Kelvin–Voigt, Zener etc.), cannot accurately describe qualitatively the dynamic behavior of real materials. The reason for the inaccurate behavior of the spring–dashpot models can be found in the stress–strain relationship defined in the time-domain by a linear differential equation of integer order. However, this differential equation can be generalized by replacing the integer order derivatives with fractional order ones. In this way the spring–dashpot models can be generalized, resulting in frequency curves having smaller slopes than those of the frequency curves relating to the original models. The models thus developed are called generalized, or fractional derivative models. They were used to characterize the rheological behavior of linear viscoelastic systems by a number of authors [2-7]. Moreover, it has been established that the fractional derivative model having only four-parameters can be used to describe the variations of dynamic properties in a wide frequency range. The Fractional Zener Model (FZM) has been improved by adding a fifth parameter[6] for a better fitting of dynamic and loss moduli at high frequencies.

Similarly, in dielectric materials, the sample is treated as a parallel or serial circuit of an ideal capacitor and an ohmic resistor. Among the abundant literature on this subject, we can refer to the precursory works. The standard and simplest model in the physics of dielectrics was provided by Debye [8] in 1912 based on a relaxation function decaying exponentially in time with a characteristic relaxation time. Since the pioneering work of Kohlrausch in 1854, introducing a stretched exponential relaxation successively rediscovered by Williams and Watts [9], important models where introduced by Cole and Cole [10], Davidson and Cole [11], Havriliak and Negami (HN) [12] and others [13-16]. The empirical five-parameter HN model describes symmetric and asymmetric broadening of the complex dielectric function. The limiting behavior of the complex dielectric function at low and high frequencies was analyzed and called universal dielectric response by Jonscher [17].

The aim of this paper is to highlight the used approaches for viscoelastic and dielectric materials through two five-parameter models [6,12]. These fractional derivatives models and empirical models don't describe the same behavior at low and high frequencies respectively with regard to the position of maximal loss. Experimental data on polymeric foams in the range 0.1 Hz-500 Hz are fitted to show the advantage of the empirical model.

2. Theory

In classic approach, mechanical models consisting of spring and dashpots, are used to describe the viscoelastic properties of viscoelastic materials [18]. A good description requires an introduction of fractional derivatives models and so the Zener model [19] was replaced by the four-parameter fractional derivative model [2]. For dielectric materials, the combination of resistors and capacitors as well as the empirical use of fractional exponents in frequency relationships has induced more efficient models such as those of Cole-Cole and HN models [17].

2.1. Zener model

For the Zener model illustrated in Fig 1, the stress-strain relationship is:

$$\left(1+\frac{\eta}{E_2}\frac{d}{dt}\right)\sigma=\left[E_1+\eta\left(1+\frac{E_1}{E_2}\right)\frac{d}{dt}\right]\varepsilon \tag{1}$$

or

$$\left(1+a\frac{d}{dt}\right)\sigma=\left[m+b\frac{d}{dt}\right]\varepsilon, \tag{2}$$

where:

$$a=\eta/E_2, \qquad m=E_1, \qquad b=\eta(1+E_1/E_2). \tag{3}$$

The complex modulus:

$$G^*(\omega)=\hat{\sigma}(\omega)/\hat{\varepsilon}(\omega), \tag{4}$$

obtained by using the Fourier transformation in Eq. (1), takes the form:

$$G^*(\omega)=\frac{m+jb\omega}{1+ja\omega} \tag{5}$$

The dynamic and loss moduli corresponding the real and imaginary part of $G^*(\omega)$ are:

$$G'(\omega)=\frac{m+ab\omega^2}{1+a^2\omega^2} \tag{6}$$

$$G''(\omega)=\frac{(b-am)\omega}{1+a^2\omega^2} \tag{7}$$

At low and high frequencies, the limits of the dynamic modulus G' are:

$$G_0=m, \qquad G_\infty=b/a. \tag{8}$$

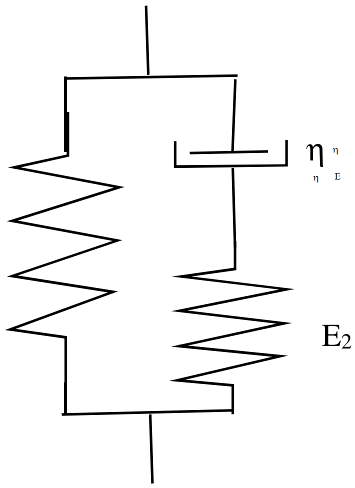


Fig.1: Classical Zener mode

2.2. Fractional Zener model

Linear viscoelasticity is certainly the field of the most extensive applications of fractional calculus, in view of its ability to model hereditary phenomena with long memory. The FZM introduced by Caputo et al. [2] can be defined from Eq. (1) where the integer derivatives are replaced by the fractional ones.

