

Study on the performance of variable-order fractional viscoelastic models to the order function parameters

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ABSTRACT

In variable-order fractional calculus, the order is allowed to be in a functional form, which provides a novel way to describe the time-dependent physical processes. In this work, the performance of variable-order fractional viscoelastic models to the order function parameters is comprehensively investigated. Firstly, the variable-order fractional constitutive model is deduced from the theory of constant-order fractional viscoelasticity. The effect of order function is studied which shows that the linear order function can effectively describe the viscoelastic properties. Then the variable-order fractional viscoelastic models are developed considering three kinds of viscoelastic behaviors, including stress-strain relationship, creep, and stress relaxation. The sensitivity of the viscoelastic behaviors to the parameters in the linear order function is analyzed in detail. Finally, the effectiveness of the proposed models is validated by comparing them to the existing experimental data of typical viscoelastic behaviors, and the laws of the order function for each case are discussed based on the fractional viscoelastic theories. It is proved that the variable-order fractional models can give an accurate description of the viscoelastic behaviors under various loading conditions, and the change of the fractional order can visualize the evolution of the mechanical properties of the material.

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1. Introduction

Fractional calculus is the mathematical theory that deals with the characteristics and applications of differential and integral operators of non-integer, real or complex orders [1]. The idea of fractional calculus was initially introduced by Gottfried Leibniz in 1695. After then, many great mathematicians contributed to developing the mathematical theories of fractional calculus in the later three hundred years, including Laplace, Fourier, Liouville, Riemann, Grünwald, Marchaud, Abel, Riesz, Weyl, and Caputo. In the last several decades, the scientific and engineering applications of fractional calculus attracted the interests of researchers, such as in the fields of material modelling, anomalous diffusion, system control, signal and image processing, biology, dielectricity, and so on [2]. Among them, the fractional order model of viscoelasticity is undoubtedly one of the most successful and widely used fields [3].

Viscoelastic properties are widely found in natural and artificial materials, such as polymers, biological tissue, soft soil, and concrete [4]. To describe the non-linear mechanical responses of viscoelastic materials, the traditional approach is to combine the spring and dashpot elements in different ways. For example, creep is a typical viscoelastic phenomenon where

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the strain of a material increases with time under constant stress. It can be depicted by the Kelvin model, which is obtained by the parallel connection of a spring and a dashpot [5]. And stress relaxation is a viscoelastic process in which the total material deformation remains constant while the stress reduces with time. The Maxwell model can well describe stress relaxation by connecting a spring and a dashpot in series [6]. In addition, for the increasingly complex viscoelastic materials in modern researches and applications, their mechanical behaviors are often not accurately described using only the integer Maxwell or Kelvin models. Therefore, many studies further combined multiple elements together, e.g., the Zener model is obtained from a spring in parallel with the Maxwell model, and the Burgers model is obtained from the Maxwell model in series with the Kelvin model [7]. However, this leads to the classical integer element models becoming formally complex and usually having a large number of model parameters.

To improve the accuracy of viscoelastic models and to reduce the number of parameters, the constant-order fractional derivative operators are introduced in the viscoelastic models. If the Hooke's law represented by the spring element is regarded as the zero order derivative and the Newton's law of viscosity of the dashpot element is the derivative of the first order, then the fractional derivative operators can describe intermediate properties between elasticity and viscosity with orders between 0 and 1. In this way, Smit and Vries [8] proposed the classical constant-order fractional viscoelastic model, which is also called the “spring-pot” element. By replacing the dashpot with “spring-pot” in the tractional element models, the fractional models are popular for describing viscoelasticity in recent decades [9]. It has been shown that the fractional order viscoelastic models can effectively characterize the time-, rate- or temperature-dependent mechanical behaviors of various materials [10,11]. In terms of physical significance, Mashayekhi et al. [12] proposed that the order in the fractional viscoelastic model for fractal materials is a rate dependent material property related to fractal and spectral dimensions. However, in most studies, the constant-order fractional derivative operators are still limited to element models with the unclear physical explanation of parameters.

The variable-order fractional calculus is a new mathematical concept put forward at the end of the 20th century. The definition of the variable-order fractional integral and differential operators was first calculated by Samko and Ross [13] in 1993 as a direct extension of the constant-order fractional calculus. Subsequently, Ingman [14], Lorenzo and Hartley [15], Coimbra [16], and other scholars independently conducted further research on the variable-order fractional operators. The variable order fractional operators allow the order to be in the functional form of independent variables, e.g., time or space. This provides a new method for describing the time dependence and historical memory of viscoelastic materials.

Ingman and Suzdalnitsky [17] were early contributors to the study of applying variable-order differential operators to the constitutive relations of viscoelastic materials. They explored the dependence of the order function on strain and strain rate through the experimental results of polymeric materials. Coimbra [18] formulated a constitutive relation for the viscoelasticity of composite materials using variable-order calculus. The variable order was physically interpreted as a measure of the rate of disorder inside materials. Therefore, the order function was considered to visually reflect the evolution of the mechanical properties of materials. In recent years, an increasing number of scholars have used variable-order fractional methods in the research of viscoelasticity, including stress-strain relationships, creep, and stress relaxation [19–21]. However, few studies have discussed the form of order functions in the variable-order fractional viscoelastic models and discussed the effects of parameters on different typical viscoelastic behaviors comprehensively. Motivated by this situation, the present work attempts to apply the variable-order fractional calculus in viscoelastic models, including the non-linear stress-strain relationship, creep, and stress relaxation. In particular, the performance of the variable-order fractional viscoelastic models to the role of the order function parameters in each condition will be discussed in detail.

The rest of this article is organized as follows. In Section 2, the fundamental theories of constant-order and variable-order fractional viscoelasticity are introduced. A comparative study will be conducted to determine the order function. Then the variable-order fractional viscoelastic models will be established in Section 3, including three conditions, stress-strain relationship, creep and stress relaxation. The sensitivity of the viscoelastic behaviors to the order function parameters will be analyzed in detail. In Section 4, the effectiveness of variable-order fractional models will be validated by comparing them to the existing experimental data of typical viscoelastic behaviors. For each case, the variation of the fractional order with relation to the change of mechanical property will be discussed. Finally, Section 5 will be the conclusion.

2. Theoretical methods

While the constant-order fractional calculus has been widely used to describe viscoelasticity, the variable-order fractional calculus is an emerging approach that has received attention only in recent decades. Many of the theoretical foundations are derived from the constant-order fractional calculus. In this section, the basic theories of constant-order fractional viscoelasticity will be introduced, followed by the derivation of variable-order fractional viscoelasticity and the determination of the order function through a comparative study.

