# Stress Relaxation for Biaxial Deformation of Filled High Polymers

J. T. BERGEN and D. C. MESSERSMITH

Armstrong Cork Company, Lancaster, Pennsylvania

# R. S. RIVLIN

Brown University, Providence, Rhode Island

## INTRODUCTION

Continuum mechanical theories of a very general character for nonlinear isotropic viscoelastic materials have been developed recently by Ericksen and Rivlin,<sup>1</sup> by Rivlin,<sup>2</sup> and by Green and Rivlin.<sup>3</sup> The constitutive equations (i.e., stress-deformation relations) which are arrived at through these theories are, in general, very complicated in character and contain large numbers of arbitrary functions of the invariants of the deformation. Before these constitutive equations could be used to make quantitative predictions for a particular material, it would generally be necessary to perform the very difficult task of determining experimentally, for that material, the arbitrary functions occurring in the constitutive equations.

However, if we content ourselves with a description of the mechanical properties of the material for some limited class of deformations, the form of the constitutive equation can be greatly simplified, and the unknowns in it which must be determined experimentally are fewer in number. In the present paper, we shall be concerned with the class of deformations in which the material is subjected to a deformation in some small time interval and the deformation is then maintained constant. The materials with which we are concerned are such that, for this class of deformations, the stress (after a time sufficiently large compared with that in which the deformation is carried out) depends only on the final state of deformation and on the time during which the material has been held in that state and is substantially independent of the path by which it was reached.

In Part I of the present paper, the constitutive equation for materials of this kind, which are isotropic in their undeformed state and incompressible, is derived for deformations of the class described above, for which the displacement gradients are small compared with unity. The physical properties of the particular material enter into the expression for the stress through a "modulus" which depends on two strain invariants and on time. By means of this constitutive equation, the forces necessary to maintain, in a cylindrical tube of the material, states of simultaneously produced torsion and simple extension are calculated. These forces depend, of course, not only on the amounts of simple extension and torsion, but also on the time that has elapsed between the instant at which the deformation was produced and that at which the forces are measured.

In Part II of this paper, experiments are described in which tubes of material are subjected to small simple extensions and to combinations of small simple extension and small torsion and are then held in these states of deformation, the tensile forces and torques applied being measured as functions of time. Five materials are employed in these experiments. They are a vulcanized, heavily filled synthetic rubber and four polyvinyl chloride compositions containing amounts of inorganic filler varying from zero to about 50% by volume. All of these materials show marked stress relaxation effects.

For each of the materials, the dependence of the modulus on the strain invariants and on the elapsed time can be calculated from the experimental results, and agreement is obtained between the values of modulus calculated from the results of different experiments. In each case it is found that the experimental results can be well represented by a modulus which depends on only one of the strain invariants and for which the strain and time dependences are separable.

## PART I. THEORETICAL

## **1. Mathematical Preliminaries**

In this paper we shall be concerned with bodies which are subjected to the following type of deformation. Initially at times t < 0, the body is undeformed. Then, in the small time interval t = 0 to  $\tau$ , the body is subjected to a deformation in which particles of the body initially at  $X_i$  (i = 1, 2, 3) in a rectangular Cartesian coordinate system  $x_1, x_2, x_3$ move to  $x_i$  in the same coordinate system and thereafter remain unchanged in position. In general, the forces which must be applied in order to maintain this state of deformation change with time.

We shall assume that the material of which the body is constituted is such that the stress components at any point of the deformed body are single-valued functions of the nine deformation gradients  $\partial x_i / \partial X_j$  and the time t and substantially independent of the path by which the deformation changes from its initial to its final value, at any rate for values of t sufficiently large compared with that time  $\tau$  during which the deformation is carried out. We shall further assume that in the undeformed state the material is isotropic and incompressible.

In a previous paper,<sup>4</sup> the independence of the stress at time t of the precise manner in which the deformation was carried out was considered to arise from the assumption that for deformations taking place in times of the order of magnitude  $\tau$ , the material behaved as an ideal elastic material. It was then seen that the stress components  $\sigma_{ij}$  in the coordinate system x at time t are given by

$$\sigma_{ij} = -p\delta_{ij} + \alpha_1 g_{ij} + \alpha_2 g_{ik} g_{kj} \qquad (1.1)$$

where  $g_{ij}$  is defined as

$$g_{ij} = (\partial x_i / \partial X_k) (\partial x_j / \partial X_k) \qquad (1.2)$$

 $\alpha_1$  and  $\alpha_2$  are single-valued functions of the strain invariants  $I_1$  and  $I_2$  defined by

 $I_1 = g_{ii}$ 

and

$$I_2 = (1/2)(g_{ii}g_{jj} - g_{ij}g_{ji})$$

and of the time t, and p is an arbitrary hydrostatic pressure.

However, it is readily seen that in order that the relation (1.1) shall be applicable for the class of deformations considered, it is not *necessary* that the material shall behave as a perfectly elastic material during the time  $\tau$  over which the deformation

is carried out. We may instead make the much wider assumption adopted in the present paper that the stress components at time t are substantially independent of the manner in which the deformation is carried out and depend only on the deformation gradients  $\partial x_i / \partial X_j$  and t; then we may still regard eq. (1.1) as the constitutive equation for our material under the class of deformations considered.

### 2. Small Deformations

We denote by  $u_i$  (i = 1, 2, 3) the components in the coordinate system x of the displacement at time t of a point of the body which is initially at  $X_i$ . Then,

$$u_i = x_i - X_i \tag{2.1}$$

We shall assume that the deformation to which the body is subjected is sufficiently small so that  $\partial u_i/\partial X_j \ll 1$ . Then, with the notation

$$e_{ij} = (1/2) \left[ (\partial u_i / \partial X_j) + (\partial u_j / \partial X_i) \right] \quad (2.2)$$

we have, from eq. (1.2),

$$g_{ij} \approx \delta_{ij} + 2e_{ij} \tag{2.3}$$

With this approximation, we may rewrite eq. (1.1) as

$$\sigma_{ij} = (-p + \alpha_1 + \alpha_2)\delta_{ij} + 2(\alpha_1 + 2\alpha_2)e_{ij} + 4\alpha_2e_{ik}e_{ki} \quad (2.4)$$

We shall employ the notation

$$\bar{p} = p - \alpha_1 - \alpha_2$$
$$\Theta = 2(\alpha_1 + 2\alpha_2)$$
$$\Phi = 4\alpha_2$$

and rewrite eq. (2.4) as

$$\sigma_{ij} = -\bar{p}\delta_{ij} + \Theta e_{ij} + \Phi e_{ik}e_{kj} \qquad (2.5)$$

Since p is undetermined if the displacements are specified, so is  $\bar{p}$ . Also, we see that  $\Theta$  and  $\Phi$  are single-valued functions of  $I_1, I_2$ , and t.

