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# Computation of the relaxation and creep functions of elastomers from harmonic shear modulus

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**Abstract** The purpose of this paper is to compute the relaxation and creep functions from the data of shear complex modulus, $G^*(i\nu)$. The experimental data are available in the frequency window $\nu \in [\nu_{\min}, \nu_{\max}]$ in terms of the storage $G'(\nu)$ and loss $G''(\nu)$ moduli. The loss factor $\eta(\nu) = \frac{G''(\nu)}{G'(\nu)}$ is asymmetrical function. Therefore, a five-parameter fractional derivative model is used to predict the complex shear modulus, $G^*(i\nu)$. The corresponding relaxation spectrum is evaluated numerically because the analytical solution does not exist. Thereby, the fractional model is approximated by a generalized Maxwell model and its rheological parameters $(G_k, \tau_k, N)$ are determined leading to the discrete relaxation spectrum $G(t)$ valid in time interval corresponding to the frequency window of the input experimental data. Based on the deterministic approach, the creep compliance $J(t)$ is computed on inversing the relaxation function $G(t)$.

**Keywords** Linear viscoelasticity · Complex modulus · Fractional derivative · Relaxation spectrum · Creep compliance · Elastomers

## 1 Introduction

In order to characterize the mechanical behaviour of viscoelastic materials, experiments can be carried out either in frequency or in time domains. In the engineering applications, the relaxation or retardation time spectrum, $G(t)$ or $J(t)$, are often required; especially for rubber-like materials, the stress-imposed problems pose a great challenge (Christensen 1980). For example, the creep compliance is needed to design elastomeric structures such as the responses of rubber tires and bond lines in adhesive joints.

Given this spectrum, it is very easy to convert one material function into another. However, this function is not directly accessible by experiment. It is possible to deduce the spectrum from noisy data of the dynamic moduli. For this purpose the integral equations supplied by linear viscoelastic theory need to be inverted (Gross 1953; Ferry 1980;

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Tschoegl 1989). This is known to be an ill-posed inverse problem (Honerkamp 1989; Honerkamp and Weese 1990, 1993; Malkin 1990; Schapery and Park 1999b).

From experimental point view, the relaxation modulus can be determined by applying a step-strain, in practice this loading cannot be performed due to infinite short ramp time (Lee and Knauss 2000; Sorvari and Malinen 2006); while the creep test can be performed with a simple dead weight system. In consequence, the relaxation modulus or creep compliance is unknown at small times, i.e. in the range time $[0, t_{\min}]$ where $t_{\min}$ is the ramp time during the relaxation loading. In order to approach the step-strain, theoretically $t_{\min}$ needs to be near zero. We note that the transient's experiments are very long tests and time consuming.

A second method (DMA: Dynamical Mechanical Analysis) is used for the characterization of the viscoelastic behaviours of polymers in which both storage $G'(\nu)$ and loss $G''(\nu)$ moduli are measured over wide range of frequency. Highly sophisticated techniques are available for that propose (Caracciolo et al. 2001), the specimen is deformed sinusoidally at small amplitudes and the stress response is measured to obtain dynamic moduli $G'(\nu)$ and $G''(\nu)$.

At small times, the relaxation and creep experiments cannot replace the vibration measurements at high frequencies. Therefore, it is practical to convert the available dynamic moduli, $G'(\nu)$ and $G''(\nu)$ data (which are easy to measure) from the frequency domain into data in the time domain in terms of the linear relaxation modulus $G(t)$ and creep compliance $J(t)$. In the field of rheology, many papers are dictated for this subject (Baumgaertel and Winter 1989, 1992; Emri and Tschoegl 1993; Tschoegl and Emri 1993; Elester et al. 1991; Roths et al. 2000; Malkin and Kuznetsov 2001 and many others).

Recently, Parrot and Duperray (2008) obtained the relaxation modulus by the direct inversion of the Fourier integral; the results (or discretization) depend on the interval of frequency (low-frequency and high-frequency domains). Indeed, these authors argued that the ratio between the lowest and highest frequencies is an important parameter and influences the processing; this method is a semi-analytic approach and no modelling is used. Schapery and Park (1999a) developed a method in which no integral transformations are used; however, this approach is not satisfactory for treating the practical problems (Olard et al. 2004). In the paper of Schapery and Park (1999b), the relaxation modulus and creep compliance are modelled by the series of Prony; the drawback of this method is the identification of the Maxwellian (or Kelvin) modes from linear viscoelastic experiment with least-squares procedures; it is known is an ill-posed problem. To reduce the number of the material parameters, the fractional derivative models (see Bagley and Torvik 1983, 1986; Koeller 1984; Rossikhin and Shitikova 2001; Pritz 2003; Beda and Chevalier 2004) can be used. In these models the number of the material parameters is four or five and the problem of their identification is maybe well conditioned. Usually, theses rheological parameters are determined from the experimental data of $G'(\nu)$ and $G''(\nu)$. The relaxation function can be obtained on inversing the complex modulus via the Mellin–Fourier integral (Soula et al. 1997) this approach was applied for the four-parameter derivative fractional model; and the relaxation function is determined in short and long times (asymptotic behaviour). But it is unknown in the transition region of short and long times. The discrete relaxation modulus $G(t)$ can be obtained from the relaxation spectrum which is not available and indirectly determined on the basis of experimental data, usually from $G'(\nu)$ and $G''(\nu)$. In this approach, the relaxation spectrum is introduced as formal conception (see, e.g., Gross 1953; Ferry 1980; Tschoegl 1989) and its solution requires an inverse solution of the Fredholm integral equation of the first or second kind. This inverse procedure is an ill-posed problem (Honerkamp 1989; Honerkamp and Weese 1990, 1993; Malkin 1990). In this context, Laun (1989) used the linear regression approach for the determination of the relaxation spectrum. However,

this method may not lead to physically meaningful spectrum. Because, as the number of Maxwell units increases some negative elastic moduli are produced and this is physically unrealistic. To overcome this difficulty, Honerkamp and Wesse (1990) considered this problem is ill-posed and applied the standard Tikhonov regularization method. The relaxation spectrum is calculated on using a large number of relaxation times. According to these authors, the regularization method is robust and is a general approach for the evaluation of the relaxation spectrum. We note that, practical realization of this method leads to rather complicated and labour-consuming operations (see Malkin and Masalova 2001). Another approach to determine the relaxation spectrum is to apply a non-linear regression method (so called IRIS method) to obtain the relaxation times, the elastic moduli and the number of Maxwell units. Baumgaertel and Winter (1989); Jackson et al. (1994) used this method and found that the elastic moduli were all positive, provided that the number of Maxwell was small. However, no details of this method are given (see Baumgaertel and Winter 1989), but it is available as a commercial software product. Later, this method was analyzed by Winter (1997) assuming that the calculation of the relaxation spectrum is well posed problem. In other words, his discrete relaxation spectrum coincides with the continuous spectrum. However, other methods discussed in the literature give different predictions concerning the time distribution for the relaxation spectrum (see, Malkin and Kuznetsov 2001). Thereby, it is necessary to introduce the concept of the continuous spectrum (Baumgaertel and Winter 1992; Jackson et al. 1994). Recently, Malkin (2006) described the continuous relaxation spectrum by a power-law form. This method leads to predict the viscoelastic properties of polymer melts. The main advantage of this approach is the reduced number of the material parameters, typically three constants. It is well known that solid materials creep according to power law or stretched-exponential law (see, Schiessel and Blumen 1993) and exhibit logarithmic creep at sufficiently long time limit (see, Lubliner and Panoskaltis 1992). The power law is maybe a special case of the fractional derivative approach.

