Elastic Tensile Recovery of a Viscoelastic Model under Power Law

By Takuji Yamaguchi, Taruaki Yanagawa and Hiroshi Kimura, Members, TMSJ

Faculty of Engineering, Fukui University, Fukui


Abstract

The Boltzman and the Schapery equations of a time-strain reduced type are used for the linear and the nonlinear models, respectively. The predicted results from both models have experimentally been discussed by two kinds of polyethylene films at room temperature.

The fractional elastic recovery (U.E.R.) of the linear model is constant regardless of the initial strain \( \varepsilon_1 \) when holding time \( t_r = 0 \), and increases with \( \varepsilon_1 \) when \( t_r > 0 \). U.E.R. of the nonlinear model is smaller than that of the linear model and the difference between them is larger at higher \( \varepsilon_1 \). If a solid shows more stress relaxation and stronger nonlinearity, its U.E.R. is smaller. The experimental results of U.E.R. are qualitatively similar to U.E.R. predicted by using the nonlinear model.

The fractional delayed elastic recovery (D.E.R.) can only be calculated from the linear model. D.E.R. of the model is constant regardless of \( \varepsilon_1 \) when \( t_r = 0 \), and decreases with \( \varepsilon_1 \) when \( t_r > 0 \). A solid exhibiting a remarkable stress relaxation has higher rate of delayed recovery and larger D.E.R. The experimental results of D.E.R. show remarkable nonlinearity and are inconsistent with the linear theory.

1. Introduction

It is known that the deformation of polymer can be divided into three components after unloading: instantaneous elastic recovery, delayed recovery and permanent set, and a fraction of the first component decreases with the increase of deformation instead of increasing other two fractions. Tensile recoveries of textile fibers and their products are important with respect to dimensional stability in processing, crease-resistance, and form retention in products. Methods for recovering-test are defined in JIS L 1073 and L 1079. The tensile recovery of various fibers has been investigated in detail by many authors. It seems that the recovery of high polymer after extension at constant rate of strain should be viscoelastic, but many investigations did not clarify how the recovery was related to the basic viscoelastic properties of materials.

In this paper, the tensile recovery behavior of a simple viscoelastic model is analyzed, the relaxation modulus of which can be expressed by a power function of time, and the relation between experimental constants of the model and the ability of recovery is studied.

It is well known that the limit of linearity in strain for crystalline polymer is less than 1% at room temperature, and hence the applicability of the linear viscoelastic theory is very limited. While many nonlinear viscoelastic theories for solid polymer have been proposed, Schapery’s theory is relatively simple to treat among them. We found that for polyethylene, polyvinylalcohol, and rigid polyvinyl chloride, viscoelastic behaviors such as single- and two-step stress relaxation and constant rate of elongation could be well expressed by the superposition equation of reduced type which was derived from the Schapery’s theory. In this paper, we adopt this theory as a nonlinear mathematical model and study the relationship between nonlinearity and recoverability.

In addition, the tensile recovery tests are carried out for polyethylenes, behaviors of which were approximately described by a power law model and the Schapery’s theory, and these results are compared with the theory to examine its applicability.

2. Analyses by a Linear and a Nonlinear Viscoelastic Theory

2.1 General theory

The superposition equation derived from the Schapery’s general superposition principle, and the elongational relaxation modulus \( E_n(t, \varepsilon) \), are

\[
\sigma(t) = \int_0^t E_0(\rho - \rho') \frac{d\varepsilon(u)}{du} du \tag{1}
\]

\[
\rho = \int_0^t \frac{ds}{\alpha_t(E(s))}, \quad \rho' = \int_0^\rho \frac{ds}{\alpha_t(E(s))} \tag{2}
\]

\[
E_n(t, \varepsilon) = E_0 \left( \frac{t}{\alpha_t(\varepsilon)} \right) \tag{3}
\]

where \( \sigma(t) \): stress at time \( t \), \( \varepsilon(u) \): strain history, \( 0 \leq u \leq t \), \( E(t) \): linear elongational relaxation modulus, \( \rho, \rho' \): reduced times to the state of standard linear strain, \( \alpha \): factor to nonlinearize.
the linear Boltzmann’s superposition equation and a positive decreasing-function of strain. Eq. (3) shows that \( E_n(t, \varepsilon) \) shifts to a shorter time as strain increases. Eq. (1) is the superposition equation by generalizing this strain dependence. When \( a_0 = 1 \), Eq. (1) reduces to a linear Boltzmann’s superposition equation, and Eq. (3) to a linear elongational relaxation modulus, respectively.