Now, let us denote the principal extensions by  $e_1, e_2$ , and  $e_3$ . We then have

$$I_1 = (1 + e_1)^2 + (1 + e_2)^2 + (1 + e_3)^2$$
  
= 3 + 2(e\_1 + e\_2 + e\_3) + (e\_1^2 + e\_2^2 + e\_3^2)

and

(1.3)

$$I_2 = (1 + e_2)^2 (1 + e_3)^2 + (1 + e_3)^2 (1 + e_1)^2 + (1 + e_1)^2 (1 + e_2)^2$$

$$= 3 + 4(e_1 + e_2 + e_3) + 2(e^2_1 + e_2^2 + e_3^2) + 4(e_2e_3 + e_3e_1 + e_1e_2) + 2(e_2e_3^3 + e_3e_2^2 + e_3e_1^2 + e_1e_3^2 + e_1e_2^2 + e_2e_1^2) + O(e^4)$$
(2.6)

where  $O(e^4)$  denotes terms of degree 4 or higher in the principal extensions.

Since the material considered is incompressible, the volume of each element remains unchanged in the deformation. We therefore have

$$(1 + e_1)(1 + e_2)(1 + e_3) = 1$$
 (2.7)

This yields

$$e_1 + e_2 + e_3 =$$
  
-( $e_2e_3 + e_3e_1 + e_1e_2$ ) -  $e_1e_2e_3$  (2.8)

Substituting from eq. (2.8) in eqs. (2.6), we obtain

 $I_1 - 3 = -4(e_2e_3 + e_3e_1 + e_1e_2) - 2e_1e_2e_3 + O(e^4)$ and (2.9)

 $I_2 - 3 = -4(e_2e_3 + e_3e_1 + e_1e_2) - 10e_1e_2e_3 + O(e^4)$ 

From the assumption that  $\partial u_i / \partial X_j \ll 1$ , it follows that  $e_1, e_2, e_3 \ll 1$ . We note that  $(I_1 - 3)$  and  $(I_2 - 3)$  are both of the second order of smallness in the principal extensions. Introducing the notation

 $J_1 = I_1 - 3$ 

and

$$J_2 = I_1 - I_2$$

 $J_1 = -4(e_2e_3 + e_3e_1 -$ 

we have

and

 $J_2 = 8e_1e_2e_3$ 

if we assume  $e_1, e_2, e_3 \ll 1$ .

 $J_1$  and  $J_2$  may be expressed as functions of the displacement gradients;  $e_1$ ,  $e_2$ , and  $e_3$  are, with the neglect of terms of higher degree than the first in the displacement gradients, the solutions for e of the equation

$$\left|e_{ij} - e\delta_{ij}\right| = 0 \tag{2.12}$$

It follows that

and

$$J_1 = 2(e_{ij}e_{ji} - e_{ii}e_{jj})$$

Since  $\Theta$  and  $\Phi$  are single-valued functions of  $I_1, I_2$ , and t, they may, of course, be regarded as single-

 $J_2 = 8|e_{ij}|$ 

valued functions of  $J_1$ ,  $J_2$ , and t. Thus, in order to define completely the constitutive equation (2.5)for a particular material subjected to deformations of the class under consideration, we must know  $\theta$ and  $\Phi$  as functions of  $J_1$ ,  $J_2$ , and t. Unless  $\Phi \gg \Theta$ , it follows from  $\partial u_i / \partial X_j \ll 1$  that we may neglect the term  $\Phi e_{ik}e_{kj}$  in comparison with  $\Theta e_{ij}$ . The constitutive equation (2.5) then becomes

$$\sigma_{ij} = \Theta e_{ij} - \bar{p} \delta_{ij} \tag{2.14}$$

and in order to define this completely we need only determine the functional dependence of  $\Theta$  on  $J_1, J_2$ , and t.

#### **3. Superposed Torsion and Simple Extension**

In this section we shall assume that the constitutive eq. (2.14) is applicable at times  $t \gg \tau$  for deformations carried out in the small time interval 0 to  $\tau$  and thereafter maintained constant.

For a straight tube of the material of uniform, circular cross section, with internal and external radii a and b, respectively, subjected simultaneously to a simple extension and torsion during the time interval 0 to  $\tau$  and thereafter maintained in this state of deformation, we shall calculate the forces which may be applied to the tube at times  $t \gg \tau$  in order to maintain this deformation. The procedure adopted is similar to that which may be employed in the case when the material is an elastic material obeying the generalized Hooke's law for an incompressible isotropic material.

If e is the fractional extension and  $\psi$  (measured in radians per unit length of the extended rod) is the amount of torsion to which the tube is subjected, then, choosing the coordinate system x with the  $x_3$ axis along the axis of the tube, we may express the displacements  $u_i$  precisely by

$$u_{1} = (1 + e)^{-1/2} [X_{1} \cos \{\psi(1 + e)X_{3}\} - X_{2} \sin \{\psi(1 + e)X_{3}\}] - X_{1}$$

$$u_{2} = (1 + e)^{-1/2} [X_{1} \sin \{\psi(1 + e)X_{3}\} - X_{2}$$

$$+ X_{2} \cos \psi(1 + e)X_{3}] - X_{2}$$

 $u_3 = eX_3$ 

If we now assume  $\psi X_3 \ll 1$  and  $e \ll 1$ , we may write eqs. (3.1) as

$$u_{1} = -\psi X_{2} X_{3} - (1/2) e X_{1}$$
  

$$u_{2} = \psi X_{1} X_{3} - (1/2) e X_{2}$$
  

$$u_{3} = e X_{3}$$
(3.2)

Introducing eqs. (3.2) into eq. (2.2) and the expres-

(2.11)

$$= I_1 - I_2$$

$$+ e_1 e_2$$
)

sions for  $e_{ij}$  so obtained into the constitutive equation (2.14), we obtain

$$\sigma_{11} = \sigma_{22} = -(1/2)\Theta e - \bar{p} \sigma_{33} = \Theta e - \bar{p} \sigma_{23} = (1/2)\Theta \psi X_1 \sigma_{31} = -(1/2)\Theta \psi X_2 \sigma_{12} = 0$$
(3.3)

and

Also, from eqs. (3.2), (2.2), and (2.13), we obtain

$$J_1 = 3e^2 + \psi^2 R^2$$

and

$$J_2 = 2e^3 + e\psi^2 R^2$$

where

$$R^2 = X_1^2 + X_2^2 \tag{3.5}$$

If no body forces are applied, the equations of equilibrium take the form

$$\partial \sigma_{ij} / \partial x_j = 0 \tag{3.6}$$

With eqs. (3.3) we obtain, with the approximation  $\partial X_t / \partial x_t = \delta_{ii}$ ,

$$\frac{\partial \bar{p}}{\partial X_1} = -(1/2)e(\partial \Theta/\partial X_1) \frac{\partial \bar{p}}{\partial X_2} = -(1/2)e(\partial \Theta/\partial X_2)$$
(3.7)

and  $\partial \bar{p} / \partial X_3 = 0$ 

whence

$$\bar{p} = -(1/2)\Theta e + \text{constant}$$
 (3.8)

The surface tractions  $F_i$  (measured per unit area of the deformed body) are given by

$$F_i = \sigma_{ij} l_j \tag{3.9}$$

where  $l_i$  are the direction cosines of the normal to the surface at the point considered. From eqs. (3.9) and (3.3) it is apparent that, if  $F_i = 0$  on the outer curved surface (R = b), we must have

$$\bar{p} = -(1/2)\Theta e \tag{3.10}$$

for R = b. With eq. (3.8), we obtain

$$\dot{p} = -(1/2)\Theta e$$
 (3.11)

throughout the tube.