2.1. Constant-order fractional viscoelasticity

The classical constant-order fractional viscoelastic model was developed by Smit and Vries [8], which is called the “spring-pot” element,

$$\sigma(t) = E\theta^\alpha D^\alpha \varepsilon(t) \quad (1)$$

where E (MPa) represents the elastic modulus and θ (s) stands for the relaxation time, which can be written into $\theta = \eta/E$. D^α is the α order fractional derivative operator and $0 \leq \alpha \leq 1$. So, Eq. (1) will equal to the Hooke's law of elasticity and the Newton's law of viscosity when the fractional order is 0 and 1, respectively.

$$\sigma(t) = E\varepsilon(t) = ED_t^0 \varepsilon(t) \quad \alpha = 0 \quad (2)$$

$$\sigma(t) = \eta d\varepsilon(t)/dt = \eta D_t^1 \varepsilon(t) \quad \alpha = 1 \quad (3)$$

Up to now, there existed many different fractional differential operators, in which the Riemann-Liouville and the Caputo definitions are the most commonly used ones. The classical Riemann-Liouville differential operator is expressed as,

$${}_0^RL_t^\alpha f(t) = \frac{1}{\Gamma(1-\alpha)} \frac{d}{dt} \int_0^t \frac{f(\tau) d\tau}{(t-\tau)^\alpha} \quad (4)$$

in which $\Gamma(*)$ is the Gamma function,

$$\Gamma(t) = \int_0^t e^{-\tau} \tau^{t-1} d\tau. \quad (5)$$

By first integrating and then differentiating, the Riemann-Liouville definition can avoid limit solving and simplify the model solution process. The Caputo differential operator is defined as,

$${}_0^CD_t^\alpha f(t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t \frac{f'(\tau) d\tau}{(t-\tau)^\alpha} \quad (6)$$

Compared to the Riemann-Liouville definition, the Caputo definition is more convenient for engineering applications because it requires less processing of initial conditions.

2.2. Variable-order fractional viscoelasticity

In reality, most materials behave time-dependent mechanical responses. The variable-order fractional viscoelastic models are proposed by several scholars indelicately around the beginning of the 21st century. The method is to turn the constant fractional order in Eq. (1) into a time-dependent function,

$$\sigma(t) = E\theta^{\alpha(t)} D^{\alpha(t)} \varepsilon(t) \quad (7)$$

Eq. (7) can describe the intermediate viscoelastic properties between Hooke solid and Newton fluid with the change of fractional order, as shown in Fig. 1.

The definitions for variable-order differential operators are usually derived from the definitions of the corresponding constant-order operators. Therefore, in describing different viscoelastic behaviors in this study, appropriate definitions of variable-order differential operators will be chosen as needed. For the viscoelastic stress-strain relationship, Coimbra [22] proposed the following variable-order Caputo differential operator,

$${}_0D_t^{\alpha(t)} f(t) = \frac{1}{\Gamma[1-\alpha(t)]} \int_0^t (t-\tau)^{-\alpha(t)} f^{(1)}(\tau) d\tau + \frac{(f(0+) - f(0-))t^{-\alpha(t)}}{\Gamma[1-\alpha(t)]} \quad (8)$$

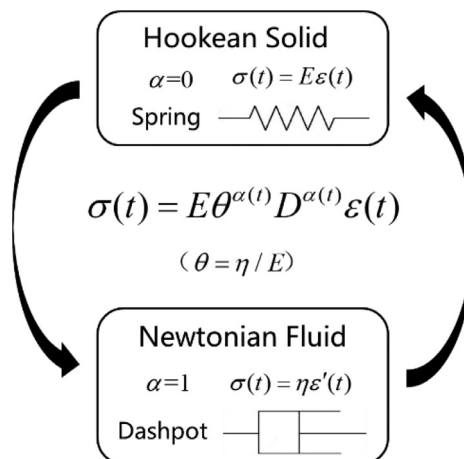


Fig. 1. Schematic representation of variable-order fractional viscoelasticity.

Eq. (8) can return to the constant-order fractional differential operator when the order is taken as a constant, which can well reflect the continuity of physical process.

The mechanical properties of materials are often tested at constant strain rate,

$$\varepsilon(t) = \dot{\varepsilon}t \quad (9)$$

Thus, the variable-order fractional differential operator in Eq. (8) can be rewritten as,

$$D^{\alpha(t)}f(t) = \frac{1}{\Gamma(1-\alpha(t))} \int_0^t (t-\tau)^{-\alpha(t)} \dot{\varepsilon} d\tau \quad (10)$$

Integration of the right part yields,

$$D^{\alpha(t)}f(t) = \frac{\dot{\varepsilon}}{\Gamma(1-\alpha(t))} \left[\frac{-(t-\tau)^{1-\alpha(t)}}{1-\alpha(t)} \right]_0^t = \frac{\dot{\varepsilon}}{\Gamma(1-\alpha(t))} \frac{t^{1-\alpha(t)}}{1-\alpha(t)} \quad (11)$$

The Gamma function has the following operational property,

$$\Gamma(x+1) = x\Gamma(x) \quad (12)$$

Combining the right part of Eq. (11) gives,

$$D^{\alpha(t)}f(t) = \frac{\dot{\varepsilon}t^{1-\alpha(t)}}{\Gamma(2-\alpha(t))} \quad (13)$$

Substitution of Eq. (13) into Eq. (7) will get,

$$\sigma(t) = E(\theta)^{\alpha(t)} \frac{\dot{\varepsilon}t^{1-\alpha(t)}}{\Gamma(2-\alpha(t))} \quad (14)$$

At constant strain rate $\varepsilon(t) = \dot{\varepsilon}t$, it can be further derived as,

$$\sigma(t) = E(\theta)^{\alpha(t)} \frac{\dot{\varepsilon}(\varepsilon(t)\dot{\varepsilon}^{-1})^{1-\alpha(t)}}{\Gamma(2-\alpha(t))} = E(\theta)^{\alpha(t)} \frac{\dot{\varepsilon}^{\alpha(t)}\varepsilon(t)^{1-\alpha(t)}}{\Gamma(2-\alpha(t))} \quad (15)$$

Finally, the stress-strain relationship of variable-order fractional viscoelasticity can be obtained,

$$\sigma(t) = E(\dot{\varepsilon}\theta)^{\alpha(t)} \frac{\varepsilon(t)^{1-\alpha(t)}}{\Gamma(2-\alpha(t))} \quad (16)$$

2.3. Determination of order function

In the application of the variable-order fractional stress-strain relationship of Eq. (16) for data simulation, there are two key points to be noted: (1) The value of the fractional order should conform to the viscoelastic interval, which is restricted to be between 0 and 1; (2) The functional form of the fractional order needs to reflect the mechanical properties evolution on the basis of variable-order fractional viscoelasticity. The standard approach in the current studies is to determine the functional form of fractional order from the perspective of data simulation. Still, few studies have discussed the impact of different order function forms on the data description.

In this work, a comparative study is conducted on four potential forms of order functions which are exponential, linear, constant, and logarithmic. Fig. 2 shows the stress-strain curves by substituting different forms of order functions into Eq. (16). The cases of order 0 and order 1 are also plotted to better clarify the reasonable interval range of the fractional order. As presented, all the four forms of order function can reflect the viscoelastic stress-strain relationship under constant strain rate within the limits of ideal solid and fluid. Among them, the curve obtained by constant order is fixed, whereas the logarithmic order curve shows a first rising and then falling trend. Therefore, these two order forms are not suitable for viscoelastic models to reflect the changing mechanical properties during deformation. In addition, while both linear and exponential order curves can show a continuous change from fluid to solid, the rise rate of exponential order curve is slower which makes it difficult to control in application. In summary, although the other three types of order functions can also reflect the viscoelastic properties, the result of the linear order function varies more smoothly over the range of the mechanical properties between solid and fluid. This suggests that the linear form of order function can describe the viscoelastic properties more effectively, especially under the conditions of constant strain rate loading. Therefore, the linear form of the variable fractional order function with time will be used in this study,

$$\alpha(t) = at + b \quad (17)$$

Then the complete form of the stress-strain relationship of variable-order fractional viscoelasticity should be,

$$\sigma(\varepsilon) = E(\dot{\varepsilon}\theta)^{at+b} \frac{\varepsilon^{1-(at+b)}}{\Gamma(2-(at+b))} \quad (18)$$

