APPLICATION OF FRACTIONAL CALCULUS METHODS TO VISCOELASTIC RESPONSE OF AMORPHOUS SHAPE MEMORY POLYMERS

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ABSTRACT

Constitutive models based on fractional calculus are utilized to investigate the viscoelastic response of thermally activated shape memory polymers (SMPs). Fractional calculus-based viscoelastic equations are fitted to experimental data existing in literature compared with traditional viscoelastic models. In addition, a fractional rheology model is applied to simulate the isothermal recovery of an amorphous SMP. The fit results show a significant improvement in the description of the strain recovery response of SMP by the fractional calculus method.

Keywords: Shape memory polymers, Fractional calculus, Viscoelasticity, Shape recovery.

1. INTRODUCTION

Shape memory polymers (SMPs) are new functional materials with a wide application prospect which have attracted widespread interests for their ability to undergo a large recoverable deformation in response to an external stimulus [1-7]. The thermally activated SMPs are shown to be the most extensively studied due to their variety, simple activated form, large recoverable deformation and rapid response. Constitutive models based on viscoelasticity are one of two kinds of main approaches to model and predict the thermomechanical behaviors of thermally activated SMPs during the past decade [8-21]. These viscoelastic models such as Kelvin and Maxwell-based models are used to describe rate-dependent behaviors of polymers on macroscopic scale and in these models, materials are assumed as a combination of springs, dashpots or frictional elements. In these rheological models, differential equations with integer order derivatives are used to characterize the viscoelastic behaviors of SMPs.

Nutting [22] earlier observed that the stress relaxation phenomenon of a number of materials could be expressed by means of fractional powers of time which was a wholly new approach. Later, Gemant [23] found some limitations of traditional viscoelastic models and suggested that time differentials of fractional order might model the stiffness and damping properties of viscoelastic materials. Furthermore, in the work of Bagley and Torvik [24], experimental results suggested that the rheological properties that give rise to viscoelasticity may have a fundamental behavior based on fractional rather than integer calculus. The introduction of fractional calculus into the theory of viscoelasticity has resulted in a powerful tool to model the static and dynamic behaviors of polymers and other materials [25-31]. The wide use of fractional calculus-based constitutive models is motivated by the advantage of few parameters required to characterize material viscoelastic behaviors and the convenience of the use of Fourier and Laplace transforms. Similarly, it is reasonable to believe that fractional models will give an adequate description of real viscoelastic behaviors of thermally activated SMPs with less number of material parameters than classical ones. However, the fractional calculus approach to viscoelasticity of SMPs is rarely reported up to now.

For this purpose, in this work the efficacy of the fractional calculus-based models will be examined by fitting experimental data compared with classic viscoelastic models. The suitability of fractional models in the description of the strain recovery response of SMPs will be investigated.

2. FRACTIONAL DERIVATIVE MODEL

2.1 'Spring-pot' by Koeller

The essential difference between the fractional rheological models and the traditional ones is that the dash-

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DOI : 10.1017/jmech.2014.98
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https://doi.org/10.1017/jmech.2014.98 Published online by Cambridge University Press

pot in the classical Kelvin and Maxwell viscoelastic models is replaced by an element termed a 'spring-pot' by Koeller [29]. The constitutive law of this fractional calculus element satisfies:

$$\sigma(t) = E^{1-\alpha}\eta^{\alpha}D^{\alpha}\varepsilon(t) = E\tau^{\alpha}D^{\alpha}\varepsilon(t) \tag{1}$$

Here σ, ε, t, E and η are the stress, strain, time, Young's modulus and viscosity, respectively, $\tau = \eta/E$ is the relaxation time, α is considered to be the memory parameter or the fractional derivative parameter, and D^{α} indicates fractional differentiation, that is

$$D^{\alpha}f(t) = DD^{\alpha-1}f(t) \tag{2}$$

where D indicates differentiation and the operation $D^{\alpha-1}$ may be expressed as a Riemann-Louiville fractional integral

$$D^{\alpha-1}f(t) = \int_0^t \frac{(t-\tau)^{-\alpha}}{\Gamma(1-\beta)} f(\tau)\, d\tau \tag{3}$$

We note that α is restricted between 0 and 1, *i.e.* $0 \le \alpha \le 1$. Obviously, the 'spring-pot' is a simple spring when $\alpha = 0$ and a simple dashpot when $\alpha = 1$. Consequently, the 'spring-pot' can better represent the nonlinear gradual change process in the rheological problems.