Introducing this result into eqs. (3.3), we obtain

$$\begin{aligned}
\sigma_{11} &= \sigma_{22} &= 0 \\
\sigma_{33} &= (3/2)\Theta e \\
\sigma_{23} &= (1/2)\Theta\psi X_1 \\
\sigma_{31} &= -(1/2)\Theta\psi X_2 \\
\sigma_{12} &= 0
\end{aligned}$$
(3.12)

It is immediately apparent from eqs. (3.9) and (3.12) that  $F_t = 0$  on the inner curved surface of the tube. On the plane end, the normal to which has direction cosines (0, 0, 1), we have

$$F_{1} = -(1/2)\Theta\psi X_{2}$$
  

$$F_{2} = (1/2)\Theta\psi X_{1}$$
  

$$F_{3} = (3/2)\Theta e$$
  
(3.13)

(3.14)

It is easily seen that this system of surface tractions is statically equivalent to a couple M and a tensile force N given by

 $M = \pi \psi \int_a^b \Theta R^3 dR$ 

and

(3.4)

$$N = 3\pi e \int_{a}^{b} \Theta R dR$$

# PART II. EXPERIMENTAL

### 4. Preliminary Remarks

In the theoretical considerations advanced in the previous sections, we have assumed that if the deformation of a body is carried out during the time interval 0 to  $\tau$  and is then held constant, the forces required to maintain the deformation at times tsufficiently large compared with  $\tau$  will be substantially independent of the path by which the deformation changes from its initial to its final value. Plainly, this assumption will not be valid for all materials. However, there are many materials for which we have good reason to expect that this assumption will be valid. These are materials for which the stress depends, in a continuous manner, on the deformation history of the material prior to the instant at which the stress is measured, and for which the effect of a previous deformation on the stress decreases to zero as the time interval between this deformation and the instant of measurement of the stress increases.

The materials employed in the experimental investigation reported in the following sections were of this type. Five different compounds were used. One of these was a vulcanized synthetic rubber compound, heavily loaded with fillers. The remaining four compounds were polyvinyl chloride, unloaded and loaded with various amounts of filler. The detailed composition of these compounds and the method of reparation of the test pieces are described in Section 6.

Tubes of these materials were subjected to simple extensions and to combinations of simple extension and torsion, the tensile force and torsional couple required to maintain these deformations at constant values being measured at times large compared with those in which the deformations were carried out.

From certain of these measurements, the dependence of  $\Theta$  in the constitutive equation (2.14) on  $J_1$ ,  $J_2$ , and t was determined by employing the formulae (3.14) for the torsional couple and the tensile force. With the values of  $\Theta$  so determined predictions were made, from the formulae (3.14), of the tensile force and torsional couple associated with other combinations of simple extension and torsion, and these were compared with the experimentally determined values. We note that, in passing from eq. (2.5) to eq. (2.14), the assumption was made that  $\Phi$  is not too large compared with  $\Theta$ . The agreement between the predicted and experimental results lends support to this assumption. It also lends support to the assumption that the materials are substantially incompressible.

In applying the results of the previous sections to particular materials, it must be borne in mind that the minimum time t for which the constitutive equation (2.14) is applicable will, in general, become greater as the time  $\tau$  during which the deformation is carried out becomes greater. Also, if for a fixed value of  $\tau$  the deformation is changed from its initial to its final value in a monotonically increasing fashion, the minimum time t for which eq. (2.14) is applicable will, in general, be lower than it would be if the deformation were first increased beyond its final value and then decreased to the final value at which it is maintained constant. Accordingly, to permit measurements which are meaningful in the context of the theory advanced in the previous sections to be made for reasonably short times t, in the experiments described below the deformation was always increased monotonically from its initial to its final value.

## 5. Description of the Apparatus

The apparatus was designed with the object of subjecting the tubular test pieces to desired amounts of simple extension or simultaneous simple extension and torsion, the deformation being then held constant while the tensile force and torsional couple are measured continuously throughout the experiment. Figure 1 shows a schematic vertical cross section of the apparatus through the axis of the test piece.

To each end of the test piece S, cylindrical steel end pieces A and B are cemented. End piece A is rigidly attached to the load cell of an Instron Tensile Tester. End piece B is attached to the crosshead C



Fig. 1. Vertical section through apparatus: (T) dynamometer; (A) and (B) end pieces; (S) test piece; (L) lever arm; (D) bearing pin; (C) crosshead of Instron Tester.

of the Instron Tester by means of bearing pins D. B is free to rotate about the axis of the specimen, but is constrained to move with the cross-head in the axial direction. A lever arm L attached to the end piece B is used to subject the test piece to torsion.

The torsional couple is measured by means of a dynamometer (represented as T in Figure 1) consisting of a thin-walled magnesium cylinder to which are cemented resistance-wire strain gages arranged to respond to tangential forces but not to bending or to axial thrust. The tensile force is measured by the load cell which is part of the Instron Tester. Both the tensile force and torsional couple are recorded continuously as functions of time through suitable amplifying and automatic recording equipment.

The test piece is subjected to simple extension by the vertical motion of the crosshead C, which is driven at a constant rate. This rate can be varied over a wide range by the synchronous motor which forms part of the Instron Tester. In the experiments in which the test piece is subjected to simple extension without torsion, the controls on the Instron Tester are so set that the extension is increased to the desired extent and then held constant. In the experiments in which the test piece is subjected to combinations of simple extension and torsion, the torsion is produced by the motion of the lever arm L. Small cables are attached to this and wound on a drum driven by a synchronous motor through a gear system which can be used to vary the rate at which the torsion is carried out.

The amount of torsion to which the test piece is subjected is controlled by the setting of an adjustable microswitch against which the lever arm L registers. The same microswitch is used to stop the motion of the crosshead C. Consequently, by simultaneous starting of the two synchronous motors, it is possible to subject the test pieces to linearly increasing amounts of simple extension and torsion in the same time interval.

The angle of twist to which the test piece is subjected is measured with an accuracy of 0.02 degree by means of an optical lever, and the extension of the tube is measured with an accuracy of  $10^{-4}$  in. by means of a dial gage. It was found by repeating experiments that, once the setting of the microswitch controlling the magnitude of the deformation has been fixed, the amounts of torsion and extension are determined with errors only slightly greater than those involved in their measurement.

### 6. Composition and Preparation of the Test Pieces

The ingredients for the vulcanized synthetic rubber compound used in the experiments are given in Table I. This is a commercial floor tile composition.