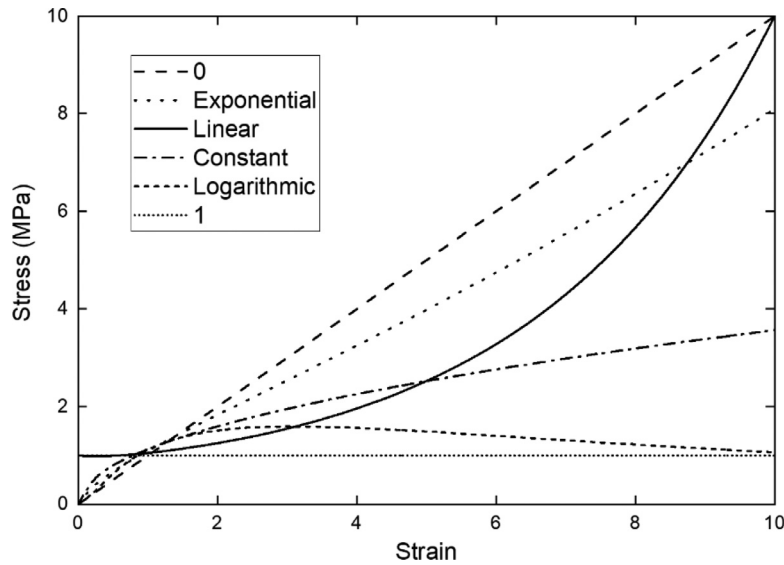


Fig. 2. Model predictions for different forms of order functions.

3. Variable-order fractional viscoelastic models

In this section, the variable-order fractional viscoelastic models will be developed based on the theoretical methods in Section 2. Three typical viscoelastic behaviors will be considered, including viscoelastic stress-strain relationship, creep, and stress relaxation. More importantly, the effects of the parameters in the order function on each model will be discussed in detail.

3.1. Variable-order fractional stress-strain model

Viscoelastic materials tend to exhibit nonlinear stress-strain responses during deformation. Eq. (18) already gives the stress-strain response of variable fractional order, where the order is a linear time function. Under constant strain rate loading, the strain is a primary function of time, so the fractional order can also be a linear function of strain,

$$\alpha(\varepsilon) = a\varepsilon + b \quad (19)$$

For the convenience of data fitting, we further substitute Eq. (19) into Eq. (18) to obtain the following variable-order fractional stress-strain model,

$$\sigma(\varepsilon) = E(\dot{\varepsilon}\theta)^{a\varepsilon+b} \frac{\varepsilon^{1-(a\varepsilon+b)}}{\Gamma(2-(a\varepsilon+b))} \quad (20)$$

Theoretically, Eq. (20) can describe the viscoelastic stress-strain relationship, and the change of fractional order can depict the mechanical property evolution in materials. Under the assumption that the form of variable order function is linear, its value is further governed by two parameters, slope a and intercept b . Therefore, these two order function parameters must significantly affect the model predictions. The sensitivity of the stress-strain relationship to the order function parameters is shown in Fig. 3.

According to Fig. 3(a), the slope parameter a mainly affects the rate of development of the stress response, and when the value of a is larger, the rate of stress increase is slower. Meanwhile, Fig. 3(b) shows that the intercept parameter b has a more pronounced effect on the initial value of the stress response, which is smaller when the value of b is larger. The results are consistent with the fractional viscoelasticity theory. In the linear order function, a larger slope or intercept would bring the value of fractional order nearer to 1, representing the material property closer to fluid, resulting in lower stress level performance.

As a result, by controlling the values of the parameters in the order function, slope a and intercept b , the variable-order fractional stress-strain model can effectively characterize the viscoelastic stress-strain responses under various conditions and reveal the trend of mechanical properties through the order function.

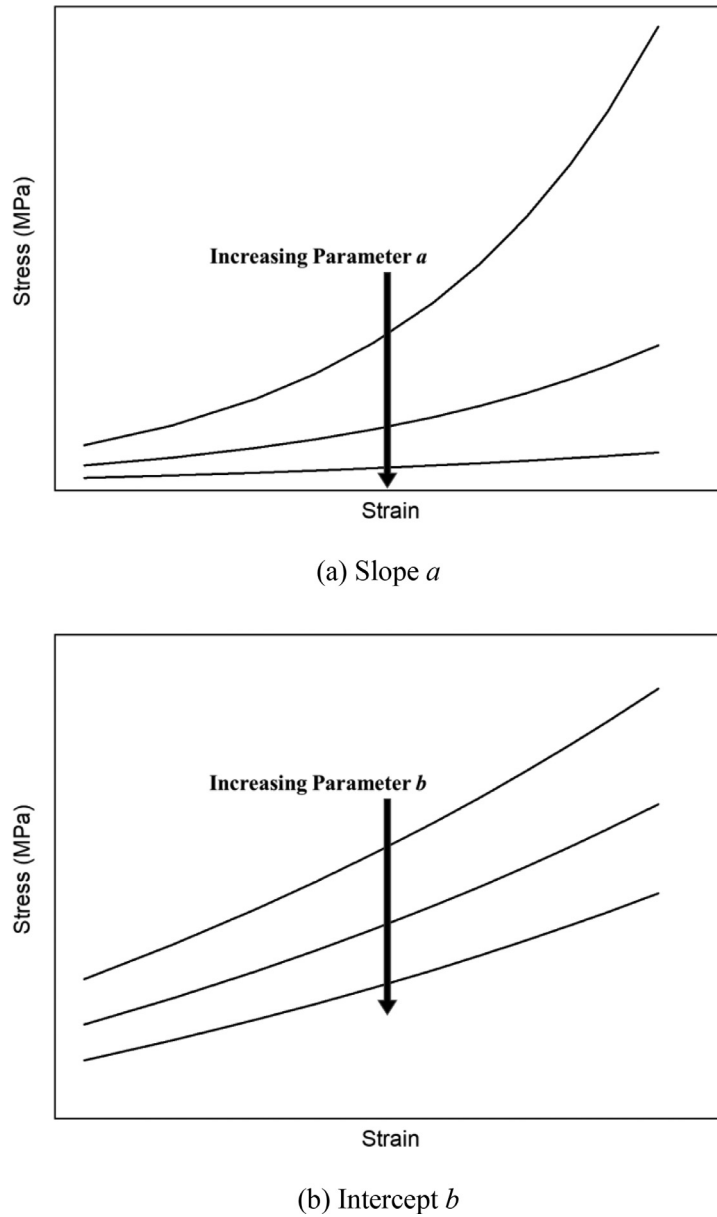


Fig. 3. Sensitivity of the stress-strain relationship to the order function parameters: (a) Slope a ; (b) Intercept b .

