Constitutive Equation for Plastic Behavior of Hydrostatic-Pressure-Dependent Polymers

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Abstract: Hydrostatic pressure dependence of the mechanical behavior of polymers is studied, using constitutive modeling of the yield surface described by the first and the second invariants of stress and the nonassociated flow rule that satisfies the incompressible hypothesis. An internal variable theory of rate-independent plasticity is presented, which incorporates isotropic hardening as a function of accumulated plastic strain. After the determination of material constants under uniaxial tension and compression, the model shows that the von Mises-type effective stress - plastic strain curves under multiaxial load are quite different from those under uniaxial load. The model is compared with the experimental results of uniaxial tension and compression obtained by Spitzig and Richmond and those of others obtained by Silano et al. under high pressure.

Key words: Constitutive equation, Polymer, Plastic behavior, Hydrostatic pressure dependence, Nonassociated flow rule

1. INTRODUCTION

In the mechanical behavior of polymers, temperature dependence, time dependence, deformation-induced anisotropy and hydrostatic pressure dependence are marked. For the time dependence, the viscoplastic constitutive equation derived by Krempl [1] and others [2,3] is applicable to the description of the stress rate dependence of the flow stress, stress relaxation and creep. The constitutive equation based on the molecular chain network theory, which reflects the microstructural changes of the rotation and orientation of the polymer chain, was proposed by Boyce et al. in order to describe the generation and the propagation behavior of necking observed during the tensile test [4,5]. The mechanical behavior of polymers under high pressure has been reported in many papers [6-11]. It is understood that the hydrostatic pressure dependence of the mechanical behavior of polymers is marked in comparison with that of metals, based on experimental results. Young's modulus, shear modulus and yield stress of polymers all increase almost linearly with the increase in pressure. The stress-strain curves of tension differ from those of compression [9,10]. The difference becomes marked with decreasing temperature [9]. The effect of hydrostatic pressure is described by the first invariant of the stress, and the yield condition under the multiaxial load condition can correct the von Mises yield condition [12,13]. Therefore, the plasticity constitutive equation for polymers must express the hydrostatic pressure dependence appropriately.

Many plasticity constitutive equation models which take into consideration the hydrostatic pressure dependence have been proposed to describe the ductile fracture behavior of metals [14-16]. The nucleation and growth of a cavity under 3-axis tension, is described by a scalar variable, and it becomes a fixed equation in these models of the plasticity constitutive equation for porous material containing cavities. However, the above-mentioned effect cannot be expressed even when it is applied to polymers as is.

In this study, a plasticity constitutive equation model, in which a yield surface and nonassociated flow law are assumed and the hydrostatic pressure dependence is considered, is proposed. The yield function is represented by the first invariant of stress and the second invariant of deviatoric stress, and the flow law is governed by the plastic potential which satisfies the incompressibility of plastic deformation. The isotropic hardening variable is defined as a function of effective plastic strain, and the concrete form is obtained from the stress-plastic strain curve of uniaxial tension. In addition, material constants of the constitutive equation are selected in order to reproduce the experimental results under uniaxial load, and the calculated results for multiaxial combined loading are shown. Finally, the calculated results are compared with the experimental data obtained under high pressure by Spitzig and Richmond [10], and other results obtained by Silano et al. [8].

2. PLASTICITY CONSTITUTIVE EQUATION

Time dependence is marked for polymers even at room temperature. However, in the structural analysis using the finite element method, time dependence may be disregarded in the first approximation. Therefore, the formulation of the plasticity constitutive equation, in which infinitesimal deformation is considered and time dependence is disregarded for simplicity, is carried out. Total strain $\varepsilon_{ij}^\varepsilon$ is assumed, for convenience, to be the sum of elastic strain $\varepsilon_{ij}^e$ and plastic strain $\varepsilon_{ij}^p$ as follows:

$$
\varepsilon_{ij} = \varepsilon_{ij}^e + \varepsilon_{ij}^p.
$$

The elastic deformation can be written in the form of Hooke’s law, as
where $E$ and $\nu$ represent Young’s modulus and Poisson’s ratio, respectively. Generally, $E$ and $\nu$ exhibit hydrostatic pressure dependence in polymers [17].

