# NON-LINEAR VISCOELASTIC BEHAVIOR OF POLYETHYLENE

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Abstract—The response of viscoelastic solids to quasi-static loading, under conditions where non-linear theories have to be used, are discussed. Creep measurements of tension and of torsion in polyethylene specimens are described. Step loading was used and the deformations were measured optically by means of travelling microscopes for the tension experiments and by the reflection of light beams from mirrors for the torsion experiments. The deformations were too large for the theory of linear viscoelasticity to hold and the constitutive relations used were of multiple integral form; some of the kernels involved have been determined. It was found that for the loading range used, two kernels were sufficient to describe pure shear deformations and three kernels were required for tension. Some experiments in which two loading steps were applied are also described and discussed.

# **INTRODUCTION**

THE mathematical description of linear viscoelastic behavior has received considerable attention in recent years (e.g. Leaderman  $[1]$ , Gross  $[2]$ , Ferry  $[3]$ ). The underlying assumption in these treatments is that linear superposition obtains. Thus ifin the one-dimensional case, the strain response of the material to a step loading is known as a function of time, i.e. the creep function is known, the response of the material to any loading history can be expressed as a convolution integral in which the kernel is this creep function. This is one form of Boltzmann's Superposition Principle; alternative forms involve kernels corresponding to a step function in strain or a delta function in either stress or strain.

Another approach is to consider the response of the material to a stress which varies sinusoidally with time. For a linear viscoelastic solid the strain will then also vary sinusoidally with time at the same frequency but will lag behind by a phase angle, which in general varies with the frequency of the oscillation. If the ratio of the stress amplitude to the strain amplitude, and this phase angle are known for all relevant frequencies, the response to any stress history can be expressed as a Fourier integral.

Thus in linear viscoelasticity there is a wide choice of formulations and as shown by Gross [2] these are all mathematically equivalent so that experimental measurements of one quantity (e.g. the creep function) can be transformed numerically to give values of any of the other viscoelastic functions.

Now most viscoelastic solids are linear for sufficiently small strains, and the problem we are concerned with in this paper is how to extend the theory to describe the non-linearities which occur for larger strains. Experimental measurements for one viscoelastic solid namely polyethylene are described and discussed in this connection.

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It should be noted that for polymers such as polyethylene the transition from linearity to non-linearity is quite smooth, or to state the situation more realistically the description "linear" and "non-linear" are to a large extent arbitrary. There are always deviations from linearity and the "non-linear" region might be defined as that region where the deviations from linearity can no longer be overlooked and this is necessarily a somewhat subjective judgment. The smallest non-linearity which is considered significant will depend on the one hand on how accurately and reproducibly measurements can be made, and on the other on the use to which the theory is being put, namely whether it is being used simply to predict the strains produced by given stresses or to lead to a better understanding of the mechanical response of the material with a view to establishing relations between its mechanical behavior and its microscopic structure.

**In** the present investigation what was required was an extension of linear theory to describe the stress-strain behavior of viscoelastic solids at strains where the deviations from linear viscoelastic behavior are not too large, i.e. where the non-linear terms could be regarded as corrections to the linear terms. There are a number of alternative mathematical descriptions which will fulfill this requirement, e.g. the use of non-linear differential equations of the form  $P\sigma = Q\epsilon$  where *P* and *Q* are non-linear differential operators with respect to time and  $\sigma$  and  $\varepsilon$  are the stress and strain respectively.

The method employed in this paper was to use a multiple "integral" representation of the stress-strain behavior. This type of representation has been discussed by Green, Rivlin and Spencer [4-6J and has been used successfully by Ward and Onat [71, Hadley and Ward [8J, and Onaran and Findley [9] to describe the non-linear mechanical response of polymers.

Ward and Onat [7] studied the behavior of oriented polypropylene monofilament in tension, and showed that the response could be represented with reasonable accuracy by the sum of a linear and a third order hereditary functional. Onaran and Findley [9] studied the response of polyvinyl chloride polymer and showed that a multiple integral functional relationship could be used to describe the results when first, second and third order stress terms were employed.