The stress-strain relationship given by Eq. (2) becomes:

$$\left(1+a\frac{d^\alpha}{dt^\alpha}\right)\sigma=\left[m+b\frac{d^\alpha}{dt^\alpha}\right]\varepsilon, \tag{9}$$

where the αth order fractional derivative of the function u(t) is defined with the gamma function as [20]:

$$\frac{d^\alpha}{dt^\alpha}u=\frac{1}{\Gamma(1-\alpha)}\frac{d}{dt}\int_0^t\frac{u(\tau)}{(t-\tau)^\alpha}d\tau. \tag{10}$$

Fortunately, by means of Fourier transformation, Eq. (9) is easy to transform in the frequency-domain by replacing d^α/dt^α by $(j\omega)^\alpha$; then the complex modulus is:

$$G^*(\omega)=\frac{m+b(j\omega)^\alpha}{1+a(j\omega)^\alpha}. \tag{11}$$

The values of the dynamic modulus for classical model at low and high frequencies holds for FZM; the introduction of G_0 and G_∞ in Eq. (11) leads to:

$$\frac{G^*-G_\infty}{G_0-G_\infty}=\frac{1}{1+a(j\omega)^\alpha}, \tag{12}$$

or

$$\frac{G^*-G_\infty}{G_0-G_\infty} = \frac{1}{1+(j\omega\tau)^\alpha}, \tag{13}$$

by introducing the relaxation time [20]:

$$\tau = a^{1/\alpha}. \tag{14}$$

For the four-parameter fractional derivative model, some mathematical manipulations on the Eq.12 result in:

$$\frac{G^*-G_0}{G_\infty-G_0} = \frac{(j\omega\tau)^\alpha}{1+(j\omega\tau)^\alpha}. \tag{15}$$

and will be discussed below.

The real and imaginary parts of $G^*(\omega)$ take the form:

$$G'(\omega) = \frac{G_0+(G_0+G_\infty)(\omega\tau)^\alpha\cos(\pi\alpha/2)+G_\infty(\omega\tau)^{2\alpha}}{1+2(\omega\tau)^\alpha\cos(\pi\alpha/2)+(\omega\tau)^{2\alpha}}, \tag{16}$$

$$G''(\omega) = \frac{(G_\infty-G_0)(\omega\tau)^\alpha\sin(\pi\alpha/2)}{1+2(\omega\tau)^\alpha\cos(\pi\alpha/2)+(\omega\tau)^{2\alpha}}. \quad 17)$$

It is worth mentioning that the frequency function defined by Eq. (13) has been
suggested empirically by Cole brothers [10] as a result of analyzing the frequency dependence of complex dielectric functions. The normalized frequency:

$$\omega_0 = \omega\tau, \quad 18)$$

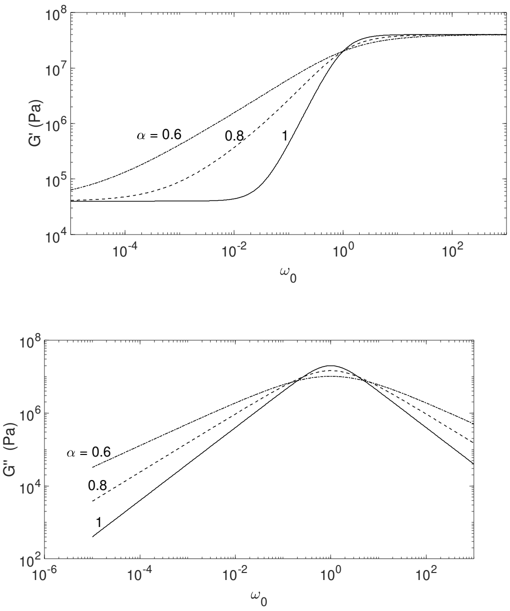
is used in the following figures.