Next, the plastic deformation is formulated. The simplest form of the yield surface with the hydrostatic pressure dependence is

$$f = (1 - \beta)\sqrt{3J_2} + \beta I_1 - \kappa = 0$$

(3)

where $J_2$ and $I_1$ represent the second invariant of deviatoric stress and the first invariant of stress, respectively. $\kappa$ represents the isotropic hardening variable. $\beta$ is a material constant. For $\beta = 0$, Eq. (3) indicates the von Mises yield surface. Figure 1 shows the yield surface in the principal stress plane, obtained using Eq. (3). $\sigma_Y$ in the figure is the yield stress of uniaxial tension.

According to the literature [10], the volume change after plastic deformation of polymers is markedly small, similar to that of the steel. That is to say, the incompressibility of plastic deformation is almost satisfied. It is possible to assume such plastic potential as follows:

$$g = \sqrt{3J_2}.$$  

(4)

Therefore, the plastic strain rate can be used to write the nonassociation flow rule as

$$\varepsilon_{ij}^p = \lambda \frac{\partial g}{\partial \sigma_{ij}}.$$  

(5)

where $\lambda$ is a scalar function of stress and a positive value which depends on the load hysteresis. From the consistency condition

$$\dot{f} = \frac{\partial f}{\partial \sigma_{ij}} \dot{\sigma}_{ij} + \frac{\partial f}{\partial \kappa} \dot{\kappa} = 0,$$  

(6)

we can easily obtain

$$\dot{\varepsilon}_{ij}^p = \frac{3}{2H} \left[ \frac{3}{2} (1 - \beta) \frac{s_{ij}}{\sqrt{3J_2}} + \beta \delta_{ij} \right] \dot{\sigma}_{ij} \frac{s_{ij}}{\sqrt{3J_2}}$$

(7)

where effective stress and effective strain are defined as follows:

$$\bar{\sigma} = (1 - \beta)\sqrt{3J_2} + \beta I_1$$

$$\bar{\varepsilon}^p = \int_0^t \left( \frac{2}{3} \varepsilon_{ij}^p \varepsilon_{ij}^p \right) dt$$

(8)

The isotropic hardening variable is assumed to be

$$\kappa = \bar{\sigma}(\varepsilon^p)$$.

(9)

The specialized form can be obtained under uniaxial stress-plastic strain. Here, two types of functions are examined:

$$\bar{\sigma} = F(b + \bar{\varepsilon}^p)^n,$$  

(10a)

$$\bar{\sigma} = \sigma_Y + (\sigma_Y^* - \sigma_Y) [1 - \exp(-c\bar{\varepsilon}^p)].$$  

(10b)

3. CALCULATED RESULTS UNDER VARIOUS MULTIAXIAL AND PROPORTIONAL LOADS

3.1. Experimental Result and Calculated Result Obtained under Uniaxial Load

Under tension and compression, stress $\sigma$ and plastic strain $\varepsilon^p$ are expressed as follows:

$$\sigma_{11} = \sigma_Y, \quad \text{other } \sigma_{ij} = 0$$

$$\varepsilon_{11}^p = \varepsilon^p, \; \varepsilon_{22}^p = \varepsilon_{33}^p = -\varepsilon^p / 2, \; \text{other } \varepsilon_{ij}^p = 0.$$  

(11a)

From Eq.(7), plastic strain is expressed as
Therefore, $\beta$ represents the difference between tension and compression under uniaxial load.

