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Creep behaviour in cellulose nitrate under superimposed tensile and hydrostatic loading

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The creep behaviour under various combinations of superimposed tensile and hydrostatic loading are quantitatively investigated using nonlinear-viscoelastic cellulose nitrate heated at 65° C. The creep strain and the creep strain-rate are seriously affected by the effect of hydrostatic pressure. The trends of behaviour under superimposed tensile and hydrostatic loading cannot be predicted by the concepts which apply to uniaxial loading. The stress–strain relation of the creep behaviour under superimposed loading is deduced from the invariant theory using an hypothesis of creep potential. The creep data obtained on the cellulose nitrate at 65° C under superimposed loading, as well as uniaxial loading, are found to fit to the deduced relation well.

1. Introduction

The mechanical behaviour of polymers has been widely investigated, mainly because of the large-scale advance and utilization of these materials. There has been much detailed study of creep deformation [1–3] and creep mechanism [4, 5] in polymers under uniaxial loading. The mechanical behaviour of polymers has been seen to be significantly influenced qualitatively by the hydrostatic pressure [6, 7], unlike most metals. The creep behaviour, in particular, under superimposed tensile and hydrostatic loading in a nonlinear viscoelastic polymer may be different from that under uniaxial loading.

In this paper, the creep behaviour under various combinations of superimposed tensile and hydrostatic loading are quantitatively investigated using nonlinear-viscoelastic cellulose nitrate heated at 65° C. Moreover, starting from the invariant theory with an hypothesis of creep potential, a stress–strain relation of the transient creep is deduced for the non-linear viscoelastic material subjected to superimposed loading, and this relationship is compared with the experimental results. As a result, although the instantaneous elastic–plastic behaviour at the instant of loading was not particularly influenced by hydrostatic pressure, the creep behaviour was so influenced. Such as phenomenon cannot be predicted by the ordinary concepts which apply under uniaxial loading. The proposed relation for transient creep based on the invariant theory gave good agreement with actual observations.

2. Stress–strain relationships

If the time-hardening theory is adopted for convenience of analysis, the creep strain-rate may be proposed by using a creep potential hypothesis [8] as follows:

$$(\dot{\epsilon}_{ij})_c = E[t+s, J_2](\partial K/\partial \sigma_{ij}), \qquad (1)$$

where ϵ_{ij} are the components of strain tensor, the dot representing differentiation with respect to time. $E[\ ]$ is a function of $(t+s)$ and J_2, t and s denote current time and material constant time introduced by the author. $J_2 = S_{ij}S_{ij}/2$ is the second invariant of the deviatoric stress tensor [9]. K, σ_{ij} and $S_{ij} = \sigma_{ij} - \frac{1}{3}\sigma_{ii}\delta_{ij}$ are a creep potential, the components of the stress tensor, and the deviatoric stress tensor of σ_{ij}, respectively [9]. Equation 1 may be expressed approximately by selecting J_2 as the creep potential as:

$$(\dot{\epsilon}_{ij})_c = B(t+s)^{\alpha}\exp(bJ_2^{1/2})(\partial J_2/\partial \sigma_{ij}), \quad (2)$$

where B, α and b are the material constants.

The instantaneous elastic–plastic strain at the instant of loading may be given as

$$(\epsilon_{ij})_0 = \frac{S_{ij}}{2G}\{1 + (J_2/k^2)^n\}, \qquad (3)$$

which is Ramberg-Osgood's relation, and is a good approximation of the non-linear stress–strain relation [10]. G, n and k in Equation 3 are the material constants.

With the components of principal stress σ_1, σ_2 and σ_3, J_2 may be expressed as follows [9]:

$$J_2 = \tfrac{1}{6}\{(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2\}. \qquad (4)$$

In the present tests, the superimposed loading of tensile stress σ_1 and hydrostatic pressure, $\sigma_2 = \sigma_3$, is applied to the uniaxial specimen. If the principal stress difference is denoted by $\Delta\sigma = \sigma_1 - \sigma_2$,

$$J_2 = (\Delta\sigma)^2/3 \qquad (5)$$

is obtained from Equation 4. Equations 2 and 3 for the principal strains may lead to the following expressions using Equation 5

$$\Delta\dot{\epsilon}_c = (\dot{\epsilon}_1)_c - (\dot{\epsilon}_2)_c = B(t+s)^\alpha \Delta\sigma \exp\,[(b/\sqrt{3})\Delta\sigma] \qquad (6)$$

and

$$\Delta\epsilon_0 = (\epsilon_1)_0 - (\epsilon_2)_0 = \frac{\Delta\sigma}{2G}\{1 + (\Delta\sigma/3k)^{2n}\}. \qquad (7)$$