The main problem in fitting experimental data (for example frequency dependence of storage modulus in a wide frequency range) by a relaxation time spectrum is the nonlinearity of the fitting procedure that presumes the ambiguity of the results of calculation. In order to avoid this difficulty, it is possible to apply a semi-inverse procedure of calculations; it can be realized by the initial rigid fixing of relaxation times distribution. Thus, only their weights should be found and the distribution might be equidistant in a logarithmic scale (Emri and Tschoegl 1993) or any other. Also, a linearization procedure for the search of parameters of a relaxation spectrum has been proposed (Malkin and Masalova 2001) that makes the results of calculations unambiguous. For solid polymers, Haupt et al. (2000) developed an original method to obtain the analytical form of the relaxation spectrum on using the four-parameter fractional derivative model. This method is applied to inverse the complex shear modulus of polyethylene in the range of frequency, $\nu \in [10^{-3}, 10^{3}]$ in Hz.

The main objective of this paper is to model the linear viscoelastic behaviour of amorphous polymers. The experimental results data are available in the wide frequency range $\nu \in [\nu_{\min} \approx 10^{-4}$ Hz, $\nu_{\max} \approx 10^{9}$ Hz] in terms of the storage $G'(\nu)$ and loss $G''(\nu)$ moduli. For elastomers, the loss factor $\eta(\nu) = \frac{G''(\nu)}{G'(\nu)}$ is asymmetrical. Therefore a five-parameter fractional derivative model is used to predict the complex shear modulus $G^*(i\nu)$. The corresponding relaxation spectrum is evaluated numerically because the analytical solution does not exist. Thereby, the fractional model is approximated by a generalized Maxwell model and its rheological parameters $(G_k, \tau_k, N)$ are determined leading to the discrete relaxation spectrum $G(t)$ which is valid in time interval corresponding to the frequency window of the input experimental data. Based on the deterministic approach, the creep compliance $J(t)$ is computed on inversing the relaxation function, $G(t)$.

## 2 The five-parameter fractional derivative model

A fractional order derivative model of viscoelasticity is known to require fewer parameters, typically four or five, to model the actual weak frequency dependence of complex modulus for engineering materials. Bagley and Torvik (1983) reached a good agreement when they fitted their fractional calculus model to measured data for an elastomer. In the range of frequency (in Hz), $\nu \in [\nu_{\min} \approx 10^{-4}, \nu_{\max} \approx 10^{9}]$ we are shown that the four-parameter fractional derivative model is not able to predict the storage and loss moduli of elastomers. Indeed, this model is not able to predict the harmonic asymmetrical loss factor of amorphous polymers. Therefore, it is necessary to extend the fractional four parameters model of Haupt et al. (2000). Based on the work of Pritz (2003) and Rossikhin and Shitikova (2001), the following five-parameter fractional derivative model is obtained

$$(1+\tau_R^{\alpha} D^{\alpha})\sigma(t) = (G_R + G\tau_C^{\beta} D^{\beta})\varepsilon(t), \tag{1}$$

where $\sigma(t)$ is the stress, $\varepsilon(t)$ is the strain, $\tau_R$ is the relaxation time, $\tau_C$ is the retardation (creep) time, $\tau_C^{\beta}\tau_R^{-\alpha} = 1+\xi, \xi = G_R G^{-1}$, $G_R$ is the nonrelaxed magnitude of the elastic modulus (instantaneous modulus of elasticity), $G$ is the relaxed magnitude of the elastic modulus, $\alpha$ and $\beta$ $(0<\alpha,\beta<1)$ are the fractional parameters, $D^{\alpha}\sigma(t)$ and $D^{\beta}\varepsilon(t)$ are the fractional derivative defined (see Caputo and Mainardi 1971) as

$$D^{\gamma} f(t) = \frac{1}{\Gamma(1-\gamma)} \int_0^t \frac{f'(s)}{(t-s)^{\gamma}} ds, \quad f(0) = 0, \tag{2}$$

and $\Gamma(\gamma) = \int_0^{\infty} s^{\gamma-1} e^{-s} ds$, is the Gamma function.

For $\alpha = \beta$, the model of (1) reduces to the four-parameter model of Haupt et al. (2000) which is considered as a generalization of the standard linear solid. The complex modulus of the model can be derived by transforming (1) into the frequency domain; we recall that the Fourier transform of a function, $f(t) = \Delta f e^{-i\omega t}$ is given (Bagley and Torvik 1983) by:

$$F.T[D^{\gamma} f(t)] = (i\omega)^{\gamma} F.T[f(t)]. \tag{3}$$

The results of applying the Fourier transform of (1) lead to the shear complex modulus:

$$G^*(i\omega) = \frac{G_R + G(i\omega\tau_C)^{\beta}}{1+(i\omega\tau_R)^{\alpha}}, \tag{4a}$$

whose real and imaginary parts give us, respectively; the storage modulus

$$\begin{aligned} &\Re E^*(i\omega) \\ &\quad = G'(\omega) \\ &\quad = \frac{G_R + G_R(\omega\tau_R)^{\alpha}\cos(\alpha\pi/2) + G(\omega\tau_C)^{\beta}\cos(\beta\pi/2) + G(\omega\tau_R)^{\alpha}(\omega\tau_C)^{\beta}\cos[(\beta-\alpha)\pi/2]}{1+2(\omega\tau_R)^{\alpha}\cos(\alpha\pi/2)+(\omega\tau_R)^{2\alpha}}, \end{aligned} \tag{4b}$$

and the loss modulus

$$\begin{aligned} &\Im E^*(i\omega) \\ &\quad = G''(\omega) \end{aligned}$$

$$= \frac{G(\omega\tau_C)^{\beta}\sin(\beta\pi/2) - G_R(\omega\tau_R)^{\alpha}\sin(\alpha\pi/2) + G(\omega\tau_R)^{\alpha}(\omega\tau_C)^{\beta}\sin[(\beta-\alpha)\pi/2]}{1 + 2(\omega\tau_R)^{\alpha}\cos(\alpha\pi/2) + (\omega\tau_R)^{2\alpha}}. \tag{4c}$$

From relationships (4b) and (4c), we find the tangent of the mechanical loss angle

$$\eta(\omega) = \tan\delta(\omega) = \frac{G''(\omega)}{G'(\omega)}$$
$$= \frac{(1+\xi)\zeta^{\beta}\tau_R^{\alpha-\beta}\sin(\beta\pi/2) - \xi\zeta^{\alpha}\sin(\alpha\pi/2) + (1+\xi)\zeta^{\alpha+\beta}\tau_R^{\alpha-\beta}\sin[(\beta-\alpha)\pi/2]}{\xi + \xi\zeta^{\alpha}\cos(\alpha\pi/2) + (1+\xi)\tau_R^{\alpha-\beta}\zeta^{\beta}\cos(\beta\pi/2) + (1+\xi)\zeta^{\alpha+\beta}\tau_R^{\alpha-\beta}\cos[(\beta-\alpha)\pi/2]}, \tag{4d}$$

where $\zeta = \omega\tau_R = 2\pi\nu\tau_R$.