More recent work by Hadley and Ward [8] on several polypropylene fibers of different molecular structures has shown that with increasing stress levels and increasing time, more than two functionals are required to describe the observed creep and recovery phenomena of these materials.

Lockett [10] has considered how the material functions for general triaxial loading can be determined experimentally for a material whose response can be adequately expressed by a constitutive equation involving multiple integrals of the first, second and third orders. Thus, in a one-dimensional test in which single step loadings are applied the values of the kernels can be found along the axis of symmetry of their arguments | i.e.  $J(t)$ ,  $K(t, t)$  and  $L(t, t, t)$ ]. These values differentiate between the "linear" and "non-linear" contributions to the deformation and (for the range of stresses and times covered) enable the strain produced by an applied step load to be determined.

The determination of the kernels for arguments which are not all equal corresponds to the study of the interaction between step loads applied at different times. An exact determination of such interactions involves very accurate and very laborious measurements, and the present investigation did not provide reliable quantitative determination of such kernels. A number of semi-qualitative results of the nature of such interactions were, however, obtained and these help us to obtain a better understanding of the nature of the non-linear response of polyethylene.

#### **THEORETICAL**

Let  $X_i$ , be the coordinates of a generic particle of a body in a fixed rectangular coordinate system at zero time and let  $x_i(\tau)$  be the coordinates at some later time  $\tau$ . A simple material is then defined as one for which the stress at that particle at time *t* depends upon the displacement gradients  $F_{ij} = \partial x_j / \partial X_i$ , at all times up to and including the time *t*. The form of the dependence of the stress upon the history of the deformation gradients is subject to two types of restrictions. First a simultaneous time-dependent rotation of the body and the reference system may be assumed not to change the stress components, and second if the body in its undeformed state possesses any symmetry properties, these will also restrict the form of the constitutive relations. In particular if the material in its initial state is isotropic this will impose severe restrictions on the form the stress-strain relation can take. The nature of these restrictions has been discussed by Green, Rivlin and Spencer [4-6]. Frechet (11] has shown that continuous non-linear functionals may be expressed to any desired degree of accuracy as a series of multiple integrals. Spencer and Rivlin [12] have found that for an initially isotropic material the number of these integrals may be reduced to five.

These constitutive relations give the stress for known histories of the deformation gradients and are appropriate for the analysis of stress-relaxation experiments, i.e. experiments where after some time *t* the deformation is kept constant and the changes in the stress components measured. The experiments carried out in the present investigation were concerned with creep measurements where the stress is maintained constant and the strains are measured. To analyse such tests an inverted set of constitutive relations where the deformation is expressed in terms of the stress history is required. Unfortunately there is no method ofinverting these relations and we must instead start from the basic assumption that the strains at any time *t* can be expressed as a function of the values of the stress at all times up to and including the time *t.* The difficulty, however, then arises that this stress functional also involves the displacement gradients, since in a fixed coordinate system the stress will change as a result of the rotation of the body.

We will not attempt to deal with this general problem here. Instead, if we confine ourselves to small rotations we may attempt to describe the non-linear behavior of the material by the use of stress-strain relations in a form similar to that used by Green and Rivlin with the strains expressed as the sum of a number of hereditary integrals describing the stress history. The material is assumed to behave in a linearly viscoelastic manner for small deformations so that the first integral will be the Boltzmann type used in the classical theory of linear viscoelasticity.