TABLE I

	Composition		
Ingredients	Wt%	Vol%	
Rubber			
GR-S 1066	18.1	35.7	
High-styrene butadiene	1.4	2.4	
	19.5	38.1	
Filler			
Hard clay	31.7	22.6	
Asbestine	18.1	10.4	
Ground limestone	13.6	9.3	
Titanium dioxide	4.5	2.1	
Asbestos	1.8	1.5	
	$\overline{69.7}$	45.9	
Chemical softeners	10.8	16.0	

Each test piece was prepared from factory-mixed unvulcanized stock which was formed by an extruder into a hollow tube of 3/4 in. i.d. and 13/32 in. o.d. The tube was fitted onto a steel mandrel of 3/4-in. diameter, then wrapped with cloth tape and vulcanized for 10 min. at  $324^{\circ}$ F. Following vulcanization, the cloth wrapping was removed, the tube and mandrel were mounted on a lathe, and the tube was ground to an external diameter of very nearly one inch. Finally the length was trimmed to about one foot.

Cylindrical steel end pieces, shown as A and B in Figure 1, were then cemented onto the ends of the specimens by means of a cold-set epoxy cement of high modulus; the test piece so formed was stored for at least two weeks at 21°C. and 50% R.H.

In addition to the vulcanized synthetic rubber compound described above, four plasticized polyvinyl chloride compounds were used in the experiments. The ingredients for these compositions are given in Table II. In each of the compounds the

TABLE I
---------

Polyviny	1 Chloride C	ompositi	ons				
	Compositions vol%						
Ingredients	No. 1	No. 2	No. 3	No. 4			
PVC (VYNW)	61.4	50.6	46.5	28.4			
Dioctyl phthalate	36.8	30.4	27.8	17.0			
Ground limestone		17.6	24.4	53.6			
Stabilizers	1.8	1.4	1.3	1.0			

ratio of the amounts (by weight) of PVC and plasticizer is approximately 3:1. However, in each of the four compounds the plasticized PVC is mixed with a different volume of filler. It is seen in Table II that the volume of filler varies from zero to about 54 vol.-%, so that the four compounds cover the range of filler contents usually encountered in commercial practice.

In preparing the test pieces, the following procedure was adopted. The ingredients were first dry-blended, then fused and mixed with a two-roll mill. After cooling, the mixture was pulverized and then preheated and extruded in the form of a tube. The tube was placed on a steel mandrel of  $^{3}/_{4}$ -in. diameter and annealed for  $^{1}/_{2}$  hr. at 250°F. to remove residual stresses. The tube was then gound to very nearly one inch in external diameter and trimmed to a length of about one foot.

Steel end pieces were then attached to the tubes in the manner employed for the vulcanized synthetic rubber compound, and the test pieces were stored at  $25^{\circ}$ C.

### 7. Conditioning of the Test Pieces

In the experiments reported in this paper, test pieces of each of the compounds described in Section 6 were subjected to various amounts of extension or combinations of extension and torsion, by use of the apparatus described in Section 5. In each state of extension or combined extension and torsion, continuous records of the dependence of tensile force and torque on time were made.

Since it is not possible to fabricate test pieces of a single compound with identical mechanical properties, it was necessary that, for each compound, a whole series of experiments be carried out on a single test piece. Also, since the mechanical properties of the materials employed depend on their deformation history, conditioning procedures were devised for each of the compounds. These were employed after each deformation, in order to restore the test piece to substantially its initial condition.

It was found that when the extension and torsion of a test piece which has been deformed are reduced to zero, a residual thrust and torque have to be exerted to maintain this state. These decay with time and their decay is closely associated with the recovery of the material to its initial state. Consequently, in the case of the PVC compounds, the conditioning procedure adopted consisted in holding the test piece in its undeformed state until the residual thrust and torque had decreased to substantially zero.

In the case of the vulcanized synthetic rubber compound, a similar procedure was adopted for conditioning the test piece between one experiment and a subsequent experiment carried out at a larger deformation with a fixed resting period of 5 min. Such a procedure could also have been used for conditioning the test piece between one experiment and a subsequent experiment at a smaller deformation, but a resting period of several days would have been required for an adequate degree of recovery to take place. Consequently, the procedure adopted consisted of heating the test piece at  $60^{\circ}$ C. for 1 hr. and then holding it in its undeformed state for 16 hr. at the temperature and humidity employed in the experiments.

# 8. Experimental Procedure and Results: Vulcanized Synthetic Rubber Compound

The experiments employing the vulcanized synthetic rubber compound were carried out at a temperature of  $21 \pm 1^{\circ}$ C. and 50% R.H. The test piece was first subjected to a simple extension of fixed amount, and a plot of tensile force versus time was obtained which covered a period of slightly more than 3 min. The experiment was then repeated with successively larger simple extensions covering the range 0.020 to 0.150 in. A typical plot of tensile force versus time obtained in this manner



Fig. 2. Typical plot of tensile force vs. time for vulcanized rubber compound in simple extension.



Fig. 3. Plot of tensile force vs. fractional extension at t = 3 min. for vulcanized rubber compound.

is shown in Figure 2. From each of these curves, the tensile force at 3 min. after the deformation was carried out was read. In Figure 3 these tensile forces are plotted against the fractional extensions to which the tube is subjected (calculated from the extensions and the length of the test piece, 11.78 in., measured between the near faces of the end pieces).

The test piece was then subjected to a series of deformations consisting of 0.02-in. extensions combined with various amounts of twist covering the range of 4-20 degrees, the twist being increased in successive deformations. For each deformation, plots of tensile force versus time and of torque versus time were obtained covering a period of slightly more than 3 min. This series of experiments was then repeated with extensions of 0.04, 0.06, 0.08, and 0.10 in. The plots of tensile force and of torque versus time obtained were similar in form to that shown in Figure 2. From each of these curves, the tensile force and torque at 3 min. was read off. In Figures 4 and 5, these data are plotted against the fractional extension and



Fig. 4. Plot of torque vs. torsion at t = 3 min. for various fractional extensions of vulcanized rubber compound.



Fig. 5. Plot of tensile force vs. torsion at t = 3 min. for various fractional extensions of vulcanized rubber compound.

amount of torsion calculated from the estension and twist, respectively, together with the measured length of the tube. For economy of presentation, the torque vs. amount of torsion curves in Figure 4 are shifted parallel to the abscissa by  $10^{-2}$  radians/ in. for successively increasing values of the fractional extension. The solid lines in these graphs represent calculations based on theoretical considerations which are discussed in the next section.

In all of the above experiments the test piece was, of course, subjected to an appropriate conditioning procedure between successive deformations, as described in section 7.