3.2. Variable-order fractional creep model

Creep is typical viscoelastic behavior, representing a continuous development in deformation with time under constant stress,

$$\sigma(t) = \sigma_0 \quad (21)$$

Thus, the variable-order viscoelastic model of Eq. (7) should be formulated into the integral form of strain,

$$\varepsilon(t) = \frac{1}{E} \theta^{-\alpha(t)} D^{-\alpha(t)} \sigma(t) \quad (22)$$

In Eq. (22), the variable-order fractional integral operator will take the definition of convolution form proposed by Lorenzo and Hartley [15],

$${}_0 D_t^{-\alpha(t)} f(t) = \int_0^t \frac{(t-\tau)^{\alpha(t)-1}}{\Gamma[\alpha(t)]} f(\tau) d\tau \quad (23)$$

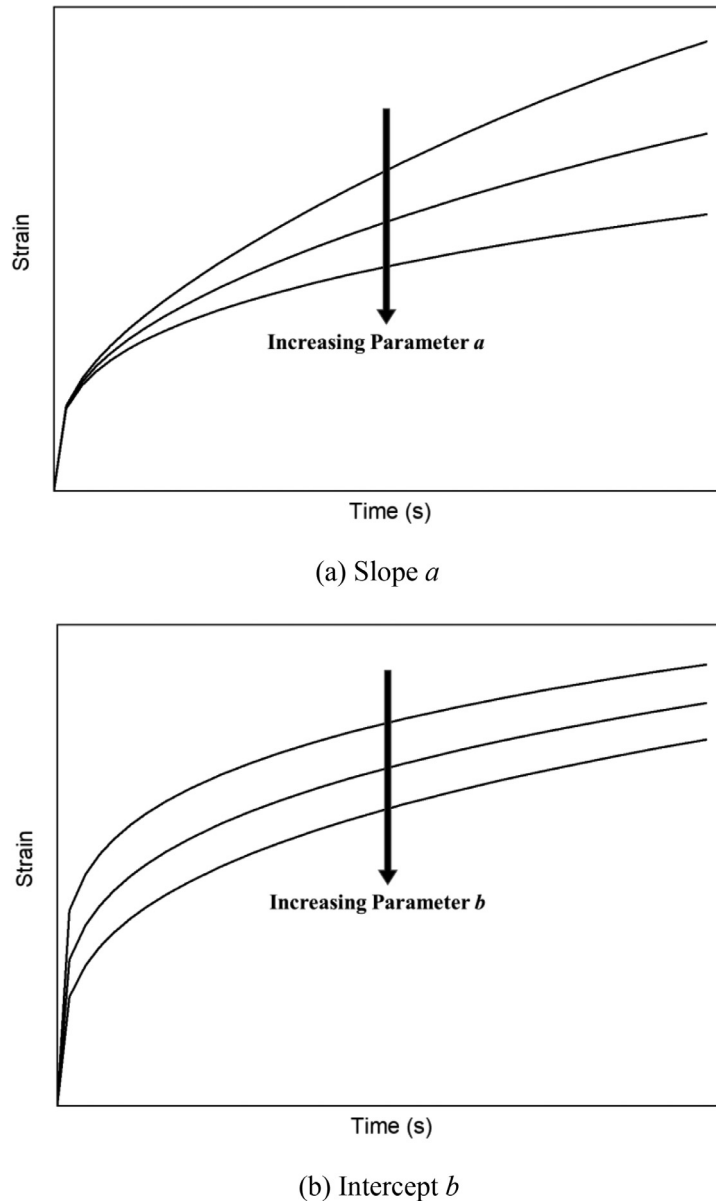


Fig. 4. Sensitivity of the creep behavior to the order function parameters: (a) Slope a ; (b) Intercept b .

Therefore, Eq. (22) can be rewritten as,

$$\varepsilon(t) = \frac{1}{E} \theta^{-\alpha(t)} \int_0^t \frac{(t-\tau)^{\alpha(t)-1}}{\Gamma[\alpha(t)]} \sigma_0 d\tau \quad (24)$$

By integrating the right part and adopting the linear order function with time, the variable-order fractional creep model can be derived,

$$\varepsilon(t) = \frac{\sigma_0}{E\theta^{at+b}} \frac{t^{at+b}}{\Gamma(1+at+b)} \quad (25)$$

Similarly, the slope parameter a and the intercept parameter b in the linear order function are essential in the performance of the variable-order fractional creep model. Fig. 4 displays the sensitivity of the creep behavior expressed in terms of strain-time curves to the order function parameters. It can be found that the slope parameter a also mainly affects the development rate of the creep strain, while its overall magnitude is mainly influenced by the intercept parameter b . More specifically, from Fig. 4(a), we can see that the rate of creep stress rise slows down as parameter a increases, while from Fig. 4(b), we can learn that the overall value of creep stress is smaller for larger parameter b . From the perspective of

fractional viscoelasticity theory, this is because a larger parameter a makes the material property approaches fluid more quickly, and a larger parameter b means that the overall property behaves more like fluid. This result demonstrates that the variable-order fractional creep model can reasonably describe the creep behavior in various cases.

3.3. Variable-order fractional stress relaxation model

Stress relaxation is a phenomenon in which the stress in a material decreases with time while the total deformation remains constant,

$$\varepsilon(t) = \varepsilon_0 \quad (26)$$

In order to maintain consistency in the definition of variable-order fractional operators, the initial condition is treated as a Heaviside function at $t = 0$,

$$\varepsilon(t) = \begin{cases} 0, & t < 0 \\ \varepsilon_0, & t \geq 0 \end{cases} \quad (27)$$

Therefore, the second term of Coimbra definition of variable-order fractional differential operator in Eq. (8) will lead to,

$$D^{\alpha(t)} f(t) = \frac{\varepsilon_0 t^{-\alpha(t)}}{\Gamma[1 - \alpha(t)]} \quad (28)$$

Eventually, Eq. (7) can be written in the form of stress relaxation,

$$\sigma(t) = E(\theta)^{at+b} \frac{\varepsilon_0 t^{-(at+b)}}{\Gamma(1 - (at + b))} \quad (29)$$

In the same way, the sensitivity of the stress relaxation behavior to the order function parameters in Eq. (29) is presented in Fig. 5. It can be seen in Fig. 5(a) that the rate of stress reduction in the stress relaxation process is significantly influenced by the slope parameter a , which is faster when a is larger. This phenomenon is similar to the previous laws. The larger a is, the faster the material properties approach to fluid, and therefore the quicker the value of stress falls.

However, the influence of the intercept parameter b on the stress relaxation process is divided into two aspects, including the initial rate and the overall value of stress. From Fig. 5(b), we can obtain that with b increases, the initial rate of stress reduction becomes lower, but the final overall stress value is smaller. These results can be explained by the fractional order viscoelastic theory. That is, when the value of b is small, the material property is more similar to solid, and the stress reduction at the beginning of the stress relaxation is more pronounced, but the final stress value will be more significant. And when b increases, the material properties gradually approach fluid, and the phenomenon in Fig. 5(b) appears. In short, the variable-order fractional method is also effective in describing stress relaxation behavior.

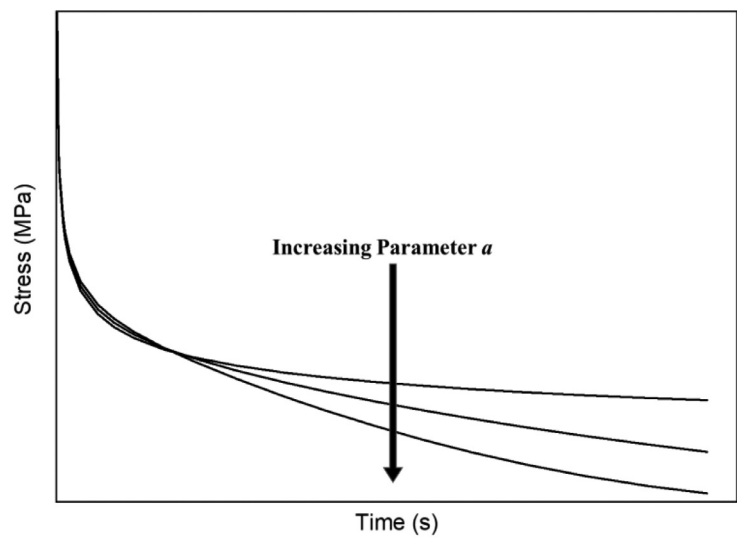
In summary, the variable-order fractional viscoelastic models are successfully developed for the conditions of stress-strain relationship, creep, and stress relaxation behaviors. The sensitivity analysis of the model fitting results to the order function parameters shows that the variable-order fractional models can provide good descriptions of viscoelastic behaviors under various loading conditions.

