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Determination of shear creep compliance of linear viscoelastic-plastic solids by instrumented indentation

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ABSTRACT

This paper presents a convenient method which is referred to as “Revised Step-Hold” method for determination of shear creep compliance of linear viscoelastic-plastic solids using instrumented indentation. Its main idea is using the revised load-depth curve which does not contain plastic deformation to determine the shear creep compliance instead of using the measured load-depth curve. The revised load-depth curve can be obtained through a three-step procedure. Applications are illustrated on two typical viscoelastic-plastic solids, namely PMMA and UPVC. The shear creep compliance of PMMA and UPVC determined by the revised step-hold method has satisfactorily good agreement with the corresponding uniaxial tensile result. Therefore, it demonstrates that the revised step-hold method is a reliable method for determination of shear creep compliance of linear viscoelastic-plastic solids.

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1. Introduction

Instrumented indentation is an efficient and convenient tool for probing mechanical properties of viscoelastic solids, such as polymers and biomaterials. For these materials, one of the most important mechanical properties is creep, which gives the time-dependent deformation. Due to the effect of creep, the unloading part of the load-depth curve exhibits a nose when the unloading rate is low [dotted curve in Fig. 1(b)], and the initial unloading stiffness, $S = (dF/dh)_u$ is negative. In this case, the widely used Oliver-Pharr method [1] is not suitable for extracting elastic modulus [2–6]. Fortunately, Tang et al. [4] and Cheng et al. [6] have put forward methods for accurately extracting elastic modulus of viscoelastic solids by removing the effects of creep.

In order to predict time-dependent deformation of viscoelastic solids, the creep compliance should be known. Based on the viscoelastic contact solution derived by Lee

and Radok [7], Hunter [8], Graham [9] and Ting [10], a number of methods for determination of shear creep compliance have been proposed [11–18]. These methods require that the deformation of materials during testing is in the regime of linear viscoelasticity. However, it is difficult to meet this requirement for viscoelastic-plastic solids such as polymethylmethacrylate (PMMA) and unplasticized polyvinyl chloride (UPVC), because the high stresses beneath the indenter tip can easily lead to these materials yielding. Lu et al. [11] and Tweedie et al. [15] pointed out that the response can be regarded as approximately linear viscoelastic when the indentation is shallow enough for plastic deformation to be negligible. However, it is difficult to identify the critical indentation load that does not cause significant plastic deformation. Menčík et al. [19,20] proposed a method to determine “apparent” shear creep compliance of linear viscoelastic-plastic solids according to the material models. Although the “apparent” shear creep compliance predicts the depth-time curve well, it contains the contribution of plastic deformation and, therefore, is not the true shear creep compliance. Seltzer et al. [21] found a linear relationship between the normalized

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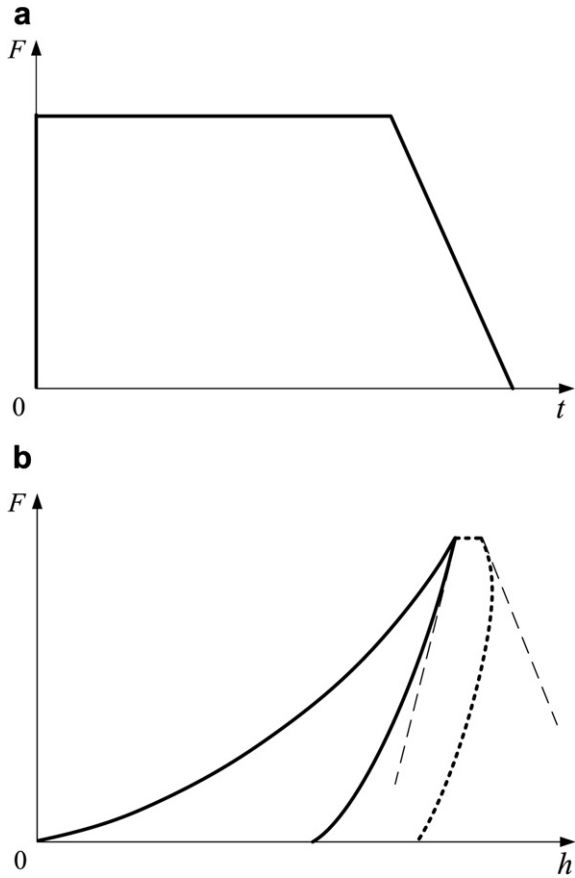