This follows the procedure used by Ward and Onat [7] and Onaran and Findley [9] and if we confine ourselves to only three multiple integrals the stress-strain relation as given by Lockett  $[10]$  may be written in matrix form as

$$
\mathbf{E}(t) = \int_{0}^{t} \left[ \mathbf{I} \psi_{1} T_{1} + \psi_{2} \mathbf{M}_{1} \right] d\tau_{1} + \int_{0}^{t} \int_{0}^{t} \left[ \mathbf{I} \psi_{3} T_{1} T_{2} + \mathbf{I} \psi_{4} T_{12} + \psi_{5} T_{1} \mathbf{M}_{2} \right] d\tau_{1} d\tau_{2} + \int_{0}^{t} \int_{0}^{t} \int_{0}^{t} \left[ \mathbf{I} \psi_{7} T_{123} + \mathbf{I} \psi_{8} T_{1} T_{23} + \psi_{9} T_{1} T_{2} \mathbf{M}_{3} + \psi_{10} T_{12} \mathbf{M}_{3} \right] d\tau_{1} d\tau_{2} d\tau_{3}
$$
\n
$$
+ \psi_{11} T_{1} \mathbf{M}_{2} \mathbf{M}_{3} + \psi_{12} \mathbf{M}_{1} \mathbf{M}_{2} \mathbf{M}_{3} \right] d\tau_{1} d\tau_{2} d\tau_{3}
$$
\n(1)

where

$$
\mathbf{E} = ||E_{ij}|| \qquad 2E_{ij} = \frac{\partial x_k}{\partial X_i} \frac{\partial x_k}{\partial X_j} - \delta_{ij}
$$
  

$$
T_{\alpha} = tr\dot{\mathbf{P}}(\tau_{\alpha}) \qquad T_{\alpha\beta} = tr[\dot{\mathbf{P}}(\tau_{\alpha})\dot{\mathbf{P}}(\tau_{\beta})]
$$
  

$$
T_{\alpha\beta\gamma} = tr[\dot{\mathbf{P}}(\tau_{\alpha})\dot{\mathbf{P}}(\tau_{\beta})\dot{\mathbf{P}}(\tau_{\gamma})] \qquad \mathbf{M}_{\alpha} = \dot{\mathbf{P}}(\tau_{\alpha})
$$
  

$$
\dot{\mathbf{P}}(\tau) = \frac{d\mathbf{P}}{d\tau}
$$

R denotes the rigid body rotation part of the displacement gradients,  $\mathbb{R}^T$  is the transpose of **R** and the  $\psi$ 's are functions of  $t - \tau_{\alpha}$  in the following way:  $\psi_1$  and  $\psi_2$  are functions of  $t-\tau_1$ ;  $\psi_3, \ldots, \psi_6$  are functions of  $t-\tau_1$  and  $t-\tau_2$ ;  $\psi_7, \ldots, \psi_{12}$  are functions of  $t-\tau_1$ .  $t - \tau_2$  and  $t - \tau_3$ . These are the functions that characterize the mechanical behavior of the material. In order to determine these functions a large number of different tests would have to be carried out; these are given in tabulated form by Lockett.

It may be worth mentioning here that the reason for using three multiple integrals rather than two is because of the assumed initial isotropy; for isotropic materials the response to a shearing force is symmetrical, i.e. a shear stress in the positive x direction will produce a shear deformation which is equal in magnitude and opposite in sign to a shear deformation produced by a similar shear stress in the negative  $x$  direction. This means that there can be no even terms in the stress-strain relations, and the multiple integrals of even order must vanish. Thus a constitutive relation involving only the first and second integrals cannot describe non-linear behavior in shear, and a third integral must be included.

#### TENSION TESTS

We consider here a one-dimensional deformation in the x direction where only the x coordinate is involved. R is then the unit matrix so that P is reduced to  $\sigma$  which has only one non-zero component,  $\sigma_{11}$ . If we now denote  $\sigma_{11}$  by  $\sigma$  and the  $E_{11}$  component of the E matrix by *E,* equation (1) may be written as

$$
E(t) = \int_0^t J(t-\tau)\dot{\sigma}(\tau) d\tau + \int_0^t \int_0^t K(t-\tau_1, t-\tau_2)\dot{\sigma}(\tau_1)\dot{\sigma}(\tau_2) d\tau_1 d\tau_2
$$
  

