Numerical modeling and experimental investigations of creep behaviour of polycarbonate

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Abstract. Polycarbonate is a typical glassy polymer widely used in engineering applications due to its high strength, light weight and recyclability. Its creep behaviour, characterized by time-dependent deformation under varying loads, is critical to predict long-term performance, especially at high temperatures or for long-term application. Classical viscoelastic models, such as Maxwell, Merchant and Burgers, are commonly employed but often difficult to accurately capture the complex nonlinear viscoelastic response of polymers. Fractional-order models are history-dependent and able to capture the entire viscoelastic behaviours. In this study, the creep behaviour of polymers is analyzed by comparing the performance of integer-order and fractional-order models. Experimental creep data are used to identify the parameters of the models and evaluate their prediction accuracy. The results show that the fractional-order model provides significantly enhanced accuracy in describing the creep behaviour under different stresses, demonstrating its superiority in modeling the viscoelastic properties of polymers.

1. Introduction

Polymeric materials are extensively utilized across various engineering fields. Polycarbonate (PC) is widely employed as a structural material in the automotive and aerospace industries. They have exceptional impact strength, excellent processability, transparency, and high heat distortion temperature [1]. The time-dependent deformation of most engineering polymers arises from its intrinsic viscoelastic properties, which are strongly influenced by the factors including temperature, humidity, physical aging, pressure, solvent concentration, and applied strain or stress levels [2]. These influences make the mechanisms of time-dependent deformation highly complex. It is a challenge to propose an accurate creep deformation model. The modeling of the time-dependent deformation behaviour of structural polymer materials remains crucial for ensuring their reliable performance in long-term applications.

Glassy polymers primarily exhibit recoverable deformation (elastic and viscoelastic) when subjected to stresses below their glass transition temperature. Under long-term constant loading at stress levels below the yield stress, they experience creep, a time-dependent deformation that can occur even before reaching the yield stress, particularly at elevated temperatures. When the applied stress exceeds the yield stress, the polymer undergoes plastic deformation, resulting in permanent strain [3]. The Maxwell model consists of a Hookean spring and a Newtonian dashpot connected in series. It can be used to predict the deformation behaviour of polymers, especially the linear viscoelastic behaviour [4]. The relationship between the stress and time-dependent deformation is linear because the nonlinear viscoelastic properties in the early stage of creep are infinitesimal and can be ignored. In fact, when the testing time is long or the applied stress is large, polymers usually exhibit nonlinear viscoelasticity. The use of additional elastic or viscous elements can better capture the time-dependent behaviour of the material and provide a theoretical basis for predicting complex behaviours such as creep behaviour. For example, the Merchant model improves the Maxwell model by capturing the immediate and delayed deformation stages. While the Burgers model, which combines the Maxwell and Kelvin-Voigt elements, is more suitable for long-term creep prediction. Gebrehiwot and Espinosa-Leal [5] applied Burgers and the generalized Maxwell models to predict the creep behaviour of polypropylene. The results showed that finite element analysis using the Burgers viscoelastic model provided better predictions, with the calculation errors below 10%.

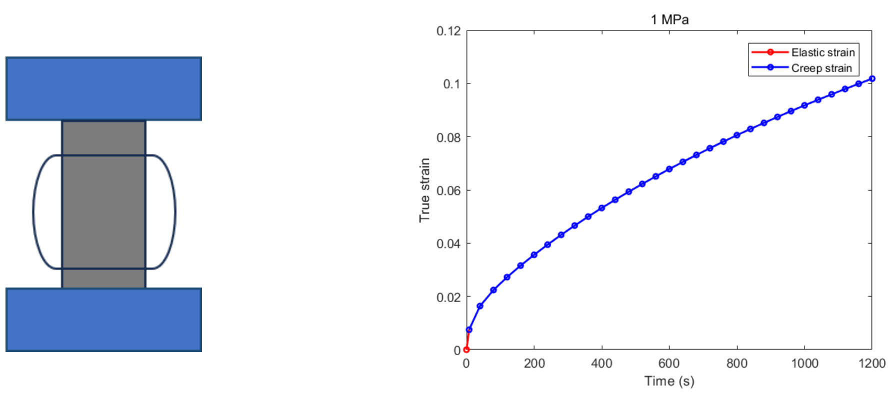
In recent decades, the fractional operator has emerged as a powerful tool for modelling viscoelastic behaviour, particularly in describing temperature and strain dependence or loading history [6]. Recent studies have shown that rheological models based on fractional calculus can fit experimental data of the viscoelastic materials using fewer elements than that required for the linear springs and dashpots used in classical models [7]. Xu and Jiang [8] investigated the three sets of creep experimental data corresponding to HDPE, PEEK and rock. The fractional viscoelastic model was applied to describe their creep behaviours. The results proved that the fractional-order model can well capture the short-term and long-term creep behaviours of viscoelastic materials.