In this four-parameter model, theoretically analyzed by Pritz [5], it is shown that there is a strict relation between the dispersion of the dynamic modulus, the loss modulus peak and the slope of the frequency curves. The variations of the dynamic and loss moduli are given respectively in Fig. 2a and Fig. 2b. It can be seen that the dynamic modulus increases from G_0 up to G_∞ with increasing frequency. The smaller α, the smaller the slope of the dynamic modulus frequency curve at the inflexion point. Similarly, the slope of the increase and decrease of the loss modulus curve is determined by a below and above their maxima, respectively. In other words, at low and at high frequency the loss modulus can be simplified respectively as follows:

$$G'' \propto \omega^\alpha, \qquad G'' \sim \propto \omega^{-\alpha}. \tag{19}$$

The peak value of the loss modulus curve and the inflexion point of the dynamic modulus curve are at the frequency of:

$$\omega_p = 1/\tau. \tag{20}$$



a)

b)

Fig. 2(a,b): Dynamic and loss moduli calculated by the four-parameter fractional Zener model

More recently, in order to describe asymmetrical loss modulus and the high-frequency behavior of polymeric damping materials, a supplementary derivation of β order by Pritz [6] leads to:

$$\frac{G^*-G_0}{G_\infty-G_0} = \frac{(j\omega\tau)^\alpha}{1+(j\omega\tau)^\beta}. \tag{21}$$

The complex modulus of this five-parameter fractional derivative model, given by Eq. (19), corresponds to the four-parameter one, Eq. (15), if $\beta = \alpha$. The loss modulus as well as the dynamic one is not influenced by β at low frequency as shown in Fig. 3. Furthermore, the deviation at high frequency introduces the asymmetry with a variable curve G'' slope. The first fractional derivative order α is quite the same for the two models: the low-frequency branches of loss modulus predicted by these models are practically superimposed.

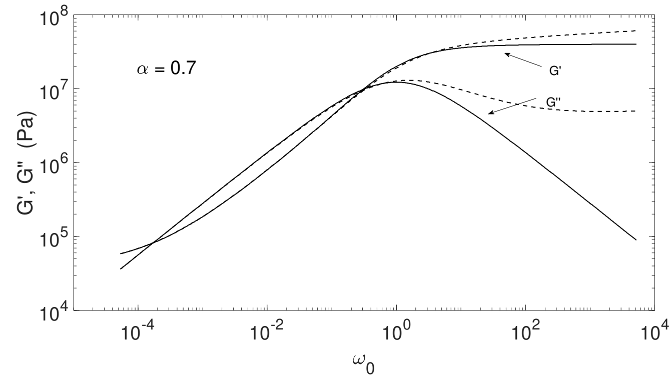


Fig. 3: Dynamic and loss moduli calculated by the five-parameter FZM (solid line β = 1, hashed line β = 0.65)

2.3. Empiric models for dielectric materials

Dielectric relaxation in solids [17] represents one of the most intensely researched topics in physics, the history of which goes back to the 18th century, and yet one whose theoretical understanding to this day escapes a satisfactory solution. The main reason might lie with the dissipative nature of the relaxation process. The standard and simplest model leads to the exponential relaxation, and correspondingly the frequency dependence of the complex permittivity is:

$$\varepsilon^* - \varepsilon_\infty = \frac{\varepsilon_0 - \varepsilon_\infty}{1 + j\omega\tau}, \qquad (22)$$

where ε_0 and ε_∞ are the "static" and "infinite" frequency dielectric constants, and τ is a characteristic relaxation time.

A generalization of the FZM was proposed by Havriliak and Negami to describe the α-dispersions in polymer system: the complex modulus G^* was empirically written as:

$$\frac{G^* - G_\infty}{G_0 - G_\infty} = \frac{1}{(1 + (j\omega\tau)^\alpha)^\beta}, \qquad (23)$$

where the parameter β controls the asymmetry of the loss modulus. The formula reduces to the Debye model, given by Eq. (22), for $\alpha = \beta = 1$ and to the Cole-Cole model for $\beta = 1$, and to Davidson-Cole model for $\alpha = 1$.

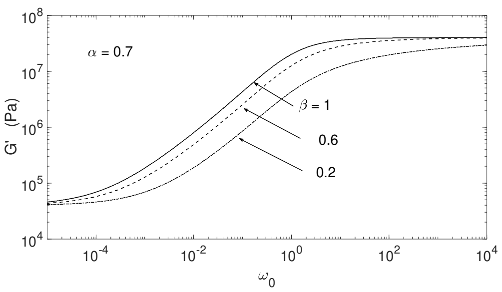
For this model the real and imaginary parts of the complex modulus are [21]:

$$G' = G_\infty + \frac{(G_0 - G_\infty)\cos(\beta\theta)}{[1 + 2\omega^\alpha\tau^\alpha\cos(\alpha\pi/2) + \omega^{2\alpha}\tau^{2\alpha}]^{\beta/2}}, \qquad (24)$$