$$
\int_0^t \int_0^t \int_0^t L(t-\tau_1, t-\tau_2, t-\tau_3)\dot{\sigma}(\tau_1)\dot{\sigma}(\tau_2)\dot{\sigma}(\tau_3) d\tau_1 d\tau_2 d\tau_3
$$
 (2)

where J, *K* and *L* characterize the mechanical properties of the material. If we define the strain as

$$
\varepsilon = \frac{\partial u}{\partial X}, \qquad u = x - X
$$

then from the definition of *E* we get

$$
E = \varepsilon + \frac{1}{2}\varepsilon^2 \tag{3}
$$

#### TORSION TESTS

Consider a thin walled hollow circular cylinder whose axis is in the  $x_1$  direction in a fixed rectangular Cartesian coordinate system. As a result of a torque T, that is applied to the cylinder, every particle in it moves in the  $x_2x_3$  plane and the motion depends only on the coordinate  $x_1$ . Now **R** here is not the unit matrix, since this type of deformation involves a rigid body rotation.

If we denote the angle of shear by  $\gamma$  then for deformations where  $\gamma < 0.10$  we ignore higher order terms, and we then find that

$$
\mathbf{R} = \begin{pmatrix} 1 & -\frac{\gamma}{2} & 0 \\ \frac{\gamma}{2} & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}
$$
 (4)

If we denote the shear stress  $\sigma_{12}$  by  $\tau$  then the stress matrix P, measured in a coordinate system which rotates with the rigid body motion, is given (after neglecting  $\gamma^2$  next to 1) by

$$
\mathbf{P} = \begin{pmatrix} \tau \gamma & \tau & 0 \\ \tau & -\tau \gamma & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{5}
$$

By substitution of P in (1) and neglecting  $y^2$  next to 1, we obtain the shear component of the strain  $E_{12} = \gamma/2$ .

#### EXPERIMENTAL

The principle of the experimental method in creep tests is to apply a constant stress and measure the resulting deformation as a function of time. There are three types of measuring techniques; these may be classified as mechanical, electrical, and optical and all three methods have been used extensively in creep measurements. With soft materials, such as plastics, there are two important sources of error; these are (a) mechanical interaction between the measuring device and the specimen and (b) slip between the ends of the specimen and the grips in the loading apparatus. Both these errors can most easily be avoided by the use of an optical method for measuring the deformations. Thus observations can be made without any mechanical contact with the specimen and the effects of slip in the specimen holders can be avoided by observing the separation between two points in the specimen, remote from the holder.

In the tension tests, cylindrical specimens ofsolid cross-section were employed and the position oftwo points 2·5 in. apart were observed with two travelling microscopes. Displacements as small as  $2 \times 10^{-4}$  in. could be detected reliably by these means.

In the torsion tests  $(Fig. 1)$  the specimens were in the form of thin-walled hollow circular cylinders so that the torsional stress was effectively uniform across the specimen. The preparation of these specimens was considerably more difficult than those used in the tension experiments, since uniform wall thickness and smooth surface finish was required. The bottom of the specimen was here held fixed and an axial torque was applied to the top of the specimen by a system of pulleys and weights. Two small mirrors which were cemented to thin pins were attached to two points on the specimen, A and B. The line AB was vertical and a  $\frac{1}{2}$  in. in length. A beam of light, the cross section of which was a long



FIG. 1. Tension-torsion test apparatus.

narrow rectangle, was reflected by the mirrors onto a scale, so that two line images were produced. The scale was in the shape of a circle with a radius of  $180/\pi$  in., with the specimen at its center so that each inch on the scale corresponded to one degree of rotation. Thus the angle ofrotation ofthe cross-section-A with respect to the cross-section-Bcould be obtained directly. The light beam was obtained by letting the light from a concentrated arc lamp (Sylvania *C25jDC)* fall on the slit formed by the edges of two razor blades. During the first two minutes after application of the load the motion of the two cross sections was recorded photographically, after which visual observations were made.