# 9. Analysis of the Experimental Results for Vulcanized Synthetic Rubber

In Section 3 expressions are derived for the tensile force N and torsional couple M which must be applied to a tube of internal and external radii aand b, respectively, in order to maintain in it simultaneously imposed simple torsion of amount  $\psi$  and simple extension of amount e. These are given [cf. eqs. (3.14)] by

and

 $N = 3\pi e \int_a^b \Theta R dR$ 

(9.1)

(9.2)

 $M = \pi \psi \int_a^b \Theta R^3 dR$ 

where R represents the distance of a generic point of the tube from its axis and  $\Theta$  is a function of the time t after the deformation is imposed on the tube, and of  $J_1$  and  $J_2$  defined [cf. eqs. (3.4)] by

 $J_1 = 3e^2 + \psi^2 R^2$ 

and

$$J_2 = 2e^3 + e\psi^2 R^2$$

We shall use the relations (9.1) in conjunction with the experimental results contained in Figures 3, 4, and 5 to determine the manner in which  $\Theta$  depends on  $J_1$ ,  $J_2$  at t = 3 min.

If the tube is sufficiently thin-walled and  $\Theta$  does not vary too rapidly with  $J_1$  and  $J_2$ , we may, to a first approximation, treat  $\Theta$  as a constant in eqs. (9.1) and obtain

 $M = (1/4)\pi\psi\Theta(b^4 - a^4)$ 

and

(9.3)  
$$N = (3/2)\pi e\Theta(b^2 - a^2)$$

where the values of  $\Theta$  correspond to the values of  $J_1$ and  $J_2$  obtained from eqs. (9.2) by taking  $R \approx b$ , viz.,

$$J_1 = 3e^2 + \psi^2 b^2$$

(9.4)

and

$$J_2 = 2e^3 + e\psi^2 b^2$$

We note that, by choosing suitable values of e and  $\psi$ , we can subject the tube to different deformations, in which  $J_1$  remains constant and  $J_2$  varies or in which  $J_2$  remains constant and  $J_1$  varies. In this way, we could, in principle, determine the manner in which  $\Theta$  depends on  $J_1$  and  $J_2$  for a given value of t from measurements of M and N at this value of t.

In the experimental test piece employed,  $(b - a)/a \approx 1/3$ . Consequently, the eqs. (9.3) would, in general, be expected to provide only a rough approximation to eqs. (9.1) unless  $\Theta$  varied very slowly with  $J_1$  and  $J_2$ . However, when the measurements of M and N (for t = 3 min.) given in Figures 3, 4, and 5 were used in conjunction with eqs. (9.3) to determine the manner in which  $\Theta$  depends on  $J_1$  and  $J_2$  for t = 3 min., the dependence on  $J_2$  was so slight as to suggest that  $\Theta$  is actually substantially independent of  $J_2$ . Consequently, with the assumption that, for a given value of t,  $\theta$ depends on  $J_1$  only, the results of the experiments on simple extension (Fig. 3) were used to determine the manner in which  $\Theta$  depends on  $J_1$  for  $t = 3 \min$ .

For simple extension, taking  $\psi = 0$  in eqs. (9.1) and (9.2) we have M = 0 and

$$N = (3/2)\pi e\Theta(b^2 - a^2)$$
(9.5)

where

$$J_1 = 3e^2$$
 and  $J_2 = 2e^3$  (9.6)

(9.7)

From the measured values of the internal and external diameters of the tube we obtained:

b = 0.499 in.

and

$$a = 0.3735$$
 in

Using these values of b and a, and the values of  $\Theta$ and e corresponding to each of the experimental points in Figure 3, we calculated the value of  $\Theta$  at t = 3 min. for each of these points as well as the values of  $J_1$  and  $J_2$  given by eqs. (9.6). The values of  $\Theta$  (at t = 3 min.) so obtained are plotted against  $J_1$  in Figure 6 on a log-linear plot. The values of  $J_2$  corresponding to the various experimental points are given in the figure.

Assuming that  $\theta$  depends on  $J_1$  only, we used the values of  $\theta$  at t = 3 min. obtained from Figure 6 in eqs. (9.1) to calculate the values of M and N at t = 3 min. for extensions of the tube of 0.02, 0.04,



Fig. 6. Plot of  $\Theta$  vs.  $J_1$  at t = 3 min. for vulcanized rubber compounds.

0.06, 0.08, and 0.10 in., with a range of values of the torsion in each case. The results of the calculations for M are plotted as the full lines in Figure 4 and the results for N are plotted as the full lines in Figure 5. It is seen that reasonably good agreement between the calculations and the experimental results is obtained, bearing out the assumption that  $\Theta$  is substantially independent of  $J_2$  over the range of values of  $J_1$  and  $J_2$  covered by the experiments.

In carrying out the calculations leading to the full lines in Figures 4 and 5, it had to be borne in mind that for each value of the fractional extension e and amount of torsion  $\psi$ ,  $J_1$ —and hence  $\Theta$ —varies throughout the thickness of the tube. The manner in which  $J_1$  varies can, of course, be found from the first of eqs. (9.2). Hence, the corresponding values of  $\Theta$  can be determined from Figure 6. By use of these, the values of M and N could have been obtained from eqs. (9.1) by numerical integration. However, it was found as an empirical fact that, for each combination of the fractional extension and amount of torsion, when  $\Theta$  was plotted against R. the radial distance from the axis of the tube, a straight line was obtained, i.e.,  $\theta$  was expressible in the form

$$\Theta = \kappa_1 + \kappa_2 R \tag{9.8}$$

where  $\kappa_1$  and  $\kappa_2$  depend, of course, on e and  $\psi$ . Introducing eq. (9.8) into eqs. (9.1), we obtain

$$M = \pi \psi [(1/4)\kappa_1(b^4 - a^4) + (1/5)\kappa_2(b^5 - a^5)]$$
  
and (9.9)

$$N = 3\pi e [(1/2)\kappa_1(b^2 - a^2) + (1/3)\kappa_2(b^3 - a^3)]$$

With the values of  $\kappa_1$  and  $\kappa_2$  obtained for each combination of  $\psi$  and e from the empirical plots of  $\Theta$  against R, the values of M and N for t = 3 min. were calculated. These are plotted as the solid lines in Figures 4 and 5.

$e \times 10^3$		1.06	1.86	2.92	4.80	6.70	9.58
$J_1  imes 10^6$		3.37	10.4	25.6	69	135	275
$J_2  imes 10^9$		2.38	12.9	49.8	221	601	1760
N, lb., for	t = 0.5 min.	0.88	1.58	2.41	3.92	5.55	7.62
	t = 1.0	0.78	1.41	2.15	3.55	5.01	6.92
	t = 3.0	0.68	1.21	1.83	3.02	4.29	5.97
	t = 10.0	0.60	1.05	1.59	2.58	3.68	5.13
$N/N_{t_0=10}$ for	t = 0.5	1.46	1.50	1,51	1.52	1.51	1.49
	t = 1.0	1.30	1.34	1.34	1.37	1.36	1.35
	t = 3.0	1.13	1.15	1.15	1.17	1.17	1.16

 TABLE III

 Tensile Force N vs. Fractional Extension e for Simple Extension of PVC Compound 1\*

\* Dimensions of test piece: b = 0.498 in., a = 0.406 in.