The primary objective of this study is to numerically simulate the nonlinear creep deformation of PC. The investigation begins with an overview of the material properties and the introduction of both classical and fractional viscoelastic models. Subsequently, creep tests are performed at three different stresses to investigate the strain variation with respect to the applied stress. Finally, the predictions of the fractional-order models are compared with those generated using the classical viscoelastic models, which requires more components to accurately fit the same experimental data.

2. Materials and methods

2.1 Specimen geometry and experimental results

The cylindrical specimens were fabricated using dried PC pellets via injection molding following the standard ASTM D3935-21 [9]. The specimen geometry is illustrated in Fig. 1(a). The specimen is with a length of 18 mm and a diameter of 10 mm. According to standard ISO 604 [10], the uniaxial compression tests were performed on an Instron machine. Based on experimental data in [2], the yield stress of PC at 140 ℃ is approximately 20 MPa. In this study, the stresses of 1, 5, and 10 MPa were applied over a duration of 1200 seconds to investigate the evolution of true strain in function of time. The time-dependent true strain response under 1 MPa is presented in Fig. 1(b).



(a) (b)

Fig.1. (a) Specimen geometry of PC; (b) experimental curves of true strain-time under 1 MPa.

The true strain-time curve depicted in Fig. 1(b) highlights the evolution of the total time-dependent strain of PC respect to time. PC initially exhibits an instantaneous elastic strain, marked in red and follows the creep strain, marked in blue. These observations are crucial for understanding the long-term viscoelastic behaviours of the polymer at elevated temperature.

The evolutions of the true creep strain respect to time under 1, 5, and 10 MPa are presented in Fig.2. Higher stress applied lead to greater strain amplitudes and higher creep rates, demonstrating a strong stress dependence of behaviour of PC. The nonlinearity of the true strain-time curve becomes more pronounced with the increasing stress.

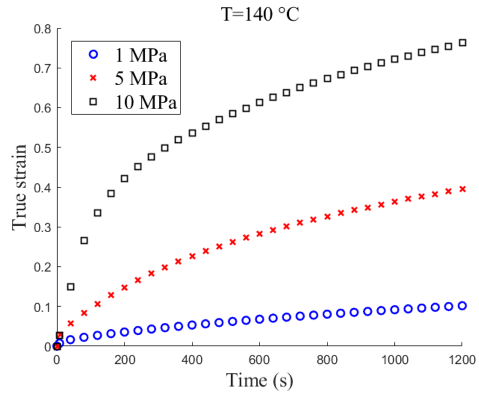


Fig.2. Evolution of the true strain of PC respect to time under different imposed stresses.

2.2. Viscoelastic model

2.2.1 Classical integer-order model

The Maxwell model is a viscoelastic model to describe the mechanical behaviours of materials, combining a spring and a dashpot connected together in series, as shown in Fig. 3 (a). The constitutive behaviour law is given as [11]

$$\varepsilon(t) = \frac{\sigma}{E} + \frac{\sigma}{\eta} t, \tag{1}$$

where σ is a constant stress, $\varepsilon(t)$ is strain, E is elastic modulus, η is viscosity.