Figures 1 (a) and (b) show a strain history and its corresponding stress. The material is elongated at a constant rate of strain \( \varepsilon_1 \) from 0 to \( t_1 \), followed by stress relaxation at a fixed strain \( \varepsilon_1 = \varepsilon_1 t_1 \) during a certain period \( t_r = t_2 - t_1 \), and is unloaded at a rate of strain \( \varepsilon_2 \) to \( \varepsilon_3 \), at that time the level of stress reaches zero. After \( t_3 \), the strain recovery is dependent on time under zero stress. The strain at \( t_3 \) is \( \varepsilon_2 = \varepsilon_1 t_1 - \varepsilon_2 (t_3 - t_2) \).

Susich et al. divided the elongational strain recovery \( \varepsilon_e \) into the instantaneous elastic recovery and the delayed elastic recovery. The sum of the latter and the recovery after \( t_3 \) was named as the delayed elastic recovery \( \varepsilon_{ie} \). In other books, \( \varepsilon_e \) and \( \varepsilon_1 - \varepsilon_e \) were named as the elastic and the plastic strain, respectively. In this study, \( \varepsilon_e \) is named as the elastic recovery by unloading, and the elastic recovery strain \( \varepsilon_{ie} \) after \( t_3 \) as the delayed elastic recovery, respectively, for calculative and experimental simplicity. Then, total strain \( \varepsilon_1 = \varepsilon_1 t_1 \), and unloading elastic strain recovery \( \varepsilon_e = \varepsilon_2 (t_3 - t_2) \). The fractional unloading elastic recovery U.E.R. is defined as

\[
U.E.R. = \frac{\varepsilon_e}{\varepsilon_1} \times 100 = r \frac{(t_3 - t_2)}{t_1} \times 100 \quad \cdots \quad (4)
\]

where \( r = \varepsilon_2/\varepsilon_1 \). There are two methods defined in JIS L 1073 for measuring elastic recovery. In Method A, \( t_r = 1 \) min, and in Method B, \( t_r = 0 \). But \( \varepsilon_1 = \varepsilon_2 \) in both methods. The reason why we put \( \varepsilon_1 > \varepsilon_2 \) is to obtain high experimental accuracy.

The solid line \( \varepsilon(t) \) in Fig. 1(a) is expressed as

\[
\varepsilon(t) = v_1 t H(t) - v_1 (t - t_1) H(t - t_1) - v_2 (t - t_2) H(t - t_2) \quad \cdots \quad (5)
\]

where \( H(t) \) is the unit step function. The following stress equation in each process is obtained by substituting Eq. (5) into Eq. (1). The symbols, \( l \) and \( n \), express the results derived from the linear and the nonlinear theory, respectively.

(a) loading process, \( 0 < t \leq t_1 \)

\[
\sigma_u(t) = v_1 \int_0^t E(t - u) \,du \quad \cdots \quad (6)
\]

(b) stress relaxation process, \( t_1 < t \leq t_2 \)

\[
\sigma_u(t) = v_1 \int_0^{t_1} E(t - u) \,du \quad \cdots \quad (7)
\]

(c) unloading process, \( t_2 < t \leq t_3 \)

\[
\sigma_u(t) = v_1 \int_0^{t_2} E(t - u) \,du - v_2 \int_0^t E(t - u) \,du \quad \cdots \quad (10)
\]

By equating right hand sides of Eq. (10) or Eq. (11) to zero and solving this for \( t, t_3 \) can be obtained.