TABLE IVTensile Force N vs. Fractional Extension e for Simple Extension of PVC Compound  $2^a$ 

$e  imes 10^3 \ J_1  imes 10^6 \ J_2  imes 10^9$		1.94 11.3 14.6	4.60 63.5 195	5.69 97 368	7.66 176 899	9.60 276 1770	11.52 398 3060
N, lb., for	t = 0.5 min.	2.22	4.90	6.11	8.05	9.60	11.25
	t = 1.0	1.97	4.40	5.48	7.21	8.66	10.20
	t = 3.0	1.66	3.73	4.64	6.15	7.40	8.70
	t = 10.0	1.39	3.16	3.93	5.22	6.32	7.40
$N/N_{t_0=10}$ , for	t = 0.5	1.60	1.55	1.55	1.54	1.52	1.52
•	t = 1.0	1.42	1.39	1.39	1.38	1.37	1.38
	t = 3.0	1.19	1.18	1.18	1.18	1.17	1.18

<sup>a</sup> Dimensions of test piece: b = 0.498 in., a = 0.406 in.

# 10. Experimental Procedure and Results for PVC Compounds

Experiments similar to those described in Section 8 were carried out on test pieces formed from each of the four PVC compounds described in Section 6. However, the temperature at which they were carried out was  $25 \pm 1^{\circ}$ C., the relative humidity remaining 50%; the conditioning procedure appropriate to PVC compounds, described in Section 7, was used. Also, the forces were measured over a time interval of slightly more than 10 min. after loading.

In one series of experiments, test pieces of each of the four PVC compounds were subjected to simple extensions of various amounts and the tensile force was measured. The values of the tensile force obtained at times 0.5, 1.0, 3.0, and 10.0 min. after deformation are given in Tables III-VI, together with the corresponding values of  $J_1$  and  $J_2$ . In each table, the measured inner and outer tube radii are given. In a second series of experiments, each of the four test pieces was subjected to various combinations of simple extension and torsion. The values of the tensile force at times 0.5, 1.0, 3.0, and 10.0 min. after loading are given in Tables VII-X. The values of  $J_1$  and  $J_2$  at the inner and outer surfaces of the tube are also given for each state of deformation. It is noted that *only* the tensile force was measured in these experiments.

In a third series of experiments, another test piece of Compound 4 was subjected to simultaneous extension and torsion; the tensile force and torque obtained at times 1.0, 3.0, and 10.0 min. after loading are given in Tables XI and XII.

From the results of the experiments on simple extension given in Tables III-VI, the values of  $\Theta$ for each of the four compounds and for t = 0.5, 1, 3, and 10 min. were obtained as functions of  $J_1$  by use of the second of eqs. (9.1), bearing in mind that in simple extension experiments  $J_1$  and  $J_2$ , and consequently  $\Theta$ , are independent of R, so that N and  $\Theta$ are related by eq. (9.5). The values of  $\Theta$  so obtained for t = 1 min. and t = 10 min. are plotted semilogarithmically against  $J_1$  as the solid circles in Figures 7 and 8, respectively. It is seen that for each of the four compounds and for each of these values of t,  $\Theta$  is a linear function of log  $J_1$  and may therefore be expressed in the form

$$\Theta = A - B \log J_1 \tag{10.1}$$

$e \times 10^3$		1.01	1.39	2.54	3.57	5.02	7.04	10.40
$J_1  imes 10^6$		3.06	5.80	19.4	38.3	76	149	325
$J_2  imes 10^9$		2.06	5.37	32.8	91	253	698	2250
N, lb., for	t = 0.5  min.	2.40	3.38	5.58	7.35	9.85	13.15	17.9
	t = 1.0	2.10	2.98	4.91	6.50	8.77	11.70	16.0
	t = 3.0	1.75	2.47	4.10	5.44	7.32	9.80	13.4
	t = 10.0	1.45	2.05	3.41	4.55	6.15	8.24	11.2
$N/N_{t_0=10}$								
for	t = 0.5	1.65	1.65	1.64	1.62	1.60	1.60	1.60
	t = 1.0	1.45	1.45	1.44	1.43	1.43	1.42	1.43
	t = 3.0	1.21	1.20	1.20	1.20	1.19	1.19	1.20

TABLE V Tensile Force N vs. Fractional Extension e for Simple Extension of PVC Compound  $3^{a}$ 

\* Dimensions of test piece: b = 0.4985 in., a = 0.406 in.

TABLE VI Tensile Force N vs. Fractional Extension e for Simple Extension of PVC Compound 4<sup>a</sup> 11.30  $e \times 10^3$ 2.944.18 5.847.18 8.54 9.93  $J_1 \times 10^6$ 25.952.4 102155 219 296383  $J_2 \times 10^9$ 50.8146 398 740 1250 1960 2890 22.7N, lb., for  $t = 0.5 \, \text{min.}$ 8.40 11.0 14.3 16.719.5 21.1t = 1.07.48 9.8 12.714.8 17.318.8 20.2 6.30 8.210.6 12.4 14.5 15.8 17.0 t = 3.0t = 10.05.336.9 9.0 10.5 12.013.314.3  $N/N_{t_{0}=10}$  for t = 0.51.58 1.59 1.59 1.591.621.581.59 t = 1.01.40 1.421.411.411.44 1.41 1.41 t = 3.01.18 1.19 1.18 1.18 1.21 1.19 1.19

\* Dimensions of test piece: b = 0.500 in., a = 0.406 in.

 $e \times 10^3$ 1.125 3.155.786.70 10.2  $\psi \times 10^3$ , radians/in. 3.7434.011.4 19.1 26.7 $J_1 \times 10^6$  at R = a6.10 51.2160 252503 R = b7.26 62.0191 311 599 R = a $J_2 \, imes \, 10^9$  at 5.44130 734 1390 4065 R = b6.75 164 909 1790 5050 N, lb., for t = 0.5 min.4.610.942.605.338.20 t = 1.02.320.84 4.18 4.827.45 t = 3.00.70 1.99 3.584.156.41 t = 10.00.60 1.72 3.53 5.53 3.10  $N/N_{t_0=10}$  for t = 0.51.57 1.51 1.49 1.511.48 t = 1.01.40 1.351.351.37 1.35t = 3.01.16 1.171.16 1.17 1.16

 TABLE VII

 Tensile Force N for Various Combinations of Simple Extension and Torsion for PVC Compound  $1^{a}$ 

<sup>a</sup> Dimensions of test piece: b = 0.498 in., a = 0.406 in.

