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Five-parameter fractional derivative model for polymeric damping materials

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Abstract

Fractional derivative models offer a powerful tool to describe the dynamic behaviour of real viscoelastic materials. A version of the fractional derivative models characterized by five parameters is presented and investigated in this paper in order to describe asymmetrical loss factor peak and the high-frequency behaviour of polymeric damping materials. The speculative derivation of the model constitutive equation containing time derivatives of stress and strain of different orders is given. The model behaviour is investigated in the frequency domain, the physical meaning of the model parameters is defined and constraints on the parameter values are made. It is shown that the asymmetry of loss peak and the high-frequency behaviour of the model are governed by the difference between the order of time derivatives of stress and strain. Moreover, it is shown that this difference is related to the high-frequency limit value of the loss factor. The model is fitted to experimental data on some polymeric damping materials to verify its behaviour.

1. Introduction

Polymeric materials are widely used for sound and vibration damping. One of the more notable properties of these materials, besides the high damping ability, is the strong frequency dependence of dynamic properties; both the dynamic modulus of elasticity and the damping characterized by the loss factor. The typical behaviour is that the dynamic modulus increases monotonically with the increase of frequency and the loss factor exhibits a wide peak [1,2]. It is rare that the loss factor peak, plotted against logarithmic frequency, is symmetrical with respect to the peak maximum, especially if a wide frequency range is considered. The

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experiments usually reveal that the peak broadens at high frequencies. In addition to this, the experimental data on some polymeric damping materials at very high frequencies, far from the peak centre, show that the loss factor–frequency curve "flattens" and seems to approach a limit value, while the dynamic modulus exhibits a weak monotonic increase at these frequencies [3–8]. These phenomena can be seen in the experimental data published by Madigosky and Lee [3], Rogers [4] and Capps [5] for polyurethanes, and moreover by Fowler [6], Nashif and Lewis [7], and Jones [8] for other polymeric damping materials.

The computerized methods of acoustical and vibration calculus require the mathematical form of frequency dependences of dynamic properties. A reasonable method of describing the frequency dependences is to find a good material model fitting the experimental data. The introduction of fractional calculus into the model theory of viscoelasticity has resulted in a powerful tool to model the dynamic behaviour of polymers and other materials [4,9–21]. In this way, the quantitative behaviour of the conventional viscoelastic models (Kelvin, Maxwell, Zener, etc.) can be improved, and a number of fractional derivative models can be developed. Of these models, the fractional derivative Zener model characterized by four parameters has proved to be especially appropriate to predict the dynamic behaviour of polymeric damping materials over a wide frequency range [4,11,18]. This model is robust and has solid theoretical basis [11], but is not able to describe the asymmetry of the loss peak and the high-frequency behaviour of the dynamic properties outlined above.

Modelling the asymmetrical loss peak is an old problem not only in polymer mechanics, but also in the field of dielectric properties of polymers. For this purpose, empirical models—mathematical formulae—have been developed which can be used in describing either the dielectric or the dynamic mechanical properties of polymers [22,23]. Among the models, the Havriliak–Negami model is especially useful and has been used intensively for the asymmetrical loss factor peak of polymeric damping materials, mainly polyurethanes [23–25]. Nevertheless, the Havriliak–Negami model cannot describe the aforementioned high-frequency behaviour of the polymeric damping materials, since this model predicts a vanishing loss factor and a finite limit value for the dynamic modulus at high frequencies. The other disadvantage of this empirical model is that it cannot be related to the general constitutive equation of viscoelastic materials. One of the fractional derivative models, used by Bagley and Torvik [10], is free from these disadvantages and able to predict an asymmetrical loss peak, but this fractional model is not correct theoretically [11]. Later Friedrich and Braun [14] suggested another empirical formula for asymmetrical loss peak, and showed that this formula could be related to the general constitutive equation of fractional derivative models, but unfortunately no attention has been paid to the loss factor in their work.

The aim of this paper is to show that a version of the fractional derivative Zener model characterized by five parameters, referred to as five-parameter fractional derivative Zener model, is able to describe not only the asymmetrical broadening of the loss factor peak, but also the peculiar high-frequency behaviour of dynamic properties of some polymeric damping materials. The model behaviour will be investigated in the frequency domain, and verified by fitting it to experimental data.

2. Need for five-parameter model

The linear dynamic properties of materials in the frequency domain are characterized by the complex modulus of elasticity. The complex shear modulus $\bar{G}$ is used in this paper to give the dynamic properties of polymeric damping materials. The definition of $\bar{G}$ is

$$\bar{G}(j\omega) = \frac{\tilde{\sigma}(j\omega)}{\tilde{\varepsilon}(j\omega)} = G_d(\omega) + jG_l(\omega) = G_d(\omega)[1 + j\eta(\omega)], \tag{1}$$

where $j = \sqrt{-1}$ is the imaginary unit, $\omega = 2\pi f$; f is the frequency in Hz, $\tilde{\sigma}(j\omega)$ and $\tilde{\varepsilon}(j\omega)$ are the Fourier transforms of the stress- and strain-time histories, respectively, G_d is the dynamic shear modulus, G_l is the shear loss modulus and η is the loss factor,

$$\eta(\omega) = \frac{G_l(\omega)}{G_d(\omega)}. \tag{2}$$

The typical frequency variations of the dynamic properties for polymeric damping materials are illustrated in Fig. 1 with the assumption that the loss peak is symmetrical with respect to logarithmic frequency (solid line). In this case, four parameters are needed as a minimum to specify mathematically the frequency dependences of the dynamic properties. The four parameters are: the frequency ω_η and the magnitude η_m of maximum of the loss factor peak (or those of the loss modulus peak), a parameter, say α, characterizing the slope of increase and decrease of loss functions measured far from the peak centre, moreover the zero frequency value of dynamic modulus, i.e., the static modulus denoted by $G_0 = G_d(0)$. It should be pointed out that the high-frequency value of the dynamic modulus and the slope of increase of $G_d(\omega)$ are dependent on the