The strain in the delayed elastic recovery process after \( t_3 \) can be derived only from the linear theory in the following way. The strain-time relation for variable stress history is given in terms of the creep compliance \( J(t) \):

\[
\varepsilon(t) = \int_0^t J(t - u) \frac{ds}{d\varepsilon} \,du \quad \cdots \quad (12)
\]

Substituting Eqs. (6), (8) and (10) into Eq. (12) yields the following general equation for the strain after \( t_3 \).
Although both viscoelastic functions \( E(t) \) and \( J(t) \) are necessary to calculate the above equation, both functions have the following relation,

\[
\int_0^t E(u)J(t-u)du = t
\]

Namely, if one is known, the other can be predicted from Eq. (14). Therefore, Eq. (13) can be expressed by either \( E(t) \) or \( J(t) \).

In the nonlinear theory, the Schapery's general superposition equation corresponding to Eq. (12) is given by

\[
\epsilon(t) = g_1(\sigma)\int_0^t J(\phi)\frac{d\sigma}{\sigma(\sigma_s)}du + \int_0^t g_2(\sigma)\frac{d\sigma}{\sigma(\sigma_s)}du \quad (15)
\]

\[
\phi = \int_0^t \frac{ds}{\sigma(\sigma_s)} \quad \sigma_s = \int_0^t \frac{ds}{\sigma(\sigma_s)} \quad (16)
\]

where \( g_1, g_2 \) and \( \sigma_s \) are functions of \( \sigma \) to nonlinearize the linear Boltzmann's superposition equation. However, since Eq. (15) is not necessarily an inverse equation of Eq. (1), the equation to predict the strain after \( t_3 \) can not be derived from the Schapery's nonlinear theory as Eq. (13).

\[
E(t) = E_i(\frac{t}{t_0})^\beta \quad (17)
\]

where \( E_i \) and \( \beta \) are material constants, and \( t_0 \) is standard time to give dimension of elasticity for \( E_i \). For simplicity, \( t_0 \) is chosen as 1 sec and is omitted from now. The value of \( \beta \) is a measure for rate of stress relaxation. The material with smaller \( \beta \) is more elastic, and its rate of stress relaxation is lower. Since material is purely elastic when \( \beta = 0 \) and is purely viscous when \( \beta = 1 \), the available value of \( \beta \) ranges from 0 to 1. The relaxation modulus of some polymeric solids is approximately described by a power law model. Equating the right hand side of Eq. (10) to zero, \( \beta \) is given as the solution of the following transcendental equation:

\[
(1-\beta) - (1-\beta)(t-t_1)^{1-\beta} - r(t-t_2)^{1-\beta} = 0 \quad (18)
\]

If unloading follows immediately after loading \( t_r = 0 \), \( t_0 \) is obtained by equating \( t_1 \) to \( t_2 \) in the above equation, and

\[
\epsilon_0 \bigg|_{t_r=0} = \frac{(1+r)^\beta}{(1+r)^\beta - 1} t_1 \quad (19)
\]

where \( p = 1/(1-\beta) \). Hence, \( t_3 \) is in direct proportion to \( t_1 \), independent of \( E_i \), and depends on \( \beta \). \( \epsilon_0 \) and U.E.R. can be written in the following concise forms:

\[
\epsilon_0 \bigg|_{t_r=0} = \frac{v_1 t_1}{(1+r)^p - 1} \quad (20)
\]

\[
\text{U.E.R.} \bigg|_{t_r=0} = \frac{r}{(1+r)^p - 1} \quad (21)
\]

U.E.R. is independent of \( t_1 \), that is, of total strain \( \epsilon_1 \). When \( t_r = 0 \) and \( r = 1 \),

\[
\text{U.E.R.} \bigg|_{t_r=0} = \frac{1}{2p-1} \quad (22)
\]

Then, U.E.R. is independent of strain rate \( v_1 \).