Adding other components or mechanisms to a constitutive model can significantly enhance its ability to accurately simulate the creep behaviour of polymer. These models combine elements such as elastic spring, viscous dashpot, and Kelvin-Voigt element to represent instantaneous elastic deformation and time-dependent viscoelastic behaviour. By integrating these frameworks, constitutive models such as Merchant model (Fig. 3(b)) and Burgers model (Fig. 3(c)), can effectively capture complex creep phenomena and give a more comprehensive understanding of the mechanical behaviour of polymers under various loading conditions.

The creep constitutive equation of the Merchant model is [12]

$$\varepsilon(t) = \frac{\sigma}{E_1} + \frac{\sigma}{E_2}\left(1 - e^{-\frac{E_2}{\eta}t}\right). \tag{2}$$

where E_1 and E_2 in the spring and Kelvin-Voigt elements, respectively.

The creep constitutive equation of the Burgers model is [4]

$$\varepsilon(t) = \frac{\sigma}{E_1} + \frac{\sigma}{\eta_2} t + \frac{\sigma}{E_2}\left(1 - e^{-\frac{E_2}{\eta_1}t}\right). \tag{3}$$

where E_1 and E_2 are elastic modulus in the Maxwell and Kelvin-Voigt elements, respectively.

2.2.2 Fractional-order model

The Abel dashpot extends the Newtonian dashpot by incorporating fractional calculus, allowing to modelling the viscoelastic behaviour that encompasses both elastic and viscous characteristics. In contrast, the Newtonian dashpot represents purely viscous behaviour, with stress directly proportional to the strain rate. The use of fractional derivatives in the Abel dashpot enables a more accurate representation of complex time-dependent behaviours in materials, such as polymers and biological tissues, which exhibit both elasticity and viscosity [13]. The constitutive relation of the Abel dashpot is given by [14]

$$\sigma(t) = \eta^{\alpha} D^{\alpha} \varepsilon(t), 0 < \alpha < 1, \tag{4}$$

where $\eta^{\alpha}$is the viscosity coefficient in fractional-order model, α is fractional-order and $D^{\alpha}\varepsilon(t)$ represents the fractional derivative of strain $\varepsilon(t)$.

In Eq. (4), the fractional derivative operator is expressed by using the Caputo definition, which is widely applied in viscoelasticity due to its efficiency for initial value problems. Its definition is provided in [15]

$$D^{\alpha}\varepsilon(t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t (t-\tau)^{-\alpha} \frac{d\varepsilon(\tau)}{d\tau} d\tau, 0 < \alpha < 1, \tag{5}$$

where $\Gamma(\chi) = \int_0^{\chi} e^{-s} s^{\chi-1} ds$ is the Gamma function.

The Abel dashpot in Eq. (4) can be used to describe both the Newtonian dashpot in a special case of $\alpha = 1$, representing the ideal fluid, and a spring in the special case of $\alpha = 0$, representing the ideal solid. The Abel dashpot exhibits a combination of elastic spring behaviour and Newtonian viscous damping characteristics.

By replacing the Newtonian dashpot in Maxwell model with the Abel dashpot, the new constitutive relation of Maxwell creep model (as shown in Fig.3 (d)) based on fractional derivative is given by [16]

$$\varepsilon(t) = \frac{\sigma}{E} + \frac{\sigma}{\eta^{\alpha}} \frac{t^{\alpha}}{\Gamma(1+\alpha)}, \tag{6}$$

Extending the fractional Maxwell model to the fractional Merchant model (as shown in Fig. 3(e)) can effectively enhance its ability to describe the creep and nonlinear behaviour of polymers. By incorporating additional fractional elements, the model can capture the complex interaction of elastic, viscous, and fractional viscoelastic effects over time. The constitutive behaviour law described by the fractional Merchant model is [12]

$$\varepsilon(t) = \frac{\sigma}{E_1} + \frac{\sigma}{\eta} \sum_{k=0}^{\infty} \frac{(-1)^k}{\Gamma(\alpha k + \alpha + 1)} \left(\frac{\eta}{E_2} t^{\alpha}\right)^{k+1}. \tag{7}$$