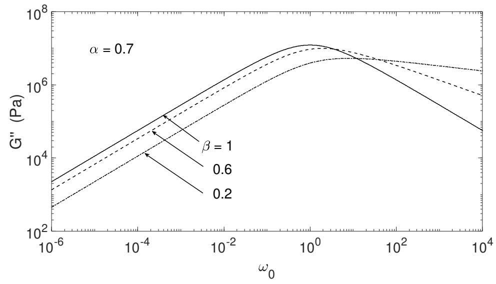
$$G'' = \frac{(G_\infty - G_0)\sin(\beta\theta)}{[1 + 2\omega^\alpha\tau^\alpha\cos(\alpha\pi/2) + \omega^{2\alpha}\tau^{2\alpha}]^{\beta/2}}, \qquad (25)$$

where:

$$\theta = tan^{-1}\frac{\omega^\alpha\tau^\alpha\sin(\alpha\pi/2)}{1 + \omega^\alpha\tau^\alpha\cos(\alpha\pi/2)}. \qquad (26)$$



a)



b)

Fig. 4(a,b): Dynamic and loss moduli calculated by the HN model

It can be seen in Fig. 4a that the smaller β, the smaller the dynamic modulus; the maximal values are obtained for $\beta = 1$ which correspond to FZM ones.

The frequency curves in Fig. 4b show the influence of the asymmetry parameter β for loss modulus at a fixed value of α. In comparing to the FZM, the loss modulus takes the form [17]:

$$G'' \propto \omega^\alpha \quad \text{and} \quad G'' \propto \omega^{-\alpha\beta}. \qquad (27)$$

At low and high frequency respectively; α and $-\alpha\,\beta$ are the slopes of logG'' vs logω at low and high frequencies respectively with regard to the position of maximal loss. These Jonscher power-law exponents are explicitly given below.

If ωτ << 1, Eq. (25) takes the form $\tan\theta \approx \omega^\alpha\tau^\alpha\sin(\alpha\pi/2)$ and G'' in Eq. (25) can be simplified as: $G'' \approx (G_\infty - G_0)\sin(\beta\theta) \approx (G_\infty - G_0)\beta\theta \approx (G_\infty - G_0)\beta\omega^\alpha\tau^\alpha\sin(\alpha\pi/2)$. Then:

$$logG'' \approx \alpha log\omega + \log[(G_\infty - G_0)\beta\tau^\alpha\sin(\alpha\pi/2)]. \qquad (28)$$

If ωτ >> 1, the similar mathematical simplifications of G'' in Eq. (25) lead to:

$$logG'' \approx -\alpha\beta log\omega + \log\left[(G_\infty - G_0)\sin(\alpha\beta\pi/2)\,/\,\tau^{\alpha\beta}\right] \qquad .(29)$$

The peak value[22] of the loss modulus curve at the frequency of:

$$\omega_p = \frac{1}{\tau}\left[sin\frac{\alpha\pi}{2+2\beta}\right]^{1/\alpha}\left[sin\frac{\alpha\beta\pi}{2+2\beta}\right]^{-1/\alpha}, \qquad (30)$$

which reduces to Eq. (19) for $\beta = 1$ corresponding to FZM. In contrast to the five-parameter FZM mentioned above in Eq. (21) and in Fig. 3, the asymmetry parameter β has an effect on the position of the loss modulus peak given by Eq. (30) and on the slope of G'' curve at high frequency as shown in Fig. 4b. It follows that the loss modulus increases with increasing α at low frequency and where the curves are parallel for $\omega < \omega_p$; in addition, the smaller β, the smaller value of the loss modulus.

3. Havriliak-Negami Model and experimental data

It was shown above that, for $\omega \ll \omega_p$, the parameters α and β have an effect on the loss modulus curve and on the dynamic modulus as shown in Fig. 4. Once the applicability of the HN model has been established for a class of materials, the dynamic properties can be predicted for wide frequency range by using data measured in a narrow range.

In a previous paper [23] a method is presented for the mechanical characterization of anisotropic foams where the objective of the experiment is to determine the static elastic parameters of an acoustic foam. In the present study, a quasi-static method [24] is implemented for shear characterization of polyurethane foams at low frequency. Porous foams are diphasic materials with coupling effects where the air flow resistivity can be neglected in shear tests.

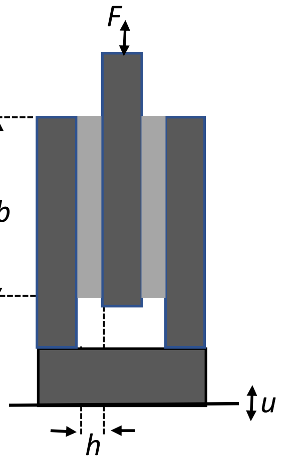


Fig. 5: Shear configuration, samples (clear grey) squeezed between a rigid plate and a U-shaped profile (black grey).

