

Improved five-parameter fractional derivative model for elastomers

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THE DYNAMIC BEHAVIOUR OF ELASTOMERS is assumed to follow a constitutive differential equation of non-integral (fractional) order. In order to describe the peculiar frequency response of the loss factor, the constitutive equation has been refined by introducing the fifth parameter to the classical fourth-order equation. The asymmetry of the loss factor in the frequency domain comes from the different time-derivative orders of the stress and strain. Either smooth asymmetry or stabilization by a plateau at high frequency can be modelled by suitable difference between the two orders of the time derivatives. The physical validity of the model is discussed and a parametrical analysis is conducted on diagrams relating the height and the width of the loss factor.

Key words: loss factor, fractional derivative, damping, elastomers, viscoelasticity.

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1. Introduction

RECENT DEVELOPMENTS IN MATHEMATICAL AREA have provided a powerful tool to describe relaxation phenomena in a complex system: the fractional operators have performed indeed a simplified description of the linear viscoelastic behaviour of polymers. Among the abundant literature on this subject, we can refer to the precursory works (BAGGLEY and TORVIK, [1] and [2]; FRIEDRICH, [3]; NONNENMACHER and GLÖCKLE, [4]) and some meaningful applications conducted on polymers (HEYMANS and BAUWENS, [5]; SCHIESEL *at al.*, [6]) or dynamic problems (ROSSIKHIN and SHITIKOVA, [7]). A more complete review may be found in METZLER and NONNENMACHER [8].

Elastomers are often used in damping applications due to their high energy-absorption abilities. The combination of high damping and large bandwidth in the frequency domain arose great practical interest. The material damping is mathematically described by the loss factor, that is the ratio between the dissipated and stored energies during harmonic loading. This loss factor is interpreted as the tangent of the phase angle separating the strain and the stress for linear materials. One traditional way to describe the material damp-

ing for a given frequency interval is based on the use of a constant loss factor. However, it has been proved that such a description cannot be inverted in the time domain and consequently, it does not satisfy the causality condition stating that the response always occurs after the excitation. Alternative models consist in taking a Newtonian, linear viscous damping. The induced dissipated energy and the loss factor are proportional to the frequency on the entire frequency range. This definition is in contradiction with the apparent stabilization of the loss factor at a high-frequency regime (i.e. sudden stress release).

Since the fundamental approach proposed by COLE and COLE [9] to describe the dielectric and mechanical properties of polymers via the equations containing fractional operators, numerous models have been extensively used to reproduce damping in the frequency domain. Four-parameter fractional derivative model (called also the fractional derivative Zener model) is frequently used in the literature.

However, this classical model appears to be inadequate to describe the experimental asymmetry of the loss factor peak in frequency. The combination between the broadening of the relaxation width and the asymmetrical frequency behaviour is successfully described by the empirical formulation proposed by HAVRILIAK and NEGAMI [10]. However, this formulation cannot be written in an explicit form of a constitutive equation relating the stress and strain histories. Recent papers have explored a new formulation including the experimental and mathematical requirements. DAVIDSON and COLE [11] proposed an empirical description of the experimental asymmetry but failed to produce a proper bandwidth evolution. BAGLEY and TORVIK [1] assumed different derivative orders of the stress and the strain in the constitutive behaviour which were not compatible with the causality condition [2]. Extensively, FRIEDRICH and BRAUN [12] suggested also the introduction of different derivative orders of strain in the constitutive behaviour. FRIEDRICH [3] also showed that the thermodynamic condition was satisfied providing that the derivative order of the stress remained smaller or equal to the one of the strain. PRITZ [13] pursued this model to provide a complete description of the loss factor peak. Nevertheless, the model initiated by Friedrich and Braun is addressed to materials showing a plateau of the loss factor at high frequency regime and a moderate asymmetry of the loss peak in the maximum vicinity.

The purpose of this paper is to present a five-parameter fractional derivative model describing either smooth or marked frequency asymmetry of the loss factor modulus. The possible existence of a plateau at a high-frequency regime is also considered. This paper is organised as follows. The second section treats the general constitutive equations containing fractional derivative operators. The complex moduli are recalled for the sake of comparison with the improved five-

parameter approach developed in Sec. 3. The influence of the second-order derivative is investigated in order to assess its efficiency in Sec. 4. Finally, the model has been applied to predict the loss factor evolution for EAR C-1002 elastomer exhibiting a plateau.

2. The fractional derivative model

2.1. General constitutive equation described by fractional derivative model

Polymer behaviour is modelled by linear viscoelastic relationships between stress and strain, in accordance with the fading memory characteristics: the stress does not only depend on the current strain, but also on the recent strain history. A particular class of material constitutive laws has been formulated through integral, hereditary or convolution form, following the superposition principle introduced by BOLTZMANN [14]. The classical hereditary models correspond to exponentially decaying memory issued from a continuous distribution of relaxation times of the generalized Maxwell model. ENELUND and OLSSON [15] gave an alternative formulation of this spectral description using a fractional derivative operator. They proposed a mathematical frame to previous empirical descriptions of constitutive equations, based on similarities observed between the mechanical and dielectric relaxations. ENELUND and LESIEUTRE [16] demonstrated that a single form of fractional derivative model is sufficient to describe the evolution equations for anelastic strains. The generic fractional order constitutive equation is retained in the following:

$$(2.1) \quad \sigma(t) + b_1 \frac{d^{\beta_1} \sigma(t)}{dt^{\beta_1}} + b_2 \frac{d^{\beta_2} \sigma(t)}{dt^{\beta_2}} + \dots + b_m \frac{d^{\beta_m} \sigma(t)}{dt^{\beta_m}} \\ = a_0 \varepsilon(t) + a_1 \frac{d^{\alpha_1} \varepsilon(t)}{dt^{\alpha_1}} + a_2 \frac{d^{\alpha_2} \varepsilon(t)}{dt^{\alpha_2}} + \dots + a_n \frac{d^{\alpha_n} \varepsilon(t)}{dt^{\alpha_n}},$$

where σ and ε may be referred equally to the deviatoric or hydrostatic parts. A set of initial conditions is added to (2.1) to render the pre-existence of the stress with respect to the strain. It is noteworthy that the number and order of time derivatives of strain and stress in (2.1) cannot be arbitrary to ensure the description of an effective material behaviour and to satisfy the causality condition formulated as $\beta_m \leq \alpha_n$ (see also ENELUND and OLSSON [15]). SAMKO *et al.* [18] expressed the basic analytical conditions of the existence and uniqueness of solutions to such fractional differential equations.

Among the several definitions of the fractional derivatives, we have followed the definition in the sense of Riemann–Liouville as it is closely related to the Laplace transform and consequently to the Fourier transform. The operator d^α of the fractional derivative of order α applied to the function $x(t)$ is defined for $0 < \alpha < 1$ as:

$$(2.2) \quad d^\alpha[x(t)] = \frac{1}{\Gamma(1-\alpha)} \frac{d}{dt} \int_0^t \frac{x(u)}{(t-u)^\alpha} du,$$

where Γ denotes the Gamma function defined for $\Re(x) > 0$. The operator d^α allows the continuation at the singularity point $t = 0$. A unique expression of the Fourier transform of the fractional derivative operator of order α th satisfies (see GEL'FAND and SHILOV [17]):

$$(2.3) \quad \mathcal{F}[d^\alpha[x(t)]] = (i\omega)^\alpha \mathcal{F}[x(t)],$$

where $(i\omega)^\alpha$ corresponds to $(\exp(i\alpha\pi/2)(\omega - i0^+)^\alpha)$ simplified to $(\exp(i\alpha\pi/2)\omega)^\alpha$ for $\omega > 0$.

So, the constitutive equation describing viscoelastic behaviour can be written in the frequency domain:

$$(2.4) \quad \bar{\sigma}(i\omega) = \bar{G}(i\omega)\bar{\varepsilon}(i\omega).$$

The complex modulus $\bar{G}(i\omega)$ is distinguished between the real and imaginary parts $G'(\omega)$ and $G''(\omega)$, defined respectively as conservative and dissipative moduli:

$$(2.5) \quad \bar{G}(i\omega) = G'(\omega) + iG''(\omega).$$

The tangent of the phase angle between stress and strain is called the loss factor $\tan \delta(\omega)$:

$$(2.6) \quad \tan \delta(\omega) = \frac{G''(\omega)}{G'(\omega)}.$$

Two aspects of the relaxation process are of primary interest when describing the damping abilities of polymeric materials: the height $\tan \delta_m$ at the extremum of the loss factor and the bandwidth W of the frequency band, taken at the half-height $\tan \delta_m/2$. These points are analyzed in the diagrams presented hereafter in Sec. 4.2.

2.2. Four-parameter fractional derivative model – Zener formulation

The fractional Zener formulation depends on the strain and the stress histories in the time domain as:

$$(2.7) \quad \sigma(t) + \tau^\alpha \frac{d^\alpha \sigma(t)}{dt^\alpha} = G_0 \varepsilon(t) + G_\infty \tau^\alpha \frac{d^\alpha \varepsilon(t)}{dt^\alpha},$$

where τ is the generalized relaxation time.

The initial condition is expressed as:

$$(2.8) \quad \frac{d^{\alpha-1}\sigma(0^+)}{dt^{\alpha-1}} = G_\infty \frac{d^{\alpha-1}\varepsilon(0^+)}{dt^{\alpha-1}}.$$

It is supposed that the generalized relaxation time τ is the only temperature-dependent parameter stemming from the time-temperature superposition, usually admitted for a large range of temperature. G_0 corresponds to the long time or relaxed modulus and G_∞ is also called the unrelaxed or instantaneous modulus. The index in moduli notation has to be interpreted as the frequency limit. It is assumed hereafter that $G_\infty/G_0 > 1$.

The complex modulus $\bar{G}(i\omega)$ of the fractional Zener model is expressed with (2.4):

$$(2.9) \quad \bar{G}(i\omega) = \frac{G_0 + G_\infty(i\omega\tau)^\alpha}{1 + (i\omega\tau)^\alpha}.$$

The loss factor modulus $\tan \delta(\omega)$ defined by (2.6) becomes:

$$(2.10) \quad \tan \delta(\omega) = \frac{(G_\infty - G_0) \sin(\alpha\pi/2)(\omega\tau)^\alpha}{G_0 + (G_\infty + G_0) \cos(\alpha\pi/2)(\omega\tau)^\alpha + G_\infty(\omega\tau)^{2\alpha}}.$$

The parameter α appears as the slope of the curve $\ln(\tan \delta)$ versus $\ln(\omega\tau)$. A qualitative physical interpretation of the parameter α can be based on the suggestion of DAVIES and LAMB [19] that single relaxation time behaviour is observed in liquids for which only pairs of molecules contribute to the relaxation. As many-body interactions become more significant, the relaxation broadens. Thus, α is a measure of the strength of coupling among the different modes of vibration in the polymer.