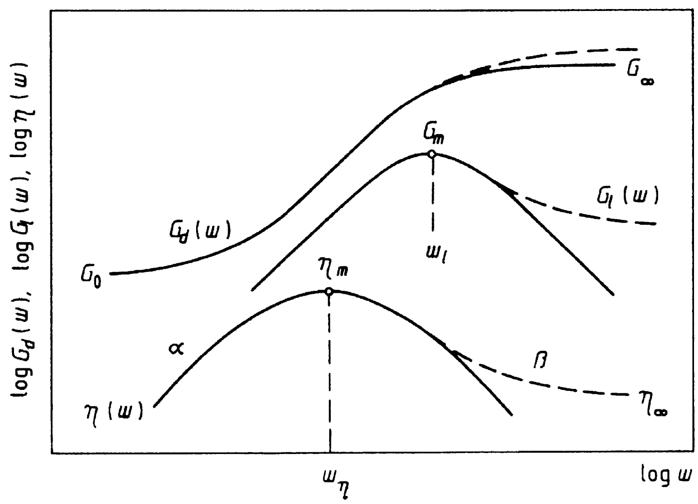

Fig. 1. The experiments made on some polymeric damping materials in a wide frequency range reveal that the loss factor peak is asymmetrical and $\eta(\omega)$ approaches a limit value, while the dynamic modulus exhibits a weak monotonic increase at high frequencies.

damping [26] and, therefore, the four parameters are enough to specify the dynamic modulus-frequency function.

The experiments, however, made on polymeric damping materials over a wide frequency range covering 8–10 decades or more, reveal that the loss factor peak is not symmetrical. Nevertheless, an approximate symmetry may often be seen if a relatively narrow range, say 2–4 decades of frequency is considered around the peak centre. Outside this range, the usual observation is that the loss factor peak broadens at high frequencies. In addition to this, the experiments made on some damping polymers far from the peak centre show that the loss factor–frequency curve "flattens" and seems to approach a limit value η_∞ at high frequencies [3–8]. The "flattening" and the high-frequency value η_∞, for example for a polymeric damping material can be observed as far from the peak centre as about 6–7 decades, and 9–10 decades of frequency, respectively [7,8]. The frequency dependence of the loss modulus is similar to that of the loss factor, while the dynamic modulus exhibits a monotonic increase in the whole frequency range of experiments. These phenomena are illustrated in Fig. 1 with a broken line, and some experimental data will be given in Section 5. (More details on the high-frequency behaviour of polymeric materials and its theoretical explanation will be given in a subsequent paper.) It is evident that at least one more parameter besides the four previous ones, say β governing the high-frequency behaviour of the material, has to be introduced for describing asymmetrical loss peak. Consequently, five parameters as a minimum are needed to model the dynamic behaviour of these polymeric damping materials over a wide frequency range.

3. Preliminaries—the four-parameter model

The fractional derivative models have been proved to be efficient in describing the dynamic behaviour of real materials, especially polymers used for sound and vibration control [4,9–21]. The development of these models is due to the recognition of the fact that the quantitative behaviour of the conventional viscoelastic models can be improved by replacing the integer order time derivatives of stress and strain in the relevant model constitutive equation with fractional order derivatives. The general form of the constitutive equation for the conventional viscoelastic models is [10]

$$\sigma(t) + b_1 \frac{\mathrm{d}}{\mathrm{d}t}\sigma(t) + b_2 \frac{\mathrm{d}^2}{\mathrm{d}t^2}\sigma(t) + \cdots + b_n \frac{\mathrm{d}^n}{\mathrm{d}t^n}\sigma(t)$$
$$= a_0\varepsilon(t) + a_1 \frac{\mathrm{d}}{\mathrm{d}t}\varepsilon(t) + a_2 \frac{\mathrm{d}^2}{\mathrm{d}t^2}\varepsilon(t) + \cdots + a_m \frac{\mathrm{d}^m}{\mathrm{d}t^m}\varepsilon(t), \tag{3}$$

where t is the time, $a_0, a_1, \ldots, a_m$ and, $b_0, b_1, \ldots, b_n$ are material constants. It is important to note that the number of time derivatives of stress and strain in Eq. (3) cannot be arbitrary to insure that the model is physically meaningful satisfying the thermodynamic requirements. It is known that the thermodynamic requirements are satisfied only if $m = n$ or $m = n + 1$ [18]. The replacement of the integer order derivatives with fractional order ones results in the general form of the constitutive equation for the fractional derivative model:

$$\sigma(t) + b_1 \frac{\mathrm{d}^{\beta_1}}{\mathrm{d}t^{\beta_1}} \sigma(t) + b_2 \frac{\mathrm{d}^{\beta_2}}{\mathrm{d}t^{\beta_2}} \sigma(t) + \cdots + b_n \frac{\mathrm{d}^{\beta_n}}{\mathrm{d}t^{\beta_n}} \sigma(t) = a_0\varepsilon(t) + a_1 \frac{\mathrm{d}^{\alpha_1}}{\mathrm{d}t^{\alpha_1}}\varepsilon(t) + a_2 \frac{\mathrm{d}^{\alpha_2}}{\mathrm{d}t^{\alpha_2}}\varepsilon(t) + \cdots + a_m \frac{\mathrm{d}^{\alpha_m}}{\mathrm{d}t^{\alpha_m}} \varepsilon(t), \tag{4}$$

where $0<\alpha_1<\alpha_2<\cdots<\alpha_m<1$ and $0<\beta_1<\beta_2<\cdots<\beta_n<1$ are material constants. The fractional derivation, say the αth order derivative of $\varepsilon(t)$, can be defined by the gamma function (Γ) as [10]

$$\frac{\mathrm{d}^{\alpha}}{\mathrm{d}t^{\alpha}} \varepsilon(t) = \frac{1}{\Gamma(1-\alpha)} \frac{\mathrm{d}}{\mathrm{d}t} \int_0^t \frac{\varepsilon(\tau)}{(t-\tau)^{\alpha}} \mathrm{d}\tau. \tag{5}$$