When it is necessary to calculate the total creep strain $\Delta\epsilon_c$, the following procedure may be adopted. Time is subdivided into small intervals, $0 \sim t_1$, $t_1 \sim t_2, \ldots, t_{m-1} \sim t_m, \ldots, t$, in which the magnitude $\Delta\dot{\epsilon}_c$ may be considered to be approximately constant. For example, $\Delta\epsilon_c(t_m)$ at arbitrary time t_m in the period $t_{m-1} \sim t_m$ is approximated as

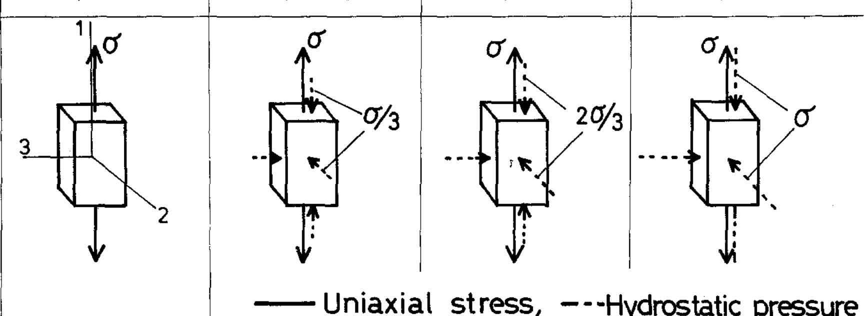
$$\Delta\epsilon_c(t_m) = \Delta\epsilon_c(t_{m-1}) + \Delta\dot{\epsilon}_c(t_{m-1})(t_m - t_{m-1}). \qquad (8)$$

3. Experimental

The uniaxial specimens were made of initially isotropic cellulose nitrate, 6 mm thick. On a surface of each specimen, a square gauge mark was cut in a region of uniform stress state. The experimental apparatus consisted of three major parts: a high pressure generator-associated oil vessel with heater; loading equipment with load cell; and instrumentation to record the amount of load and deformation. Detailed description of the shape of the specimen and of the apparatus is given in [11].

The creep tests were performed under superimposed loading of axial tensile stress, σ_1, and hydrostatic pressure $\sigma_2 = \sigma_3$, as shown in Table I, at 65° C. Table I shows the stress states of an element

TABLE I Stress states of an element of the specimen for different test conditions

	Test condition $A = \sigma_{ii}/\Delta\sigma$			
	$A = 1$	$A = 0$	$A = -1$	$A = -2$
Stress state				
σ_1	σ	$2\sigma/3$	$\sigma/3$	0
$\sigma_2 = \sigma_3$	0	$-\sigma/3$	$-2\sigma/3$	$-\sigma$
$\Delta\sigma = \sigma_1 - \sigma_2$	σ	σ	σ	σ
$\sigma_{ii} = \sigma_1 + \sigma_2 + \sigma_3$	σ	0	$-\sigma$	-2σ

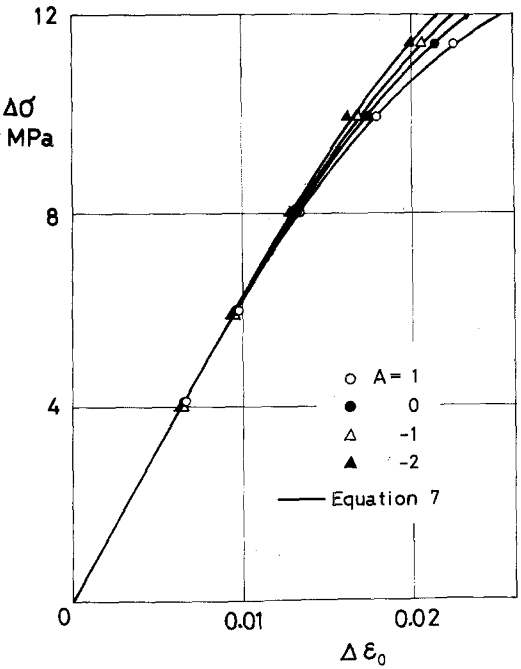


Figure 1 Relationship between $\Delta\sigma$ and $\Delta\epsilon_0$ at the instant of loading.