2.3. Havriliak–Negami empirical formulation

A generalization of the fractional Zener model was proposed by HAVRILIAK and NEGAMI in [10] to describe the α -dispersions in polymer system: the complex modulus $\bar{G}(i\omega)$ was empirically written as:

$$(2.11) \quad \frac{\bar{G}(i\omega) - G_\infty}{G_0 - G_\infty} = \frac{1}{(1 + (i\omega\tau)^\alpha)^\beta}$$

where the parameter β controls the asymmetry of the loss factor. The parameters α and β are taken as $0 < \beta < \alpha < 1$. The relevant parameter fractional derivative order α ranges from about 0.4 to 0.8 and the calibrating asymmetry parameter β ranges from 0.1 to 0.4.

The loss factor modulus was calculated by HARTMANN, LEE and LEE [20]:

$$(2.12) \quad \tan \delta(\omega) = \frac{(1 - G_0/G_\infty) \sin(\beta\theta)}{[1 + 2 \cos(\alpha\pi/2)(\omega\tau)^\alpha + (\omega\tau)^{2\alpha}] + (1 - G_0/G_\infty) \cos(\beta\theta)}$$

with

$$(2.13) \quad \theta = \arctan \frac{(\omega\tau)^\alpha \sin(\alpha\pi/2)}{1 + \cos(\alpha\pi/2)}.$$

The extremum of the loss factor corresponds to the normalized frequency $\omega_{n_{\max}}$ satisfying the equation:

$$\begin{aligned} [\sin(\alpha\pi/2 - \beta\theta) - \omega_{n_{\max}}^\alpha \sin(\beta\theta)](1 + 2\omega_{n_{\max}}^\alpha \cos(\alpha\pi/2) + \omega_{n_{\max}}^{2\alpha})^{\beta/2} \\ = (1 - G_0/G_\infty) \sin(\alpha\pi/2). \end{aligned}$$

The height $\tan \delta_m$ and the bandwidth W are calculated after substitution of $\omega_{n_{\max}}$ into (2.12).

3. Improved five-parameter fractional derivative model

3.1. Classical asymmetry description in the constitutive law

As observed in the Havriliak–Negami formulation of the loss factor (2.12) and (2.13), the asymmetry results mathematically from a different order exponent at the numerator and the denominator in $\tan \delta(\omega)$. A simplified way to obtain this difference is to consider a different derivation order of the stress and strain (Bagley and Torvik [2]):

$$(3.1) \quad \sigma(t) + \tau^\beta \frac{d^\beta \sigma(t)}{dt^\beta} = G_0 \varepsilon(t) + G_\infty \tau^\alpha \frac{d^\alpha \varepsilon(t)}{dt^\alpha}$$

with $\beta > \alpha$ to ensure a finite instantaneous modulus calculated as $\lim_{\omega \rightarrow \infty} \bar{G}(i\omega) = G_\infty$. This model fits correctly the experimental data but has to be dismissed for not satisfying the causality condition $\beta < \alpha$.

A conventional way to increase the number of time derivative is to add a second fractional derivative of strain in the second member of the four-parameter fractional derivative model (FRIEDRICH and BRAUN [12] and PRITZ [13]):

$$(3.2) \quad \sigma(t) + \tau^\beta \frac{d^\beta \sigma(t)}{dt^\beta} = G_0 \varepsilon(t) + G_\infty \tau^\beta \frac{d^\beta \varepsilon(t)}{dt^\beta} + (G_\infty - G_0) \tau^\alpha \frac{d^\alpha \varepsilon(t)}{dt^\alpha}$$

with $\beta < \alpha$ to describe a meaningful behaviour. However, this model is restricted to a polymer category presenting an undefined value for the instantaneous modulus G_∞ and a limit value of the loss factor $\tan \delta(\omega) \approx \tan[(\alpha - \beta)\pi/2]$ when $\omega \rightarrow \infty$. It can be observed that broadening of the loss factor is poor in the vicinity of the peak (PRITZ [13], Fig. 3).

3.2. Formulation of the improved model

To increase the time derivative orders, it is chosen to multiply the classical four-parameter fractional derivative equation at the α th order (2.8) by the distributive derivative operator $(1 + \tau^\beta d^\beta)$ with $\beta < \alpha$. The relaxation time τ is preserved for the sake of simplicity. The constitutive equation resulting from this operation can be written as:

$$(3.3) \quad (1 + \tau^\alpha d^\alpha)(1 + \tau^\beta d^\beta)\sigma(t) = (G_0 + G_\infty \tau^\alpha d^\alpha)(1 + \tau^\beta d^\beta)\varepsilon(t).$$