2.2 Fractional Poynting-Thomson model

A large variety of fractional rheological models for viscoelastic behaviors can be obtained from corresponding classic viscoelastic models by replacing the dashpot with the above 'spring-pot'. Here, the fractional Poynting-Thomson model is taken to exposit the characteristic of fractional calculus-based models. The standard linear solid model consists of either a Kelvin unit and a spring in series or a Maxwell unit and a spring in parallel, the former is also called Poynting-Thomson model. Caputo and Mainardi [32] generalized it from the integer order derivatives to the fractional order ones. The fractional Poynting-Thomson model with four parameters E_0, E_1, α and τ_1, is characterized by the mechanical model shown in Fig. 1 and may be expressed as:

$$E_0(D^{\alpha} + 1/\tau_1^{\alpha})\,\varepsilon(t) = (D^{\alpha} + 1/t_1^{\alpha})\,\sigma(t) \tag{4}$$

where t_1 is defined by the relationship:

$$t_1 = \tau_1 / \sqrt[\alpha]{1 + E_0/E_1} \tag{5}$$

For this model, by use of Laplace transform, inverse Laplace transform and series expansion $1/(1+x) = \sum_{n=0}^{\infty}(-x)^n$, the expressions of creep function and relaxation modulus are obtained [33] as follows:

$$J(t) = \frac{1}{E_0}\left\{1 + \frac{E_0}{E_1}\cdot E_{-\alpha,1}\left[-\left(\frac{t}{\tau_1}\right)^{-\alpha}\right]\right\} \tag{6}$$

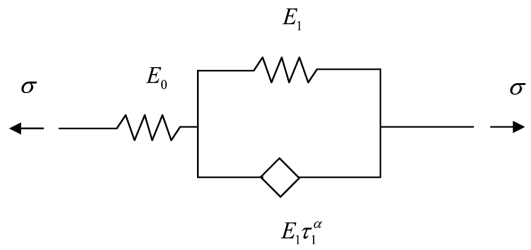


Fig. 1 The Fractional Poynting-Thomson model.

$$G(t) = E_0\left\{1 - \frac{E_0/E_1}{1 + E_0/E_1}\cdot E_{-\alpha,1}\left[-\left(\frac{t}{t_1}\right)^{-\alpha}\right]\right\} \tag{7}$$

where $E_{\alpha,\beta}[x]$ is the Mittag-Leffler function:

$$E_{\alpha,\beta}[x] = \sum_{n=0}^{\infty}\frac{x^n}{\Gamma(\alpha n + \beta)} \tag{8}$$

3. CURVE FITTING OF STATIC AND DYNAMIC TEST DATA USING FRACTIONAL MODELS

3.1 Background

As mentioned earlier, thermoviscoelastic modeling is a main approach to mechanical behaviors of SMPs. The static and dynamic test is needed in the approach. In this section three sets of static and dynamic data of SMPs available in the literature are taken to study whether the fractional models fit experimental data adequately. The first two sets of data are the relaxation moduli for a SMP obtained by Hong *et al.* [15], and for polyurethane based SMPs got by Heuchel *et al.* [16]. The other one is the storage modulus and loss factor data for an epoxy network given by Diani *et al.* [34]. These sets of data are chosen because they exhibit a variety of material behavior characteristics of various SMPs thus investigating the efficiency of fractional calculus-based models. Besides, these data are readily available in corresponding references.

