Comparison of the effects of hydrostatic pressure and the third invariant of deviatoric stress tensor on non-linear viscoelastic deformation in cellulose nitrate

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The behaviour of polymers is qualitatively known to be remarkably influenced by the hydrostatic pressure on the general plastic deformation, unlike most metals. However, as polymers behave differently in simple tension and compression, they may be influenced by the effect of the third invariant of deviatoric stress tensor as well as hydrostatic pressure. In this paper, a comparison of the effects of the hydrostatic pressure and the third invariant of deviatoric stress tensor on the non-linear viscoelastic deformation of cellulose nitrate are discussed quantitatively, following experiments of torsion of tubular specimens and simple tension of uniaxial specimens. As the result, the effect of the third invariant of deviatoric stress tensor on the non-linear viscoelastic deformation in cellulose nitrate was found to be much smaller than that of the hydrostatic pressure.

1. Introduction

The concept of non-linear viscoelastic behaviour of polymers has received widespread attention and application. The mechanical behaviour of polymers used as matrix for composite materials has received particular attention. The behaviour of polymers, unlike that of most metals, is qualitatively known to be significantly influenced by the hydrostatic pressure in creep deformation [1] or elastic-plastic deformation [2, 3]. However, as the polymers behave differently in simple tension and compression [4], the behaviour of polymers may also be influenced by the effect of the third invariant of deviatoric stress tensor. No work has investigated the comparison of the effects of the third invariant of deviatoric stress tensor and the hydrostatic pressure on the elastic-plastic or nonlinear viscoelastic polymers.

In this paper a quantitative comparison of the effects of the third invariant and the hydrostatic pressure on the non-linear viscoelastic deformation in cellulose nitrate is made following experiments on the tubular specimens subjected to either simple tension, or torsion or combined tension with hydrostatic stresses. It was found that the effect of the third invariant on the non-linear viscoelastic deformation in cellulose nitrate was less significant than that of the hydrostatic pressure.

2. Basic concepts of stress and strain tensors

2.1. Invariants of stress and strain tensors In a given element in a polymer, the stress is characterized by a symmetric stress tensor in rectangular cartesian co-ordinates with axes x, y, and zas follows [5]

$$\mathbf{T} = |\sigma_{ij}| = |\sigma_{ij} - \sigma\delta_{ij}| + |\sigma\delta_{ij}| \tag{1}$$

where σ_{ij} for $i = j(\sigma_i)$ and for $i \neq j(\tau_{ij})$ are the normal and shear components. $|\sigma_{ij}|$ and $|\sigma_{ij} - \sigma \delta_{ij}|$ are the stress and the deviatoric stress tensors, respectively. δ_{ij} and σ denote the Kronecker delta and the mean stress respectively.

The invariants of stress or deviatoric stress tensors and the second invariant of strain tensor are defined as follows [5]. The hydrostatic pressure, or the first invariant of stress tensor:

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$$A = \sigma_{ii} = \sigma_x + \sigma_y + \sigma_z. \tag{2}$$

The second invariant of deviatoric stress tensor (or the equivalent stress):

$$S = \{ [(\sigma_x - \sigma_y)^2 + (\sigma_y - \sigma_z)^2 + (\sigma_z - \sigma_x)^2 + 6(\tau_{xy}^2 + \tau_{yz}^2 + \tau_{xz}^2)]/2 \}^{1/2}.$$
 (3)

The third invariant of deviatoric stress tensor:

$$B = \{(\sigma_x - \sigma)(\sigma_y - \sigma)(\sigma_z - \sigma) - (\sigma_x - \sigma)\tau_{yz}^2 - (\sigma_y - \sigma)\tau_{xz}^2 - (\sigma_z - \sigma)\tau_{xy}^2 + 2\tau_{xy}\tau_{yz}\tau_{xz}\}^{1/3}.$$
 (4)

The second invariant of deviatoric strain tensor (or the equivalent strain):

$$E = \frac{1}{3} \{ 2 [(\epsilon_x - \epsilon_y)^2 + (\epsilon_y - \epsilon_z)^2 + (\epsilon_z - \epsilon_x)^2 + 6(\gamma_{xy}^2 + \gamma_{yz}^2 + \gamma_{xz}^2)] \}^{1/2},$$
 (5)

where ϵ_i and γ_{ij} represent the strains in the direction of axes and the shear strains.