Three derivative orders appear after the development of (3.3):

$$(3.4) \quad \begin{aligned} \sigma(t) + \tau^\alpha \frac{d^\alpha \sigma(t)}{dt^\alpha} + \tau^\beta \frac{d^\beta \sigma(t)}{dt^\beta} + \tau^{\alpha+\beta} \frac{d^{\alpha+\beta} \sigma(t)}{dt^{\alpha+\beta}} \\ = G_0 \varepsilon(t) + G_0 \tau^\beta \frac{d^\beta \varepsilon(t)}{dt^\beta} + G_\infty \frac{d^\alpha \varepsilon(t)}{dt^\alpha} + G_\infty \tau^{\alpha+\beta} \frac{d^{\alpha+\beta} \varepsilon(t)}{dt^{\alpha+\beta}}. \end{aligned}$$

In the frequency domain, the constitutive equation and the complex modulus may be simplified by the complex term $(1 + (i\omega\tau)^\beta)$ which is the Fourier transform of the derivative operator $(1 + \tau^\beta d^\beta)$. The foregoing analysis of the model described by the Eq. (3.4) is identical to the initial four-parameter model (2.8).

As the time derivative in the constitutive equation (3.4) is increased twice from the four-parameter model, it is suggested to remove the terms at the derivation order $(\alpha + \beta)$ (similar results may be obtained by suppression of orders α or β , but the parameter interpretation is more intricate). The constitutive differential equation is simplified to:

$$(3.5) \quad \sigma(t) + \tau^\alpha \frac{d^\alpha \sigma(t)}{dt^\alpha} + \tau^\beta \frac{d^\beta \sigma(t)}{dt^\beta} = G_0 \varepsilon(t) + G_0 \tau^\beta \frac{d^\beta \varepsilon(t)}{dt^\beta} + G_\infty \frac{d^\alpha \varepsilon(t)}{dt^\alpha}.$$

The complex modulus is calculated as:

$$(3.6) \quad \bar{G}(i\omega) = \frac{G_0 + G_0(i\omega\tau)^\beta + G_\infty(i\omega\tau)^\alpha}{1 + (i\omega\tau)^\alpha + (i\omega\tau)^\beta}.$$

This improved five-parameter model provides the instantaneous modulus G_∞ for high-frequency regime when the condition $\beta < \alpha$ is imposed. The relaxed modulus G_0 is retrieved at low frequencies.

A normalized frequency $\omega_n = \omega\tau$ is introduced in order to simplify the moduli and loss factor expressions. The conservative and dissipative moduli are given by the definition (2.5) as a function of the dispersion modulus $c = (G_\infty/G_0) > 1$:

$$(3.7) \quad \frac{G'(\omega_n)}{G_0} = \frac{1+(c+1)\cos(\frac{\alpha\pi}{2})\omega_n^\alpha + 2\cos(\frac{\beta\pi}{2})\omega_n^\beta + c\omega_n^{2\alpha} + \omega_n^{2\beta} + (c+1)\cos(\frac{(\alpha-\beta)\pi}{2})\omega_n^{\alpha+\beta}}{1+2\cos(\frac{\alpha\pi}{2})\omega_n^\alpha + 2\cos(\frac{\beta\pi}{2})\omega_n^\beta + \omega_n^{2\alpha} + \omega_n^{2\beta} + 2\cos(\frac{(\alpha-\beta)\pi}{2})\omega_n^{\alpha+\beta}},$$

$$\frac{G''(\omega_n)}{G_0} = \frac{(c-1)(\sin(\frac{\alpha\pi}{2})\omega_n^\alpha + \sin(\frac{(\alpha-\beta)\pi}{2})\omega_n^{\alpha+\beta})}{1+2\cos(\frac{\alpha\pi}{2})\omega_n^\alpha + 2\cos(\frac{\beta\pi}{2})\omega_n^\beta + \omega_n^{2\alpha} + \omega_n^{2\beta} + 2\cos(\frac{(\alpha-\beta)\pi}{2})\omega_n^{\alpha+\beta}}.$$

The loss factor $\tan \delta(\omega)$ is determined by the definition (2.6):

$$(3.8) \quad \tan \delta(\omega_n) = \frac{(c-1)(\sin(\frac{\alpha\pi}{2})\omega_n^\alpha + \sin(\frac{(\alpha-\beta)\pi}{2})\omega_n^{\alpha+\beta})}{1+(c+1)\cos(\frac{\alpha\pi}{2})\omega_n^\alpha + 2\cos(\frac{\beta\pi}{2})\omega_n^\beta + c\omega_n^{2\alpha} + \omega_n^{2\beta} + (c+1)\cos(\frac{(\alpha-\beta)\pi}{2})\omega_n^{\alpha+\beta}}.$$

3.3. Thermodynamic requirements

The model parameters have to satisfy some thermodynamic requirements to enable a realistic behaviour description. The dissipativity condition stating that energy should be removed rather than introduced, is formulated as $\Im(\bar{G}(i\omega)) > 0$ for $\omega > 0$ and $\Im(\bar{G}(i\omega))$ an odd function. The positivity condition imposing a positive relaxed stiffness is formulated as $\Re(\bar{G}(0)) > 0$. \Re and \Im extract respectively the real and imaginary parts of a complex function. As the instantaneous modulus G_∞ and the relaxed modulus G_0 are positive and the calculated dispersion modulus $c = G_\infty/G_0$ is greater than 1, the above-mentioned conditions are satisfied. The causality condition is satisfied since the same orders of derivation are imposed for the stress and the strain (3.5).