4. Model validation

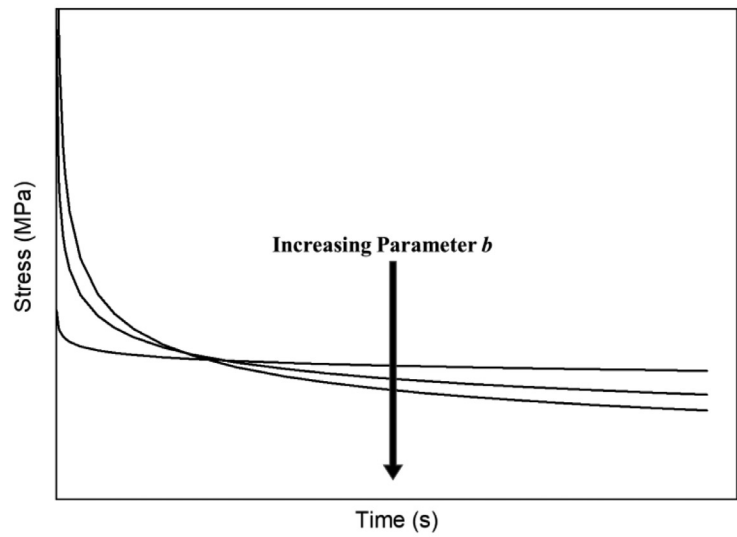
In this section, the variable-order fractional models will be validated by comparing them to the experimental data of typical viscoelastic behaviors in the literature. The MATLAB software will be used to fit the data and obtain the model parameters by least square method. The goodness of fit will be assessed by the values of the root mean square error (RMSE) and the coefficient of determination (R-square). In each case, the comparison between model simulation and experimental data will be plotted to show the best-fitting results. Order function curves will also be plotted to visualize the order changes during each viscoelastic behavior for a variety of loading conditions.

4.1. Large deformation stress-strain behavior

A series of constant strain rate experiments were performed on poly(ethylene terephthalate) (PET) by Boyce et al. [23] to obtain the finite deformation stress-strain behavior above the glass transition. The uniaxial compression stress-strain data of PET at the temperatures of 90, 100, and 105 °C for the strain rate of 0.1/s are selected for validation. We first tried to use the constant-order fractional model to describe the experimental data. The model parameters of the constant-order fractional stress-strain model for PET are presented in Table 1, and the comparative results are shown in Fig. 6. In Table 1, it can be found that when the temperature rises, the elastic modulus E decreases while the relaxation time θ increases. Also, the value of fractional order α is closer to 1 with rising temperature, which is all in accordance with the fact that the mechanical response of PET is softer at higher temperatures. However, Fig. 6 illustrates that although the constant-order fractional model can well describe the stress-strain behavior of PET at small strains less than 1, in the case of large deformation, the stress-strain curves of PET will significantly rise due to strain hardening which cannot be reflected by the



(a) Slope a



(b) Intercept b

Fig. 5. Sensitivity of the stress relaxation behavior to the order function parameters: (a) Slope a ; (b) Intercept b .

Table 1
Model parameters of constant-order fractional stress-strain model for PET.

T (°C)	E (MPa)	θ (s)	α
90	10.01	0.0017	0.13
100	4.278	0.1429	0.17
105	1.94	1.693	0.227

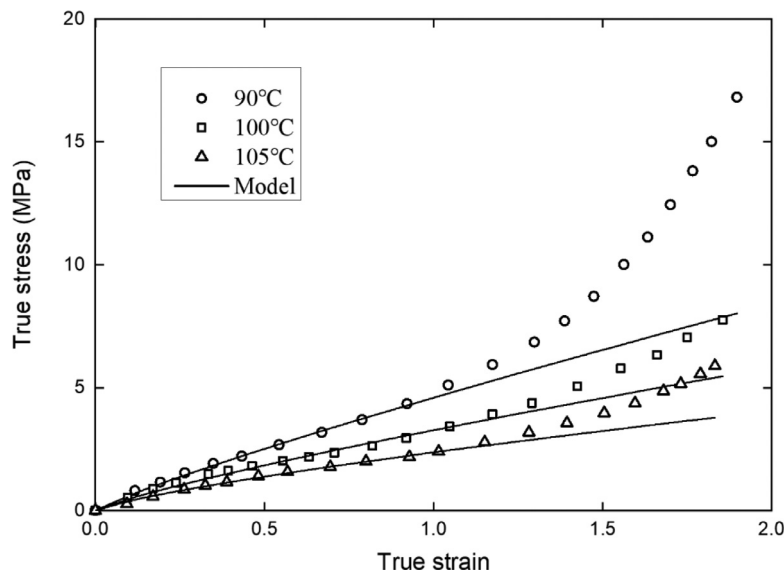


Fig. 6. Comparisons between the experimental data and the constant-order fractional model for the stress-strain behavior of PET at various temperatures.

Table 2

Model parameters of variable-order fractional stress-strain model for PET at large deformations.

T (°C)	E (MPa)	θ (s)	a	b	RMSE	R ²
90	11.36	0.0027	−0.1	0.22	0.3165	0.9951
100	5.673	0.188	−0.1	0.26	0.05662	0.999
105	5.4	0.2475	−0.1	0.328	0.249	0.9646

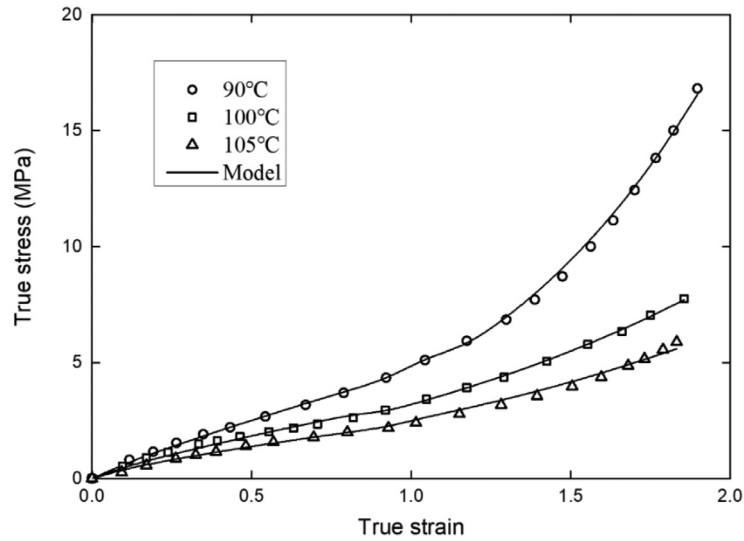
constant-order fractional model. Therefore, the large deformation behavior of PET should be described by the variable-order fractional stress-strain model.