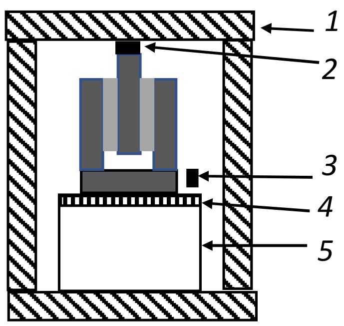


Fig. 6: Shear measurement setup (1- supporting frame, 2- piezoelectric force transducer (PCB 209B01), 3- inductive gauging sensor (Keyence EX-201:305) , 4- driving plate, 5- Electrodynamic shaker (Bruel & Kjaer 4808)

A simplified representation of the shear setup is given in Fig. 5. The brick-shaped samples, having a thickness h and a square section area $S = b^2$, are placed between a rigid plate and a U-shaped profile. In this test, the measured shear stiffness *F/u* is twice the stiffness of each sample where F and u are defined in Fig. 5. The U-shaped device is harmonically translated by an electrodynamic shaker using a 0.1–500 Hz sweep-sine as illustrated in Fig. 6. An inductive displacement sensor is used to measure the displacement $u^*(\omega)$ of the driving plate. A piezoelectric force transducer placed between the top of the middle plate and the supporting frame is used to measure the transmitted force $F^*(\omega)$. A FFT analyzer computes the following mechanical impedance:

$$H^*(\omega) = F^*/u^*. \quad (32)$$

Under the assumption that the thickness h of the sample is small compared to the other dimension b of the sample, the complex shear modulus $G^*(\omega)$ is given by [23]:

$$G^*(\omega) = H^*(\omega)h/2S. \quad (33)$$

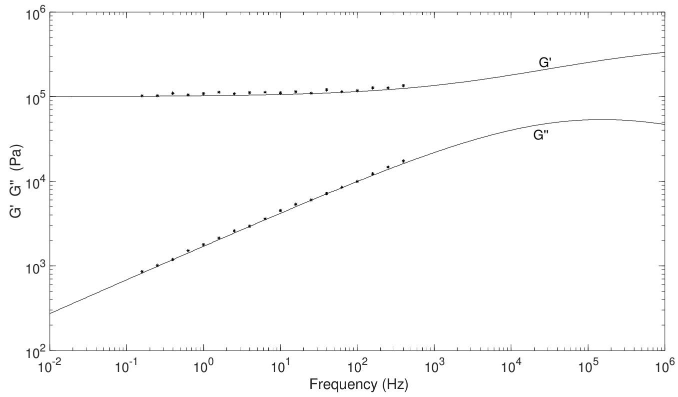


Fig.7: Dynamic and loss moduli, solid line for HN model, * for measured values of polyurethane foam.

Five pairs of samples having the dimensions h = 2mm and b = 8 mm are squeezed by means of sand papers to avoid any slippage at the four interfaces. A fit of HN model to the average data measured at 20 °C from Eq. (33) gives the following parameters: $G_0 = 100\ kPa$, $G_\infty = 450\ kPa$, $\tau = 10^{-5}\ s$, $\alpha = 0.4$ and $\beta = 0.84$. As can be seen in Fig. 7, a good fit is obtained over the entire frequency range for both dynamic and loss moduli. The sound absorbing properties of this foam have applications sought in the audible band, less than 20 kHz, and at a higher frequency for light thin structure damping. For this foam, the useful frequency range is very small compared to ω_p where the simplified G'' expression in Eq. (28) leads straightforward to the slope α. For $\beta = 1$, HN and five-parameter FZM are both reduced to four- parameter FZM. Furthermore, for Zener models, the β parameter has no influence on loss modulus as well as the dynamic one at low frequencies. Despite the empiric origin of HN model not related to constitutive equations, the β value obtained above has improved the fitting of the experimental data.

4. Conclusion

This article presented the main models in viscoelastic and dielectric materials to predict the frequency dependences of properties for a wide range. Generally, the introduction of a new parameter leads to a better fitting of dynamic and loss moduli. However, this improvement can be better adapted to a class of materials depending on the used model.

The five-parameter HN model was analyzed and compared to the five-parameter fractional derivatives model. It is shown that the fifth parameter in fractional model can modify the viscoelastic only at high frequencies while the one in HN model affect the prediction in a wide frequency range. A method for studying the viscoelastic properties of polymeric foams at low frequency less than 500 Hz has been developed. The complex shear modulus is predicted on a large frequency range until 100 kHz by using the HN model. As a result of this investigation, this model is successfully adapted to viscoelastic behavior of polyurethane foams.

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