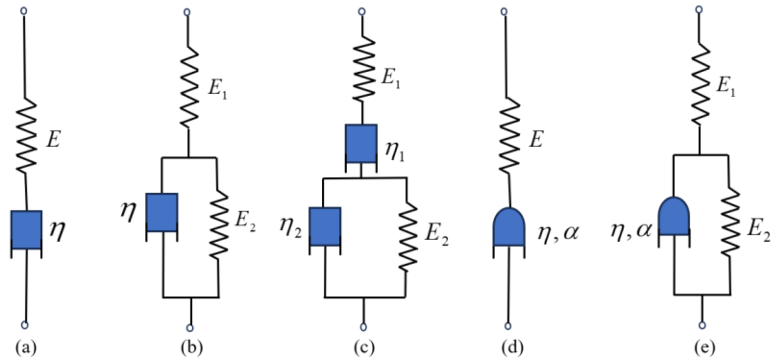


Fig.3. Representation of the viscoelastic models: (a) Maxwell model; (b) Merchant model; (c) Burgers model; (d) fractional Maxwell model; (e) fractional Merchant model.

3. Results and discussion

3.1. The sensitivity of the fractional-order in the fractional Merchant model

The objective of this study is to investigate the sensitivity of the fractional-order α on the description of viscoelastic behaviour of materials. Fig. 4 shows the true strain-time curves with different values of the α under a stress of 10 MPa. As α increases, the true strain increases faster. When α is 0, the fractional Merchant model can be considered as a linear elastic model. When α is 1, the fractional Merchant model is a Merchant model. When $0 < \alpha < 1$, the true strain of the model increases slowly with time, neither remaining unchanged like the linear elastic model nor increasing rapidly like the Merchant model. The above analysis shows that the fractional merchant model is more applicable and can well reflect the nonlinear viscoelastic behaviours.

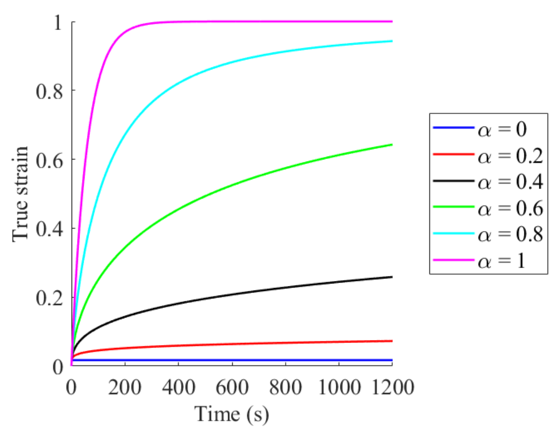
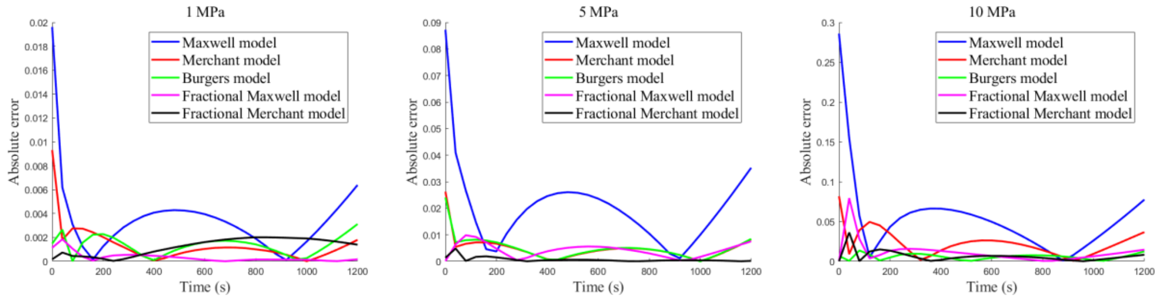


Fig.4. Evolution of true strain vs. time of PC described by fractional Merchant model with different α values.

3.2 Comparison with different integer-order and fractional-order models

In this study, the integer-order models such as the Maxwell, Merchant and Burgers models and the fractional-order models such as fractional Maxwell and fractional Merchant models were employed to characterize the creep behaviour of PC. The fitting effects of two fractional-order models and three integer-order models on true strain-time experimental data are compared under the same temperature and pressure condition. Fig. 4 presents the absolute errors between the experimental data and the numerical models for different stresses of 1, 5, and 10 MPa. The Maxwell model exhibits the largest absolute error, as it can only describe linear elastic behaviour.