Differentiating Eq. (22) by \( \beta \) yields,

\[
\frac{\partial}{\partial \beta} (\text{U.E.R.})_{t_r=0} = \frac{r(1+r)^p \ln(1+r)}{(1-\beta)^2(1+r)^p - 1} < 0 \quad (23)
\]

Hence, \( \text{U.E.R.} \bigg|_{t_r=0} \) of the material with large \( \beta \) is low. Inserting \( t_0 = t_3 - t_2 \) into Eq. (18) yields

\[
t_r(1+t_r+t_1)^{1-\beta} - t_r^\beta(1+t_1^\beta) = 0 \quad (24)
\]

Expanding the left hand side of Eq. (24) in a series and assuming \( t_r \) is sufficiently larger than \( t_1 \) and \( t_2 \), the following approximated equations can be obtained:

\[
t_r = \left( 1 - \frac{\beta}{r} \right) t_1^{1-\beta} \quad (25)
\]

\[
\text{U.E.R.} = \left( 1 - \beta \right) \left( \frac{t_1^\beta}{t_r^\beta} \right)^{1-\beta} \quad (26)
\]

If \( t_r \) is sufficiently large, U.E.R. is an increasing function of \( t_1 \), namely of \( \epsilon_1 \), and a decreasing function of \( t_r \), since \( 0 < \beta < 1 \) for a viscoelastic power law material.

Differentiating Eq. (26) with respect to \( \beta \) yields the following equation,

\[
\frac{\partial}{\partial \beta} (\text{U.E.R.})_{t_r=0, t_3}
\]

\[
\text{U.E.R.} = \frac{\ln \left( \frac{t_1}{t_r} \right)}{(1-\beta)^2(1-\beta)^p - 1} \quad (27)
\]

If \( r \leq 1 \), the above equation is negative so that U.E.R. of the material of large \( \beta \) is low.

The exact distribution function \( H(\ln \tau) \) of relaxation time \( \tau \) of the linear power law material is as follows:

\[
H(\ln \tau) = \frac{E_1}{\Gamma(\beta)} \tau^{\beta-1} \quad (28)
\]

where \( \Gamma(\beta) \) is a gamma function. Thus the curve of \( \log H \) vs. \( \log \tau \) shows a straight line of a negative slope \( -\beta \).

Differentiating Eq. (28) by \( \beta \) yields

\[
\frac{\partial}{\partial \beta} H(\ln \tau) = \frac{E_1^{\beta-1}}{\Gamma(\beta)} \left[ \ln \frac{\tau^{\beta} - 1}{2\beta} + 2 \int_0^\infty \frac{x}{(x^2 + \beta^2)(e^{\beta x} - 1)} dx \right] \quad (29)
\]
Since $0 < \beta < 1$, the right hand side of Eq. (29) is positive in the range of $r \geq 1$. Thus $H(\ln r)$ is an increasing function of $\beta$ in that range. If $E_1$ is either equal to or greater than others, a material of larger $\beta$ has always higher $\log H$ of wedge type in the range $r \geq 1$. Since the value of $E_1$ does not contribute to $U.E.R.$, a material having higher $\log H$ in that range and a steeper slope of wedge shows smaller $U.E.R.$.

Solving Eq. (18) by the method of regular falsi, examples of calculated $U.E.R.$ are shown in Fig. 2. In the numerical calculation, we set $r = 0.1$, and the adopted values of $\beta$ (0.103 and 0.091) correspond to the values of the high- and the low-density polyethylene, respectively. The results in Fig. 2 explain the above analysis. Namely, when $t_r = 0$, $U.E.R.$ is independent of $E_1$ and constant. When $t_r > 0$, $U.E.R.$ increases with increased $E_1$ and decreased $t_r$. The model of small $\beta$ shows high $U.E.R.$.