The admissibility of the proposed model with the second law of thermodynamics is required, i.e., the mechanical loss angle $\tan\delta(\omega) \geq 0$ and both $G'(\omega)$ and $G''(\omega)$ are positive values of $\omega$ (Bagley and Torvik 1986). For the simulation, the parameters $\xi,\ \alpha,\ \beta$ and $\tau_R$ are varied in the frequency range, $[10^{-4},\ 10^{9}]$; the minimum and maximum values of relaxation times for elastomers are estimated respectively by: $\tau_R^{\min} = 10^{-9}$ s and $\tau_R^{\max} = 0.1$ s; $G_R$ is the static modulus which is taken as an arbitrary constant; $G$, is the modulus at high-frequency; for elastomers, the number $\xi$ satisfies the relation, $\xi < 1$. When $\beta < \alpha$ the loss factor $\eta(\omega)$ is negative, hence the model violate the second law of thermodynamics. Therefore, the values of the rheological parameters which give the loss factor is not positive are discarded in the identification procedure. Because these parameters lead to obtain the relaxation (retardation) function nonmonotonic, this is physically unrealistic. In consequence, we impose $\beta \geq \alpha$, typically results with parameters of $\beta = 0.9$; $\alpha = 0.6,\ 0.7,\ 0.8$ and $\xi = 10^{-5},\ 10^{-1}$ are shown in Figs. 1a, 1b, the abscissa axis is represented by $x = \log\nu$; the behaviour of the four-parameter standard linear solid ($\alpha = \beta = 0.9$) is plotted for the sake of comparison. It is shown that the simulation results of the loss factor are always positive in the frequency range; and the proposed model is thermodynamically admissible especially in the frequency window $[10^{-4}, 10^{9}]$ corresponding to the experimental domain results data used in this paper. When $\alpha = \beta$ the shape of loss factor is symmetric; in contrast, it is asymmetric for $\alpha \neq \beta$. One can see that the loss factor can be approximated by $\eta_{\infty} \cong \tan[(\beta-\alpha)\pi/2]$ at high frequencies ($\omega \to \infty$), it is a function of the difference $(\beta - \alpha)$ and independent on frequency; at low frequencies ($\omega \to 0$) the loss factor is approximated by $\eta(\omega) \cong 0$; the maximum of the loss factor is affected by the value of $\xi$, it becomes asymmetric and is inversely proportional to $\xi$.

## 3 Identification of the material parameters

The experimental results data of literature are used for the validation of the model: the polyisobutylene rubber (Tobolsky and Catsiff 1956) and a polyurethane rubber which is commercially referenced as GE. SMRD (Fowler 1989). In this paper, theses materials are designed respectively by the abbreviation M1 and M2. The plots of the experimental loss factors and simulations of (4d) are shown in Figs. 2a, 2b; it is shown that the curve of the experimental loss factor is asymmetric. In consequence, the four-parameter standard linear solid is not appropriate for the prediction of the behaviour of these polymers. Using the limit

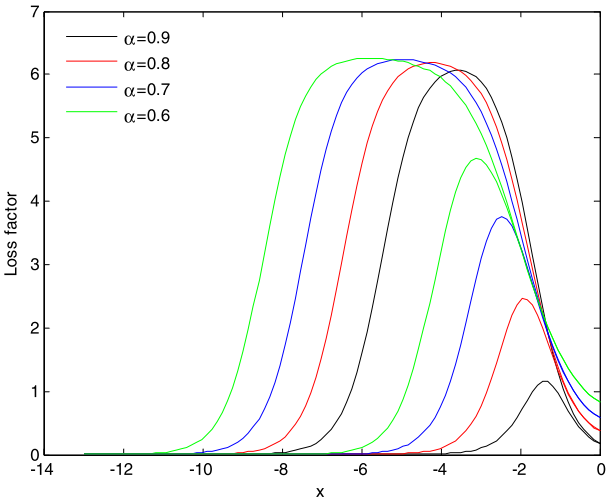


**Fig. 1a** Simulation of the loss factor obtained from (4d) using the five-parameter fractional model ($\beta = 0.9$), and four-parameter standard linear solid ($\alpha = \beta = 0.9$), the relaxation time, $\tau_R^{\min} = 10^{-9}$ s; *solid lines* correspond to $\xi = 10^{-5}$ and *dashed lines* for $\xi = 10^{-1}$

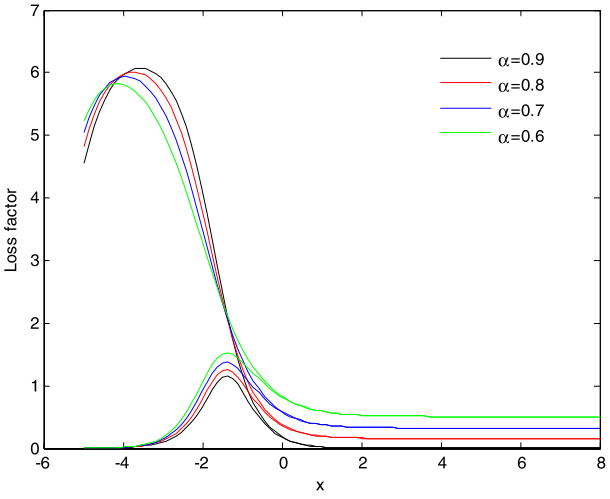


**Fig. 1b** Simulation of the loss factor obtained from (4d) using the five-parameter fractional model ($\beta = 0.9$), and four-parameter standard linear solid ($\alpha = \beta = 0.9$), the relaxation time, $\tau_R = 10^{-1}$ s; *solid lines* correspond to $\xi = 10^{-5}$ and *dashed lines* for $\xi = 10^{-1}$

value of the loss factor, $\eta_\infty$ at high frequency, the number of the material parameters can be reduced. Indeed, the parameter $\beta$ is eliminated in (4b) and (4c) as follows:

$$\beta \cong \alpha + 2\arctan \eta_\infty/\pi, \tag{5a}$$

$$\cong \alpha + k. \tag{5b}$$

Introducing (5b) into (4b) and (4c), one obtains

$$\frac{G'(\omega)}{G_R} \cong \frac{1 + (\omega\tau_R)^\alpha \cos(\alpha\pi/2) + (1+\xi)\xi^{-1}(\omega\tau_R)^\alpha \omega^k [\cos((\alpha+k)\pi/2) + (\omega\tau_R)^\alpha \cos(k\pi/2)]}{1 + 2(\omega\tau_R)^\alpha \cos(\alpha\pi/2) + (\omega\tau_R)^{2\alpha}}, \tag{6a}$$

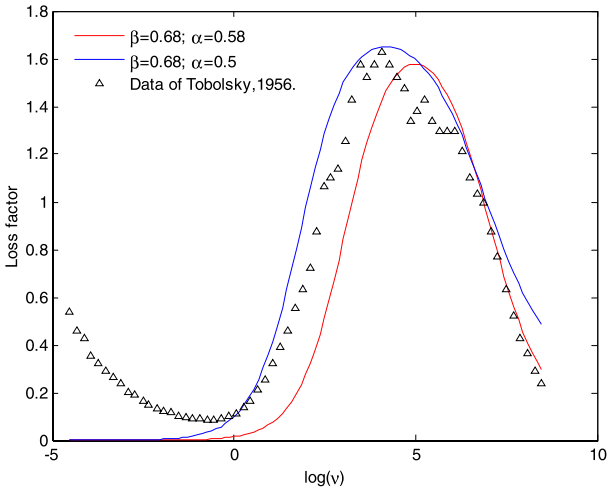


**Fig. 2a** Comparison the experimental results data of the loss factor and theoretical curves computed from (4d), for M1 the numerical values of the parameters $\xi = 0.0065$, $\tau_R = 410^{-9}$ s are used