Fig. 1. Schematic illustrations of (a) step-hold-unload loading profile and (b) load-depth curves of indentation tests (dotted curve is the typical holding and unloading curve of viscoelastic solid).

“apparent” shear creep compliance J^{app}/J_0 and the plastic depth ratio h_0^p/h_0^e (J^{app} is “apparent” shear creep compliance, J_0 is instantaneous shear creep compliance, h_0^p and h_0^e are the plastic and elastic depth at the beginning of holding, respectively). According to this linear relationship, they obtained the shear creep compliance by linear extrapolation. As each linear extrapolation only determines the shear creep compliance at the given time, a series of extrapolations are needed to determine the shear creep compliance function. The procedure of Seltzer’s method is complicated. Thus, simple methods for determining shear creep compliance of linear viscoelastic-plastic solids are needed.

In the present work, a Berkovich indenter indenting linear viscoelastic-plastic solids with a step-hold-unload loading profile [Fig. 1(a)] is considered. It is assumed that elastic-plastic deformation is dominant and viscoelastic deformation can be neglected during the fast loading segment, and that there is only viscoelastic deformation during the holding segment. Therefore, plastic and viscoelastic deformations can be studied separately. A three-step procedure is used to remove the plastic deformation from the measured load-depth curve. Then, a revised load-depth curve which does not include plastic deformation is obtained, and the shear creep compliance can be accurately determined by using the revised load-depth curve.

2. Theoretical background

2.1. Formulae for extracting elastic modulus

The most often used method for extracting elastic modulus of elastic-plastic solids was established by Oliver and Pharr [1]. They put forward the formula for calculating contact depth

$$h_c = h_u - \varepsilon \frac{F_u}{S} \quad (1)$$

where h_u and F_u are the depth and load at the beginning of unloading, respectively; ε is a constant related to indenter shape ($\varepsilon = 0.72$ for a conical indenter and $\varepsilon = 0.75$ for a paraboloid of revolution); S is the contact stiffness at the beginning of unloading. The projected contact area A can be determined from the contact depth. For a perfect Berkovich indenter, $A = 24.5h_c^2$. The contact stiffness and projected contact area are then used to determine the reduced modulus E_r using the following equation

$$E_r = \frac{\sqrt{\pi}}{2\beta} \frac{S}{\sqrt{A}} \quad (2)$$

where β is a correction factor for indenter shape ($\beta = 1.034$ for Berkovich indenter and $\beta = 1.012$ for Vickers indenter [22,23]). Taking into account the effect of a non-rigid indenter, the reduced modulus is defined as

$$\frac{1}{E_r} = \frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i} \quad (3)$$

where E and ν are the elastic modulus and Poisson’s ratio of the sample, respectively, and E_i and ν_i are the elastic modulus and Poisson’s ratio of the indenter, respectively. For a diamond indenter, $E_i = 1141$ GPa and $\nu_i = 0.07$. Hence, the elastic modulus E of elastic-plastic solids can be determined once E_r and ν are known.

However, for viscoelastic-plastic solids, the deformation is time-dependent and more complex. Due to the effect of creep, a platform appears during holding, and the unloading part of load-depth curve is more convex [dotted curve in Fig. 1(b)] than that for elastic-plastic materials. This leads to overestimating the contact stiffness. If the unloading rate is sufficiently low, a nose may be evident and the contact stiffness is negative. Using the measured contact stiffness S to determine the contact depth and elastic modulus could introduce considerable error. Feng et al. [2] have proposed that the true (elastic) contact stiffness S_e can be calculated from the measured contact stiffness S using the following equation

$$\frac{1}{S_e} = \frac{1}{S} + \frac{\dot{h}_h}{|\dot{F}_u|} \quad (4)$$

where \dot{h}_h is the creep rate (dh/dt) at the end of holding, \dot{F}_u is the unloading rate (dF/dt) at the beginning of unloading. Replacing S in Eqs. (1) and (2) with S_e , the contact depth and the reduced modulus can be accurately determined.