Starting with Eq. (4), the fractional version of any conventional viscoelastic model, e.g., Kelvin, Maxwell, Zener, etc., is easy to develop. Among the models, the fractional Zener model has been found to be efficient to predict frequency variations like those drawn by solid line in Fig. 1 (the symmetrical loss factor peak). The constitutive equation for this model can be derived from Eq. (4) with the assumption that all parameters are zero with the exception of a_0, a_1, b_1, α_1 and β_1, then

$$\sigma(t) + b_1 \frac{\mathrm{d}^{\beta_1}}{\mathrm{d}t^{\beta_1}} \sigma(t) = a_0\varepsilon(t) + a_1 \frac{\mathrm{d}^{\alpha_1}}{\mathrm{d}t^{\alpha_1}} \varepsilon(t). \tag{6}$$

This equation contains five parameters, but four of them are enough for the symmetrical loss peak. Bearing in mind that all the four members in Eq. (6) are necessary to model the dynamic behaviour of a solid material exhibiting one loss peak, there is only one way to reduce the number of the parameters, that is to assume that: $\beta_1 = \alpha_1 = \alpha$. Then, the model parameters can be written as

$$b_1 = \tau^{\alpha}, \tag{7}$$

$$a_0 = G_0, \tag{8}$$

$$a_1 = G_{\infty}\tau^{\alpha}, \tag{9}$$

where τ is the relaxation time and the parameters G_0 and G_{∞} have modulus dimension. Using these notations, Eq. (6) is

$$\sigma(t) + \tau^{\alpha} \frac{\mathrm{d}^{\alpha}}{\mathrm{d}t^{\alpha}} \sigma(t) = G_0\varepsilon(t) + G_{\infty}\tau^{\alpha} \frac{\mathrm{d}^{\alpha}}{\mathrm{d}t^{\alpha}} \varepsilon(t). \tag{10}$$

This equation yields the constitutive equation of the conventional Zener model if $\alpha = 1$. Therefore, the model represented by Eq. (7) is rightly referred to as fractional Zener model, or more precisely as the four-parameter fractional derivative Zener model.

The complex modulus of the model can be derived by transforming Eq. (10) into the frequency domain. The derivation is easy to perform, since [10]

$$F \frac{\mathrm{d}^{\alpha}}{\mathrm{d}t^{\alpha}} \varepsilon(t) = (\mathrm{j}\omega)^{\alpha} F\varepsilon(t), \tag{11}$$

where F denotes the Fourier transform. The derivation results in

$$\bar{G}(j\omega) = G_0 \frac{1 + d(j\omega\tau)^\alpha}{1 + (j\omega\tau)^\alpha}, \tag{12}$$

where

$$d = \frac{G_\infty}{G_0}. \tag{13}$$

The rearranged form of Eq. (12) will be useful in this paper:

$$\bar{G}(j\omega) = G_0 + G_0(d-1)\frac{(j\omega\tau)^\alpha}{1 + (j\omega\tau)^\alpha}. \tag{14}$$

The components of the complex modulus are:

$$G_d(\omega) = G_0 \frac{1 + (d+1)\cos(\alpha\pi/2)\omega_n^\alpha + d\omega_n^{2\alpha}}{1 + 2\cos(\alpha\pi/2)\omega_n^\alpha + \omega_n^{2\alpha}}, \tag{15}$$

$$G_l(\omega) = G_0 \frac{(d-1)\sin(\alpha\pi/2)\omega_n^\alpha}{1 + 2\cos(\alpha\pi/2)\omega_n^\alpha + \omega_n^{2\alpha}}, \tag{16}$$

$$\eta(\omega) = \frac{(d-1)\sin(\alpha\pi/2)\omega_n^\alpha}{1 + (d+1)\cos(\alpha\pi/2)\omega_n^\alpha + d\omega_n^{2\alpha}}, \tag{17}$$

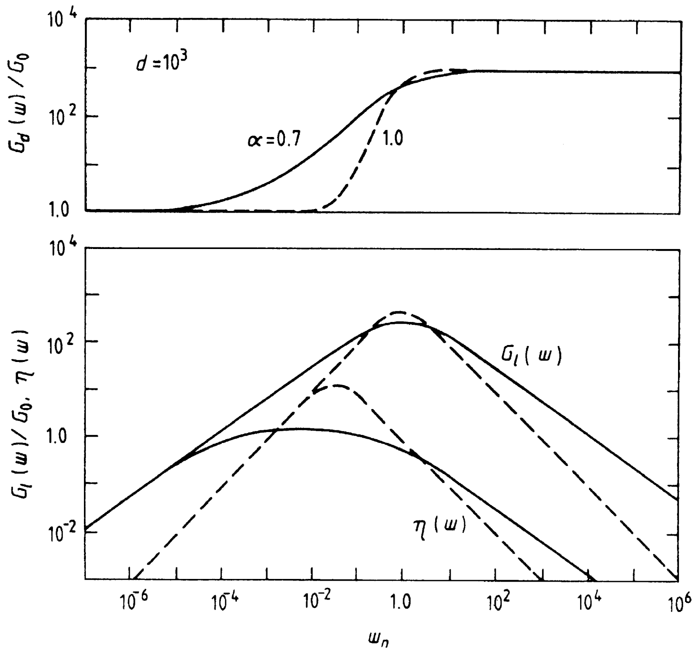

Fig. 2. Frequency variations of the dynamic modulus, loss modulus and loss factor predicted by the four-parameter fractional Zener model (—) and the conventional Zener model (----), respectively.