2.2. Difference of the third invariant due to stress states

A graphical representation of the stress state in a given element is provided by Mohr's circle. Suppose that at this point the directions of the coordinate axes coincide with the principle directions, then Mohr's circle is expressed as Fig. 1, where σ_1 , σ_2 and σ_3 are principal stresses, σ_n and τ_n are normal and shear stresses. It is possible to express the interrelationship between the principle stresses by means of Lode's (or Nadai's) parameter ([6] pp. 15–17) for $\sigma_1 > \sigma_2 > \sigma_3$

$$\mu = 2 \frac{\sigma_2 - \sigma_3}{\sigma_1 - \sigma_3} - 1.$$
 (6)

This characterizes the position of the intermediate principal stress σ_2 in Mohr's circle and loses its meaning only in the case of hydrostatic pressure $\sigma_1 = \sigma_2 = \sigma_3$.

For the same values of μ , Mohr's circles are



Figure 1 Mohr's circle.

similar. It is evident that at a fixed value of μ the character of the stress is precisely defined independently of a multiplicative factor of the principal stresses and an additive hydrostatic pressure. In this sense, it is possible to regard the parameter μ as representing the shape of the stress tensor or deviatoric stress tensor. The parameter μ varies in the range -1 to +1; thus

for simple tension

$$(\sigma_1 > 0, \sigma_2 = \sigma_3 = 0), \mu = -1,$$

for simple compression

$$(\sigma_1 = \sigma_2 = 0, \sigma_3 < 0), \mu = +1,$$

for pure shear

$$(\sigma_1 > 0, \sigma_2 = 0, \sigma_3 = -\sigma_1), \mu = 0$$

Therefore, the parameter μ depends on the difference in stress states, but is independent of the hydrostatic pressure.

The parameter μ is a function of the second invariants S and the third invariant of deviatoric stress tensors B([6] pp. 10–11), and bears a simple relationship to the angle ω which is the parameter

$$\mu = \sqrt{3} \cot \left(\omega + \frac{1}{3}\pi\right),\tag{7}$$

where

$$-\cos 3\omega = \frac{27 B^3}{2 S^3}.$$
 (8)

The angle ω is sometimes called the angle of the nature of the stress state; thus

for simple tension,

$$\omega = \frac{1}{3}\pi,$$

for simple compression,

$$\omega = 0,$$

for pure shear,

$$\omega = \frac{1}{6}\pi$$

Accordingly, as the second invariant S is independent of the stress state, only the third invariant B depends on the nature of the stress state, or the difference of stress states. Since polymers generally behave differently in simple tension and simple compression, the behaviour should be influenced by the third invariant of deviatoric stress tensor B.

Figure 2 Stress states at the element of specimens.

Name of experiment	Exp.1	Exp. 2	Exp. 3	Exp.4
Schematic stress state	<u> ~ ()~</u> ,	C (C (C (C (C (C (C (C (C (C (▲ a ↓ a	
Stress tensor	a 0 0 0 0 0 0 0 0	0 <u>a</u> a <u>[</u> 3 0 0 0 0 0	a 0 0 0 0 0 0 0 0	$\begin{bmatrix} a - \frac{a}{3} & 0 & 0 \\ 0 & -\frac{a}{3} & 0 \\ 0 & 0 & -\frac{a}{3} \end{bmatrix}$
Deviatoric stress tensor	$\frac{2}{3}a 0 0$ $0 - \frac{a}{3} 0$ $0 0 - \frac{a}{3}$	$\begin{array}{c} 0 & \frac{a}{2} & 0 \\ \frac{\sqrt{3}}{2} & 0 & 0 \\ 0 & 0 & 0 \\ \end{array}$	$ \frac{2}{3}a 0 0 \\ 0 - \frac{a}{3} 0 \\ 0 0 - \frac{a}{3} $	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$
Value of A	a	0	a	0
Value of B	3 <u>√2</u> a	0	$\frac{3\sqrt{2}}{3}a$	$\frac{3\sqrt{2}}{3}a$
Value of S	а	a	a	a

3. Experimental procedure

The experimental apparatus consisted of three major systems: a high-pressure generator associated with an oil vessel and heater; a loading system with load cell; and instruments to record the amount of load and deformation. A detailed description of the apparatus is given by Ohashi [7].