The comparisons between the variable-order fractional stress-strain model and the experimental data for the large deformation of PET are shown in Fig. 7(a). Compared with Fig. 6, the variable-order fractional model can describe the complete stress-strain curves, including the rising part at large deformation. The model parameters and the goodness of fit during simulation are presented in Table 2. Likewise, the elastic modulus E is decreasing and the relaxation time θ is increasing when the temperature rises. The difference is that in this case, the constant fractional order is still used at the small deformation stage where the strain is less than 1, while the linearly decreasing order function is used at the large deformation stage, as presented in Fig. 7(b). In theory, the decreasing fractional order intuitively reflects the strain hardening of PET at large deformation. And from an overall perspective, the value of the order is generally larger at higher temperatures, also reflecting that the mechanical response of PET is closer to fluid at higher temperatures. Therefore, it can be concluded that the variable-order fractional stress-strain model can give an accurate description of the viscoelastic stress-strain relationship of PET. The law of linear order function can effectively capture the strain hardening behavior of PET at large deformation as well as the evolution of mechanical properties at different temperatures.

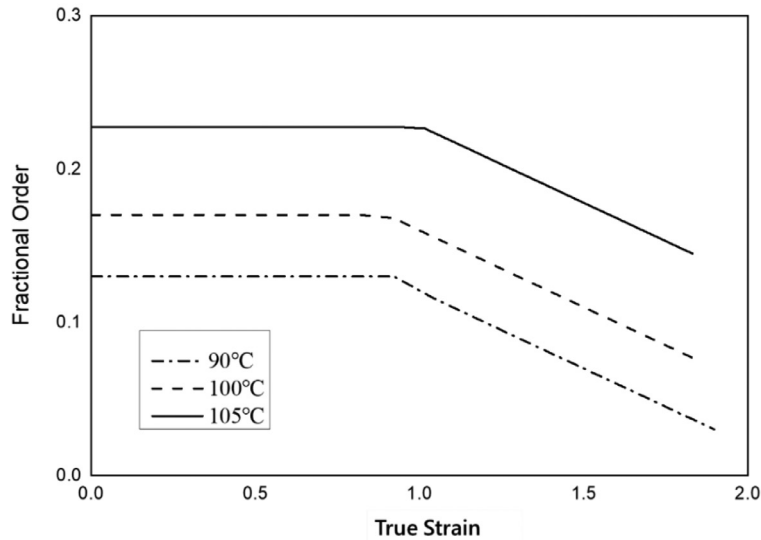
To validate the advantages of the proposed methods, a comparative study is further conducted on the variable-order fractional stress-strain model as a representative, since the creep and stress relaxation models are derived from the stress-strain relationships. Khajehsaeid et al. [24] introduced an integer visco-hyperelastic model to describe the large deformation viscoelastic behaviors by paralleling an elastic spring with a Maxwell element, see Fig. 2 in Ref. [24]. Under constant strain rate uniaxial deformation, their model is given by

$$\sigma_{11} = 2A(\lambda^2 - \lambda^{-1})[\exp(a(I_1 - 3)) - b \ln(I_1 - 2)] + \int_1^\lambda \frac{\partial [2\tilde{A}(\exp(\tilde{a}(I_1 - 3)) - \tilde{b} \ln(I_1 - 2))(\xi^2 - \xi^{-1})]}{\partial \xi} \exp\left(-\frac{\lambda - \xi}{\dot{\varepsilon}_{11}\theta(\dot{\varepsilon})}\right) d\xi \quad (30)$$

$$\theta = \theta_{ref} \left(\frac{\|\dot{C}\|_{ref}}{\|\dot{C}\|} \right)^\eta \quad (31)$$



(a) Simulation results



(b) Order curves

Fig. 7. Comparisons between the experimental data and the variable-order fractional stress-strain model for the large deformation behavior of PET at various temperatures: (a) Simulation results; (b) Order curves.

in which $\zeta = \dot{\epsilon}_{11} \tau + 1$ is a dummy variable, $\|\dot{\mathbf{C}}\| = \dot{\epsilon}_{11} \sqrt{4\zeta^2 + 2\zeta^{-4}}$ is the magnitude of the time rate of right Cauchy–Green deformation tensor and $\theta_{ref} = \varphi_{ref} \|\dot{\mathbf{C}}\|_{ref}^{-\eta}$ is the relaxation time on the reference path where φ_{ref} is a material parameter. Therefore, there are eight parameters to be determined in the visco-hyperelastic model which are $A, a, b, \tilde{A}, \tilde{a}, \tilde{b}, \varphi_{ref}, \eta$.

It is without doubt that the variable order fractional model has a simpler form compared to Eq. (30). The comparison of the variable order fractional model with the visco-hyperelastic model on the stress-strain behavior of polyurea is plotted in Fig. 8, and the goodness of fit is provided by the RMSE and R-square values in Table 3. As is generally known, the smaller the value of RMSE, or the closer the value of R-square is to 1, the better the model works. Thus, both the RMSE and R-square values in Table 3 indicates that the variable order fractional model achieves better results than the visco-hyperelastic model. In consequence, by comparison with the integer order model, the variable-order fractional model is demonstrated to benefits from the formal simplicity and higher accuracy, as well as the ability to characterize the mechanical properties of macroscopic intermediate materials.

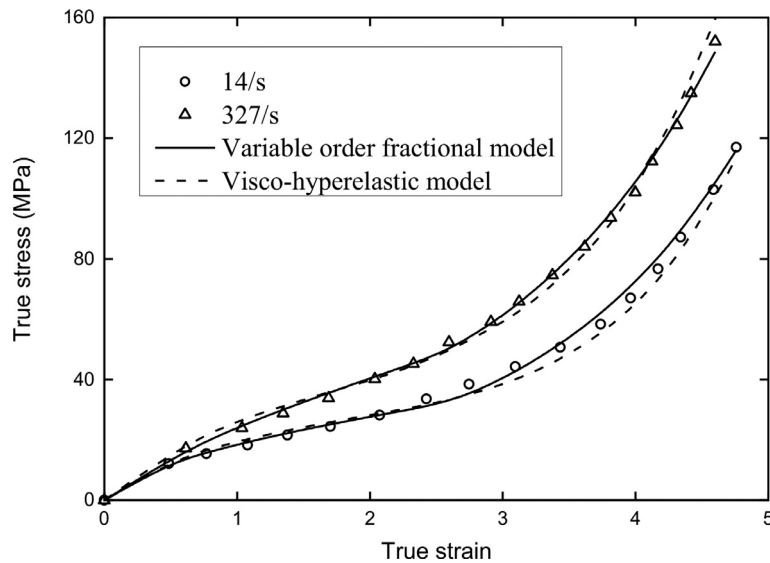


Fig. 8. Comparison of the variable order fractional model with the visco-hyperelastic model on the stress-strain behavior of polyurea.

Table 3

The goodness of fit of models for the stress-strain behavior of polyurea.

	Strain rate (1/s)	Variable order fractional model	Visco-hyperelastic model
RMSE	14	2.13	3.057
	327	1.592	3.267
R ²	14	0.997	0.997
	327	0.999	0.996

Table 4

Model parameters of variable-order fractional creep model for PP.