where ω_n is the normalized frequency,

$$\omega_n = \omega\tau. \tag{18}$$

The variations of $G_d(\omega)/G_0$, $G_l(\omega)/G_0$ and $\eta(\omega)$ are plotted against the normalized frequency in Fig. 2 in a log–log co-ordinate system for $\alpha = 0.7$ and $d = G_\infty/G_0 = 10^3$ by way of example. The behaviour of the conventional Zener model ($\alpha = 1$) is shown too for the sake of comparison. It can be seen that the dynamic modulus monotonically increases with increasing frequency, but $G_d(\omega)$ has an upper limit value. Both the loss modulus and the loss factor have a peak, which are symmetrical with respect to logarithmic frequency. The maximum in the loss modulus occurs at $\omega_n = 1$, i.e., at the frequency of

$$\omega_l = 1/\tau. \tag{19}$$

Therefore, the normalized frequency can be defined as

$$\omega_n = \omega/\omega_l. \tag{20}$$

The model behaviour below and above the loss modulus peak is clear from the low- and high-frequency approximations, respectively, of Eqs. (15)–(17). The approximations with the assumption that $G_\infty/G_0 \gg 1$ result in

$$G_d(\omega) \cong G_0, \tag{21}$$

$$G_l(\omega) \cong G_\infty \sin(\alpha\pi/2)\omega_n^\alpha, \tag{22}$$

$$\eta(\omega) \cong \frac{G_\infty}{G_0} \sin(\alpha\pi/2)\omega_n^\alpha, \tag{23}$$

if $\omega_n \ll 1$, and

$$G_d(\omega) \cong G_\infty, \tag{24}$$

$$G_l(\omega) \cong \frac{G_\infty \sin(\alpha\pi/2)}{\omega_n^\alpha}, \tag{25}$$

$$\eta(\omega) \cong \frac{\sin(\alpha\pi/2)}{\omega_n^\alpha}, \tag{26}$$

if $\omega_n \gg 1$. It can be seen from this brief study that all parameters in the model have a clear physical meaning, namely: G_0 is the static modulus of elasticity, G_∞ is the high-frequency limit value of dynamic modulus, and the parameter α governs the increase and decrease of the loss functions at low and high frequencies, respectively. Note that Eq. (19) offers another interpretation for the meaning of parameter τ, that is $1/\tau$ is the frequency of maximum in the loss modulus peak.

The four-parameter fractional Zener model has solid theoretical basis, it is related to the general fractional derivative constitutive equation of viscoelastic materials as discussed above, moreover the model is causal and satisfies the thermodynamic constraints [11]. The model has successfully been fitted to experimental data on a wide variety of materials, especially polymers for vibration damping [4,9,11,18]. Nevertheless, the four-parameter model is not able to predict asymmetrical loss peak.

It seems to be reasonable to try to modify the four-parameter model by assuming that the order of time derivative of stress and strain is different in Eq. (6), that is $\alpha_1 \neq \beta_1$, in order to create asymmetrical loss peak. This assumption leads to the constitutive equation:

$$\sigma(t) + \tau^{\beta} \frac{\mathrm{d}^{\beta}}{\mathrm{d}t^{\beta}} \sigma(t) = G_0 \varepsilon(t) + G_{\infty} \tau^{\alpha} \frac{\mathrm{d}^{\alpha}}{\mathrm{d}t^{\alpha}} \varepsilon(t), \tag{27}$$

where the notations $\alpha = \alpha_1$ and $\beta = \beta_1$ have been introduced. The complex modulus of the model is

$$\bar{G}(\mathrm{j}\omega) = G_0 \frac{1 + d(\mathrm{j}\omega\tau)^{\alpha}}{1 + (\mathrm{j}\omega\tau)^{\beta}}. \tag{28}$$

This model was used by Bagley and Torvik [10] to fit experimental data to a damping material exhibiting asymmetrical loss peak. Nevertheless, it was later shown theoretically that this version of the fractional Zener model was not correct, because it does not satisfy the thermodynamic constraints [11].

4. The five-parameter model

4.1. Constitutive equation

As a result of the above survey of the four-parameter fractional Zener model and its modification, it can be concluded that the number of time derivatives in Eq. (6) has to be increased to create a physically meaningful five-parameter model. The number of time derivatives of strain is reasonable to increase, bearing in mind that with the conventional viscoelastic models (i.e., in case of integer order derivatives) the number of time derivatives of stress must not be larger than that of strain to satisfy the thermodynamic constraints as mentioned in Section 3. Consequently, the new constitutive equation is

$$\sigma(t) + b_1 \frac{\mathrm{d}^{\beta_1}}{\mathrm{d}t^{\beta_1}} \sigma(t) = a_0 \varepsilon(t) + a_1 \frac{\mathrm{d}^{\alpha_1}}{\mathrm{d}t^{\alpha_1}} \varepsilon(t) + a_2 \frac{\mathrm{d}^{\alpha_2}}{\mathrm{d}t^{\alpha_2}} \varepsilon(t), \tag{29}$$

where $\alpha_2 > \alpha_1$. This equation contains seven parameters, but two of them are unnecessary for our purpose. In order to reduce the number of parameters, it is assumed by the analogy of deriving the four-parameter model that: $\alpha_1 = \beta_1 = \beta$. The parameters can be written as

$$b_1 = \tau^{\beta}, \tag{30}$$

$$a_0 = G_0, \tag{31}$$

$$a_1 = G_1 \tau^{\beta}, \tag{32}$$

$$a_2 = G_2 \tau^{\alpha}, \tag{33}$$

where G_1 and G_2 have modulus dimension, and $\alpha = \alpha_2$. Using these notations, Eq. (29) is

$$\sigma(t) + \tau^\beta \frac{d^\beta}{dt^\beta}\sigma(t) = G_0\varepsilon(t) + G_1\tau^\beta \frac{d^\beta}{dt^\beta}\varepsilon(t) + G_2\tau^\alpha \frac{d^\alpha}{dt^\alpha}\varepsilon(t), \tag{34}$$