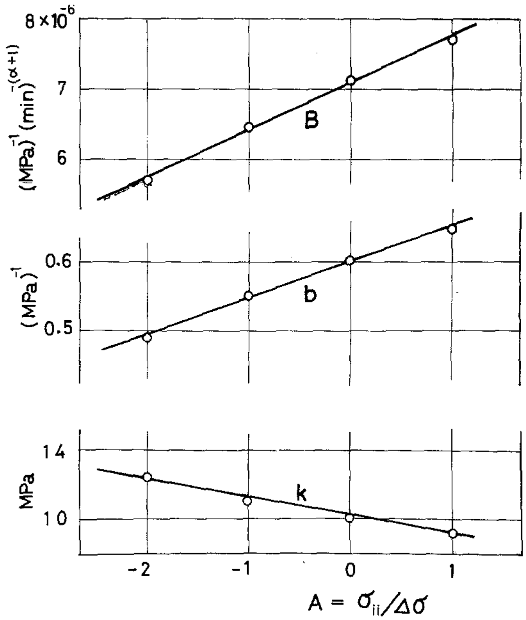


Figure 2 Relationship between A and the material constants affected by A.

of the specimen for various values of $A = \sigma_{ii}/\Delta\sigma$, where A is the dimensionless first invariant of the stress tensor, and $\sigma_{ii} = \sigma_1 + \sigma_2 + \sigma_3$ and $\Delta\sigma = \sigma_1 - \sigma_2$. Each load was applied so as to obtain constant values of $\Delta\sigma$ of 4, 6, 8, 10 and 11.5 MPa for each value of A. Axial elongation and cross contraction between gauge marks engraved on the specimen were measured to within 0.005 mm from photographs of the gauge marks using a magnifying projector. The accuracy of strain thus obtained is within about 2×10^{-4}. The principal strain difference $\Delta\epsilon = \epsilon_1 - \epsilon_2$ was calculated in the natural strain system $\epsilon_\beta = \ln(1 + e_\beta)$, where $e_\beta (\beta = 1, 2)$ are the conventional engineering strains.

4. Results and discussions

4.1. Instantaneous elastic-plastic strain

Fig. 1 shows the relations between the applied stress, $\Delta\sigma$, and the instantaneous elastic-plastic strain, $\Delta\epsilon_0$, at the instant of loading, obtained from the experiments for each value of A at 65° C, where it can be seen that a proportional dependence is not assumed. Each point is plotted using average values of three seperate test results. $\Delta\epsilon_0$ may depend on the value of A or on the effect of hydrostatic pressure. When Ramberg–Osgood's relation (Equation 7) is used as a good approximation of the instantaneous strain, the material constants in Equation 7 were determined from the experimental results shown in Fig. 1, as $G = 315$ MPa and $n = 1.85$ for each value of A. However, the plastic constant k varied with A, and the relationship between k and A is shown in Fig. 2, which also shows the relations between the material constants dependent on A and the values of A as shown in Table I. The solid curves in Fig. 1 show the relations between $\Delta\epsilon_0$ and $\Delta\sigma$ obtained from Equation 7 using the corresponding material constants G, n and k for each value of A.

4.2 Transient creep behaviour

The curves in Fig. 3 show the experimental creep relations between the strain $\Delta\epsilon = \Delta\epsilon_0 + \Delta\epsilon_c$ and time for each value of A at either $\Delta\sigma = 11.5$ or 8 MPa at 65° C. Each curve is plotted using the average values of three separate test results. The solid curves in Fig. 4a and b show the relations between the creep strain-rate, $\Delta\dot{\epsilon}_c$, and time, corresponding respectively to the thick and thin curves in Fig. 3. Although the instantaneous elastic-plastic strain in Fig. 1 is not so significantly influenced by the value of A, the creep strain or the creep strain-rate is, with lapse of time after loading. Furthermore, taking the extremes $A = 1$ and $A = -2$ in Fig. 4, the curves for $A = 1$ indicate that the creep rate is sensitive to the stress level $\Delta\sigma$ in tension, but the creep curves for compression ($A = -2$) show little variation of creep rate with stress. These

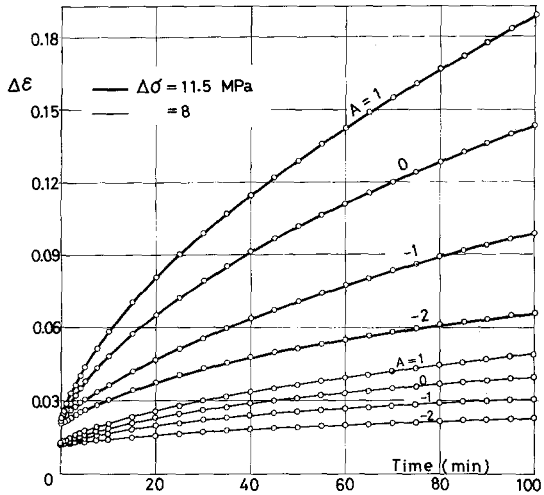


Figure 3 Experimental creep relations between the strain $\Delta\epsilon$ and time for each value of A, with $\Delta\sigma = 11.5$ and 8 MPa.

phenomena may not be unexpected in uniaxial creep experiments. Since the above features of creep behaviour are obtained for cellulose nitrate at 65° C, it is difficult to conclude that all these features would also apply to the creep behaviour of many polymers. However, since the stress–strain relations of polymers in elastic-plastic deformation are affected by hydrostatic pressure [6, 7], it is supposed that the phenomena similar to those mentioned above may appear in the creep behaviour of many polymers.