3.4. Asymptotic expansion of conservative and loss moduli

Low frequency regime. The behaviour at low frequencies is described via the asymptotic expansion of formulas (3.7) and (3.8) for the normalized frequency ω_n tending to zero:

$$(3.9) \quad \begin{aligned} G'(\omega_n) &\approx G_0, \\ G''(\omega_n) &\approx G_\infty \sin(\alpha\pi/2)\omega_n^\alpha, \\ \tan \delta(\omega_n) &\approx (G_\infty/G_0) \sin(\alpha\pi/2)\omega_n^\alpha. \end{aligned}$$

These responses at low frequencies are identical to the ones given by the classical four-fractional derivative model at the α th order. It follows that the ascending branches of the loss factor for low frequencies are the same for the four- and five-parameter models. Approximations given by (3.9) are also identical to the expansions of the Havriliak–Negami moduli and the loss factor for slow regime.

High frequency regime. The high frequency regime is analyzed by expansions for the normalized frequency ω_n tending to infinity:

$$\begin{aligned}
 (3.10) \quad & G'(\omega_n) \approx G_\infty, \\
 & G''(\omega_n) \approx G_\infty \sin((\alpha - \beta)\pi/2)\omega_n^{-(\alpha-\beta)}, \\
 & \tan \delta(\omega_n) \approx \sin((\alpha - \beta)\pi/2)\omega_n^{-(\alpha-\beta)}.
 \end{aligned}$$

The conservative and dissipative factors are bounded for a high-frequency regime.

These results are compared to the similar expansions derived for the four-fractional derivative model at the α th order which are recalled below:

$$\begin{aligned}
 (3.11) \quad & G'(\omega_n) \approx G_\infty, \\
 & G''(\omega_n) \approx G_\infty \sin(\alpha\pi/2)\omega_n^{-\alpha}, \\
 & \tan \delta(\omega_n) \approx \sin(\alpha\pi/2)\omega_n^{-\alpha}.
 \end{aligned}$$

It appears that for five-parameter model β balances the effect of the parameter α in order to reproduce the asymmetrical descending branch of the loss factor, whose slope is characterised by the exponent $-(\alpha - \beta)$. This balance effect is absent in the case of Zener fractional model.

Another meaningful comparison is conducted with the Havriliak–Negami approximations for $\omega_n \rightarrow \infty$:

$$\begin{aligned}
 (3.12) \quad & G'(\omega_n) \approx G_\infty, \\
 & G''(\omega_n) \approx G_\infty \sin(\alpha\beta\pi/2)\omega_n^{-\alpha\beta}, \\
 & \tan \delta(\omega_n) \approx \sin(\alpha\beta\pi/2)\omega_n^{-\alpha\beta}.
 \end{aligned}$$

It appears that in the Havriliak–Negami model, the frequency asymmetry of the loss factor is obtained by multiplication of the two derivative orders.

3.5. Loss factor properties

The loss factor is plotted in logarithmic scale as a function of the normalized frequency ω_n in Fig. 1 for a constant dispersion modulus c and constant fractional derivative order α . It is shown that either smooth or marked frequency asymmetry may be obtained. The loss factor may also be stabilized in a plateau value at high frequencies. This latter characteristic can not be predicted by the Havriliak–Negami model.

The maximum value of the loss factor occurs at the normalized frequency which is a solution of the following equation:

$$\begin{aligned}
 (3.13) \quad 0 = & -\beta \sin(\alpha\pi/2) + (\sin(\alpha\pi/2) + \sin((\alpha - \beta)\pi/2)\omega_n^\beta) \\
 & \times \left[(\alpha + \beta)(1 - (1 + c) \cos((\alpha - \beta)\pi/2)\omega_n^{\alpha+\beta}) \right. \\
 & \left. - ((1 + c) \cos(\alpha\pi/2) + 2c\omega_n^\alpha)\alpha\omega_n^\alpha - 2(\cos(\beta\pi/2) + \omega_n^\beta)\beta\omega_n^\beta \right].
 \end{aligned}$$

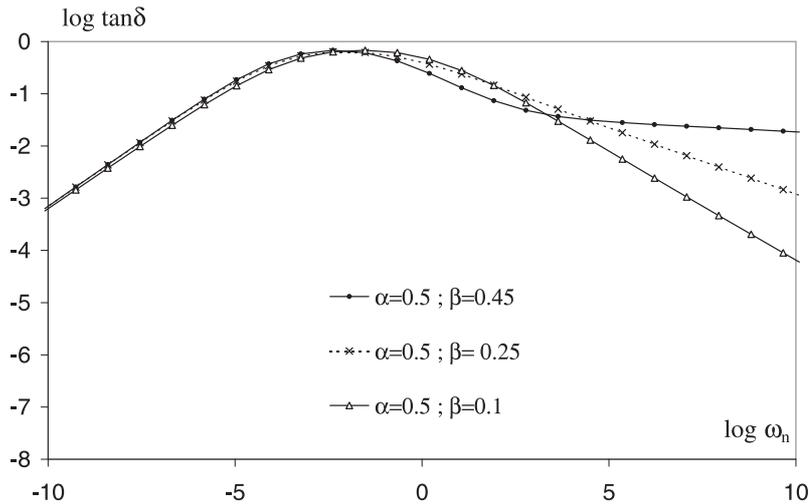


FIG. 1. Effect of the parameter β on the loss factor at given dispersion modulus c .