The torsion machine was designed to enable combined tension-torsion tests to be carried out. All the moving parts had to have very low frictional losses and special bearings were obtained for this purpose. Both in the tension and the torsion tests it was necessary for the load to be released uniformly. In the tension machine this was done by a hydraulic jack and in the torsion machine it was done manually.

Since the interpretation of the results required comparison of a number of tests it was important to be able to carry out experiments on identical specimens and be sure that at the beginning of the test the material was in the same unstrained condition. It was found that the polyethylene specimens did not return to their original shape after large deformations, at least not within a reasonable time, and a number of different methods of annealing were therefore tried Annealing in air resulted in the specimens becoming brittle and colored, presumably due to oxidation, and a nitrogen atmosphere was therefore used. It was found that annealing for 12 hr at a temperature of  $100^{\circ}$ C was sufficient to bri  $\cdot$  the material back

to its initial state. The effectiveness ofthe annealing was tested by whether or not reproducible results could be obtained in the stress-strain measurements. Each specimen was annealed a number of times at different stages of its machining and then again after each test.

The first step in the investigation was to carry out simple creep tests under constant loads, so that

$$
\sigma(t) = c_i H(t) \tag{6}
$$

where  $H$  denotes the Heaviside function

$$
H(t) = \begin{cases} 0 & t < 0 \\ 1 & t \ge 0 \end{cases}
$$

The strain response in tension given by (2) is thus reduced to

$$
E(t; i) = c_i J(t) + c_i^2 K(t, t) + c_i^3 L(t, t, t)
$$
\n(7)

and in torsion it is

$$
E_{12}(t; i) = c_i M(t) + c_i^3 N(t, t, t)
$$
\n(8)

Equation (7) can be written in a formal way as

$$
y_i = a_{ij}z_j \qquad (j = 1, 2, 3)
$$
  
\n
$$
y_i = E(t; i) \qquad a_{ij} = (c_i)^j
$$
  
\n
$$
z_1 = J(t), \qquad z_2 = K(t, t), \qquad z_3 = L(t, t, t)
$$
  
\n(9)

In order to determine the three unknown  $z_j$ , we have to carry out three experiments with three different loads, thus in  $(9)$  i also has the range 1, 2, 3. The solution of  $(9)$  is

$$
z_j = a_{ji}^{-1} y_i \tag{10}
$$

where  $a_{ii}^{-1}$  is an element in the inverse matrix of  $a_{ii}$ .

If sufficiently accurate experimental data from creep experiments are available the kernels  $z_1$ ,  $z_2$ , and  $z_3$  can be calculated from equation (10).

It was found that the unsmoothed creep measurements gave reliable values for  $z_1$ but that the scatter in the values of  $z_2$  was large and the values of  $z_3$  were even less reliable. In order to obtain meaningful results it was clearly necessary to "smooth" the experimental data. Instead of doing this, it was decided to approximate the observed results with an analytical curve which fell within the experimental scatter. Values of  $y_i$  could then be computed to any degree of accuracy from the analytical expression and used to solve equation (9). The sole justification for this numerical procedure was that it yielded kernels which described the experimental results well, and this will be discussed in the next section of this paper. An analytical curve which was found to fit the experimental observations was of the form

$$
\log f = a + b \log t \tag{11}
$$

where

$$
f = \varepsilon - h/\varepsilon^2. \tag{12}
$$

 $(h)$  is a disposable parameter which is chosen to make the  $log$ -log plot of f against *t* as close to a straight line as possible.) To obtain the values of  $\varepsilon$  from corresponding values of  $f$  we must solve the cubic equation (12) and take the appropriate root.

The calculations for the torsion tests were the same as for the tension tests. The only difference is that for torsion we need only two creep curves in order to determine the two kernels, whereas in tension we need three creep curves for the three kernels.

Experiments involving two-step loading were then carried out, the accuracy of the tests was not sufficiently high for reliable quantitative results to be obtained. Some qualitative conclusions could however be obtained and these are discussed in the next section.