The values of material constants in Equation 6 were determined from the solid curves in Fig. 4 to be $B = 7.7 \times 10^{-6}$ [MPa]$^{-1}$ [min]$^{-(\alpha+1)}$, $s = 1.0$ [min], $\alpha = -0.35$, $b = 0.65$ [MPa]$^{-1}$, for $A = 1$. (As B and b were affected by the value of A, they are also shown in Fig. 2.) The dashed curves in Fig. 4a and b show the relations between $\Delta\dot{\epsilon}_c$ and time calculated from Equation 6 using the corresponding material constants for each value of A. These curves agree well with the corresponding experimental results (solid curves) especially at the early stage after the instant of loading. The proposed relation for creep behaviour which takes account of material constant time s as a new parameter in Equation 6, gives good agreement with actual observations under not only superimposed loading but also uniaxial loading.

In Fig. 2, the dependence of the material constants B, b and k on A, or hydrostatic pressure, may be assumed to be proportional, at least in the range of A shown in this figure, and it is seen that the plastic-creep strain will tend to be zero for smaller values of A, and only the elastic strain may remain.

5. Conclusions

The main results are:

(1) The instantaneous elastic-plastic strain of nonlinear viscoelastic cellulose nitrate is influenced by hydrostatic pressure: The strain decreases with increasing hydrostatic pressure.

(2) The creep strain of nonlinear-viscoelastic cellulose nitrate is remarkably influenced by hydrostatic pressure, unlike most metals. Moreoever,

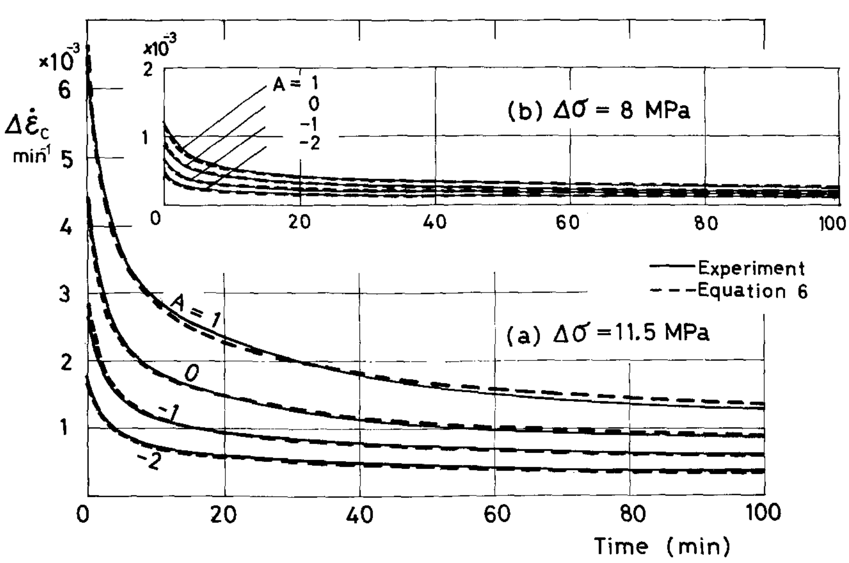


Figure 4 Relationship between creep strain-rate and time, (a) $\Delta\sigma = 11.5$ MPa, (b) $\Delta\sigma = 8$ MPa.

those features affected by hydrostatic pressure become larger for increasing stress levels, $\Delta\sigma$. Such trends cannot be predicted by the ordinary concept of creep behaviour under uniaxial loading.

(3) The proposed invariant theory of creep behaviour shows good agreement with actual observations under not only superimposed loading but also uniaxial loading over a wide range of stress and time.

(4) The material constants affected by hydrostatic pressure (k, B, b) may almost be approximated by a linear function of the value of the dimensionless first invariant of the stress tensor.

(5) Although the features of creep behaviour mentioned above are obtained for the cellulose nitrate, it may be considered that the similar phenomena may appear in the creep behaviour for many polymers.

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