2.2. Formulae for determining shear creep compliance

Considering a smooth rigid indenter pressed against a linear viscoelastic half-space, and assuming the Poisson’s

ratio is time-independent, Lee and Radok [7], Hunter [8], Graham [9] and Ting [10] derived the load-depth relationships for indentations in linear viscoelastic solids, which can be briefly written as

$$h^{(n+1)/n}(t) = \frac{1-\nu}{4C_n} \int_0^t J(t-\tau) \frac{dF(\tau)}{d\tau} d\tau \quad (5)$$

where $J(t)$ is shear creep compliance; ν is the time-independent Poisson's ratio; C_n is a constant related to indenter shape, $n = 1$, $C_1 = \tan \alpha/\pi$ for a conical indenter, and α is the included half-angle; $n = 2$, $C_2 = 2\sqrt{R}/3$ for a spherical indenter, where R is the radius of the spherical indenter. They also pointed out that Eq. (5) is valid so long as the contact area does not decrease with time. Hence, Eq. (5) is applicable to the loading and holding segments of an indentation.

The use of a specific loading profile can simplify Eq. (5) and derive the explicit expression for shear creep compliance. For example, a step-hold loading profile is adopted in the load-controlled instrumented indentation. The indentation load is expressed as

$$F(t) = F_0 H(t) \quad (6)$$

where F_0 is the maximum load and $H(t)$ is the Heaviside unit step function. Substituting Eq. (6) into Eq. (5), the explicit expression for shear creep compliance is easily obtained

$$J(t) = \frac{4 \tan \alpha}{\pi(1-\nu)F_0} h^2(t) \quad (7)$$

The shear creep compliance can then be calculated once the variation of depth during the holding segment is known.

3. Methods

Eq. (7) is derived from Eq. (5) which is obtained by assuming the sample is linear viscoelastic. If Eq. (7) is used to calculate the shear creep compliance of linear viscoelastic-plastic solids, considerable error will be introduced when significant plastic deformation occurs during indentation tests. The approach that Tang et al. [4] used to accurately extract elastic modulus by replacing the measured contact stiffness with the true (elastic) contact stiffness suggests a solution: we can use the revised load-depth curve which does not include plastic deformation

to accurately determine the shear creep compliance of linear viscoelastic-plastic solids instead of using the measured load-depth curve.

A step-hold-unload loading profile as shown in Fig. 1(a) is used in indentation tests. It is assumed that (1) elastic-plastic deformation is dominant and viscoelastic deformation is negligible during the fast loading; (2) in the holding segment, only viscoelastic process occurs; (3) the total indentation depth is comprised of the elastic part h_e , the plastic part h_p and the viscoelastic part h_v . Based on these assumptions, the plastic deformation can be removed from the measured load-depth curve through the following three steps:

- (i) Extract the reduced modulus E_r from the measured load-depth curve [Fig. 2(a)] by replacing S in Eqs. (1) and (2) with S_e .
- (ii) Simulate a pure elastic load-depth curve for loading segment [dotted curve in Fig. 2(b)] using Sneddon's solution revised by Hay et al. [24]. The revised solution is expressed as

$$h_e(t) = \sqrt{\frac{\pi}{2\gamma E_r \tan \alpha} F(t)} \quad (8)$$

where $F(t)$ is the load for loading segment, γ is a correction factor that depends on the included half-angle α and Poisson's ratio ν

$$\gamma = \pi \frac{\frac{\pi}{4} + 0.15483073 \frac{1-2\nu}{4(1-\nu)} \cot \alpha}{\left[\frac{\pi}{2} - 0.83119312 \frac{1-2\nu}{4(1-\nu)} \cot \alpha \right]^2} \quad (9)$$