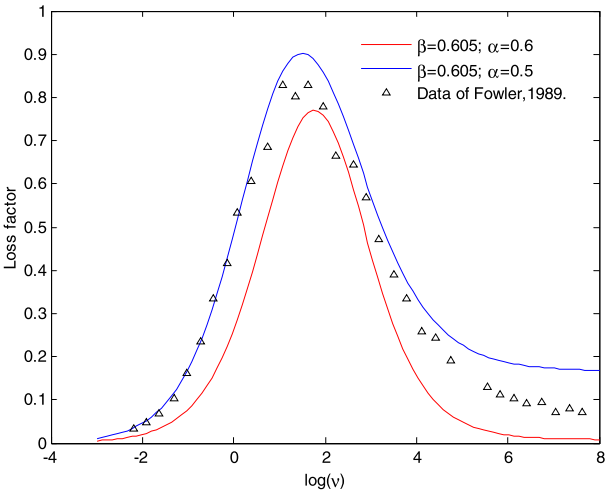


**Fig. 2b** Comparison the experimental results data of the loss factor and theoretical curves computed from (4d), for M2 the numerical values of the parameters $\xi = 0.045$, $\tau_R = 210^{-4}$ s are used

and

$$\frac{G''(\omega)}{G_R} \cong \frac{-(\omega\tau_R)^{\alpha}\sin(\alpha\pi/2) + (1+\xi)\xi^{-1}(\omega\tau_R)^{\alpha}\omega^{k}[\sin((\alpha+k)\pi/2) + (\omega\tau_R)^{\alpha}\sin(k\pi/2)]}{1 + 2(\omega\tau_R)^{\alpha}\cos(\alpha\pi/2) + (\omega\tau_R)^{2\alpha}}, \tag{6b}$$

where $(G_R, \xi, \tau_R, \alpha)$ are the four materials parameters which need to be obtained. To this end, the numerical values of $(\alpha, \beta, \xi, \tau_R)$ used in the simulation (see Figs. 2a, 2b) guided us in the identification procedure of the material parameters. Indeed, these parameters are determined by a stochastic Monte Carlo technique, each parameter is statistically varied in given limits corresponding to the maximum value $V_{\max}$ and a minimum value $V_{\min}$, these values are physically estimated as follows:

$$G_R^{\min} \le G_R \le G_R^{\max}; \quad \xi^{\min} \le \xi \le \xi^{\max}; \quad \alpha^{\max} \le \alpha \le \alpha^{\min}; \quad \tau_R^{\max} \le \tau_R \le \tau_R^{\min}, \tag{7}$$

The values of $(G_R, G, \tau_R, \alpha)$ are generated stochastically via an arbitrary random numbers $0 \leq r_i \leq 1$ with $i = 1, \ldots, 5$ using

$$p(i) = r_i V_{\max}(i) + (1 - r_i) V_{\min}(i). \tag{8}$$

The column $V_{\max}(i)$, $V_{\max}(i)$ and $p(i)$ contain respectively, the maximum, minimum and stochastic values of the parameters. The quadratic error norm is defined as

$$error = \sqrt{\sum_{i=1}^{i=M} [(G'_{meas.}(\omega_i) - G'_{theo.}(\omega_i))^2 + (G''_{meas.}(\omega_i) - G''_{theo.}(\omega_i))^2]}, \tag{9}$$

where $M$ is the number of experimental data; $G'_{meas.}(\omega_i)$ and $G''_{meas.}(\omega_i)$ are the experimental data of the loss and storage moduli which are shown in Figs. 3a, 3b; and $G'_{theo.}(\omega_i)$, $G''_{theo.}(\omega_i)$ are the corresponding theoretical values (see (4b) and (4c)). This method is implemented in the commercial package MATLAB, several ten thousand parameter sets were tested and evaluated; we stop the procedure of iteration when the error is minimized. Therefore, the optimized values of $(G_R, G, \tau_R, \alpha)$ are determined, and the results of simulations are shown in Figs. 3a, 3b; one can see that the predictions of the model are quite satisfactory.

## 4 Determination of the transient functions

### 4.1 Relaxation modulus

The relaxation function can be obtained on inversing the shear complex modulus by the Mellin–Fourier integral. On considering the four-parameter derivative fractional model, this approach was developed in Soula et al. (1997); the asymptotic response behaviour (in the sort and long times) of the relaxation modulus is determined. In other words, this function is unknown in the transition region in which a jump of three or then decades is observed for the relaxation modulus of polymers. Haupt et al. (2000) developed an original method to evaluate the discrete relaxation spectrum which is based on the so-called cumulative relaxation spectrum. This method is successfully applied to inverse the shear complex modulus of polyethylene in the frequency range $\nu \in [10^{-3}, 10^3]$. Our objective is to calculate the discrete relaxation spectrum corresponding to the five-parameter fractional derivative model. In our knowledge this problem is not treated in the literature. Assuming that the model of (1) can be approximated by a simple discrete generalized Maxwell model (see, e.g., Schiessel and Blumen 1993), the relaxation modulus is given by:

$$G(t) = G_R + \sum_{k=1}^{k=N} G_k \exp(-t/\tau_k), \tag{10a}$$

where $G_R$ represents the equilibrium or the residual modulus at the fully decaying state, that is, when all relaxable stress is fully relaxed, $\tau_k$ are the relaxation time and the $G_k$ values are corresponding weights, and $N$ is the number of Maxwell units. The knowledge of the set $(G_k, \tau_k)$ is very useful because it allows one to predict the behaviour of the material in any standard experiment. Applying the Fourier transform for (10a), the real and imaginary parts

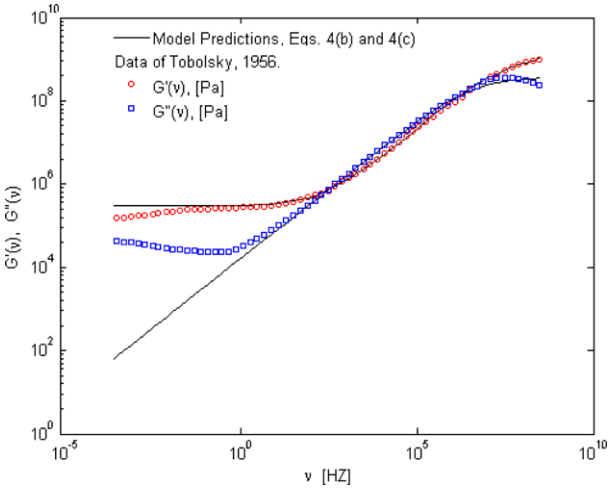


**Fig. 3a** Comparison of the experimental results data of the storage and loss moduli corresponding to M1, with the predictions of the proposed model given by (4b) and (4c)