where $\alpha > \beta$. Now the model has six parameters, which are easy to reduce to five by considering the fact that Eq. (34) must yield the four-parameter fractional Zener model if $\alpha = \beta$. This requirement is satisfied by the choice of

$$G_1 = G_0 \tag{35}$$

and

$$G_2 = G_\infty - G_0. \tag{36}$$

Therefore, five parameters ($G_0, G_\infty, \alpha, \beta$ and τ) have remained, and the final form of the model constitutive equation is

$$\sigma(t) + \tau^\beta \frac{d^\beta}{dt^\beta}\sigma(t) = G_0\varepsilon(t) + G_0\tau^\beta \frac{d^\beta}{dt^\beta}\varepsilon(t) + (G_\infty - G_0)\tau^\alpha \frac{d^\alpha}{dt^\alpha}\varepsilon(t). \tag{37}$$

The model represented by Eq. (37) is referred to as the five-parameter fractional derivative Zener model, or shortly as the five-parameter fractional model.

The complex modulus for the five-parameter model is easy to derive by means of Eq. (11), the result is

$$\bar{G}(j\omega) = G_0 + G_0(d-1)\frac{(j\omega\tau)^\alpha}{1 + (j\omega\tau)^\beta}. \tag{38}$$

The comparison of Eqs. (38) and (14) suggests that the five-parameter model can be derived from the four-parameter model simply by replacing the α exponent in the denominator of Eq. (14) with β. Friedrich and Braun [14] introduced in this way the complex modulus of the five-parameter model and showed that the transformation of Eq. (38) into the time-domain results in the constitutive equation (37).

It should be noted that the constitutive equation (37) contains formally the same parameters as Eq. (10), but the meaning of some parameters occurring in both equations is not the same. The investigation of the model behaviour in the time-domain has revealed that τ is the relaxation time [14], but the value of τ is evidently different from that in the four-parameter model. The parameter G_0 is naturally the static modulus, and it is very probable that G_∞ is related to the high-frequency behaviour of dynamic modulus, but the precise meaning of G_∞ is not known yet. The meaning of G_∞, moreover α and β will be cleared up by investigating the model behaviour in the frequency domain.

Note further that the derivation of the model constitutive Eq. (37) imply a restriction on the relation between α and β, that is: $\alpha > \beta$. It will also be shown by investigating the model behaviour that this restriction is absolutely necessary to insure that the five-parameter model is physically meaningful.

4.2. Model behaviour

The dynamic and loss moduli are derived from Eq. (38) to investigate the model behaviour in the frequency domain. Some mathematical manipulations on the real and imaginary parts of Eq. (38) result in

$$G_d(\omega) = G_0 + G_0(d-1)\,\frac{\cos(\alpha\pi/2)\omega_n^{\alpha} + \cos[(\alpha-\beta)\pi/2]\omega_n^{\alpha+\beta}}{1 + 2\cos(\beta\pi/2)\omega_n^{\beta} + \omega_n^{2\beta}}, \tag{39}$$

$$G_l(\omega) = G_0(d-1)\,\frac{\sin(\alpha\pi/2)\omega_n^{\alpha} + \sin[(\alpha-\beta)\pi/2]\omega_n^{\alpha+\beta}}{1 + 2\,\cos(\beta\pi/2)\omega_n^{\beta} + \omega_n^{2\beta}}, \tag{40}$$

$$\eta(\omega) = \frac{(d-1)\{\sin(\alpha\pi/2)\omega_n^{\alpha} + \sin[(\alpha-\beta)\pi/2\}\omega_n^{\alpha+\beta}}{1 + 2\cos(\beta\pi/2)\omega_n^{\beta} + \omega_n^{2\beta} + (d-1)\{\cos(\alpha\pi/2)\omega_n^{\alpha} + \cos[(\alpha-\beta)\pi/2)]\omega_n^{\alpha+\beta}\}}, \tag{41}$$

where ω_n is defined by Eq. (18).

The frequency variations of the dynamic and loss moduli and the loss factor were studied numerically with special respect to the role of the difference between α and β, bearing in mind that $\alpha > \beta$. Typical results with parameters of $\alpha = 0.7$; $\beta = 0.69$, 0.65, 0.60, and $d = 10^3$ are shown in Fig. 3. The behaviour of the four-parameter fractional Zener model ($\alpha = \beta = 0.7$) is shown too in Fig. 3 for the sake of comparison.

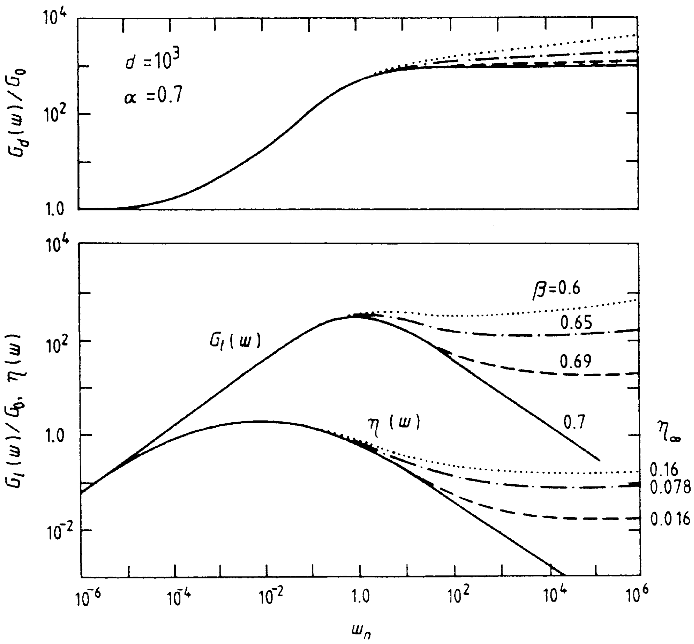

Fig. 3. Frequency variations of dynamic modulus, loss modulus and loss factor predicted by the five-parameter fractional Zener model (- - - -, —· –, ···) and the four-parameter fractional Zener model (—), respectively.