This equation is solved numerically in $\omega_{n_{\max}}$, the corresponding loss factor peak is calculated as $\tan \delta_m = \tan \delta(\omega_{n_{\max}})$ by using (3.8). The bandwidth W in logarithmic scale is expressed as the ratio of the two frequencies associated with the half-height of the loss factor maximum $\tan \delta_m/2$. Equation (3.13) may be simplified to a second-order equation for small values of β or close derivation orders α and β ; the positive solution ω_n^α is then expressed as a function of the dispersion modulus and the derivative order.

4. Efficiency of the improved five-parameter model

The improved five-parameter model allows a good analytical description of the glass transition centered at the loss factor peak $(\omega_{n_{\max}}, \tan \delta_m)$. Large classes of polymeric materials may be described by the new parameters set (α, β, c) . The extreme values of the dispersion modulus c are taken between 10 and 10^4 to describe the glass transition in polymers. Experimental evidence proves that the parameter α varies between 0.4 and 0.8.

4.1. Conservative and dissipative moduli

Figure 2 shows that the inflexion point of the conservative modulus $G'(\omega_n)$ appears at almost the same normalized frequency ω_n whatever is the combination (α, β) . It is noteworthy that the high frequency value G_∞ is always retrieved.

The maximum value of the dissipative modulus $G''(\omega_n)$ drawn in Fig. 3 is shifted towards the higher frequencies and the peak is slightly increased with aug-

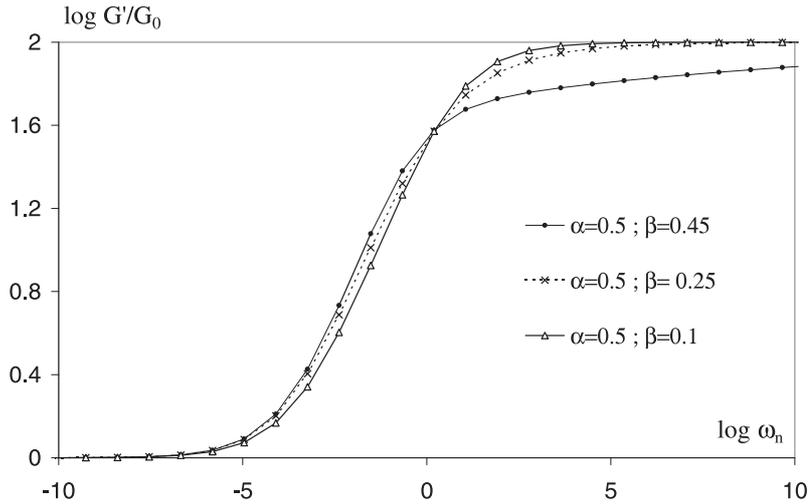


FIG. 2. Effect of the parameter β on the conservative modulus at given dispersion modulus c .

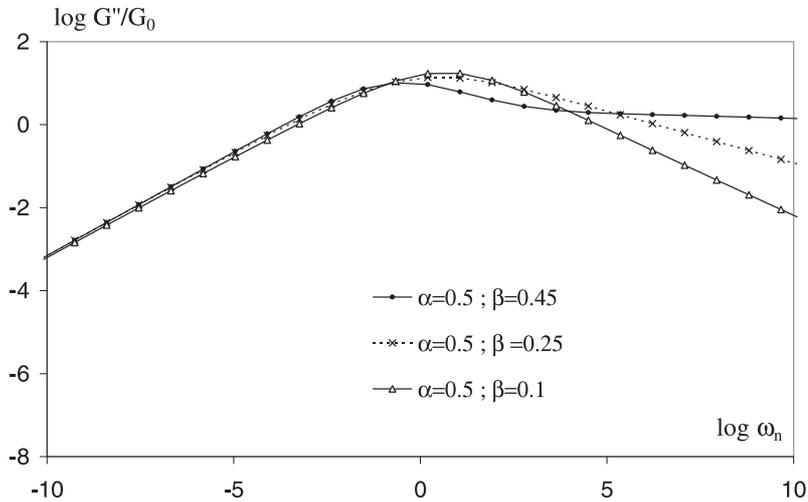


FIG. 3. Effect of the parameter β on the dissipative modulus at given dispersion modulus c .

menting $(\alpha - \beta)$. For close derivative orders, $(\alpha - \beta)$ tends to zero and $\omega_n^{-(\alpha-\beta)}$ tends to 1 at high frequency regime, inducing a quasi-constant dissipative value estimated by $G_\infty \sin((\alpha - \beta)\pi/2)$. As experimental evidence shows the saturation value of the loss factor $\tan \delta_\infty$ to be around 0.01, the high frequency limit $\tan \delta_\infty \approx \sin((\alpha - \beta)\pi/2)$ gives the maximal difference

$$(4.1) \quad (\alpha - \beta)_{\text{lim}} = 2/\pi \arcsin(0.01).$$

The saturation value of the dissipative modulus $G'''(\omega_n)$ is estimated as $0.1G_\infty$.