σ (MPa)	E (MPa)	θ (s)	a	b	RMSE	R ²
15	420.4	23,680	$-6.15\text{e-}5$	0.1189	$3.18\text{e-}4$	0.9953
20	220.4	12,200	$-3.55\text{e-}5$	0.2101	$5.24\text{e-}4$	0.9982
21.5	142.8	9513	$-1.16\text{e-}5$	0.3002	$2.36\text{e-}3$	0.9867

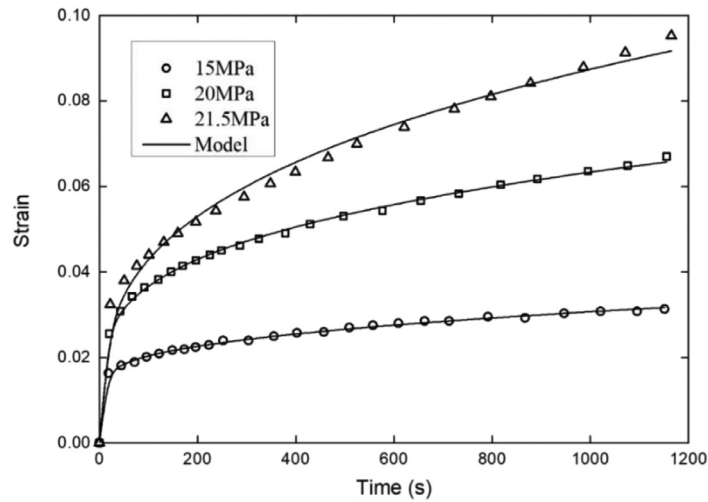
4.2. Creep

Drozdov et al. [25] carried out the creep test on polypropylene (PP) at the ambient temperature of 23 °C. The experimental data at the required stresses of 15, 20, and 21.5 MPa are chosen to verify the variable-order fractional creep model. The model parameters are given in Table 4, in which the elastic modulus E and the relaxation time θ both decrease with the applied stress increases. The comparisons between the experimental data and the model fitting results, as well as the order curves during simulation, are drawn in Fig. 9(a) and 9(b), respectively.

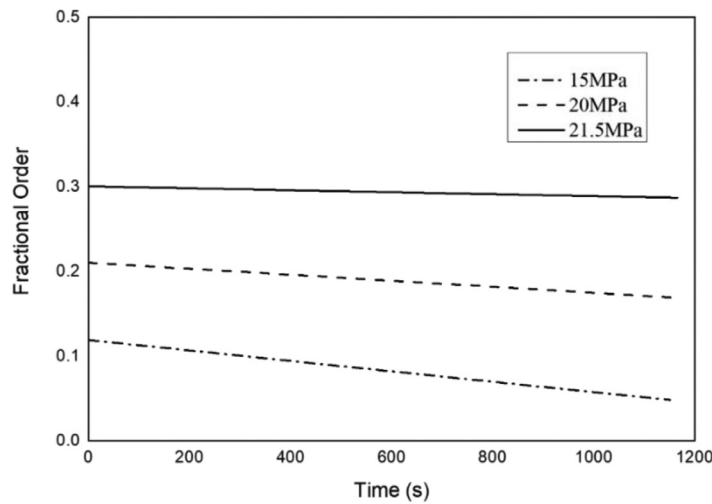
From Fig. 9(a), the model simulation results are in good agreement with the creep data of PP under various stresses. And Fig. 9(b) shows that the fractional order decreases linearly with time during creep. Meanwhile, the overall value of the fractional order becomes more significant with the increase of the applied stress. According to the fractional viscoelastic theory, the regulation of order curves suggests that the mechanical response of PP becomes harder during creep but relatively softer at larger applied stress. Hence, the comparative study on the creep data of PP once again proves the excellent fitting of the variable-order fractional model, in which the order function has the ability to effectively reflect the change of mechanical properties of materials during creep.

4.3. Stress relaxation

To validate the variable-order fractional stress relaxation model, the experimental data of Poly(methyl methacrylate) (PMMA) obtained by Mathiesen [26] are chosen for the comparative study. The samples of PMMA were loaded at a rate of 3.0/min to the final strain of 1.0, then held for 180 s to get the stress relaxation data of different temperatures. The model parameters of the variable-order fractional stress relaxation model for the experimental data at the temperatures



(a) Simulation results



(b) Order curves

Fig. 9. Comparisons between the experimental data of PP and the variable-order creep model at various stresses: (a) Simulation results; (b) Order curves.

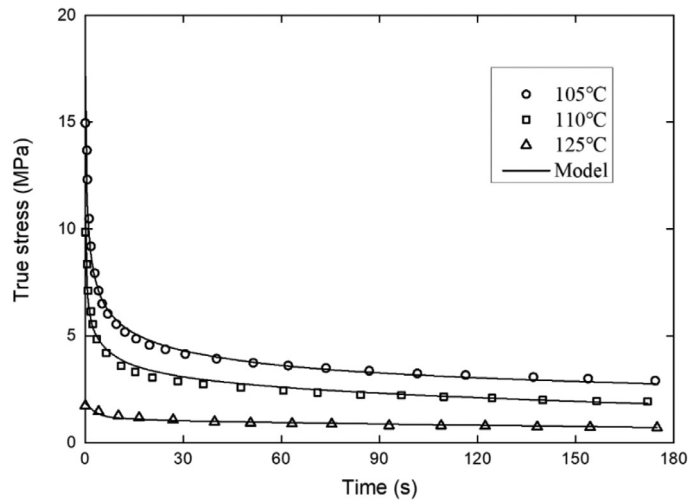
Table 5

Model parameters of variable-order fractional stress relaxation model for PMMA.

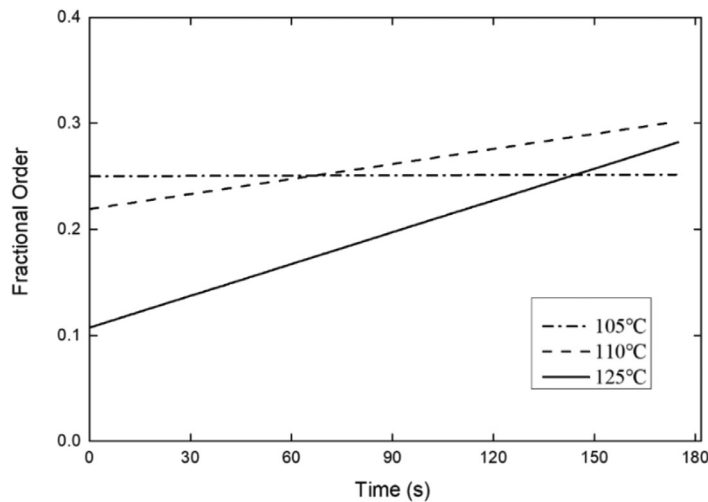
T (°C)	E (MPa)	θ (s)	a	b	RMSE	R ²
105	4.038	86.68	1e-5	0.25	0.8895	0.9385
110	2.797	100	4.732e-4	0.2191	0.4205	0.9686
125	0.8734	200	1e-3	0.1073	0.08492	0.9242

of 105, 110, and 125 °C are presented in Table 5. As the temperature increases, the elastic modulus E decreases, and the relaxation time θ increases which is the same law as that obtained for the stress-strain behavior.

The model comparison results are shown in Fig. 10(a), which shows that the proposed variable-order fractional method can also depict the stress relaxation behavior of PMMA very well. The order variations at different temperatures are plotted in Fig. 10(b). It is shown that the value of the fractional order is initially smaller at higher temperature, but increase faster during the subsequent stress relaxation process. This is due to the fact that the initial value of the stress is larger at lower temperatures, and therefore the relaxation is faster in the initial stage, i.e., the material softens faster. And as the stress relaxation process develops, the mechanical properties of PMMA eventually become softer at higher temperatures.