Fig. 3 demonstrates that there is a striking difference between the behaviours of the five- and the four-parameter models at high frequencies. Nevertheless, the low-frequency behaviour of these models is practically the same, as one can see from the low-frequency approximation of Eqs. (39)–(41), resulting in

$$G_d(\omega) \cong G_0, \tag{42}$$

$$G_l(\omega) \cong G_\infty \sin(\alpha\pi/2)\omega_n^\alpha, \tag{43}$$

$$\eta(\omega) \cong \frac{G_\infty}{G_0} \sin(\alpha\pi/2)\omega_n^\alpha, \tag{44}$$

provided that $\omega_n \ll 1$ and $G_\infty/G_0 \gg 1$. The same relations have been derived in Section 3 for the four-parameter model. Therefore, it can be concluded that the difference between α and β practically has no effect on the behaviour of the five-parameter model at low frequencies.

In contrast to this, the behaviour of the five-parameter model at high frequencies is fundamentally affected by the value of $\alpha - \beta$. The larger the difference between α and β is, the more prominent its effect is on the model behaviour. It can be seen in Fig. 3 that the loss peak broadens at high frequencies. In addition to the peak broadening, there is another fundamental difference between the high-frequency behaviour of the five- and four-parameter models. It is known that the dynamic modulus of the four-parameter model approaches a limit value (G_∞) at high frequencies, while the relevant loss functions approach zero (Fig. 2). In contrast to this behaviour, the dynamic modulus of the five-parameter model monotonically increases at high frequencies without upper limit, and the loss modulus, after a slight decrease above the peak maximum starts to increase like the dynamic modulus, and the loss factor approaches a limit value η_∞. All these are clear from the high-frequency approximations of Eqs. (39)–(41) resulting in

$$G_d(\omega) \cong G_\infty \cos[(\alpha - \beta)\pi/2]\omega_n^{\alpha-\beta}, \tag{45}$$

$$G_l(\omega) \cong G_\infty \sin[(\alpha - \beta)\pi/2]\omega_n^{\alpha-\beta}, \tag{46}$$

$$\eta_\infty \cong \tan[(\alpha - \beta)\pi/2], \tag{47}$$

provided that $\omega_n \gg 1$ and $G_\infty/G_0 \gg 1$. It can be read from these equations that the slope of the high-frequency increase of the dynamic and the loss moduli is the same, and this slope is determined by the value of $\alpha - \beta$. Moreover, it can be seen that $\alpha - \beta$ is related to the high-frequency value of the loss factor.

The latter fact provides a useful tool to estimate the difference between α and β for polymeric damping materials. It is known from the experiments that the high-frequency value of loss factor of these materials is usually smaller than 0.1 [3–8]. Using this value, it can be predicted by Eq. (47) that

$$\alpha - \beta < 0.06 \tag{48}$$

with polymeric damping materials. Therefore, Eqs. (45)–(47) can further be simplified as

$$G_d(\omega) \cong G_\infty \omega_n^{\alpha-\beta}, \tag{49}$$

$$G_l(\omega) \cong \frac{\pi}{2} G_\infty (\alpha - \beta)\omega_n^{\alpha-\beta}, \tag{50}$$

$$\eta_\infty \cong \frac{\pi}{2}(\alpha - \beta). \tag{51}$$

It is interesting to note that the value of $\alpha - \beta$ practically has no effect on either the maximum value or the position of the loss factor peak. On the contrary, the loss modulus peak is more sensitive to the value of $\alpha - \beta$ than the loss factor peak is. The loss modulus peak is shifted toward higher frequencies and the peak magnitude slightly increases with the increase of $\alpha - \beta$. In addition to this, the loss modulus peak may disappear if $\alpha - \beta$ exceeds a certain value, e.g., $\alpha - \beta > 0.1$, but this value is not realistic with solid polymeric damping materials as mentioned before.

As a result of this study, the physical meaning of the parameters α, β and G_∞ in the model constitutive equation (37) can be given. It is clear that α governs the low frequency increase of the loss modulus and the loss factor. The value of β, or more precisely the deviation of β from α, governs the asymmetry of the loss peak and the high-frequency behaviour of dynamic properties. Finally, the parameter G_∞ is related to the high-frequency value of the dynamic modulus, but here, in contrast to the four-parameter model, G_∞ is not the limit value of $G_d(\omega)$. The precise meaning of G_∞ can be defined by means of Eq. (39), that is G_∞ is a value of the dynamic modulus which occurs above the loss modulus peak at a frequency determined by the solution of equation as follows:

$$\frac{G_d(\omega)}{G_\infty} = 1 = \frac{\cos(\alpha\pi/2)\omega_n^\alpha + \cos[(\alpha - \beta)\pi/2]\omega_n^{\alpha+\beta}}{1 + 2\cos(\beta\pi/2)\omega_n^\beta + \omega_n^{2\beta}}. \tag{52}$$

Fig. 4 shows the graphical solution of this transcendental equation for some values of α and $\alpha - \beta$ which are characteristic of polymeric damping materials. It can be read from Fig. 4 that G_∞ is a value of dynamic modulus at a frequency between about $10\omega_l$ and $100\omega_l$.

