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On approximate description of non-linear viscoelastic material

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Abstract- An approximate description of non-linear viscoelastic behavior is proposed and applied in analysis of creep phenomena of a polyvinyl chloride. The description is based upon the concept of regions of linear and non-linear behavior, and superposition of linear and non-linear strain components. It is shown that creep and recovery creep can be described by introducing smaller number of terms in nonlinear functional representation. Hardening of the polyvinyl chloride in additional creep was also observed.

1. Introduction

A non-linear functional representation due to Green and Rivlin /1,2,3/ has been applied to describe a non-linear viscoelastic behavior of different high polymers or soils by numerous authors /4,5,6,7,1,9,10/. For sufficiently high stress and strain levels, high polymers are markedly non-linear and the linear laws cannot be applied even at the room temperature. The integral representation of Green and Rivlin constitutes a powerful tool in the analysis of experimental results due to its general formulation, applicable for any

complex program and for more or less strong non-linearity. The data already published, show applicability of this representation to non-linear viscoelastic behavior. On the other hand, an extensive experimental program is required to determine material functions even if a few terms of representation are retained. It is therefore of essential importance to work out some simplified versions of this general representation that could be used in practical applications. One of such propositions was recently made by Pipkin and Rogers /11/.

In the present paper, an approximate description of the non-linear viscoelastic behavior is proposed which is based on the concept of regions of linear and nonlinear behavior 1. A boundary between both regions depends on the stress state and resembles a familiar boundary between elastic and elastic-plastic regions from the theory of plasticity. It will be shown that a smaller number of terms in, the non-linear representation sufficies to describe experimental data if the distinction is made between linear and non-linear regions. Only one-dimensional stress state is considered for which an experimental verification is presented. Creep tests on polyvinyl chloride were performed for different stress histories.

^{1/} A similar concept of several regimes in which different linear operators describe the viscoelastic behavior was discussed by Onat /12/. The viscoplastic behavior of soils was also considered in /10/ on the basis of the non-linear integral representation, and the notion of yield surface separating viscoelastic and viscoplastic regimes was introduced.

2. Approximate description

The Green - Rivlin representation can be presented for the one-dimensional stress state in a form of the following infinite integral series / cf. /4/ /

$$\mathcal{E}(t) = \int_{0}^{t} J_{A}(t-T_{A}) \dot{G}(T_{A}) dT_{A} + \int_{0}^{t} J_{2}(t-T_{A}, t-T_{2}) \dot{G}(T_{A}) \dot{G}(T_{2}) dT_{A} dT_{2} + \\ + \cdots + \int_{0}^{t} \cdots \int_{0}^{t} J_{N}(t-T_{A}, \cdots t-T_{N}) \dot{G}(T_{A}) \cdots \dot{G}(T_{N}) dT_{A} \cdots dT_{N} + \cdots$$

Eq. / 2.1 / has been applied by numerous authors /4,5,6,7,8, 9,10/ for description of creep tests of high polymers and soils. The experiments were limited to one- or two-step histories. In order to discuss this representation in more detail, let us consider the one-step loading history which can be described as follows

$$T \le 0$$
, $G(T) = 0$; $T > 0$, $G(T) = G_0 = const$ / 2.2 / Eq. / 2.1 / for the program / 2.2 / yields

$$\mathcal{E}_{c}(t,G_{o}) = J_{4}(t)G_{o} + J_{2}(t,t)G_{o}^{2} + \cdots + J_{N}(t,\cdots t)G_{o}^{N} + \cdots / 2.3 /$$

Dividing / 2.3 / by G_{o} , we obtain

$$\frac{\mathcal{E}_{c}(t,G_{o})}{G_{o}} = J_{1}(t) + J_{2}(t,t)G_{o} + \cdots J_{N}(t,\cdots t)G_{o}^{N-1} + \cdots / 2.4 /$$

The left-hand side of / 2.4 / presents the well known creep compliance, widely used in the analysis of rheological behavior of different materials. The creep compliance defined as

$$J_c(t,G_o) \stackrel{\text{def}}{=} \frac{\mathcal{E}_c(t,G_o)}{G_o}$$
 /2.5/

permits to establish easily whether the tested material exhibits linear or non-linear viscoelastic pehavior. For the linear material, the creep compliance is indepedentent of the stress level and is a function of time only, $J_C = J_C(t)$. In this case, it descress entirely the mechanical behavior of a linear material, and can be used in order to calculate other rheological functions.