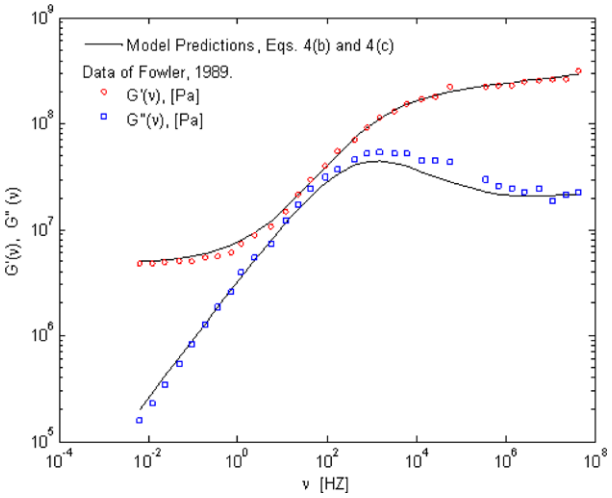


**Fig. 3b** Comparison of the experimental results data of the storage and loss moduli corresponding to M2, with the predictions of the proposed model given by (4b) and (4c)

of the complex modulus may be calculated versus the circular frequency $\omega = 2\pi\nu$ as

$$G'(\omega) = G_R + \sum_{k=1}^{k=N} G_k \frac{\omega^2\tau_k^2}{1+\omega^2\tau_k^2}$$
$$= G_R + \sum_{k=1}^{k=N} G_k \frac{\omega^2}{\nu_k^2+\omega^2}, \tag{10b}$$

and

$$G''(\omega) = \sum_{k=1}^{k=N} G_k \frac{\omega\tau_k}{1+\omega^2\tau_k^2}$$
$$= \sum_{k=1}^{k=N} G_k \frac{\omega}{\nu_k^2+\omega^2} \quad \text{with } \nu_k = \frac{1}{\tau_k}. \tag{10c}$$

When the number of Maxwell unit approaches infinity, the summation given in (10a) is replaced by an integral

$$G(t) = G_R + \int_0^\infty G(\tau)\exp(-t/\tau)d\tau, \tag{11}$$

where $G(\tau)$ is a continuous function over relaxation time $\tau$; setting $\nu = \frac{1}{\tau}$ in (11), we obtain

$$\begin{aligned} G(t) &= G_R + \int_0^\infty \frac{G(1/\nu)}{\nu^2}\exp(-\nu t)d\nu \\ &= G_R + \int_0^\infty h(\nu)\exp(-\nu t)d\nu, \end{aligned} \tag{12}$$

$h(\nu)$ is called the relaxation spectrum which is correlated to the shear complex modulus (see Haupt et al. 2000) as follows:

$$h(\nu) = \frac{1}{2\pi i}\left(\frac{G^*(\nu e^{-i\pi})}{\nu e^{-i\pi}} - \frac{G^*(\nu e^{+i\pi})}{\nu e^{+i\pi}}\right) \quad \text{with } (i^2 = -1), \tag{13}$$

and the so-called cumulative relaxation spectrum is defined as

$$H(\nu) = \int_0^\nu h(z)dz. \tag{14}$$

Substituting (4a) into account (13), one obtains the relaxation spectrum

$$h(\nu) = \frac{G(\nu\tau_C)^\beta(\nu\tau_R)^\alpha\sin((\beta-\alpha)\pi) + G(\nu\tau_C)^\beta\sin(\beta\pi) - G_R(\nu\tau_R)^\alpha\sin(\alpha\pi)}{\pi\nu[1 + 2(\nu\tau_R)^\alpha\cos(\alpha\pi) + (\nu\tau_R)^{2\alpha}]}. \tag{15}$$

Replacing $\alpha = \beta$ and $G_R = 0$ in (15), we retrieve the relaxation spectrum of the four- parameter standard linear solid (see Haupt et al. 2000). Introducing (15) into (14), we obtain:

$$H(\nu) = \int_0^\nu \frac{G(z\tau_C)^\beta(z\tau_R)^\alpha\sin((\beta-\alpha)\pi) + G(z\tau_C)^\beta\sin(\beta\pi) - G_R(z\tau_R)^\alpha\sin(\alpha\pi)}{\pi z[1 + 2(z\tau_R)^\alpha\cos(\alpha\pi) + (z\tau_R)^{2\alpha}]}dz, \tag{16}$$

the analytic solution of (16) does not exist. To determine the Maxwellian modes $(G_k, \tau_k)$, we introduce the linear cumulative relaxation spectrum, $H(\nu) \approx H_{lin}(\nu)$ which is defined as a series of step functions:

$$H_{lin}(\nu) = \sum_{k=1}^{k=N} G_k\Theta(\nu - \nu_k), \tag{17}$$

where $\Theta(\nu - \nu_k)$ is the Heaviside step function. According to Ferry (1980), the storage modulus $G'(\omega)$ of the Maxwell model is linked to the relaxations spectrum by

$$G'(\omega) = G_R + \int_0^\infty h(\nu)\frac{\omega^2}{\nu^2 + \omega^2}d\nu. \tag{18}$$

Introducing (17) into (18), one obtains

$$G'(\omega) = G_R + \sum_{k=1}^{k=N} G_k \int_0^{\infty} \delta(\nu - \nu_k) \frac{\omega^2}{\nu^2 + \omega^2} d\nu$$
$$= G_R + \sum_{k=1}^{k=N} G_k \frac{\omega^2}{\nu_k^2 + \omega^2}, \tag{19}$$

where $\delta(\nu - \nu_k)$ is the Dirac delta function; one can see that (10b) is retrieved. The parameter $G_R \neq 0$ is assumed (see, (4a)) in the identification procedure of the parameters $G_k, \tau_k$ and $N$. Now, one needs to determine the cumulative relaxation spectrum, $H(\nu) \approx H_{lin}(\nu)$ it is evaluated numerically as follows: the heights of the steps correspond to the weights, $G_k$ and their locations to the inverse relaxations, $\tau_k$ of the generalized Maxwell model. The integration of (16) is evaluated on using the Gauss integration method

$$H(\nu_i) = \int_{\nu_{\min}}^{\nu_i} h(z) dz, \quad \text{with } \nu_0 = 0 \qquad \text{and} \qquad H(\nu_0) = 0, \quad i \in [1, N]. \tag{20a}$$

To determine the discrete relaxation spectrum, $G(t)$ i.e., the fractional model is approximated by a finite sequence of Maxwell units; we choose the parameters $\nu_{\min}, \nu_{\max}$ such that, $0 < \nu_{\min} < \nu_{\max}$ and $N$ is the number of Maxwell units in the approximate model. The parameters $\nu_{\min}$ and $\nu_{\max}$ correspond to the range of frequency values of $\nu$ for which the approximation will be accurate. We define the sequence of points $\nu_1, \ldots, \nu_N$ by requiring that they satisfy $\nu_1 = \nu_{\min}, \nu_N = \nu_{\max}$ and that there exists a ratio $r$ such that for each, $i = 1, \ldots N$ $\nu_i / \nu_{i-1} = r$, i.e. the points are geometrically spaced. The unique solution to these requirements is given by the formula $\nu_i = \nu_{\min}(\nu_{\max}/\nu_{\min})^{i-1/N-1}$. In consequence, it is easy to calculate numerically the values of elastic moduli, $G_k$ and their corresponding relaxation times, $\tau_k$ as follows:

$$0 \leq \nu \leq \nu_1: \qquad H_{lin}(\nu) = G_1, \quad \tau_1 = 1/\nu_1; \tag{21a}$$
$$\nu_1 \leq \nu \leq \nu_2: \qquad H_{lin}(\nu) = G_2, \quad \tau_2 = 1/\nu_2; \tag{21b}$$
$$\nu_{i-1} \leq \nu \leq \nu_i: \qquad H_{lin}(\nu) = G_i, \quad \tau_i = 1/\nu_i; \tag{21c}$$
$$\vdots$$
$$\nu_{N-1} \leq \nu \leq \nu_N: \qquad H_{lin}(\nu) = G_N, \quad \tau_N = 1/\nu_N. \tag{21d}$$