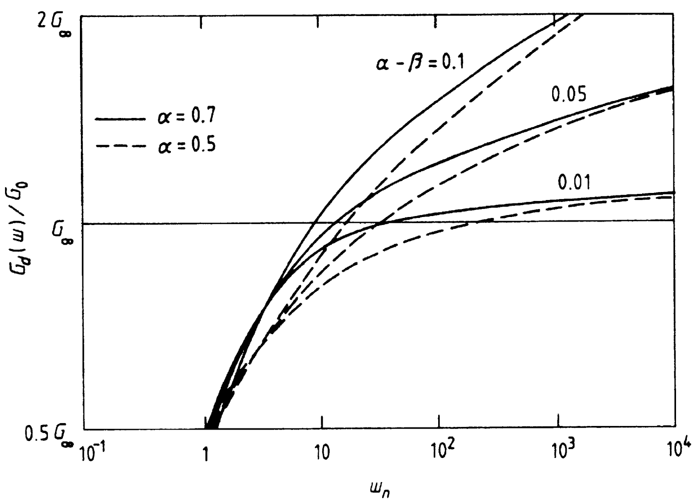

Fig. 4. G_∞ is a value of dynamic modulus at a frequency somewhere between $10\omega_l$ and $100\omega_l$ with polymeric damping materials (ω_l is the frequency of maximum in loss modulus).

Finally, in relation to the high-frequency behaviour it should be noted that the five-parameter model is not intended to be used beyond a certain frequency. The model is aimed to be used within a frequency range where the asymmetry of the loss peak and the peculiar high-frequency behaviour of some polymeric materials can be observed.

4.3. Thermodynamic requirements

It is known from thermodynamics applied to real solid materials loaded dynamically that the internal work and the dissipated energy must be positive [11]. It has been proved that these requirements are satisfied if both the dynamic modulus and the loss modulus are positive for all frequencies [11], i.e.,

$$G_d(\omega) \geqslant 0 \quad \text{and} \quad G_l(\omega) \geqslant 0, \tag{53}$$

for

$$0 < \omega < \infty .$$

It is clear that a physically meaningful model must satisfy the thermodynamic requirements. On the basis of conditions (53), restrictions on the model parameters can be developed. It can be seen from Eq. (39) that the dynamic modulus of the five-parameter model is positive for all frequencies regardless of the value of α and β, if

$$G_0 \geqslant 0, \tag{54}$$

$$G_\infty \geqslant 0, \tag{55}$$

$$d \geqslant 1 \tag{56}$$

and

$$\tau \geqslant 0. \tag{57}$$

It follows from the physical meaning of the model parameters discussed above that conditions (54)–(57) are satisfied and, therefore, the dynamic modulus is positive for all frequencies. On the contrary, the loss modulus can be negative for some frequencies if $\alpha < \beta$, even if conditions (54)–(57) are satisfied. The fact that the energy loss is negative at high frequencies if $\alpha < \beta$, is clear from Eqs. (46) and (47). Therefore, it can be concluded that the five-parameter fractional Zener model is physically meaningful only if

$$\alpha > \beta. \tag{58}$$

This conclusion supports the constraint implied in the derivation of the model constitutive equation. Moreover, constraints (54)–(58) are in complete agreement with those developed by Friedrich and Braun [14] as a result of investigating the model behaviour in the time domain.

5. Model and experimental data

The fitting of a model to the relevant experimental data is the true test of the model behaviour. The five-parameter fractional Zener model was fitted to experimental data on some polymeric

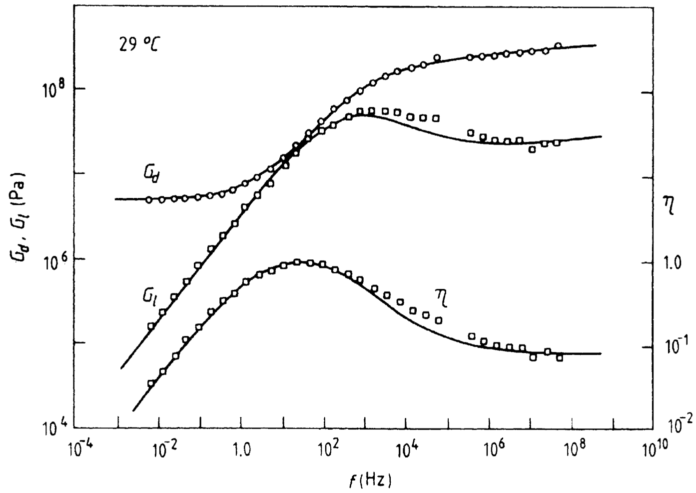

Fig. 5. Dynamic shear properties of damping material GE.SMRD. □, ○, Experimental data from Ref. [6] (some overlapping data have been omitted for the sake of clarity); —, calculated by the five-parameter fractional Zener model with the parameters given in Table 1.

damping materials exhibiting dynamic behaviour outlined in the paper. The data were taken from the literature after a careful consideration of the frequency range, the scatter and the reliability of the data, and after the evaluation of the experimental method applied to produce the data. The materials chosen for the fitting procedure include a polyurethane rubber [3], two commercially available damping polymers known as GE.SMRD [6], and EAR C-1002 [7,8], respectively. In addition to these organic polymers, an inorganic polymer known as Corning glass developed for high temperature damping [10] was chosen for the fitting. In all cases, the fitting of the five-parameter fractional Zener model to the experimental data was remarkably good. Results for two materials are presented and discussed here briefly by way of example.

Fig. 5 shows the experimental values of the dynamic shear properties of the damping material GE.SMRD taken from Ref. [6]. The dynamic properties were measured by the composite beam method at several temperatures, and the data covering about 10 decades of frequency were determined by means of the frequency–temperature equivalence principle. The low scatter and the smoothness of the reduced frequency curves testify the reliability of the experimental data. The frequency dependences of the dynamic properties undoubtedly reveal a material behaviour to be described by the five-parameter fractional model. The loss factor peak is asymmetrical, it broadens above the maximum and $\eta(\omega)$ approaches a value of approx. 0.08 at high frequencies. The loss modulus exhibits a similar behaviour, however, the relevant peak is less prominent than the loss factor peak. The increase of the dynamic modulus is monotonic and no upper limit can be seen in the frequency range of the experiment.