Figure 2 shows stress-strain curves of high-density polyethylene (HDPE) and polypropylene (PP) under uniaxial load. The dotted line in the figure represents our experimental result. The solid line and chain line indicate results calculated from Eqs. (10a) and (10b), respectively. The following material constants were used.

$$
E = 0.982 \text{GPa}, \quad \beta = 0.12 \\
F = 37.5 \text{MPa}, \quad b = 1.0 \times 10^{-4}, \quad n = 0.244 \\
\sigma_y = 7.0 \text{MPa}, \quad \sigma_y^* = 20.0 \text{MPa}, \quad c = 40.0
$$

(12a)

The values of $F$, $\beta$, $\sigma_y$, $\sigma_y^*$ and $c$ were determined using the stress-plastic strain curve for uniaxial tension. Next, the value of $\beta$ was determined in order to clarify the compression behavior.

The experimental result can be described with good accuracy, regardless of the type of function, Eq. (10a) or Eq. (10b), used. Equation (10a) indicates a stress-strain relation after yielding, which coincides well with experimental results for small strain, although it predicts a large stress value for large strain.

### 3.2 Calculated Result for Multiaxial Combined Loading

Here, the calculated results for biaxial tension, biaxial compression and simple shear, as an example of multiaxial combined loading, are shown.

Under biaxial tension and biaxial compression, stress $\sigma$ and plastic strain $\varepsilon^p$ can be expressed as follows:

$$
\sigma_{11} = \sigma_{22} = \sigma, \quad \text{other } \sigma_{ij} = 0 \\
\varepsilon_{11}^p = \varepsilon_{22}^p = \varepsilon^p, \quad \varepsilon_{33}^p = -2\varepsilon^p, \quad \text{other } \varepsilon_{ij}^p = 0
$$

(13a)

From Eq. (7), plastic strain rate is expressed as

$$
2\dot{\varepsilon}^p = \sqrt{\frac{1}{H} \sigma \dot{\sigma}} \quad \text{biaxial tension} \\
\sqrt{1 - \frac{3\beta}{H} \sigma \dot{\sigma}} \quad \text{biaxial compression}
$$

(13b)

Under simple shear, shear stress $\tau$ and plastic shear strain $\gamma^p$ can be expressed as follows:

$$
\sigma_{12} = \sigma_{21} = \tau, \quad \text{other } \sigma_{ij} = 0 \\
\varepsilon_{12}^p = \varepsilon_{21}^p = \gamma^p / 2, \quad \text{other } \varepsilon_{ij}^p = 0
$$

(14a)

The solid lines and chain lines in Fig. 3 represent results calculated from Eqs. (13) and (14) using Eqs. (10b) and (10a), respectively. The results of uniaxial tension and uniaxial compression calculated using Eq. (11) are also shown for reference. The material constants of Eq. (12b) were employed in this calculation.

A prediction which markedly differs from the results for the combined loading condition is elucidated. It is necessary to examine the validity of the theory by comparing it with the experimental result obtained under the same load condition, because the calculated result for biaxial...
Fig. 3. Stress-plastic strain curves under multiaxial stress.

compression is very different from that for biaxial tension. There is not a large difference observed between the results calculated with Eq. (10a) and with Eq. (10b) under multiaxial combined loading. These results also agree with the calculated result for uniaxial tension with $\beta = 0$ (von Mises yield surface).

3.3. Comparison of Calculated Result and Experimental Result under High Pressure

Here, the result calculated using the present model and various experimental results previously reported [8,10] under high pressure are compared.

Figure 4 shows the result calculated using this constitutive equation model and the experimental results for uniaxial tension and uniaxial compression under various hydrostatic pressures obtained by Spitzig and Richmond [10]. The symbols □ □ □ □ □ □ and □ □ □ □ □ □ represent uniaxial tension and compression under $p = 0.101$, 138, 276, 552, 828, and 1104 MPa, respectively. The solid line and dashed line represent the results calculated with Eq. (10b) under uniaxial tension and uniaxial compression, respectively. The chain line and chain double-dashed line show the results calculated using Eq. (10a) under tension and compression, respectively.