Since the elastic-plastic deformation is dominant and the viscoelastic deformation is negligible during fast loading, the measured depth of loading segment only contains the elastic part h_e and the plastic part h_p . The plastic depth is simply the difference between the measured depth and simulated (pure elastic) depth. At the end of loading, a maximum plastic depth h_0^p exists [Fig. 2(b)]

$$h_0^p = h_0^{ep} - h_0^e \quad (10)$$

where h_0^{ep} and h_0^e are the measured depth and the simulated depth at the end of loading, respectively. Because only a viscoelastic process occurs during holding and plastic deformation is irreversible during unloading, the plastic deformation neither increases nor decreases during holding and unloading segments. Thus, the measured

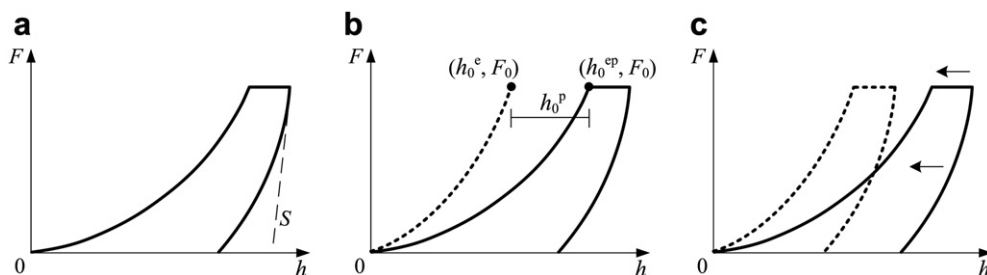


Fig. 2. Schematic illustrations for obtaining the revised load-depth curve which does not contain plastic deformation. Solid curve is the measured load-depth curve and dotted curve is the revised load-depth curve.

depth of holding and unloading segments contains a constant plastic depth h_0^p .

- (iii) Remove the plastic depth from the measured curve of holding and unloading segments using the following expression

$$h_{re}(t) = h(t) - h_0^p \quad (11)$$

and integrate it with the simulated (pure elastic) curve. Then, a revised load-depth curve which does not include plastic deformation [dotted curve in Fig. 2(c)] is obtained

$$h_{re}(t) = \begin{cases} h_e(t) & \text{for loading} \\ h(t) - h_0^p & \text{for holding unloading} \end{cases} \quad (12)$$

Replacing the measured depth $h(t)$ in Eq. (7) with the revised depth $h_{re}(t)$, it comes to

$$J(t) = \frac{4 \tan \alpha}{\pi(1-\nu)F_0} h_{re}^2(t) \quad (13)$$

The shear creep compliance of linear viscoelastic-plastic solids can then be accurately determined using Eq. (13). This method is referred to as the “Revised Step-Hold” method in order to distinguish it from the prevalent step-hold method.

4. Experiments

Both indentation creep tests and uniaxial tensile creep (conventional) tests were performed. The materials used were PMMA and UPVC (Anheda Plastic Products Co., Ltd., Suzhou, China). For uniaxial tensile creep tests, the PMMA and UPVC plates were processed into dumbbell-shaped specimens according to ISO 527-2. For indentation creep tests, these plates were processed into small specimens measuring 20 mm × 20 mm × 4 mm. PMMA has a glass transition temperature of 105 °C, while the glass transition temperature of UPVC is 87 °C. All specimens were annealed at 120 °C (for PMMA) and 102 °C (for UPVC) for 2.5 h in air and then cooled slowly to room temperature by switching off the power to the oven.