The smooth frequency curves inspired the author to try to read off the model parameters directly from the experimental data. The value of α could easily be determined from the frequency

Table 1
Parameter values determined for the five-parameter fractional Zener model

Material	G_0 (Pa)	G_∞ (Pa)	d	η_∞	α	β	f_l (Hz)	τ (s)
GE.SMRD	5×10^6	1.8×10^8	36	0.08	0.605	0.554	760	2.09×10^{-4}
EAR C-1002	8×10^5	1.256×10^9	1570	0.012	0.566	0.558	2.2×10^6	7.23×10^{-10}

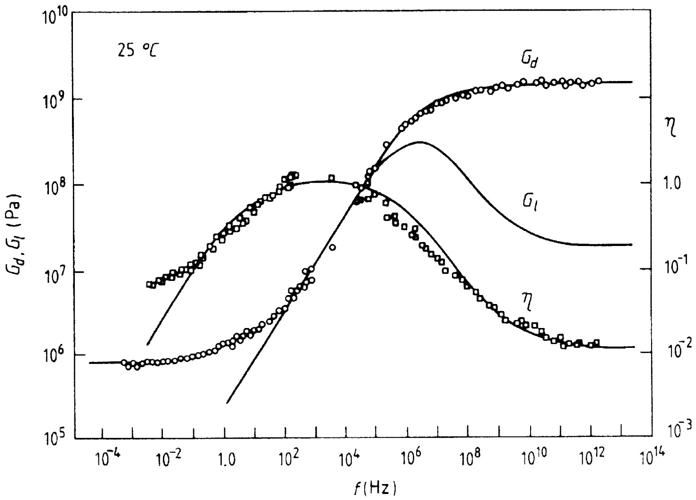

Fig. 6. Dynamic shear properties of damping material EAR C-1002. □, ○, Experimental data from Ref. [7] (some overlapping and erroneous data have been omitted for the sake of clarity); —, calculated by the five-parameter fractional Zener model with the parameters given in Table 1.

increase of the loss modulus, and β was calculated by Eq. (47) from the high-frequency limit value of the loss factor. Moreover, the static modulus G_0 could quite reliably be read off from Fig. 5, but only approximate values for G_∞ and the f_l frequency of the loss modulus peak could be determined from the experimental data. These parameter values were used to start the usual fitting procedure. The refined parameters are given in Table 1, and the model behaviour calculated by these parameters is shown in Fig. 5. The fitting of the five-parameter fractional model to the experimental data is very convincing.

The experimental values of the dynamic shear modulus and the relevant loss factor determined for the damping material EAR C-1002 are given in Fig. 6. The data have been taken from Ref. [7]. The data cover a very wide frequency range extending from 10^{-3} up to 10^{12} Hz. The wide frequency range is the result of the use of different measurement techniques and the application of the frequency–temperature equivalence principle. The low-frequency data were measured by a direct stiffness method, and the high-frequency data were determined by the well-known composite beam method used in a wide temperature range. It should be noted that this material was the subject of an international Round Robin test [8], and the results of this test support the reliability of data given in Fig. 6. It can be seen that the experimental values of the dynamic modulus have low scatter and the relevant frequency curve is pretty smooth. In contrast to this, the loss factor data exhibit somewhat larger scatter, as usual, but the peak asymmetry and the asymptotic high-frequency behaviour of the loss factor is unambiguous with this material too. The experimental values of the dynamic Young's modulus and the relevant loss factor are also available for this material up to 10^{14} Hz [7,8], and these data definitely support the frequency variations of the shear dynamic properties seen in Fig. 6.

The fitting procedure was started again by estimating the parameter values from the experimental frequency curves. The approximate value of α, in this case, was determined from the maximum slope of the dynamic modulus–frequency curve, since this slope is nearly identical with that of the frequency increase of the loss modulus if $G_\infty/G_0 \gg 1$ [26]. The value of β was calculated by the aforementioned method from the high-frequency loss factor ($\eta_\infty \cong 0.012$). Quite a reliable value for G_0 and G_∞ could be read from the smooth dynamic modulus–frequency curve. Finally, the frequency f_l of the loss modulus peak was estimated from the frequency f_η of the loss factor peak by using the relationship derived for the four-parameter fractional Zener model [18]:

$$f_l = f_\eta \left(\frac{G_\infty}{G_0} \right)^{1/2\beta}. \tag{59}$$

The refined parameter values determined by the fitting procedure are given in Table 1, and the model behaviour is shown in Fig. 6. It can be seen that the five-parameter fractional Zener model is able to describe the dynamic behaviour of this damping material over a range covering 16 decades of frequency.

6. Conclusions

A modified version of the fractional derivative Zener model characterized by five-parameters has been derived and investigated in this paper in order to describe the asymmetry of the loss factor peak and the high-frequency behaviour of dynamic properties experienced with some polymeric damping materials. A speciality of this five-parameter model is due to the fact that the relevant constitutive equation contains time derivatives of stress and strain of different orders. A speculative derivation of the constitutive equation has been presented, and the physical meaning of the model parameters has been cleared up. The behaviour of the five-parameter fractional model has been investigated in the frequency domain and restrictions on the parameter values have been developed. As a result of this investigation the following main conclusions can be drawn.

(a) The five-parameter fractional model is physically meaningful only in that case if the order of time derivative of strain is larger than that of stress.
(b) The five-parameter fractional model predicts an asymmetrical loss factor peak and that the loss factor approaches a limit value, while the loss modulus and dynamic modulus increase by the same power function at high frequencies.
(c) The asymmetry of the loss factor peak and the model high-frequency behaviour is governed by the difference between the order of time derivatives of strain and stress.
(d) The difference between the order of time derivatives of strain and stress is related to the high-frequency limit value of the loss factor.

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