3.2 Fitting Relaxation Modulus Data

The fractional Poynting-Thomson model has been proved usable to represent the relaxation behaviors of some polymeric materials accurately over the entire time range of the response [25-27]. Here, we consider its applicability for describing the relaxation modulus of SMPs based on the optimization software package 1stOpt [35]. However, the Mittag-Leffler function in Eq. (7) is a sum of an infinite series which is unavailable in 1stOpt, thus the method in Ref. [33] is adopted here, and the sum is truncated in order to make computation possible. In like manner an N-term truncated series is utilized to replace the infinite series in Eq. (7), and the truncation condition given by $\left|\sum_{n=0}^{N}\frac{x^n}{\Gamma(\alpha n+\beta)} - \sum_{n=0}^{N+M}\frac{x^n}{\Gamma(\alpha n+\beta)}\right| \le 10^{-6}$ is suitable to

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guarantee their uniform convergence result, where M is even, $M \geq 10$. The fractional relaxation modulus is fitted to the first two sets of data with 1stOpt using the Levenberg-Marquardt and Universal Global Optimization algorithms. As a result, $N = 39$ for the data in Ref. [15] and $N = 29$ for the data in Ref. [16] respectively, in addition the corresponding parameters E_0, E_1, α, τ_1 are presented in Table 1.

The classical technique to fit creep or relaxation data by using Prony series is also adopted in Refs. [15,16]. It is a series of exponential functions in the following form:

$$E = \sum_{i=1} E_i \exp\left(-\frac{t}{\tau_i}\right) \tag{9}$$

The fractional fit and the exponential fit to the first two sets of data are shown in Figs. 2 and 3 respectively. These figures indicate the fractional Poynting-Thomson model accurately represents the relaxation behavior of the polyurethane SMPs over the entire time range of the response. Moreover, the fractional Poynting-Thomson model gives a slightly better fit to the data in both cases than the Prony series model. Compared with 17 and 5 parameters used in the exponential fit to these data respectively, only 4 parameters are sufficient in the fractional fit. Consequently the fractional Poynting-Thomson model is capable of presenting the relaxation data with fewer parameters.

3.3 Fitting Dynamic Modulus Data

Fractional derivative models offer a powerful tool to describe the dynamic behaviors of real viscoelastic materials [27,36]. Pritz [31] derived a fractional derivative Zener model characterized by five parameters in order to depict the asymmetry of the loss factor peak and the high-frequency behavior of dynamic properties experienced with some polymeric damping materials. Now this model is taken to fit the third set of data aiming at its validity. Its storage modulus G' and loss factor tanδ are

$$G' = G_0 + G_0(d-1)\frac{\cos(\alpha\pi/2)\omega_n^{\alpha} + \cos\left[(\alpha-\beta)\pi/2\right]\omega_n^{\alpha+\beta}}{1 + 2\cos(\beta\pi/2)\,\omega_n^{2\beta} + \omega_n^{2\beta}} \tag{10}$$

$$\begin{aligned}\tan\delta = {} & (d-1)\left\{\sin(\alpha\pi/2)\,\omega_n^{\alpha}\right. \\ & \left. + \sin\left[(\alpha-\beta)\pi/2\right]\omega_n^{\alpha+\beta}\right\} \Big/ \left\{1 + 2\cos(\beta\pi/2)\,\omega_n^{2\beta}\right. \\ & \left. + \omega_n^{2\beta} + (d-1)\left\{\cos(\alpha\pi/2)\,\omega_n^{\alpha}\right.\right. \\ & \left.\left. + \cos\left[(\alpha-\beta)\pi/2\right]\omega_n^{\alpha+\beta}\right\}\right\}\end{aligned} \tag{11}$$

where $\omega_n = \omega\tau$, $\omega = 2\pi f$, f is the frequency in Hz, while G_0, d, τ, α, β are five parameters. As before, this model is fitted to the data in Ref. [34] with 1stOpt using the Levenberg-Marquardt and Universal Global Optimization algorithms. It results in the parameters given in Table 2. The data as well as the fit are given in Figs. 4 and 5. Both the generalized Maxwell model and the

Table 1 Parameters for the fractional Poynting-Thomson model fitted to relaxation data.