4.1. Indentation creep tests

The indentation creep tests were performed at room temperature (23 °C) using a MTS Nano Indenter XP system

(MTS Nano Instruments, Oak Ridge, TN) with a Berkovich indenter, which can be modeled as an equivalent cone with a included half-angle of 70.3°. An approximate step-hold-unload loading profile was adopted. The load was increased quickly to the maximum in 2 s, held at the maximum load for 300 s and finally decreased linearly to zero in 50 s. The maximum load was 18 mN for PMMA and 13 mN for UPVC. Each indentation creep test was repeated 5 times.

4.2. Uniaxial tensile creep tests

An Instron 5848 MicroTester (Instron, Canton, MA) was used for the uniaxial tensile creep tests. The testing temperature was the same as that for the indentation creep tests. An approximate step-hold loading profile was adopted. The load was increased quickly to the maximum in 2 s and then held at the maximum load for 300 s. Since the yield stresses of PMMA and UPVC are about 50 MPa and 45 MPa, respectively, the maximum stress for holding was set to 35 MPa to avoid plastic deformation. Each uniaxial tensile creep test was repeated 5 times.

5. Results and discussion

5.1. Indentation creep data

The revised load-depth curve that does not include plastic deformation can be obtained through the three step procedure proposed in Section 3. Both the revised and the measured load-depth curves of PMMA and UPVC are plotted in Fig. 3. The revised and measured load-depth curves of PMMA are close to each other. This demonstrates that the plastic deformation is not significant. Conversely, the remarkable difference between the revised and measured load-depth curves of UPVC indicates that significant plastic deformation occurs during the experiments.

5.2. Shear creep compliance

Both the prevalent step-hold method [Eq. (7)] and the revised step-hold method [Eq. (13)] were used to determine the shear creep compliance of PMMA and UPVC. For the prevalent step-hold method, the measured load-depth data were used; and for the revised step-hold method, the

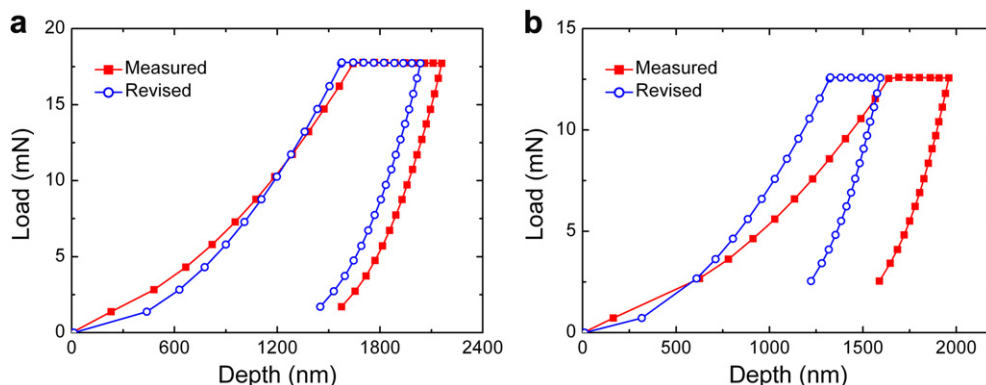


Fig. 3. Comparison between the revised load-depth curves and the measured load-depth curves of (a) PMMA and (b) UPVC.

revised load-depth data were used. The shear creep compliance determined by uniaxial tensile creep (conventional) test is regarded as the reference value, which can be calculated by

$$J(t) = 2(1 + \nu) \frac{\varepsilon(t)}{\sigma_0} \quad (14)$$

where σ_0 is the stress for holding, $\varepsilon(t)$ is the strain at time t , and ν is the time-independent Poisson's ratio.

The indentation results were compared with the reference values to verify the reliability of the revised step-hold method. From Fig. 4(a) and (b), we find that the shear creep compliance of PMMA determined by the prevalent step-hold method is about 11% greater than the reference value, and that determined by the revised step-hold method is about 5% less than the reference value. These results determined by both methods are close to the reference value mainly because the plastic deformation of PMMA is not significant during testing. It demonstrates that when plastic deformation is not significant, both the prevalent step-hold method and the revised step-hold method can be used to determine shear creep compliance. However, the result determined by the revised step-hold method is more accurate.