4.2. Height-width diagrams

Diagrams relating the loss factor height $\tan \delta_m$ and the bandwidth W are presented in Figs. 4 to 6. The log-log plot of the height-width diagrams allow the qualitative interpretation of the second derivative order effect.

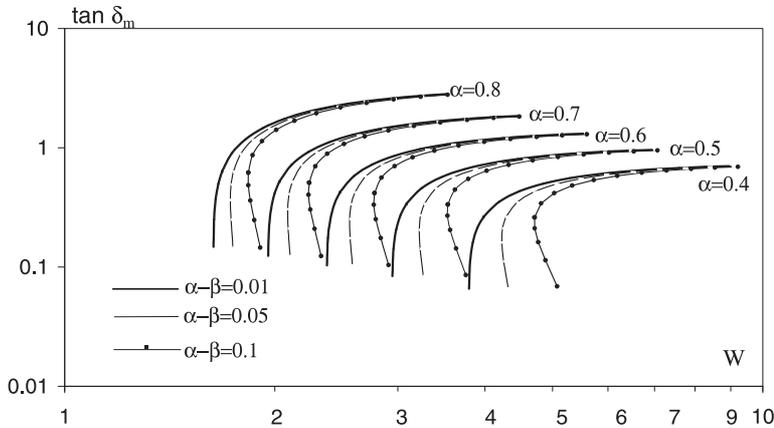


FIG. 4. Effect of the parameter β on the height-width diagram – small difference case.

The family of curves drawn in Fig. 4 for variable dispersion modulus c shows similar shape for α taken between 0.4 to 0.8 and differences $(\alpha - \beta)$ varying from 0.01 to 0.1. The bandwidth broadening corresponds to a loss factor stabilized in a plateau at high-frequency regime. The increase in α induces a higher $\tan \delta_m$ and a lower bandwidth W .

For fixed small derivative parameter β , the log-log curves $(\omega_{n_{\max}}, \tan \delta_m)$ drawn in Fig. 5 are parallel to the reference curve describing the classical four-

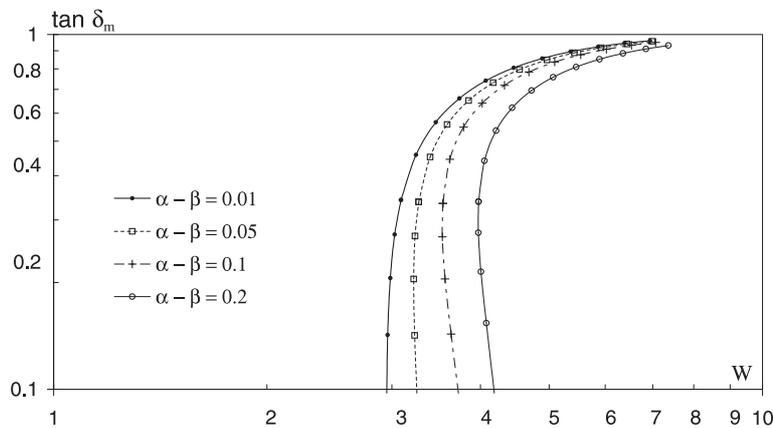


FIG. 5. Effect of the parameter β on the height-width diagram for $\alpha = 0.5$.

parameter fractional derivative model for β close to 0. The bandwidth broadening is more regular since the loss factor asymmetry is observed on the almost complete descending branch, as in the Havriliak–Negami model.

The log-log $(\omega_{n_{\max}}, \tan \delta_m)$ curves plotted in Fig. 6 for constant dispersion modulus c appear as parallel straight lines graduated with a multiplying factor of $10^{0.3}$.

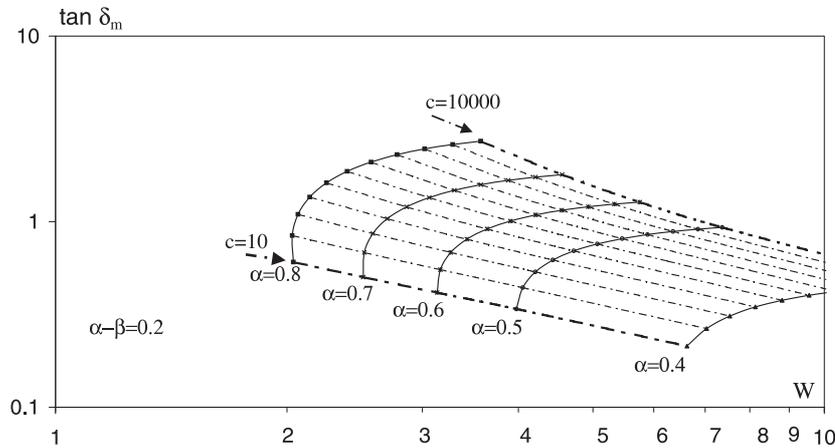


FIG. 6. Effect of the dispersion modulus c on the height-width diagram.

Since the precise fitting of experimental curves $\tan \delta(\omega_n)$ by the parameter set (α, β, c) given by (3.8) is not the only numerical way to achieve the constitutive behaviour, the height-width diagram $(\tan \delta_m, W)$ is aimed at verifying qualitatively the relations predicted by the theoretical model and allows a quick comparison between the experimental data on elastomers. As a consequence, these diagrams are essentially used to make a rough estimate of the set (α, β, c) from the measurements performed on elastomers during the Dynamic Mechanical Thermal Analysis (DMTA).