2.3 Delayed elastic recovery of linear power law model

If $E(t)$ is expressed by Eq. (17), the solution of Eq. (14) is

$$f(t) = \sin \frac{\beta \pi}{E_1 \pi} t^\beta$$

From Eqs. (13), (17) and (30), the strain in the delayed elastic recovery process is

$$\varepsilon(t') = -\sin \frac{\beta \pi}{E_1 \pi} \left[ v_1(t_3-t_1) \int_0^1 \left( \frac{1+t'}{t_3-t_1} \right)^\frac{1}{u} -1 \right]^u \frac{1}{u} \frac{d u}{u}$$

$$-v_1(t_3-t_1) \int_0^1 \left( \frac{1+t'}{t_3-t_1} \right)^\frac{1}{u} -1 \right]^u \frac{1}{u} \frac{d u}{u}$$

where $t' = t-t_3$. $\varepsilon(t')$ is independent of $E_1$ as in case of $\varepsilon_e$. Some numerical results from Eq. (31) are shown in Figs. 3–5. For calculation, values of $v_1$ and $v_2$ are set as 1/60 and 1/600 (1/sec), respectively, and values of $\beta$ is are equal to those in Fig. 2. Fig. 3 shows that the recovery curve for the model of small $\beta$ is low, though this model has high $U.E.R.$ From Fig. 4, it is recognized that the rate of delayed recovery increases with the increase of $E_1$. As clarified by Fig. 5, while the recovery curve rapidly converges to zero when $t_r = 0$, the amount of recovery markedly decreases but the rate of recovery increases when $t_r = 0.5$ hr. Further increase of $t_r$ brings about small change of recovery and its rate.

The fractional delayed elastic recovery is defined as follows:

$$D.E.R. = \frac{\varepsilon(t') - \varepsilon(t') \times 100 - \varepsilon(t') \times 100}{\varepsilon_1}$$

In Fig. 6 $D.E.R.$ at 10,000 sec is shown. While $D.E.R.$ at $t_r = 0$ is independent of $E_1$ as the case of $U.E.R.$ in Fig. 2, $D.E.R.$ when $t_r > 0$ decreases with the increase of $E_1$ and the decrease of $t_r$. $D.E.R.$ of a model of smaller $\beta$ is greater than that of the model of larger $\beta$, and this result is directly opposite to the case of $U.E.R.$ The fractional total elastic recovery $T.E.R.$ ($= U.E.R.+D.E.R.$) at 10,000 sec is independent of $E_1$. In Fig. 7, $T.E.R.$ at that time is plotted against $t_r$. It is recognized that $T.E.R.$ exponentially decreases with the in-
crease of $t_r$, and its dependence on $t_r$ is appreciable if $\beta$ is large.

### 2.4 Unloading elastic recovery of a nonlinear power law model of exponential type

It seems that the above analytical results are different from the behavior of actual polymer because of the limited applicability of the linear theory. It has been reported that the elongational viscoelastic behavior of polyethylene could be approximately described by Eq. (1), and the experimental equation for $a_e$ was given by\(^{10,11}\)

$$a_e = \begin{cases} 1 & \epsilon \leq \epsilon_0 \\ \exp[-\alpha(\epsilon - \epsilon_0)] & \epsilon > \epsilon_0 \end{cases} \quad \cdots \cdots (33)$$

where $\epsilon_0$ was linear limit of strain, and $\alpha$ a constant regarded as measure of nonlinearity. The material of larger $\alpha$ represents the stronger nonlinear behavior. Then $\alpha$ is named a degree of nonlinearity. The power law model having $a_e$ given by Eq. (33) is termed as a nonlinear power law model of exponential type. For simplicity it is assumed that $\epsilon_0 = 0$, and $a_e$ is described only by an exponential function of $\epsilon$ in the range of application of Eq. (1). The following transcendental equation can be derived from Eq. (11),

$$\int_0^{t''} \left( 1 + \frac{1}{r} + \alpha \nu_1 t_r - \frac{1}{r} \exp(-\alpha \nu_1 t'') \right) d\nu = \exp(-\alpha \nu_1 t'_2) \int_0^{t''} \exp(\alpha \nu_1 u) - 1 \right)^{-\beta} d\nu = 0$$

where $t'' = t - t_2$. From the solution of Eq. (34), $t_3 = t_3'' + t_2$ can be obtained.