From Fig. 4(c) and (d), it is obviously that the shear creep compliance of UPVC determined by the prevalent step-hold method is much greater (about 65%) than the reference value, and the result determined by the revised step-hold

method has satisfactorily good agreement with the reference value with a relative error of about 5%. In the cases of significant plastic deformation, the prevalent step-hold method fails to determine shear creep compliance accurately. To sum up, the prevalent step-hold method can approximately determine shear creep compliance when the plastic deformation is not significant, but fails when the plastic deformation is significant. Whether or not plastic deformation is significant, the revised step-hold method can be used to accurately determine shear creep compliance of linear viscoelastic-plastic solids.

5.3. Prediction of deformation

The holding stress σ_0 for uniaxial tensile creep tests was 35 MPa. The variation of strain $\varepsilon(t)$ during uniaxial tensile creep tests can be predicted by Eq. (14) once the shear creep compliance $J(t)$ is known. The strain-time curves predicted using the shear creep compliance determined by the revised step-hold method are plotted in Fig. (5). Comparison with the experimental curve shown in the same diagram indicates that the differences are about 5% for PMMA and UPVC. This consistently demonstrates that the shear creep compliance determined by the revised step-hold method can be used to predict the variation of deformation. In other words, the revised step-hold method is a reliable method for determination of the shear creep compliance of linear viscoelastic-plastic solids.

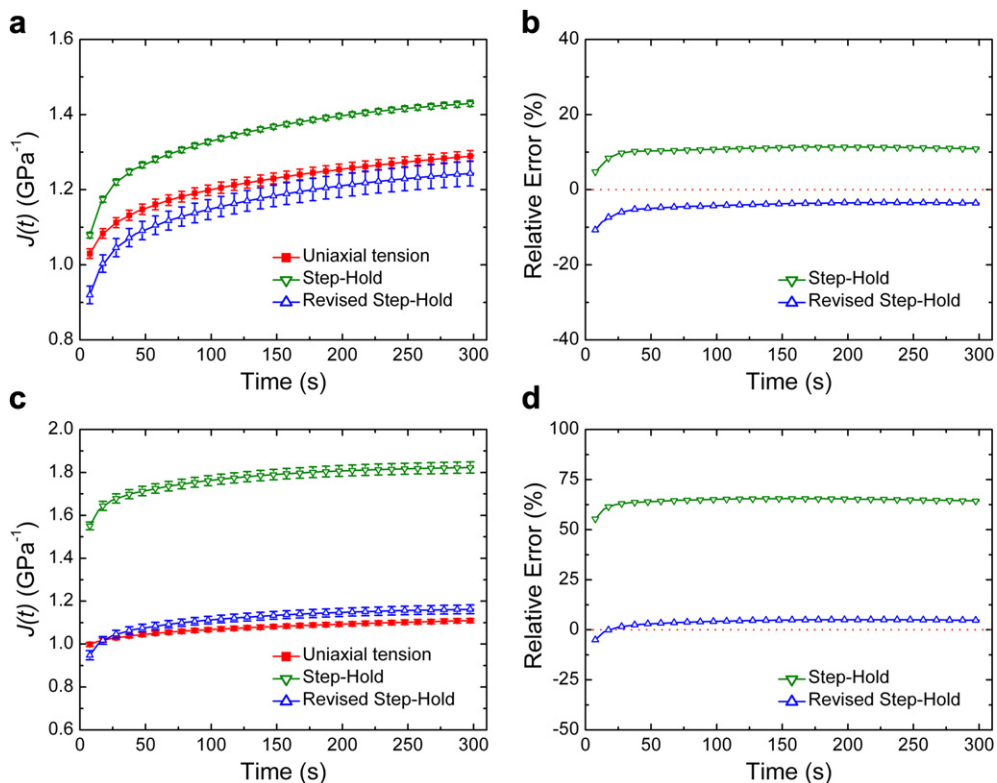


Fig. 4. The shear creep compliance determined by the revised step-hold method and the prevalent step-hold method is compared with the uniaxial tensile result which is the reference value. (a) Shear creep compliance and (b) corresponding relative errors of PMMA; (c) shear creep compliance and (d) corresponding relative errors of UPVC.