The cumulative relaxation spectrum and its approximation are shown in Figs. 4a, 4b. The number $N$ is varied, and the predictions of (10b) and (10c) are compared with the experimental data for each value of $N$; to avoid the oscillations in the numerical simulations of the elastic moduli $G'(\omega)$ and $G''(\omega)$, the number of Maxwell units $N$ is optimized. In Figs. 5a, 5b, we have shown that for, $N = 25$, the waves disappeared and the simulations are quite satisfactory. One can see that the Maxwell model ($N = 25$) is failed to predict the elastic moduli at high frequencies for the material M2; in consequence, the Maxwell model is not appropriate to replace the five-parameter fractional model; therefore, it needs to be approximated by a more complex discrete model. In Figs. 6a, 6b, the relaxation function computed from (10a) and compared with the formula of Schwarzl; this approximation is used because

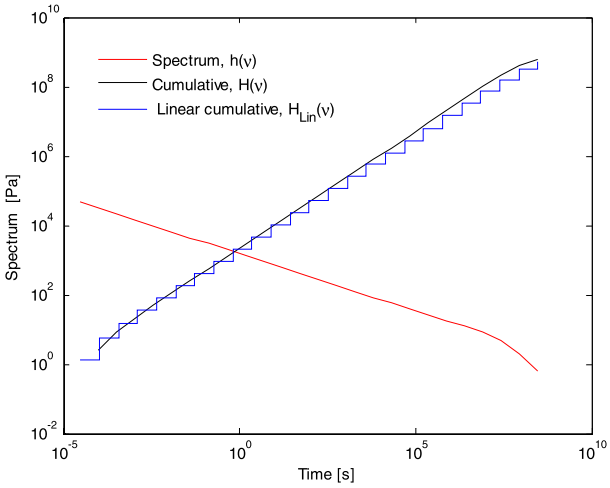


**Fig. 4a** The relaxation spectrum, $h(\nu)$, and the corresponding cumulative spectrum, $H(\nu)$ and $H_{lin}(\nu)$ of M1 for $N = 25$ units

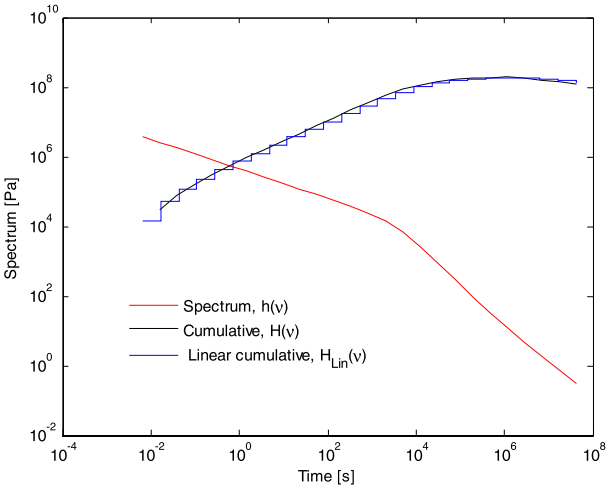


**Fig. 4b** The relaxation spectrum, $h(\nu)$, and the corresponding cumulative spectrum, $H(\nu)$ and $H_{lin}(\nu)$ of M2 for $N = 25$ units

it is the best method to compute the relaxation modulus from the corresponding frequency-dependent function (see Emri et al. 2005); the Schwarzl relaxation modulus is given by

$$\begin{aligned} G(t_i) \approx\ & G'(\nu_i) - 0.0080G''(\nu_i/16) - 0.00719G''(\nu_i/8) + 0.00616G''(\nu_i/4) \\ & - 0.467G''(\nu_i/2) + 0.0918G''(\nu_i) + 0.0534G''(2\nu_i) \\ & - 0.08G''(4\nu_i) + 0.0428G''(8\nu_i)|_{\nu_i = 1/t_i} \end{aligned} \tag{22}$$

with

$$t_i = (1/\nu_{\max})(\nu_{\min}/\nu_{\max})^{i-1/N-1}, \qquad \nu_i = \nu_{\min}(\nu_{\max}/\nu_{\min})^{i-1/N-1} \quad \text{and} \quad i = 1 \ldots N;$$

the storage $G'(\omega)$, and loss $G''(\omega)$ moduli are calculated respectively from (4b) and (4c) tacking into account $\omega_i = 2\pi\nu_i$. For the material M1, the relaxation modulus presents two

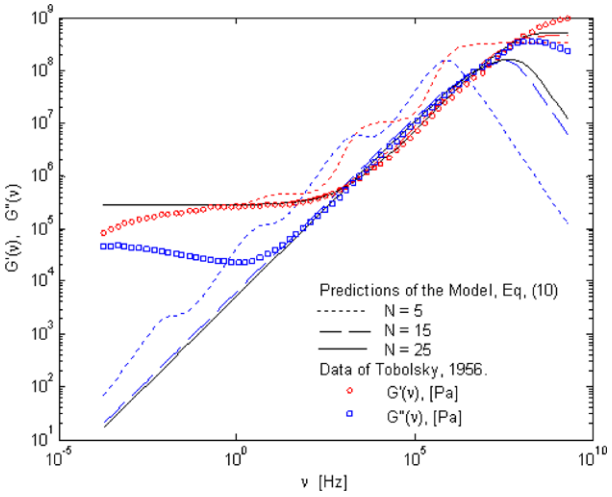


**Fig. 5a** Comparison of the experimental results data of the storage and loss moduli corresponding to M1, with the predictions of (10b) and (10c): (1) $N = 5$ units, (2) $N = 15$ units, (3) $N = 25$ units

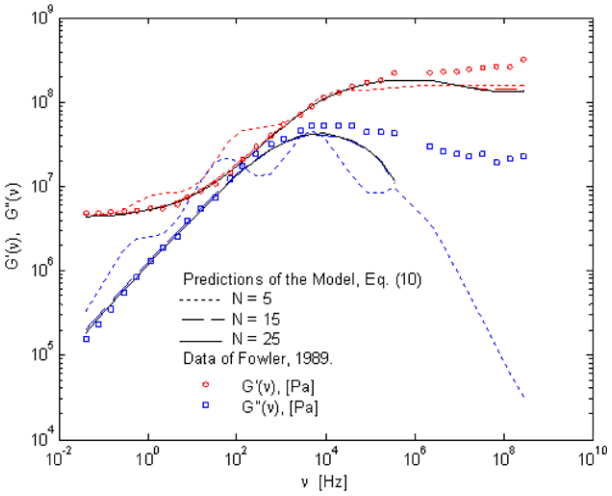


**Fig. 5b** Comparison of the experimental results data of the storage and loss moduli corresponding to M2, with the predictions of (10b) and (10c): (1) $N = 5$ units, (2) $N = 15$ units, (3) $N = 25$ units

transition regions with time; the proposed model reproduces very well the first one and failed in the second. Because the model is originally conceived for polymers which have one transition region; therefore, the model may be needed to extend with increasing the number of material parameters more than five-parameters.