The numerical solution $t_3''$ of Eq. (34) was obtained by a digital computer FACOM 230-28 at Fukui University by the regular falsi method. The values of U.E.R. calculated from Eq. (4) into which results of $t_3$ are substituted are shown in Fig. 8. For calculation, the values of $V_1$ and $V_2$ are put as the same as those in Fig. 3, and $\alpha$ and $\beta$ are equal to those obtained in a previous paper\(^{10}\) for polyethylene (Table 2.). When $t_r = 0$, U.E.R. decreases with the increase of $\epsilon_1$, and differs from the result by the linear theory. Its dependence on $\epsilon_1$ is remarkable for a model of stronger nonlinearity. When $t_r > 0$, U.E.R. $-\epsilon_1$ curves have the maximum near $\epsilon_1 =
2-3% and do not monotonically increase with the increase of $\varepsilon_1$ in contrast to Fig. 2 predicted from the linear theory. U.E.R. of a model of larger $\alpha$, i.e. of stronger nonlinearity, decreases appreciably down from that of a linear model. From there results, it is recognized that a degree of nonlinearity $\alpha$ has an effect to reduce U.E.R. of the linear model and that an increase in $\varepsilon_1$ enhances effect of $\alpha$. Therefore, the shape of U.E.R. curves is changed from Fig. 2 to Fig. 8.

3. Experiment

3.1 Test specimen

Low density polyethylene Yukalon K-3212 (Mitsubishi Petrochemical Co.) and high density polyethylene Hizex 5000-S (Mitsui Petrochemical Co.) were used as raw material. Pellets of these polymers were molded into sheets of thin film by hot press. The molding and annealing conditions listed in Table 1 are the same as those used in previous papers[10-12,24]. Densities and degrees of crystallinities of film obtained in a previous paper are listed in Table 2 as a reference. Some constants of experimental equations such as $E_1$, $\alpha$, $\beta$ and $\varepsilon_0$ determined from previous data[10] for stress relaxation are also listed in Table 2. The values of $\alpha$ and $\beta$ for Hizex are greater than those for Yukalon. Hence, the nonlinearity of Hizex is strong and its rate of stress relaxation is high.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Hizex 5000-S</th>
<th>Yukalon K-3212</th>
</tr>
</thead>
<tbody>
<tr>
<td>melting</td>
<td>at 180°C for 10 min.</td>
<td>at 140°C for 10 min.</td>
</tr>
<tr>
<td>solidifying</td>
<td>at 125°C for 1 hr.</td>
<td>at 105°C for 1 hr.</td>
</tr>
<tr>
<td>cooling</td>
<td>gradually to room temp.</td>
<td>same as Hizex</td>
</tr>
<tr>
<td>annealing</td>
<td>at 125°C for 2 hr.</td>
<td>at 105°C for 2 hr.</td>
</tr>
</tbody>
</table>

3.2 Method

The film was cut into strips of length 10 cm and of width 1 cm. The specimens were clamped in an Instron tensile testing machine (MODEL TM-M) with the gauge length of 5 cm between clamps. The cross head speeds when loading and unloading were chosen such that the strain rates of $v_1 = 1/60$ (sec$^{-1}$) and $v_2 = 1/600$ (sec$^{-1}$) were obtained. These strain rates were used in the former theoretical calculations. The holding times at a constant strain before unloading were 0, 0.5, 1 and 2 hr. The method for measurement of deformation in delayed elastic recovery is as follows: The maximum and the minimum load cams attached on a recorder of the Instron tester were adjusted to 1 g and 0 g, respectively. After loading followed by stress relaxation, the cross head moves up at the speed of $V_2$ to reduce the load to the adjusted minimum. It stops there and holds the sample at a constant strain until the load due to constrictive stress increases to the adjusted maximum. Then, the cross head again moves up automatically to reduce the load to the minimum limit and stops for a while. These processes are repeated. The displacement of cross head is detected by a differential transformer and its amplified output is recorded on another pen-recorder. The slow speed of crosshead $V_2$ was selected to...
4. Experimental Results and Discussions