Fractional-order models demonstrate higher accuracy and superior predictive capability compared to integer-order models.



(a) (b) (c)

Fig.5. Absolute errors of experimental results compared with numerical results obtained by various integer-order and fractional-order models under different stresses.

3.3 The fitting parameters and error analysis

The identified values of the parameters of different integer-order and fractional-order models for the creep behaviour of PC are presented in Table 1. The model performance was evaluated comprehensively by using two indicators: the root mean square error (RMSE) to assess prediction accuracy for individual data points and the coefficient of determination (R^2) to evaluate the model's ability to capture overall data trends. A lower RMSE indicates higher accuracy in predicting individual values, while a higher R^2 (closer to 1) reflects a better overall fit.

Table 1: Fitting parameters for creep models.

Maxwell model						
σ (MPa)	E (MPa)	η (MPa · s^α)			R^2	RMSE
1	51	13550			0.9672	0.0049
5	57	17475			0.9413	0.0256
10	35	21658			0.8314	0.0744
Merchant model						
σ (MPa)	E_1 (MPa)	E_2 (MPa)	η (MPa · s^α)		R^2	RMSE
1	107	8	7407		0.9939	0.0021
5	191	12	7433		0.9962	0.0065
10	122	15	4894		0.9766	0.0277
Burgers model						
σ (MPa)	E_1 (MPa)	E_2 (MPa)	η_1 (MPa · s^α)	η_2 (MPa · s^α)	R^2	RMSE
1	674	38	15522	3309	0.9967	0.0015
5	208	12	999540	7354	0.9963	0.0065
10	1478	23	36440	2566	0.9905	0.0177
Fractional Maxwell model						
σ (MPa)	E (MPa)	η (MP · s^α)	α		R^2	RMSE
1	881	746	0.59		0.9996	0.0005
5	51673	646	0.54		0.9976	0.0048
10	87074	185	0.36		0.9975	0.0091
Fractional Merchant model						
σ (MPa)	E_1 (MPa)	E_2 (MPa)	η (MPa · s^α)	α	R^2	RMSE
1	5814	72	71	0.0065	0.9997	0.0005

5	3683	1407	6	0.71	0.9999	0.0011
10	467168	498	9	0.61	0.9986	0.0067

Based on the value of these two indicators, the integer-order models exhibit improved fitting accuracy for nonlinear viscoelastic behaviour with an increased number of springs and dashpots. The fractional-order models provide a more accurate fitting results compared to the integer-order models. The fractional Merchant model achieves a smaller fitting error than the fractional Maxwell model, demonstrating its superior ability to characterize the experimental data and effectively capture the creep behaviour under various stress levels.

4. Conclusion

This study investigates the creep behaviour of PC under uniaxial compression at different stress levels of 1, 5 and 10 MPa, comparing the performance of the integer viscoelastic models (Maxwell, Merchant, and Burgers models) with their fractional counterparts (fractional Maxwell and fractional Merchant models). Experimental results revealed significant nonlinearity and stress-dependent behaviour, particularly under higher stress levels. The prediction with these constitutive behaviour laws on the creep data was compared to investigate their fitting efficiency and accuracy.

The fractional-order models, incorporating fractional derivatives, demonstrated superior accuracy in describing the true strain-time relationship across all stress levels. These models effectively capture the nonlinear and time-dependent characteristics of PC's creep behaviour, outperforming integer-order models in terms of fitting accuracy and physical relevance. The fractional Maxwell and Merchant models, in particular, showed excellent agreement with the experimental data, highlighting their potential as robust tools for modeling complex viscoelastic behaviours.

These findings emphasize the advantages of the fractional viscoelastic models in accurately predicting the long-term viscoelastic behaviour of polymers, especially under various stress conditions. The study provides valuable insights for developing more effective modeling approaches for engineering applications involving polymers, paving the way for improved material design and analysis.

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