### 4.2 Creep compliance

Using the standard relation of linear viscoelasticity (see, e.g., Gross 1953; Ferry 1980; Tschoegl 1989), the creep compliance, $J(t)$ can be obtained from the relaxation modulus, $G(t)$ as follows:

$$t = \int_0^t G(t-s)J(s)ds = \int_0^t J(t-s)G(s)ds. \tag{23}$$

Equation (23) is called the Volterra equation of first kind, and its inversion is known to be an ill-posed problem. Sorvari and Malinen (2007) applied the Tikhonov regularization method

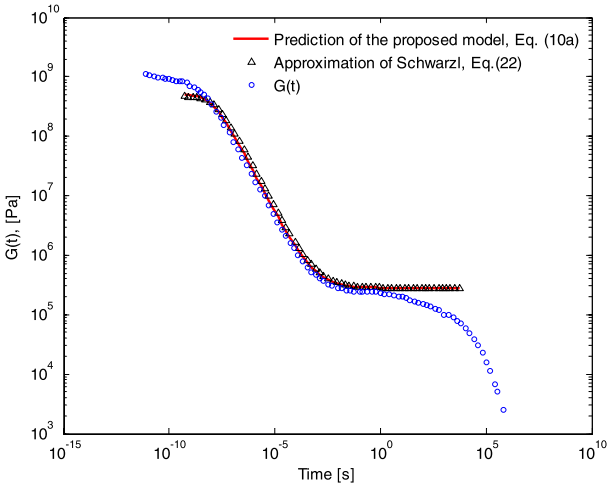


**Fig. 6a** Comparison of the relaxation modulus calculated from (10a) with the approximate function of Schwarzl given by (22) and experimental results data corresponding to M1

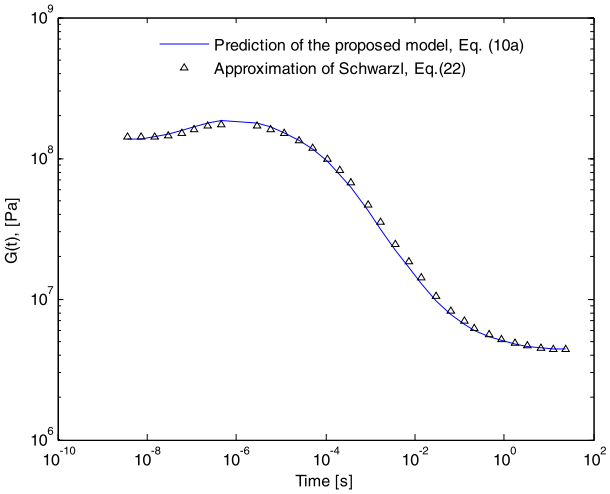


**Fig. 6b** Comparison of the relaxation modulus calculated from (10a) for M2 with the approximate function of Schwarzl given by (22)

to obtain the creep function, $J(t)$; however, it is not easy to determine the regularization parameter. Equation (23) can be split into three parts:

$$t = \int_0^{t_{\min}} G(t-s)J(s)ds + \int_{t_{\min}}^{t_{\max}} G(t-s)J(s)ds + \int_{t_{\max}}^{t} G(t-s)J(s)ds, \qquad (24)$$

the first and third terms require respectively extrapolation in the time range $t = 0$ to $t_{\min}$ and $t = t_{\max}$ to $t$, while experiment data are only available in the intermediate window (second term). It is obvious that this extrapolation process is highly arbitrary; indeed, the relaxation modulus is unknown in time interval $[0, t_{\min}]$ and the value $G(0^+)$ is may be indefinite (see Pritz 2005). Therefore, the exact solution of (23) cannot be found in the time range $[0, t]$; in consequence, many approximate solutions may be possible for the creep compliance $J(t)$.

In this paper, we compute the creep compliance in time range $[t_{\min}, t_{\max}]$; to this end, we assume

$$t \approx t_{\min} + \int_{t_{\min}}^{t_{\max}} G(t-s)J(s)ds, \tag{25}$$

$$\Delta t_k = t_k - t_{\min} \approx \int_{t_{\min}}^{t_{\max}} G(t_k - s)J(s)ds, \tag{26}$$

$$\Delta t_k \approx \sum_{l=1}^{l=k-1} \int_{t_{l-1}}^{t_l} G(t_k - s)J(s)ds \quad \text{with } t_0 = t_{\min}, \text{ and } k \geq 2, \tag{27}$$

The value of the creep function in the time interval $[t_{l-1}, t_l]$ is assumed to be $J(s) = J_l$, substituting (10a) into the integral of (26), we obtain:

$$\Delta t_k \approx \sum_{l=1}^{l=k-1} J_l \int_{t_{l-1}}^{t_l} G(t_k - s)ds = \sum_{l=1}^{l=k-1} \left[ G_\infty (t_l - t_{l-1}) + \sum_{n=1}^{n=25} \tau_n G_n e^{-\frac{t_k}{\tau_n}} \left( e^{\frac{t_l}{\tau_n}} - e^{\frac{t_{l-1}}{\tau_n}} \right) \right] J_l. \tag{28}$$

Equation (28) can be expressed as follows:

$$\Delta t_k \approx \sum_{l=1}^{l=k-1} A_{kl} J_l \quad \text{with } l \leq k-1; \tag{29}$$

Therefore, (29) can be written in a matrix form, $Ax = b$, where the elements of the matrix $A$ are the values of the relaxation modulus evaluated for different times; the vectors $x$ and $b$ contains respectively the unknown values of the creep compliance $J_l$, and times values $\Delta t_k$:

$$A_{kl} = \left[ G_R(t_l - t_{l-1}) + \sum_{n=1}^{n=25} \tau_n G_n e^{-\frac{t_k}{\tau_n}} \left( e^{\frac{t_l}{\tau_n}} - e^{\frac{t_{l-1}}{\tau_n}} \right) \right], \quad \text{if } l \leq k-1, \text{ else } A_{kl} = 0, \tag{30a}$$

$$x_l = J_l, \tag{30b}$$

$$b_k = \Delta t_k, \tag{30c}$$

for

$$k = 2, \ldots M, \quad l = 1, \quad k-1 \quad \text{and} \quad t_l = t_{\min}(t_{\max}/t_{\min})^{l/M}. \tag{30d}$$

The results obtained from (29) depend on the time step; increasing the number $M$, i.e., refining this step leads to augment the time of computation. We note that the relaxation or creep functions are unknown at small times corresponding to high frequencies in harmonic regime; therefore, it is judicious to determine the creep compliance in the time window which is impossible to measure by standard tests. However in this paper, the full input interval data is considered; for the computation, the minimum and maximum times are respectively chosen, $t_{\min} = 1/\nu_{\max}, t_{\max} = 1/\nu_{\min}$ and $M = 400$; (29) leads to solve linear algebraic system

equations:

$$\begin{pmatrix} A(2,1) & 0 & 0 & \dots & . & 0 \\ A(3,1) & A(3,2) & 0 & \dots & . & 0 \\ \vdots & \vdots & \vdots & \dots & \vdots & \vdots \\ A(M,1) & A(M,2) & A(M,3) & \dots & A(M,M-2) & A(M,M-1) \end{pmatrix} \begin{pmatrix} J_2 \\ J_3 \\ J_4 \\ \vdots \\ J_M \end{pmatrix} = \begin{pmatrix} \Delta t_2 \\ \Delta t_3 \\ \Delta t_4 \\ \vdots \\ \Delta t_M \end{pmatrix}; \tag{30e}$$

for $t \in [t_0, t_1]$, the value of the creep compliance is given by: $J_1 \approx [G_R + \sum_{n=1}^{n=25}(1 - t_1/\tau_n)G_n]^{-1}$; the results obtained from (30e) are compared with the predictions of the approximated creep compliance of Schwarzl (see Emri et al. 2005) which is given by

$$\begin{aligned} J(t_i) \approx\ & J'(\nu_i) + 0.496J''(\nu_i/2) + 0.0651[J'(\nu_i/2) - J'(\nu_i/4)] \\ & + 0.0731[J'(\nu_i) - J'(\nu_i/2)] \\ & + 0.111[J'(2\nu_i) - J'(\nu_i)] + 0.03[J'(16\nu_i) - J'(8\nu_i)] \\ & + .00683[J'(64\nu_i) - J'(32\nu_i)]|_{\nu_i = 1/t_i}. \end{aligned} \tag{31}$$