In Fig. 9 are shown experimental results of $U.E.R.$ with calculated curves from the nonlinear theory. All of the observed $U.E.R.$ decrease with the increase of strain $\varepsilon_1$, and this tendency is marked when $t_r = 0$. While the observed $U.E.R.$ appreciably decreases by increasing $t_r$ from 0 to 0.5 hr, further increase in $t_r$ only reduces the level of $U.E.R.$ $U.E.R.$ for Yukalon of smaller $\alpha$ and $\beta$ is greater than that for Hizex, and more weakly depends on $\varepsilon_1$. Those tendencies are qualitatively similar to the calculated results from a nonlinear power law model. The observed curves are all lower than the calculated curves, and the discrepancy between both curves is larger for Hizex of stronger nonlinearity than for Yukalon. The reasons for these results may be explained as follows: For calculation, it is assumed that the function $a(\varepsilon)$ is reversible even when the process changes from loading to unloading and is independent of strain history and depends on the value of strain. However, in practice, unrecoverable deformation, namely plastic deformation, arises in process of loading and stress relaxation. Therefore, it is considered that $a(\varepsilon)$ at unloading may disagree with that at loading, and the observed $U.E.R.$ may be less than the calculated values. Also, it is supposed that the effect on $a(\varepsilon)$ may be much larger for Hizex than for Yukalon because of its large plastic deformation.

Figure 10 shows the delayed elastic recovery when $t_r = 2$ hr. The rate of delayed recovery becomes high as $\varepsilon_1$ increases, and its tendency is marked for curves of Yukalon. This dependency on $\varepsilon_1$ is qualitatively similar to that of the calculated curves for a linear power law model shown in Fig. 4. While the rate of recovery for Yukalon of small $\beta$ is higher than that for Hizex of large $\beta$, the rate of recovery in the calculated curves is high for the model of large $\beta$. It is difficult to explain this contradiction clearly, because the delayed recovery can not be calculated from the nonlinear model. As stated before, the degree of nonlinearity $\alpha$ has an effect to reduce $U.E.R.$ of a linear model and its effect increases as $\alpha$ increases. From these analogical considerations, the following inference may be drawn. The delayed recovery of polyethylene, which represents nonlinear behavior, may be less than the value predicted from a linear theory. This difference may be larger for Hizex because of high nonlinearity. Therefore, the rate of recovery for Hizex may be decreased compared with Yukalon. Also, the plastic deformation, which arises in process of loading and stress relaxation, may have some effect to reduce delayed elastic recovery.

In Fig. 11 the effect of $t_r$ on the delayed recovery for Hizex is shown. If $t_r$ increases from 0 to 0.5 hr, the rate of recovery rapidly increases. However, further increase in $t_r$ slightly lessens the amount of recovery and does not change the rate of recovery. These trends are similar to the calculated curves in Fig. 2. However, the observed curves show slower recovery than calculated curves.

Figure 12 shows the effect of $\varepsilon_1$ on $D.E.R.$ at 2 hr after unloading. When $t_r = 0$, $D.E.R.$ increases with the increase...
of $\varepsilon_1$, and this trend is considerable for Hizex of strong nonlinearity. When $t_r > 0$, D.E.R. is independent of $\varepsilon_1$ and $t_r$. D.E.R. of Yukalon of low $\beta$ is greater than that of Hizex. These experimental results disagree with the calculated results from a linear theory. This contradiction may be due to nonlinearity.

In Fig. 13 the dependency of T.E.R. on $t_r$ is shown. T.E.R. decreases exponentially with $t_r$. It is different from the results of linear calculation, and shows the T.E.R. dependency on $\varepsilon_1$. Hizex shows more remarkable effect of $\varepsilon_1$, and the poorer recovery than Yukalon.

5. Conclusion

It is concluded theoretically and experimentally that the material having less stress relaxation, namely is more elastic, and having weak nonlinearity, shows good elastic recovery during unloading. The model and the nonlinear theory which are used in this paper are very simple and do not always hold to other materials. But it is considered that the above conclusion may be generally applied to any model theory. While, the delayed elastic recovery could not be predicted from the linear theory because of the strong nonlinearity of materials. However, there are no proper theories to predict nonlinear delayed recovery. Hence, further development in phenomenology of nonlinear viscoelasticity is hoped.

Reference