Data	E_0 (MPa)	E_1 (MPa)	α	τ_1(s)
In Ref. [15]	38.186	21.162	0.225	50.152
In Ref. [16]	1.546	0.200	0.283	7.712×10^5

Table 2 Parameters for the fractional model fitted to dynamic data.

G_0 (Pa)	d	τ(s)	α	β
1.813×10^6	2.116×10^2	6.355×10^{-3}	0.772	0.657

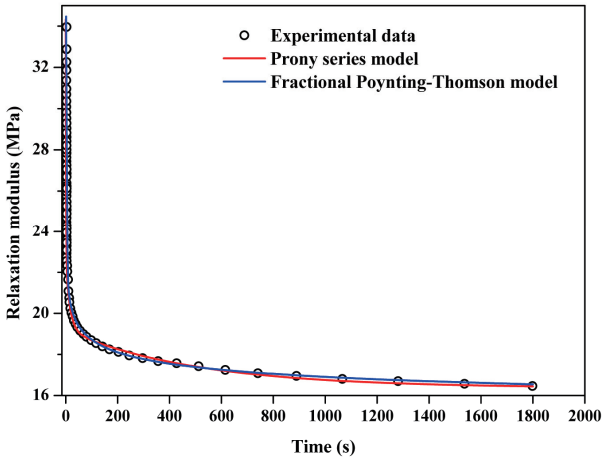


Fig. 2 Comparison of the experimental data of SMPs with the fractional Poynting-Thomson model and the Prony series model.

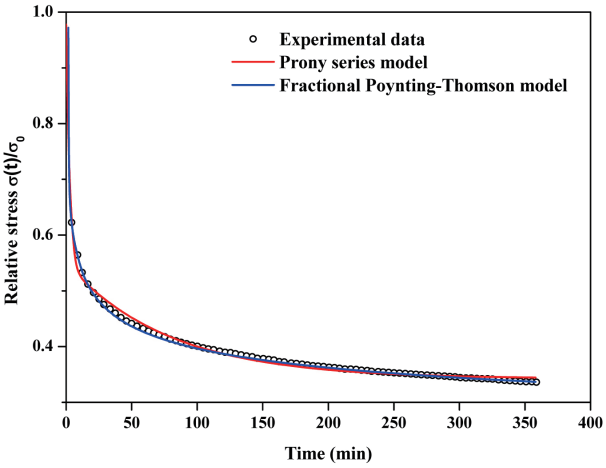


Fig. 3 Comparison of the experimental data of the polyurethane based SMPs with the fractional Poynting-Thomson model and the Prony series model.

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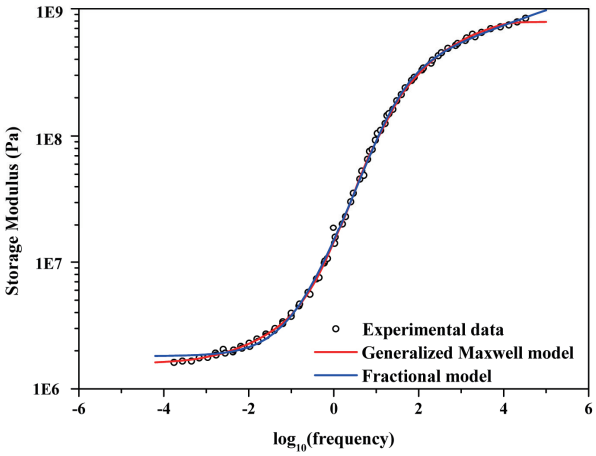


Fig. 4 Comparison of the experimental storage modulus data of the epoxy with the fractional model and the generalized Maxwell model.