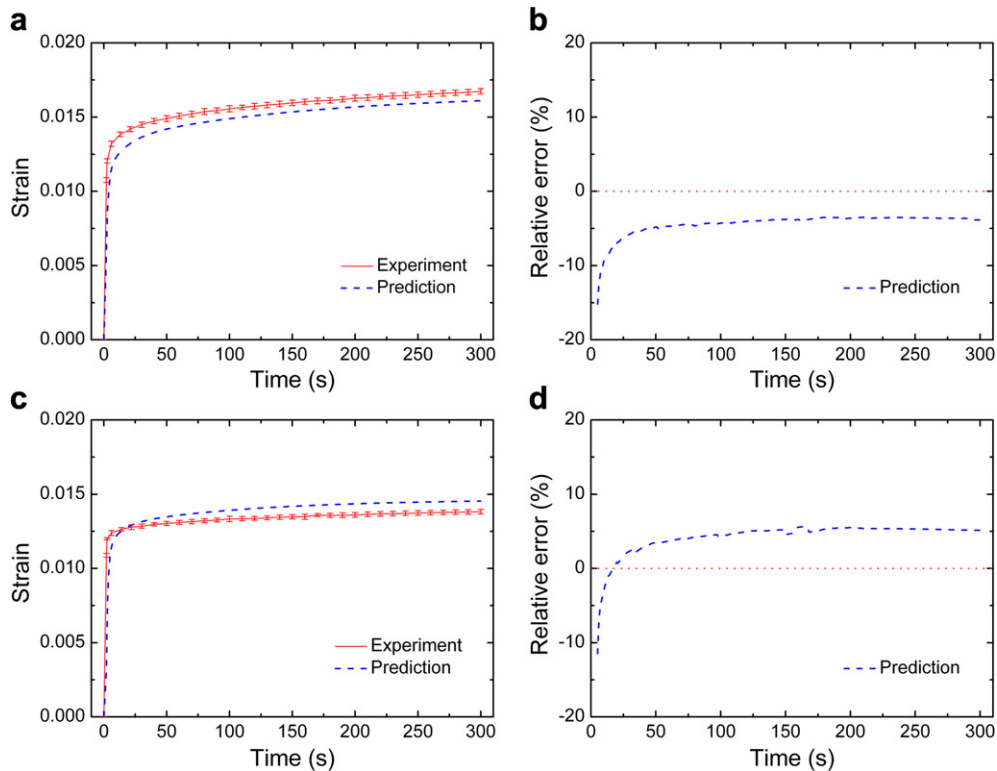


Fig. 5. The strain-time curves predicted using the shear creep compliance that determined by the revised step-hold method are compared with the experimental curves. (a) Strain-time curves and (b) corresponding relative errors of PMMA; (c) strain-time curves and (d) corresponding relative errors of UPVC.

6. Conclusions

This work provides a convenient method, the revised step-hold method, for determination of the shear creep compliance of linear viscoelastic-plastic solids. It adopts a step-hold-unload loading profile. Based on the assumptions that (1) the elastic-plastic deformation is dominant and the viscoelastic deformation is negligible during fast loading; (2) in the holding segment, only viscoelastic process occurs; (3) the total indentation depth is comprised of the elastic part, the plastic part and the viscoelastic part; the revised load-depth curve which does not contain plastic deformation is obtained through a three-step procedure. Then, the shear creep compliance is determined using the revised load-depth data. Two typical viscoelastic-plastic solids, namely PMMA and UPVC, were used in the experiments to verify this method. The shear creep compliance determined by the revised step-hold method has satisfactorily good agreement with the reference value. This demonstrates that the revised step-hold method can be used to determine the shear creep compliance of linear viscoelastic-plastic solids using instrumented indentation.

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