\[
E = 1140 + 5p \text{ MPa, } \beta = 0.035 \\
F = 40.0 \text{ MPa, } b = 1.0 \times 10^{-4}, n = 0.172 \quad (\text{HDPE})
\]
\[
\sigma_Y = 13.0 \text{ MPa}, \quad \sigma_Y^* = 28.5 \text{ MPa}, \quad c = 30.0
\]

\[
E = 2350 + 3p \text{ MPa, } \beta = 0.050 \\
F = 100 \text{ MPa, } b = 1.0 \times 10^{-4}, n = 0.114 \quad (\text{PC})
\]
\[
\sigma_Y = 40.0 \text{ MPa}, \quad \sigma_Y^* = 6898 \text{ MPa}, \quad c = 100
\]

Fig. 4. Tensile and compressive stress-strain curves at various hydrostatic pressures.

Here, the pressure dependence of the modulus of longitudinal elasticity reported in the literature [10] was used. The values of $F$, $b$, $n$, $\sigma_Y$, $\sigma_Y^*$ and $c$ were determined in order to describe the stress-plastic strain curve of uniaxial tension
under atmospheric pressure. Finally, the value of $\beta$ was
determined in order to express the compression behavior
under atmospheric pressure. HDPE in Fig. 4(a) and Fig. 2(a), are materials of different grades, made by various
companies, which show different mechanical behaviors.

The experimental results show a drop in stress immediately
after yielding. The specimens show strain localization
(necking) immediately after yielding and propagation of the
localization region with strain hardening. This model re-
produces the experimental results under high pressure until
yielding comparatively well, as evident from the figure.

The initial yield stress at high pressure is overestimated, un-
der close observation, particularly in the case of PC. This
can be improved appropriately, taking account of the dif-
ference between the hydrostatic pressure dependence under
initial yield stress and subsequent yield stress. There is not
a large difference between the results calculated using Eq.
(10a) and Eq. (10b).

Figure 5 shows the torsion test result of POM under
various hydrostatic pressures, obtained by Silano et al. [8],
and results calculated using the constitutive equation model.
The symbols $\square$, $\square$, $\square$, and $\square$ represent the experimental results for
$\rho=0.101, 200, 400$, and $600$ MPa, respectively. Solid
and chain lines represent the results calculated using Eq.
(10a) and Eq. (10b), respectively. In the calculation, the
following material constants were used.

$$
G = 955 + p \text{ MPa}, \quad \beta = 0.0277 \\
F = 182 \text{MPa}, \quad b = 1.0 \times 10^{-4}, \quad n = 0.366 \\
\sigma_Y = 3.36 \text{MPa}, \quad \sigma_Y^* = 89.0 \text{MPa}, \quad c = 23.0
$$

(16)

The experimental results indicate a true softening with
increasing strain. This is because the failure of specimens
occurred by buckling. This model also describes the
torsion test results under high pressure until softening com-
paratively well. The value of stress obtained using Eq.
(10a) tends to be larger than that obtained using Eq. (10b)
der large strain.

![Fig. 5. Shear stress - strain curves at various hydrostatic pressures.](image)

4. CONCLUSIONS

Using the yield surface rule that takes into account the
hydrostatic pressure dependence and nonassociation flow, a
plasticity constitutive equation model for polymers was for-
mulated. The following conclusions were made on the
basis of a comparison of results calculated using this model
and conventional experimental results.

1. This model was able to accurately reproduce experimen-
tal results of uniaxial tension and uniaxial compression.
   There was not a large difference in the calculation results for
   other loads, when a functional type could accurately describe
   the stress-plastic strain curve of uniaxial tension.

2. The present model predicted a markedly different curve
   for von Mises-type equivalent stress-strain under multiaxial
   combined loading. Under the plane stress condition, cal-
   culation results of biaxial tension and biaxial compression
   were very different.

3. The constitutive equation appropriately reproduced ex-
   perimental results under high pressure. However, it was
   necessary to modify this model accurately to describe the
difference between hydrostatic pressure dependence of initial
   yield stress and subsequent yield stress under high pressure.

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