### **RESULTS**

The measurements of creep in both tension and torsion of polyethylene were fitted to equations of the form of  $(11)$  and  $(12)$ . The values of the strain at different times were then determined from the analytic curves and inserted in equation (7) to give the form of the kernels  $J(t)$ ,  $K(t, t)$  and  $L(t, t, t)$  for the tensile response of this material; equation (8) was used to find the kernels  $M(t)$  and  $N(t, t, t)$  for the response in torsion. The resulting curves are shown in Fig. 2.



FIG. 2. The kernels for tension and torsion tests.

It may be seen that the values of all five functions increase in magnitude with increasing time and that the second kernel  $K(t, t)$  is negative in sign over the whole time range. Figures 3 and 4 show a comparison between the experimental results in tension and the values calculated from the kernels given in Fig. 2. Figures 6 and 7 show a similar comparison for the torsion tests.

In each case only the results for the lowest and highest load used in the experiments are shown in the figures, the results were obtained for a number of intermediate loads and these showed a gradual transition between the two extremes given here.

In Fig. 3 it may be seen that for a tensile load of  $254 \frac{\text{lb}}{\text{in}^2}$  the viscoelastic behavior is almost linear and can be adequately described by the first kernel  $J$ , the contribution of  $K$ 



FIG. 3. Comparison between experimental and calculated strain (tension) ( $\sigma_0 = 253.6 \text{ lb/in}^2$ ).



FIG. 4. Comparison between experimental and calculated strain (tension) ( $\sigma_0 = 760.9$  lb/in<sup>2</sup>).

and L are small and cancel each other out. Similarly, Fig. 6 shows that for a torsion load of  $127 \frac{\text{lb}}{\text{in}^2}$  the material is behaving in an essentially linear viscoelastic manner and the contribution of the N kernel is very small.

Figure 4 shows the experimental results in creep for an initial load of  $760-91$ b/in<sup>2</sup>. Here the non-linearity is very pronounced and the deviation between the linear viscoelastic contribution, given by the  $J$  kernel, and the observed results may be seen to increase with increasing time. The calculated curve with all three kernels gives excellent agreement

# 392 J. M. LIFSHITZ and H. KOLSKY

with the experimental results except for the very highest strains  $($  >6 percent) where small deviations occur. These may be due to *the* fact that the load was kept constant so that the *stress* increased *with* decreasing cross-sectional area of the specimen. This effect. which becomes increasingly important as the strain increases. was not allowed for in the caJculations. It may be seen from Fig. 4 that although the third integral *(L kernel) contributes* much more to the calculated strain than second integral  $(K$  kernel) all three are required to predict the observed strain accurately. Attempts to describe the behavior purely in terms of *the* J and *L* kernels were found to give unsatisfactory agreement with the observed behavior. These results are in contrast with those found for oriented polyisopropylene by



FIG. 5. Deviation from linearity in tension tests.



FIG. 6. Comparison between experimental and calculated strains (torsion) ( $\tau_0 = 126.8 \text{ lb/in}^2$ ).

Ward and Onat  $|7|$  who were able to obtain satisfactory agreement with only two integrals  $(J \text{ and } L)$ .

Comparing Figs. 6 and 7 which give the torsional results we see that at the lower load  $\tau_0 = 126.8 \text{ lb/in}^2$  the nonlinearity is negligible whereas for the highest load  $\tau_0 = 527.1 \text{ lb/in}^2$ the contribution of the third integral, N, is quite large and its relative effect increases with increasing time.

It should perhaps be emphasized here that while the first kernels,  $J$  for tension and  $M$  for torsion, have been determined completely, the second kernel  $K$  has only been determined on the bisector of its arguments and the third kernels. L for tension and *N* for torsion. have only been found along the trisectors of their arguments.



FIG. 7. Comparison between experimental and calculated strains (torsion) ( $\tau_0 = 527 \cdot 1 \text{ lb/in}^2$ ).



FIG. 8. Deviation from linearity in torsion tests.

**In** order to illustrate the effect of increasing non-linearity with the time of application of the load, the creep results have been plotted in a different form in Figs. 5 and 8.