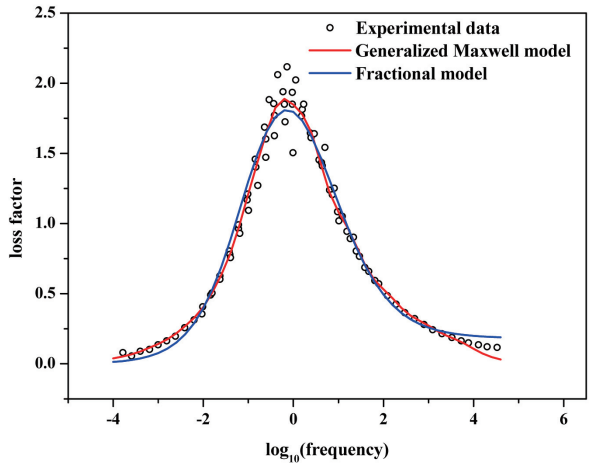


Fig. 5 Comparison of the experimental loss factor data of the epoxy with the fractional model and the generalized Maxwell model.

five-parameter fractional model fit the experimental data very well. The accuracy of fit by the fractional model is not so excellent as the generalized Maxwell model in Fig. 5. It is mainly because the number of parameters in the fractional model is not enough, and it may be improved by adding one or two parameters. However, the advantage of the fractional model is that the model needs fewer parameters compared with the generalized Maxwell model in which 12 pairs of relaxation times and relaxation moduli are defined for the epoxy.

In order to describe the static and dynamic behaviors of viscoelastic materials with high accuracy, often ten or twenty parameters are needed in the traditional viscoelastic models [15,34]. Whereas owing to the vantage of 'spring-pot' mentioned in Section 2.1, it is possible for fractional models with few parameters to characterize the rheological properties.

4. FRACTIONAL MODEL FOR ISOTHERMAL RECOVERY OF POLYURETHANE BASED SMPS

It is not the most important purpose of this work to show that the fractional models of viscoelastic behaviors fit experimental data accurately, which has been done previously and can be found in the literatures [37,38]. One of the motivations for the use of the fractional models applied to SMPs is the superiority to characterize their thermomechanical behaviors. As can be seen from the data fitting in Section 3, it is true that so few parameters are required for fractional models to present the static and dynamic modulus. This lays a sound foundation for describing thermomechanical behaviors.

Now we consider the effectiveness of the fractional models for a shape-memory cycle, especially the free recovery of polyurethane based SMPs. Heuchel *et al.* [16] used the standard linear solid (SLS) model to obtain the apparent shape recovery rate R_r^{app}.

$$R_r^{app} = \frac{\varepsilon_d - \varepsilon(t)}{\varepsilon_d} \tag{12}$$

where $\varepsilon(t)$ is the time-dependent strain and ε_d is the deformed strain of the sample after relaxation at the beginning of the recovery experiment at $t = 0$. Based on the solution of the SLS model in the stress- free case (σ, $\dot{\sigma} = 0$), they put forward a new expression of the time dependent strain.

$$\varepsilon(t) = \varepsilon_d^* \exp\left(-\frac{t}{\tau^*}\right) \tag{13}$$

where ε_d^*, τ^* are two fit parameters. Therefore, the apparent shape recovery rate R_r^{app} with two fit parameters of the effective relaxation time τ^* and the ratio $\varepsilon_d^* / \varepsilon_d$ was obtained as follows [16].

$$R_r^{app} = 1 - \frac{\varepsilon_d^*}{\varepsilon_d} \exp\left(-\frac{t}{\tau^*}\right) \tag{14}$$

In Section 3.2, the fractional Poynting-Thomson model is slightly better to fit the relaxation data. Similarly, according to the solution of the fractional Poynting-Thomson model in the stress-free case, the time dependent strain can be revised as

$$\varepsilon(t) = \varepsilon_d \, E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] \tag{15}$$

where α is the fractional derivative parameter and τ is the effective relaxation time, while $E_\alpha[x]$ is the particular case of Eq. (8) when $\beta = 1$. As a result, the apparent shape recovery rate is

$$R_r^{app} = 1 - E_\alpha\left[-\left(\frac{t}{\tau}\right)^\alpha\right] \tag{16}$$

There are two parameters in both Eqs. (16) and (14). The fitting parameters in Eq. (16) are listed in Table 3, where T_{prog} denotes the programming temperature. The fit of the two equations to experimental data is compared in Fig. 6, where the symbols ■, ● and ▲ represent the experimental data, the dot curve and the

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Table 3 Parameters for the fractional Poynting-Thomson model fitted to free recovery rates.