Similar results are obtained (but not shown here) for t = 0.5 min. and t = 3 min. Thus, for each of the four polyvinyl chloride compounds employed, A and B are independent of the extension to which the test-piece is subjected and are functions of t. It is noted that in the case of the unfilled PVC Compound 1, B = 0. Both A and B increase with the amount of filler. From the experimental results for combinations of simple extension and torsion given in Tables VII-X, the values of  $\Theta$  for t = 0.5, 1, 3, and 10 min. were obtained for each of the four compounds as functions of  $J_1$ . In calculating these values of  $\Theta$ , the variation of  $\Theta$  over the thickness of the tube was neglected, so that the second of eqs. (9.3) could be used. The values of  $\Theta$  so obtained for t = 1 min. and t = 10 min. are plotted against the values of  $J_1$  at the outer surface of the tube, as the open circles in Figures 7 and 8. It is seen that they lie on the same straight lines as do the values for  $\Theta$ 

obtained from the simple extension measurements. (It may be remarked that these points would be only slightly shifted if the values of  $\Theta$  were plotted against the values of  $J_1$  at the inner surface of the

5.22

1.53

1.38

1.17

9.05

7.71

6.59

1.53

1.37

1.17

TABLE	VIII
-------	------

Tensile Force N for Various Combinations of Simple Extension and Torsion for PVC Compound 2ª  $e \times 10^3$ 1.09 3.36 5.728.81 10.34  $\psi \times 10^3$  radians/in. 3.70 11.45 19.2 26.739.3  $J_1 \times 10^6$ at R = a5.8297.5 159 350 575 R = b108 704 6.96 190 410  $J_2 \times 10^9$ at R = a5.05106 722 2400 4840 R = b6.29 143 897 2930 6170 t = 0.5 min.5.89 N, lb., for 1.273.60 8.00 10.05 t = 1.01.14 3.21 5.287.18 t = 3.00.96 2.734.496.12

0.80

1.59

1.43

1.20

<sup>a</sup> Dimensions of test pieces; b = 0.498 in., a = 0.406 in.

t = 10.0

t = 0.5

t = 1.0

t = 3.0

TABLE IX

2.32

1.55

1.38

1.18

3.70

1.59

1.43

1.21

Tensile Force N for Various Combinations of Simple Extension and Torsion for PVC Compound 3ª

$e \times 10^3$		1.21	3.47	6.21	8.44	10.92
$\psi \times 10^3$ radia	ans/in.	3.86	11.76	20.0	27.9	39.7
$J_1 \times 10^6$ at	R = a	6.85	58.9	182	342	617
	R = b	8.09	70.5	215	407	749
$J_2 \times 10^9$ at	R = a	6.51	162	888	2285	5440
	R = b	8.02	203	1100	2830	6880
N, lb., for	t = 0.5  min.	2.84	7.19	11.4	14.8	18.5
	t = 1.0	2.50	6.33	10.1	13.2	16.45
	t = 3.0	2.03	5.23	8.43	11.0	13.8
	t = 10.0	1.65	4.37	7.03	9.20	11.50
$N/N_{t_0=10}$ for	t = 0.5	1.72	1.65	1.62	1.61	1.61
-	t = 1.0	1.51	1.45	1.44	1.43	1.43
	t = 3.0	1.23	1.20	1.20	1.20	1.20

<sup>a</sup> Dimensions of test piece: b = 0.4985 in., a = 0.406 in.

TABLE X

Те	nsile Force N for Vari	ous Combinations	of Simple Extens	ion and Torsion fo	r PVC Compound	4ª
e × 10 <sup>3</sup>		3.66	3.56	5.75	8.21	11.29
$\psi   imes  10^3$ radia	nns/in.	3.61	11.0	18.4	25.4	32.3
$J_i   imes  10^6$ at	R = a	42.3	58.0	155	308	554
	R = b	43.5	68.3	184	363	643
$J_2 \times 10^9$ at	R = a	106	161	701	1980	4820
	R = b	110	198	867	2430	5820
N, lb., for	t = 0.5 min.	9.60	8.85	13.2	16.4	20.8
	t = 1.0	8.50	7,90	11.7	14.6	18.7
	t = 3.0	7.10	6.60	9.70	12.3	15.8
	t = 10.0	5,90	5.50	8.10	10.5	13.3
$N/N_{40-10}$ for	t = 0.5	1.63	1.61	1.63	1.56	1.59
	t = 1.0	1.44	1.44	1,44	1.39	1.46
	t = 3.0	1.20	1.20	1.20	1.17	1.10

<sup>a</sup> Dimensions of test piece: b = 0.500 in., a = 0.406 in.

 $N/N_{t_0=10}$  for

		¥	$= 11.2 \times 1$	0 <sup>-s</sup> radians/	$\psi = 18.7 \times 10^{-3}$ radians/in.			
$e \times 10^3$		0.58	1.56	2.27	4.95	1.08	2.22	6.51
$J_1   imes  10^6$ at	R = a	21.7	<b>28.0</b>	36.1	94.2	61.1	72.4	185
	R = b	32.4	38.6	46.8	105	90.9	102	215
$J_2 \times 10^{\circ}$ at	R = a	12.4	39.8	39.8 70.3	345	64.8	150	927
	R = b	18.6	56.5	94.6	398	96.9	216	1120
N, lb., for	t = 1  min.	1.63	4.10	5.72	11.2	2.55	4.90	13.6
	t = 3	1.38	3.45	4.75	9.4	2.15	4.09	11.4
	t = 10	1.20	2.90	3.95	7.9	1.85	3.42	9.6
M, inlb., for	t = 1	2.14	1,95	1.85	1.62	3.15	2.95	2.60
	t = 3	1.67	1.63	1.55	1.39	2.62	2.43	2.15
	t = 10	1.45	1.38	1.28	1.12	2.20	2.03	1.80
$N/N_{t_0=10}$ for	t = 1	1.36	1.41	1.45	1.42	1.38	1.43	1.42
	t = 3	1.15	1.19	1.20	1.19	1.16	1.20	1.19
$M/M_{t,-10}$ for	t = 1	1.48	1.41	1.44	1.45	1.43	1.45	1.44
	t = 3	1.15	1.18	1.21	1.24	1.19	1.20	1.19

TABLE XI	
Fensile Force N and Torque M for Various Combinations of Simple Extension and Torsion for PVC Compound 4, Secon	ıd
Test Piece <sup>a</sup>	

\* Dimensions of test piece: b = 0.500 in., a = 0.406 in.

TABLE XII

Tensile Force N and Torque M for Various Combinations of Simple Extension and Torsion for PVC Compound 4, Second Test Piece<sup>a</sup>

		Ý	= 26.0 radians	/in.	$\psi = 33.2$ radians/in.			
$e \times 10^3$		1.52	4,40	8.93	2.00	4.72	9,90	
$J_1 \times 10^6$ at	R = a	118	169	351	194	248	476	
	R = b	176	227	408	288	342	570	
$J_2 \times 10^{\circ}$ at	R = a	176	661	2420	379	1070	3740	
	R = b	264	914	2930	567	1510	4670	
N, lb., for	t = 1  min.	3.15	8.75	17.2	3.62	9.20	17.7	
	t = 3	2.65	7.30	14.6	3.05	7.70	15.0	
	t = 10	2.30	6.10	12.2	2.62	6.50	13.0	
M, inlb., for	t = 1	4.20	3.80	3.45	4.90	4.85	3.95	
	t = 3	3.35	3.22	2.88	4.10	4.10	3.35	
	t = 10	2.90	2.70	2.42	3.40	3.40	2.85	
$N/N_{to=10}$ for	t = 1	1.37	1.43	1.41	1.38	1.41	1.36	
•	t = 3	1.15	1.20	1.20	1.16	1.18	1.15	
$M/M_{t_0-10}$ for	t = 1	1.45	1.41	1.43	1.44	1.43	1.47	
÷	t = 3	1.16	1.19	1.19	1.21	1.21	1.18	

• Dimensions at test piece: b = 0.500 in., a = 0.406 in.

tube.) Similar results were obtained for t = 0.5 min. and t = 3 min. (these data not shown here). It thus appears that the eq. (10.1) for  $\Theta$  in which A and B are dependent on t and the compound employed but not on  $J_1$  or  $J_2$  is valid for combinations of simple extension and torsion.