(a) Simulation results



(b) Order curves

Fig. 10. Comparisons between the experimental data of PMMA and the variable order stress relaxation model at various temperatures: (a) Simulation results; (b) Order curves.

Consequently, this result suggests that the variable-order fractional stress relaxation model is also capable of revealing the evolution of mechanical property during the stress relaxation process.

In conclusion, it is demonstrated that the variable-order fractional constitutive method can effectively describe the typical viscoelastic behaviors, including stress-strain relationship, creep, and stress relaxation. For each case, the order functions in the variable-order fractional viscoelastic models have appropriate laws that help to reveal the time-dependent mechanical property evolution of viscoelastic materials intuitively.

5. Conclusion

In this work, the variable-order fractional viscoelastic models are developed considering three kinds of typical viscoelastic behaviors, including stress-strain relationship, creep, and stress relaxation. The effect of order function is studied which shows that the order function in a linear form can effectively describe the viscoelastic properties. The sensitivity analysis of the viscoelastic behaviors concerning the parameters in the linear order function indicates that the variable-order fractional models can describe the viscoelastic behaviors for different loading requirements. The effectiveness of the variable-order fractional models is validated by comparing them to the existing experimental data of typical viscoelastic behaviors, where the laws of the order functions in each case are obtained as follows.

- (i) In describing the viscoelastic stress-strain relationship, the fractional order at the small deformation stage is a constant. In contrast, at the large deformation stage, it becomes a linearly decreasing function, and the overall value is more significant at higher temperatures.
- (ii) When describing the creep behavior, the variable fractional order decreases linearly with time, and the overall value becomes larger when the applied stress increases.
- (iii) For the stress relaxation behavior, the initial value of the fractional order is smaller at higher temperatures, but then increases faster to a larger final value.

From the perspective of fractional viscoelastic theories, it is demonstrated that the variable-order fractional models can effectively reveal the experimental data of viscoelastic behaviors. The trend of the order function can give an intuitive reflection of the mechanical property evolution in viscoelastic materials.

Data availability

Data will be made available on request.

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References

- [1] K. Diethelm, V. Kiryakova, Y. Luchko, et al., Trends, directions for further research, and some open problems of fractional calculus, *Nonlinear Dyn.* (2022) 1–26.
- [2] H.G. Sun, Y. Zhang, D. Baleanu, et al., A new collection of real world applications of fractional calculus in science and engineering, *Commun. Nonlinear Sci. Numer. Simul.* 64 (2018) 213–231.
- [3] C. Zopf, S.E. Hoque, M. Kaliske, Comparison of approaches to model viscoelasticity based on fractional time derivatives, *Comput. Mater. Sci.* 98 (2015) 287–296.
- [4] J.H. Snoeijer, A. Pandey, M.A. Herrada, et al., The relationship between viscoelasticity and elasticity, *Proc. R. Soc. A* 476 (2243) (2020) 20200419.
- [5] B.K. Bihari, N.P.N. Rao, M. Gupta, et al., A study on creep behavior of composite solid propellants using the kelvin-Voigt model, *Cent. Eur. J. Energ. Mater.* 14 (3) (2017) 742–756.
- [6] J.H. Gao, X.X. Yang, Study of the viscoelasticity of chopped aramid fiber reinforced rubber composite, *Key Eng. Mater. Trans. Tech Publications Ltd* 827 (2020) 385–391.
- [7] D.L. Chen, P.F. Yang, Y.S. Lai, A review of three-dimensional viscoelastic models with an application to viscoelasticity characterization using nanoindentation, *Microelectron. Reliab.* 52 (3) (2012) 541–558.
- [8] W. Smit, H. De Vries, Rheological models containing fractional derivatives, *Rheol. Acta* 9 (4) (1970) 525–534.
- [9] N. Bhangale, K.B. Kachhia, J.F. Gómez-Aguilar, Fractional viscoelastic models with Caputo generalized fractional derivative, *Math. Methods Appl. Sci.* (2021) 1–12.
- [10] F. Mainardi, G. Spada, Creep, relaxation and viscosity properties for basic fractional models in rheology, *Eur. Phys. J. Spec. Top.* 193 (1) (2011) 133–160.
- [11] E. Stanisauskis, S. Mashayekhi, B. Pahari, et al., Fractional and fractal order effects in soft elastomers: strain rate and temperature dependent nonlinear mechanics, *Mech. Mater.* 172 (2022) 104390.
- [12] S. Mashayekhi, M.Y. Hussaini, W. Oates, A physical interpretation of fractional viscoelasticity based on the fractal structure of media: theory and experimental validation, *J. Mech. Phys. Solids* 128 (2019) 137–150.
- [13] S.G. Samko, B. Ross, Integration and differentiation to a variable fractional order, *Integr. Transforms Spec. Funct.* 1 (4) (1993) 277–300.
- [14] D. Ingman, J. Suzdalnitsky, M. Zeifman, Constitutive dynamic-order model for nonlinear contact phenomena, *J. Appl. Mech.* 67 (2) (2000) 383–390.
- [15] C.F. Lorenzo, T.T. Hartley, Variable order and distributed order fractional operators, *Nonlinear Dyn.* 29 (1) (2002) 57–98.
- [16] C.F.M. Coimbra, Mechanics with variable-order differential operators, *Ann. Phys.* 12 (11–12) (2003) 692–703.
- [17] D. Ingman, J. Suzdalnitsky, Application of differential operator with servo-order function in model of viscoelastic deformation process, *J. Eng. Mech.* 131 (7) (2005) 763–767.
- [18] L.E.S. Ramirez, C.F.M. Coimbra, A variable order constitutive relation for viscoelasticity, *Ann. Phys.* 519 (7–8) (2007) 543–552.
- [19] A. Burlon, G. Alotta, M. Di Paola, et al., An original perspective on variable-order fractional operators for viscoelastic materials, *Meccanica* 56 (4) (2021) 769–784.
- [20] X. Liu, D. Li, C. Han, et al., A Caputo variable-order fractional damage creep model for sandstone considering effect of relaxation time, *Acta Geotech.* 17 (1) (2022) 153–167.
- [21] M. Di Paola, G. Alotta, A. Burlon, et al., A novel approach to nonlinear variable-order fractional viscoelasticity, *Philos. Trans. R. Soc. A* 378 (2172) (2020) 20190296.
- [22] L.E.S. Ramirez, C.F.M. Coimbra, On the selection and meaning of variable order operators for dynamic modeling, *Int. J. Differ. Equ.* (2010) 846107.
- [23] M.C. Boyce, S. Socrates, P.G. Llana, Constitutive model for the finite deformation stress-strain behavior of poly (ethylene terephthalate) above the glass transition, *Polymer* 41 (6) (2000) 2183–2201.
- [24] H. Khajehsaeid, J. Arghavani, R. Naghdabadi, et al., A visco-hyperelastic constitutive model for rubber-like materials: a rate-dependent relaxation time scheme, *Int. J. Eng. Sci.* 79 (2014) 44–58.
- [25] A.D. Drozdov, A.L.H. Leijre, J.C. Christiansen, Viscoelasticity, viscoplasticity, and creep failure of polypropylene/clay nanocomposites, *Compos. Sci. Technol.* 69 (15–16) (2009) 2596–2603.
- [26] D. Mathiesen, D. Vogtmann, R.B. Dupaix, Characterization and constitutive modeling of stress-relaxation behavior of poly (methyl methacrylate)(PMMA) across the glass transition temperature, *Mech. Mater.* 71 (2014) 74–84.