T_{prog} (°C)	α	τ (min)
40	0.658	8.743
50	0.744	29.247
60	0.611	89.933

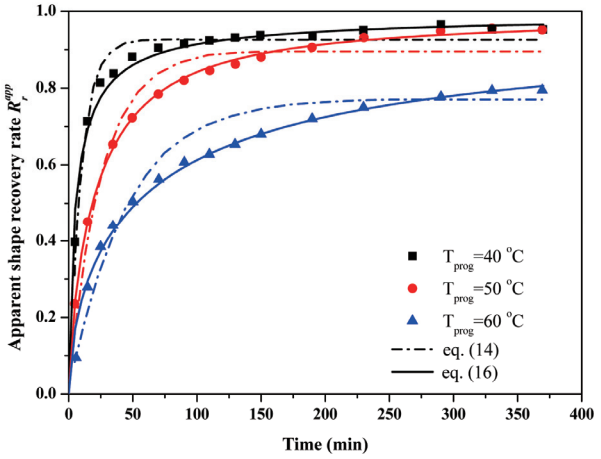


Fig. 6 Comparison of the experimental free recovery rates of the polyurethane based SMPs with the fractional Poynting-Thomson model and the SLS model.

solid curve correspond to the fit of Eqs. (14) and (16) to the experimental data, respectively, and the color differentiates the data and the fit at different programming temperatures T_{prog}. For example, the red symbol is the experimental data at T_{prog} = 50°C and the blue solid curve is the fit of Eq. (16) to the experimental data at T_{prog} = 60°C. It is conspicuous that the fractional model provides a remarkable improvement for description of the shape recovery behavior of SMPs.

5. DISCUSSION

This research suggests that the fractional models have the ability to fit a wide range of data with few parameters. Thus, the fractional models with fewer parameters can characterize the static and dynamic response of SMPs compared with the classical viscoelastic models. Additionally, it is more effective to capture the strain recovery response of SMPs during the free recovery by the fractional calculus method. These findings lead us to believe this method has considerable potential for thermoviscoelastic modeling of SMPs. Although some incipient researches have been provided in the study, many significant problems are still subject to further research, such as, what kind of fractional models can predict strain recovery response of SMPs during isothermal and nonisothermal free recovery, how these models can be utilized in the case of three-dimensional finite deformation of SMPs, and so on.

6. CONCLUSIONS

To characterize the static and dynamic response of SMPs, constitutive models based on fractional calculus are fitted to several sets of experimental data. It is shown that the fractional models can give a similar or better fit with fewer parameters by comparison with the classic viscoelastic models. In the frame of thermoviscoelastic modeling approaches, an accurate fit lays a foundation for describing the thermomechanical behaviors of thermally activated SMPs. On this basis, a fractional rheology model is applied to simulate the isothermal recovery of an amorphous SMP. The fit results show a significant improvement in depicting the strain recovery response of SMPs by the fractional calculus method.

ACKNOWLEDGEMENTS

This work is supported by a Project Funded by the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD), the Research Fund of State Key Laboratory of Mechanics and Control of Mechanical Structures (Grant No. 0214G02), the Funding of Jiangsu Innovation Program for Graduate Education (No.CXLX13_136) and the Fundamental Research Funds for the Central Universities.

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(Manuscript received June 10, 2014,
accepted for publication September 6, 2014.)

https://doi.org/10.1017/jmech.2014.98 Published online by Cambridge University Press