4.3. Parameters identification

The dispersion modulus c can be read off from the ratio between the minimum G_0 and maximum G_∞ values of the conservative modulus G' . The first fractional derivative order α can be measured as the slope of the ascending branch of the loss factor versus the normalized frequency curve in logarithmic scale. For smooth asymmetry of the loss peak, the second-derivative order is extracted from the slope of the descending branch corresponding to $(\beta - \alpha)$ (see also the expansion (3.10)). For marked asymmetry of the loss factor continued by a plateau, a great difference $(\beta - \alpha)$ has to be found. In that latter case, the bandwidth estimate is unattainable; the loss factor is considered to be quasi-constant at

high frequency regime and may be approached by

$$(4.2) \quad \tan \delta_\infty \approx \sin((\alpha - \beta)\pi/2).$$

The first estimate of the parameters set (α, β, c) is introduced as the set of initial values in a generalized non-linear regression Maple procedure [21], programmed to identify the loss factor expression (3.8).

A material exhibiting a marked asymmetry of the loss factor is chosen in view to compare the present model with the classical approaches by Pritz and Havriliak–Negami. The damping polymer EAR C-1002 has been chosen, its dynamic shear properties are taken from the data base established by NASHIF and LEWIS [22]. Table 1 summarizes the identified parameters of the three models and Fig. 7 illustrates the obtained fitting of experimental data of the loss factor.

Table 1. Parameter values for the EAR C-1002.

	α	β	c	$\tau(s)$
Present model	0.615158	0.58082	856.8	$2.0467 \cdot 10^{-6}$
Pritz model	0.612924	0.59241	426.4	$6.3027 \cdot 10^{-5}$
Havriliak–Negami model	0.616342	0.5204	554.2	$1.2034 \cdot 10^{-5}$

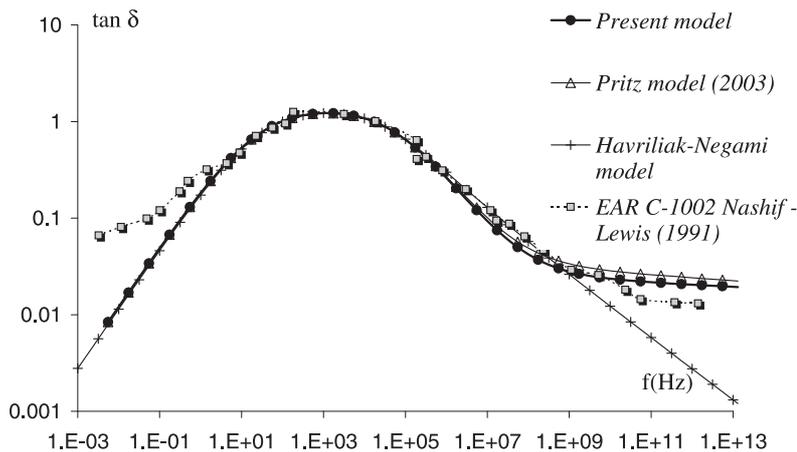


FIG. 7. Parameters identification with the present model, experimental data for the EAR C-1002.

The first fractional derivative order α is quite the same for the three models: the low-frequency branches of loss factor predicted by these models are practically superimposed. The asymmetry parameter β is slightly different as it does not play the same role in the constitutive equations: the Havriliak–Negami model considers the parameter β as a mathematical weighting factor, see (2.11). In the

present model, the parameter β appears as a secondary differentiation order of the stress and strain.

The main improvement of the present model as compared to the Havriliak–Negami one is the best description of the plateau (Fig. 7). This figure shows also that this new approach is as effective as Pritz's five-parameter fractional derivative constitutive law [13] in describing the marked asymmetry of the loss factor.

However, as illustrated by Fig. 1, the present model allows also to describe the quasi-symmetrical loss factor which may be difficult to obtain using the Pritz expression. The improved five-parameter model appears as a more efficient description of both the symmetric and asymmetric evolutions of the loss factor.

5. Conclusion

In this paper, the description of the asymmetrical loss peak in logarithmic frequency domain has been attempted through an improved five-parameter fractional derivative model. A meaningful constitutive equation relating the stress and strain histories has been found; the two different orders of derivation are characterised via the loss factor curve. The main derivative order α governs the low-frequency regime of the dissipative and conservative moduli and the loss factor. The deviation of β from α governs the asymmetry of the loss peak and the high frequency behaviour of the dynamic properties. The classical relaxed and unrelaxed moduli, other relevant characteristics of the material behaviour, are retrieved as the dynamic modulus limits. The generalized relaxation time which is the only temperature-dependent parameter, is raised alternatively to the two different exponents describing, in some sense, two coupling modes of vibration in the entanglements of the polymeric chains.

The improved five-parameter model allows a slight or marked frequency asymmetry in logarithmic scale, when the difference between the orders of time derivative is respectively great or small. When the loss factor shows a plateau at a high-frequency regime, the difference between the orders of time derivatives may be estimated as a function of the plateau value. The dissipative modulus remains bounded, even for high frequencies. Quantitative trends in the damping behaviour of elastomers may be retrieved and explained in the theoretical height-width diagrams.

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