The conclusion is further supported by the third series of experiments in which a test piece of PVC Compound 4 was subjected to combinations of simple extension and torsion and both the tensile force and torque were measured. From the results of these experiments (given in Tables XI and XII), the values of  $\theta$  at t = 1, 3, and 10 min. were obtained as functions of  $J_1$ , the variation of  $\Theta$  over the thickness of the tube being neglected so that eqs. (9.3) could be used. For each state of deformation, two values of  $\Theta$  were calculated, one from the measured torque and the other from the measured tensile force. These values of  $\Theta$  are plotted against the corresponding values of  $J_1$  at r = b in Figure 9, the values obtained from the torque measurements being shown as open circles and those obtained from the tensile force measurements as crosses. Note that in Figure 9, the points for t = 1 and t = 3min. are displaced by amounts 2.0 and 0.8 lb./in.<sup>2</sup>, respectively, parallel to the ordinate, for ease of



 $0 \frac{1}{2} \frac{1}{3} \frac{1}{4} \frac{1}{5} \frac{1}{10} \frac{1}{20} \frac{1}{20} \frac{1}{30} \frac{1}{40} \frac{1}{50} \frac{1}{1000} \frac{1}{1000$ 

Fig. 8. Plots of  $\Theta$  vs.  $J_1$  at t = 10 min. for the four PVC compounds.

display. We see that for each value of t, the values of  $\Theta$  obtained from the tensile force and torque measurements agree.

We conclude that for each of the PVC compounds and for each time t in the range covered by the experiments, the results of the experiments on combined simple extension and torsion are well represented by assuming that  $\Theta$  takes the form of eq. (10.1) where A and B depend on the compound and on t.

#### 11. Dependence of $\theta$ on Time

If the time dependence of  $\Theta$  is such that it may be expressed in the form

$$\Theta = \varphi(t)\Theta^*(J_1, J_2) \tag{11.1}$$

where  $\varphi(t)$  is independent of the deformation and  $\Theta^*$  is independent of t, then from eq. (9.1) we see that

$$M = \pi \psi \varphi(t) \int_a^b \Theta^* R^3 dR$$



Fig. 9. Plots of  $\Theta$  vs.  $J_1$  at various values of t for PVC compound 4: (O) values from torque measurements; (+) values from tensile force measurements.

and (11.2)

$$N = 3\pi e\varphi(t) \int_a^b \Theta^* R dR$$

Thus, if  $M_{t_0}$  and  $N_{t_0}$  denote the values of M and N at some reference time  $t_0$ , we have

$$M/M_{t_0} = N/N_{t_0} = \varphi(t)/\varphi(t_0)$$
 (11.3)

It can easily be shown that this is in fact the case for the experiments described in this paper.

We first consider the results for the four PVC compounds given in Tables III-X; taking  $t_0 = 10$  min., we calculate  $N/N_{t_0=10}$  for t = 0.5, 1.0, 3.0, and 10.0 min. for each of the compounds and for each of the deformations. The results are given in Tables III-X. It is seen that  $N/N_{t_0=10}$  is, within experimental error, independent of the deformation for each of the compounds.

Again, from the results given in Tables XI and XII,  $N/N_{t_0=10}$  and  $M/M_{t_0=10}$  are calculated for the second test piece of PVC Compound 4 for t = 1 and 3 min. and are given in Tables XI and XII. Again, we see that

$$N/N_{t_0=10} = M/M_{t_0=10}$$
(11.4)

and these ratios are independent of the deformation.

We note that, for a given value of t, the ratios  $N/N_{t_0=10}$  and  $M/M_{t_0=10}$  differ only slightly for the

four PVC compounds. This suggests that the form of the function  $\varphi(t)$  in eqs. (11.2) is determined primarily by the high-polymeric matrix rather than by the filler.

Similar results have also been obtained for the synthetic vulcanized rubber compounds, but are not given in detail here.

#### References

1. Rivlin, R. S., and J. L. Ericksen, J. Ratl. Mech. & Anal., 4, 323 (1955).

2. Rivlin, R. S., J. Ratl. Mech. & Anal., 4, 681 (1955).

3. Green, A. E., and R. S. Rivlin, Arch. Ratl. Mech. & Anal., 1, 1 (1957).

4. Rivlin, R. S., Q. Appl. Math., 14, 83 (1956).

#### Synopsis

A constitutive equation is derived for stress-relaxation in isotropic incompressible viscoelastic solids at small constant deformations. This is used to calculate the torque and tensile force necessary to maintain simultaneous torsion and simple extension in a straight circular tube. Measurements are made of the dependence of the tensile force and torque on the amount of torsion and fractional extension in tubes of filled high-polymers. It is shown that by means of the theory the results of certain of the experiments can be predicted from others.

#### Résumé

Une équation est déduite pour le relâchement de force dans le cas de solides isotropiques, incompressibles et viscoélastiques, pour de petites déformations constantes. Cette équation est employée pour calculer la force de torsion et de tension nécessaire pour maintenir une torsion et une simple extension simultanément dans un tube circulaire droit. Des mesures ont été faites au sujet de la dépendance de la force de tension et de torsion sur la grandeur de la fraction de tension et de torsion dans des tubes remplis de hauts polymères. On a montré que, par l'intermédiaire de la théorie, les résultats de certaines de ces expériences peuvent être prévus à partir d'autres.

#### Zusammenfassung

Eine grundlegende Gleichung für die Spannungsrelaxation in isotropen, inkompressibeln, visko-elastischen Festkörpern bei kleiner, konstanter Verformung wird abgeleitet. Sie wird zur Berechnung der Torsions- und Dehnungskraft verwendet, die notwendig ist um eine gleichzeitige Torsion und einfache Dehnung in einem geraden Rohr mit kreisförmigem Querschnitt aufrecht zu erhalten. Messungen der Abhängigkeit der Dehnungs- und Torsionskraft vom Betrag der Torsion und relativen Dehnung werden an Rohren aus gefüllten Hochpolymeren ausgeführt. Es wird gezeigt, dass mit Hilfe der Theorie die Ergebnisse gewisser Versuche aus anderen Versuchen vorausgesagt werden können.

Received March 23, 1959