The four kinds of experiments shown in Fig. 2 were performed in an oil vessel using thin-walled tubes and tensile specimens of cullulose nitrate, respectively, at 55 and 65° C. Fig. 2 shows the stress states, the stress tensors, and the values of ∞ the invariants in an element of specimens for experiments 1 to 4. The shaded portions correspond to the elements within the specimens. A square network of 5 mm distance is incised on a surface of each specimen to measure the displacements and the angle of torsion. The specimens used were confirmed to be isotropic from preliminary tests.

Experiment 1 corresponds to the simple tension of the tubular specimen, the outer diameter is 50 mm and wall thickness 3 mm. Experiment 2 corresponds to the torsion of the tubular specimen, and experiment 3 refers to the simple tension of uni-axial specimens made of the same cellulose nitrate as the tubes. Experiment 4 relates to the simple tension superimposed by oil pressure (dashed line in Fig. 2). The increasing rate of stress \dot{S} in all proportional loading were maintained constant at 0.1 and 0.5 MPa min⁻¹ for all experiments.

4. Expressions of strain components

The axial tensile strain, ϵ_x , the cross contraction or the circumferential strain, ϵ_y , and the shear strain, γ_{xy} , can be obtained from the displacements measured on the recorded film of deformed networks within an accuracy of 0.005 mm for axial displacement and an accuracy to 0.02° in angle between axes x and y. They are calculated from the following formulas calculating finite strain [8]

$$\epsilon_{\mathbf{x}} = \frac{(1+e_{\mathbf{x}})^2 - 1}{2}, \ \epsilon_{\mathbf{y}} = \frac{(1+e_{\mathbf{y}})^2 - 1}{2}, \gamma_{\mathbf{x}\mathbf{y}} = (1+e_{\mathbf{x}})(1+e_{\mathbf{y}})\sin\theta_{\mathbf{x}\mathbf{y}},$$
(9)

where e_x and e_y are conventional engineering strains, and θ_{xy} denotes an angle between the axes x and y. The second invariant of strain tensor E was calculated from Equation 5 by using the strain components in Equation 9.

5. Results and discussion

Figs. 3 and 4 show the relations between S and E obtained from the experiments for $\dot{S} = 0.1$ and 0.5 MPa min⁻¹ at 55° C. Figs 4 and 5 show the relations between S and E for $\dot{S} = 0.1$ and 0.5 MPa min⁻¹ at 65° C. The chain curve in each figure



Figure 3 Relations between S and E obtained from the experiments for $\dot{S} = 0.1$ MPa min⁻¹ at 55° C.



shows that of simple compression for each value of \dot{S} , for comparison. Each experimental result expressed by the corresponding symbol is plotted by using average values of the three separate test results. From these figures, by considering the section of stress level $S \simeq 12$ MPa for Figs. 3 and 4, or of stress level $S \simeq 7$ MPa for Figs. 5 and 6, for example, the following remarks are obtained from comparing the values of A and B shown in Fig. 2. The difference $(E_1 - E_2)$ of E between experiments 1 and 2 in each figure is due to the difference of A and B, where the subscripts of E denote the experiment number. The difference $(E_3 - E_4)$ in E between experiments 3 and 4 is only due to the difference of A. E_1 agrees well



Figure 4 Relations between S and E obtained from the experiments for $\dot{S} = 0.5$ MPa min⁻¹ at 55° C.

with E_3 because of the same condition of stress state. Accordingly, the difference $(E_1 - E_4)$ is due to the effect of A, and the difference $(E_4 - E_2)$ is due to the effect of B. Such a trend is independent of the increasing rate of stress \dot{S} and of the test temperature, but the difference of deformation on the third invariant B increases with deformation E. From the above discussion, it is clear that the effect of B is much smaller than that of A.

6. Concluding remarks

The effects of two stress invariants on the nonlinear viscoelastic deformation were discussed with special reference to cellulose nitrate. It was found that the effect of the third invariant of the devia-

Figure 5 Relations between S and E obtained from the experiments for $S = 0.1 \text{ MPa min}^{-1}$ at 65° C.



Figure 6 Relations between S and E obtained from the experiments for $\dot{S} = 0.5$ MPa min⁻¹ at 65° C.

toric stress tensor on the non-linear viscoelastic deformation of cellulose nitrate was much smaller than that of the hydrostatic pressure. Such a trend did not depend on the increasing rate of stress and the test temperature.

Although the above result is obtained for cellulose nitrate, it may by applicable to other polymers which exhibit differences of behaviour in simple tension and compression comparable with those in cellulose nitrate shown in these figures.

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