To enable us to evaluate the creep compliance of (31); we use the standard interconversion relations (Ferry 1980; Tschoegl 1989) in harmonic regime

$$J'(\nu_i) = \frac{G'(\nu_i)}{[G'(\nu_i)]^2 + [G''(\nu_i)]^2}, \tag{32}$$

and

$$J''(\nu_i) = \frac{G''(\nu_i)}{[G'(\nu_i)]^2 + [G''(\nu_i)]^2}. \tag{33}$$

The harmonic compliances obtained from (32) and (33) are shown in Figs. 7a, 7b; for M2 the experimental results data are available. The transient creep compliances calculated from (30) and (31) are shown in Figs. 8a, 8b; tacking the value of $M = 10^3$, one can see that the creep compliance $J(t)$ can be computed; the obtained results approximated very well the predictions creep compliance $J(t)$ of Schwarzl.

## 5 Conclusion and perspectives

A five-parameter derivative model is considered as a generalization of the four-parameter standard linear solid. The experimental data of literature are used for the validation of the model. To this end, the consistent intervals for each parameter $(\alpha, \beta, \xi, \tau_R)$ are determined. The model is reliable to predict the viscoelastic behaviour of elastomers in the wide range frequency. The corresponding cumulative relaxation spectrum of the five-parameter model is evaluated numerically on assuming that is equivalent to a discrete generalized Maxwell

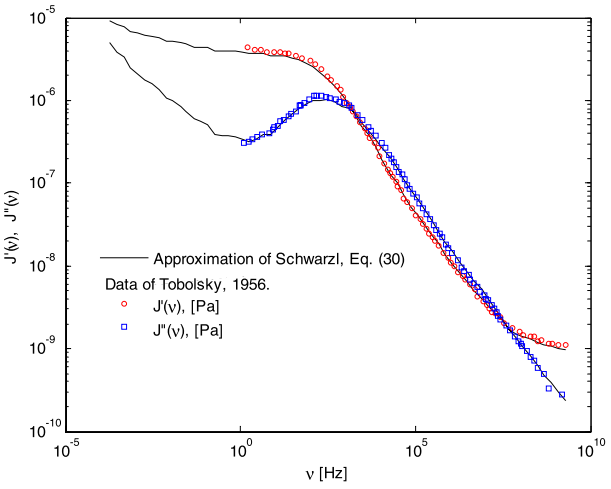


**Fig. 7a** The storage and loss compliances calculated from (32) and (33), and experimental results data corresponding to M1

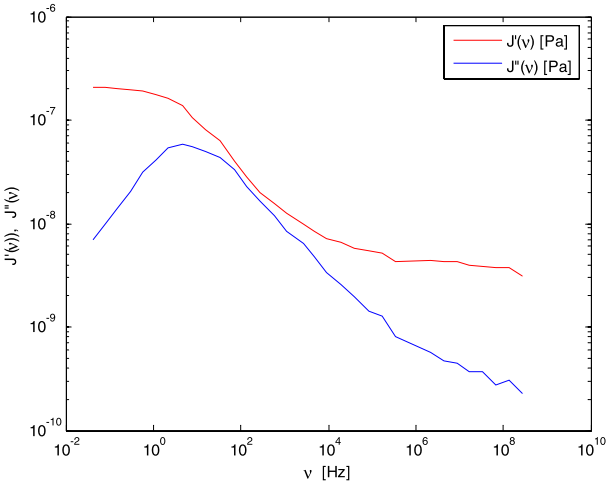


**Fig. 7b** The storage and loss compliances calculated from (32) and (33) corresponding to M2

model. Therefore, the Maxwellian modes are determined via the linear cumulative relaxation spectrum. It is shown that on increasing smoothly the Maxwell units, the results of fitting elastic moduli $G'(\omega)$ and $G''(\omega)$ are very good agreement with experimental results data in the range of intermediate frequency. Indeed, one can assert that at high frequencies the discrete generalized Maxwell model is not the best approximation of the five-parameter fractional model. The creep compliance is calculated on inversing the discrete relaxation modulus. In order to confirm the validity of our results, the simulation data of the discrete relaxation and creep compliance are compared with the predictions approximate functions of Schwarzl. It is shown that the comparison is quite satisfactory.

One advantage of this approach is no prior assumption is made concerning the number of Maxwell units, the values of relaxation times and their weights. However, the relaxation times distribution depends on the number of Maxwell units $N$, $\nu_{\min}$ and $\nu_{\max}$. In numerical point of view, the inversion of the relaxation modulus in the wide range of frequency leads to use a great computing time to determine the creep compliance. In consequence, the results

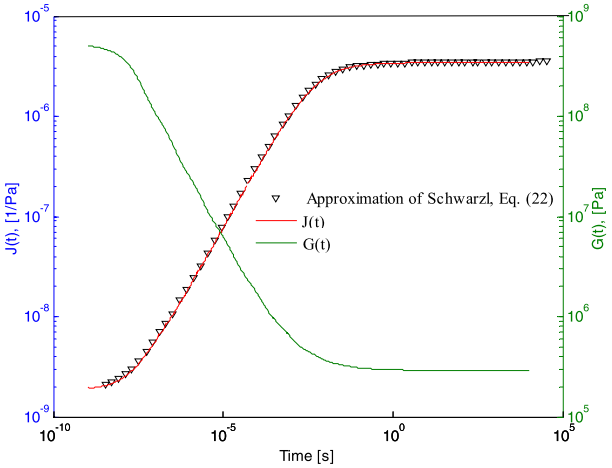


**Fig. 8a** Comparison of the creep compliance calculated from (30) with the approximate function of Schwarzl for M1, (31)

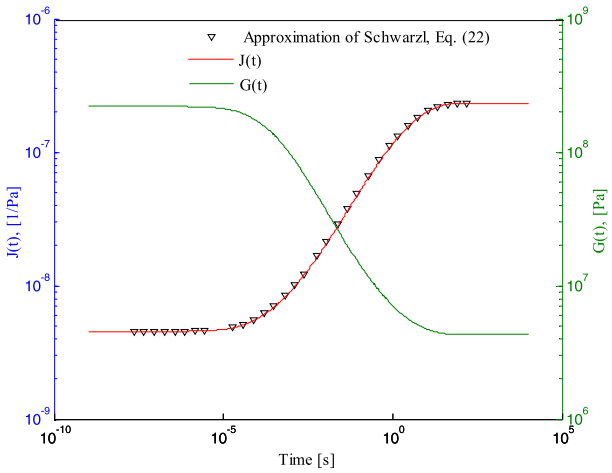


**Fig. 8b** Comparison of the creep compliance calculated from (30) with the approximate function of Schwarzl for M2, (31)

of inversion depend on the integration step. This point needs to be clarified. Indeed, the calculation of the creep compliance developed in this paper needs to be checked. To this end, a comparison with the experimental data obtained from standard creep tests is hoped.

In the future work, it is necessary to take into account the influence of noise on experimental data of dynamic moduli for the determination of the rheological parameters of the generalized Maxwell model. This noise may be affecting the results of creep function. A small perturbation on the values of the relaxation modulus can induces large errors in the computation of the creep compliance.

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