For a linear viscoelastic solid subjected to step loads  $\sigma(1)$ ,  $\sigma(2)$ , ...,  $\sigma(i)$  etc. at time  $t = 0$ . the strains  $E(t; 1), E(t; 2), \ldots, E(t; i)$ ... at any time *t* will be proportional to the loads, i.e.  $E(t; i)/\sigma(i)$  is constant. If we now define a function

$$
a(t;i) = \frac{E(t;i)\sigma(1)}{E(t;1)\sigma(i)}; \qquad i = 1, 2, 3, 4, 5
$$

This will be unity for all *t* and all values of i when the material is linearly viscoelastic. The non-linearity will be shown by deviation of this quantity from unity.

Figure 5 shows the function  $a(t)$  plotted as a function of time for four different tensile loads, a load of 253.6 lb/in<sup>2</sup> has been used to obtain the reference values  $\sigma(1)$  and  $E(t; 1)$ . Figure 8 shows a similar set of curves obtained from the torsion tests. It may be seen from Fig. 5 that the non-linearity for tensile loads increases rapidly not only with increasing load but also for anyone load with increasing time. This effect is not so marked in torsion tests where the slopes of the curves actually decrease slightly with increasing time of application of the load.

The difference between the stress fields in the tension and torsion tests lies in the presence of hydrostatic components in the former which are absent in the latter. The finite value of the *K* kernel in tension as well as the large differences in non-linear behavior between the two types of test must result from the presence of these hydrostatic components in tension, and presumably ultimately occur because of the density changes produced by these components.

The results of some two-step loading experiments are shown in Fig. 9. In the first set of experiments a constant tensile load  $\sigma_0$  was applied and a second load  $\sigma_1$  was added at



FIG. 9. The effect of prestraining the material on subsequent creep ( $\sigma_1 = 260.6 \text{ lb/in}^2$ ).

time  $t_1$ . A few tests were first carried out with fixed values of  $\sigma_0$  and  $\sigma_1$  and only the time  $t_1$  was varied. Some further tests were then carried out in which the value of  $\sigma_0$  was changed. In Fig. 9 two sets of observations are shown. In curves A, B, and C the value of  $\sigma_0$ is 353.5 lb/in<sup>2</sup> in D, E, F, and G  $\sigma_0 = 507.2$  lb/in<sup>2</sup>. In all cases  $\sigma_1 = 260.5$  lb/in<sup>2</sup>. In all the figures the strain  $\epsilon - \epsilon_0$  is plotted against time,  $\epsilon_0$  being the value the strain would have had at time *t* if the additional load had not been added.

It may be seen from Fig. 9 that with increasing time of preloading, i.e. with increasing  $t_1$ , the material becomes "stiffer" for additional loads, in other words the larger the prestraining the less the creep produced by a given additional load. Further by comparing, for example, curves B and G which had the same strain  $\varepsilon_0$  at the instant that the additional load  $\sigma_1$  was added, we see that the response for B is much smaller than that for G. Thus when we reach the same strain with a small load applied for a long time, the material is stiffer to additional loads than if the same strain is reached with a larger load for a shorter time.

This increased stiffening is presumably associated with the orientation of the long chain molecules along the direction of stretch, (the "cold-drawing" of polyethylene is an extreme example of this where an extremely stiff fiber is produced). The results given here indicate that the effectiveness of such orientation is not purely a question of the macroscopic strain produced but depends on the manner in which this was reached; it becomes more effective as the length of the loading time is increased.

In Fig. 10 the responses of a specimen to two double-step loads are compared. In both programs the final load was  $767.5 \frac{\text{lb}}{\text{in}^2}$ . For the experiment shown in curve A a load of



FIG. 10. The response to two double-step loading programs.

 $507·2$  lb/in<sup>2</sup> was applied and one minute later when the strain was  $2·15$  percent, a second load of  $260.5 \frac{\text{lb}}{\text{in}^2}$  was added. In the second experiment, curve B, a load of  $353.5 \frac{\text{lb}}{\text{in}^2}$ was applied and one hour later when the strain was again about 2 percent, the second load of 414<sup>.0</sup> Ib/in<sup>2</sup> was added. It may be seen that the strains would appear to be approaching the same limit but that a one hour delay is maintained between the two curves as creep progresses.

# **CONCLUSIONS**

The results of creep experiments in tension and torsion have been described and the experimental curves have been interpreted in terms of multiple integrals. The determination of the kernels from these experimental results inevitably involves a certain amount of curve-fitting and it may perhaps be held that this is all that such treatments can achieve. If this were so it could still be argued that a knowledge of these kernels summarizes in a very convenient form, the creep response of the material over a wide range of stresses and times. The authors, however, feel that a knowledge of the shape of these kernel functions throws considerable light on the way in which the material departs from linear viscoelastic behavior as the deformations become larger. Thus for example the very different nature of the nonlinearity in shear and simple extension is clearly seen from the analysis of the experimental results.

The experiments on two-step loading have also produced some interesting results on how the nature of the mechanical response changes with degree of preloading which is the essence of its non-linear response.

Clearly considerably more experimental work is required in this field before a better understanding of the complete non-linear response can be expected.

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Résumé—La réaction des solides viscoélastiques à des charges quasi-statiques, sous des conditions où des théories non-lineaires ont ete employees. est discutee. Des mesures de fluage de tension et de tension sur des specimen de polyéthylène sont décrites. Des chargements à étapes sont employés et les déformations mesurées optiquement au moyen de microscopes pour les experiences de tension et par la reflection de rayons de lumiere sur des mirroirs pour les expériences de torsion. Les déformations sont trop grandes pour que la théorie de viscoélasticité linéaire puisse se maintenir et les relations constitutives employées sont de forme multiple et intégrale; quelques un des noyaux élevés ont été déterminés. Il a été constaté que pour la gamme de chargements employés deux noyaux etaient suffisants pour decrire les deformations de cisaillement pur et trois noyaux etaient requis pour la tension. Quelques expériences dans lesquelles deux étapes de chargement étaient appliquées sont également décrites et discutées.

**Zusammenfassung-Es** wird beschrieben wie viskoelastische Flussigkeiten auf quasi-statische Belastung reagieren, wenn die herrschenden Bedingungen so sind, dass nichtlineare Theorien angewandt werden mussen, Kriechmessungen der Spannung und Verdrehung von Polyathylenmustern werden beschrieben, Belastung war stufenweise und die Verformung wurde optisch gemessen, Spannung mittels beweglichen Mikroskopes und Verdrehung mittels Spiegeln und reflektierter Lichtstrahlen. Die Verformungen waren fUr die Theorie der linearen Viskoelastizitat zu gross und die Materialbeziehungen waren in der Form von Vielfachintegralen; manche Kerne wurden bestimmt. Es wurde festgestellet, dass im gegebenen Belastungs-Bereich, zwei Kerne genügen, um Scherungs-Verformungen zu beschreiben, und drei Kerne für die Spannungs-Verformung, Weitere Experimente in denen zwei Belasungsstufen angewandt wurden werden auch beschrieben,

Абстракт-Рассматривается ответная реакция вязко-эластичных твёрдых тел на квази-статическую нагрузку при условиях, где должны оыть употреблены нелинейные теории. Описываются ползучие измерения напряжения и кручения в полиэтиленовых образцах. Применялась ступенчатая загрузка и деформации измерялись оптически посредством передвигающихся микроскопов для опытов с напряжением и отражением световых лучей от зеркал для экспериментов с кручением. Деформации были слишком велики для удержания теории линейной вязко-эластичности и были применены конститутивные отношения формы кратного интеграла; определены некоторые вовлечённые ядра. Было найдено, что для нагрузок применяемого диапазона для описания деформаций чистого сдвига достаточно двух ядер и для описания напряжения требуется три ядра. Описаны и рассмотрены также некоторые опыты, в которых применялись две ступени нагрузки.