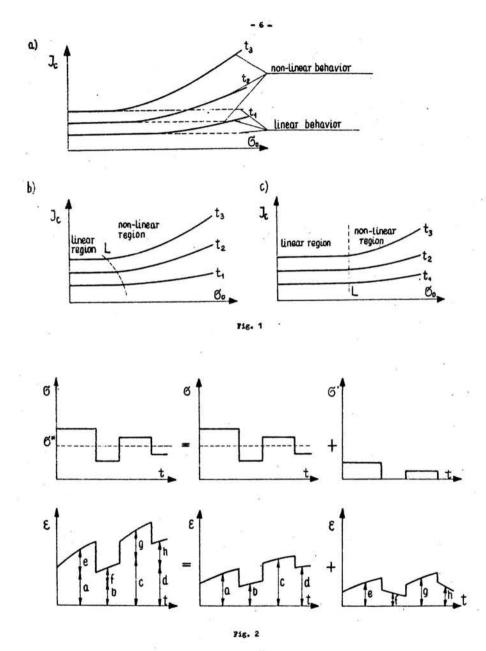
From equation / 2.4 / it follows that the creep compliance resulting from Green - Rivlin representation is a nonlinear function of stress. Moreover, Eq. / 2.4 / shows that the non-linearity is of a polynomial type. Thus, application of the representation /2.1 / starts from the question whether the observed non-linearity of a tested material is in a qualitative agreement with the polynomial type. This is usually done by drawing the calculated creep compliance versus stress for different instants t; / Fig. 1 a /. For a linear material, the results must lie along straight lines parallel to axis. Similary to the creep compliance, the recovery compliance or the additional compliance for more complex stress histories can be defined, and used to determine non-linearity with respect to the prescribed stress history. For linear material, any compliance must be identical to the creep compliance independly of the stress history.

When the non-linear behavior of the polynomial type is observed, the second question arises as how many terms of /2.1 / should be included in order to describe the test results. Their number depends on the required accuracy and ability to perform sufficiently large number of tests necessary to determine kernel functions of /2.1 /. Remembering that the non-linear kernel functions can be represented as hypersurfaces in the space of their arguments, it is seen that the one-step program furnishes only information on some sections of these hypersurfaces. To determine entirely the kernel functions, a very extensive test program must be carried out, even if only two first non-linear terms are retained / for details see Lockett /13//.

A difficulty in application of / 2.1 / arises particulary in cases where the transition from linear to non-linear behavior occurs more or less sharply at a certain stress level and experimental compliance functions are represented by curves sharply bending upwards starting from some critical stress value. If we want to apply the polynomial / 2.4 / to describe this effect, many higher order terms should be introduced and lower order terms deleted; thus, the number of tests, necessary to determine kernel functions, increases enormously. The concept of linear and non-linear regions may essentially simplify the material description in such cases.

Consider the relationship $J_c = J_c(c_o)$ schematically presented in Fig 1 b. For real materials, the linear or weakly non-linear region is bounded by the curve L. Let us first assume that this boundary line can be treated as a straight line parallel to the J_c - axis / Fig. 1 c /. This assumption is equivalent to a postulate that the non-linear behavior occurs above a certain value of stress $c = c_o$. For c_o the material exhibits the linear viscoelastic response. The critical stress c_o can now be treated as a material constant, not affected by the stress history. For multiaxial stress states c_o will correspond to a hypersurface in the stress space bounding the region of linear behavior. A more general case presented in Fig. 1 b will not be discussed.

To describe the mechanical behavior in the non-linear region, further assumption will be introduced: if the stress history G(T) exceeds at n instants t_i the value of G^* , the total effect at the instant $t_k \geqslant t_i$ is equal to the sum of linear effects caused by the stress history G(T) and non-linear effects caused by the history of the excess stress $G'(T) = G(T) - G^*$. Both histories G(T) and G'(T) are defined in the whole time interval $0 < T \leqslant t_K$. In other words, the total solution is equal to the sum of two independent solutions: linear, determined by the history of G(T) and non-linear determined by the history of G'(T). The proposed pro-



cedure is schematically presented in Fig. 2.

The linear behavior obeys the Boltzmann Superposition Principle, and can be described by any form of linear viscoelastic constitutive equation. For the case of creep-tests the integral representation has the form

$$\mathcal{E}^{l}(t) = \int_{0}^{t} J(t-\tau) \dot{G}(\tau) d\tau \qquad / 2.6 /$$

Eq. / 2.6 / describes the mechanical behavior entirely whenever an arbitrary stress history $\mathcal{G}(\mathcal{T})$ does not exceed at any instant t; the critical stress \mathcal{G}^* . Taking into account that the non-linear effects can depend on the whole stress history $\mathcal{G}^{\dagger}(\mathcal{T})$, ie. the material exhibit the non-linear memory effects, as the constitutive equation for non-linear part the Green - Rivlin representation will be applied. This representation, now, has the form

According to the assumed superposition of strains, the equation valid for 6 > 6* takes the following form:

$$\begin{split} \mathcal{E}(t) &= \int_{0}^{t} J(t-\tau) \dot{G}(\tau) d\tau + \int_{0}^{t} K_{1}(t-\tau_{1}) \dot{G}'(\tau_{1}) d\tau_{1} + \\ &+ \int_{0}^{t} K_{2}(t-\tau_{1},t-\tau_{2}) \dot{G}'(\tau_{1}) \dot{G}'(\tau_{2}) d\tau_{1} d\tau_{2} + \cdots \\ &/ 2.8 \text{ /} \end{split}$$

Equation / 2.8 / reduces to / 2.6 / when $|G(\tau)| < |G^*|$ In consequence, / 2.8 / constitutes the description of nonlinear material with distinct linear region. Before applying Eq. / 2.8 / to describe the experimental data. two additional requirements will be discussed:

- i/ validity of the equation independly of the sign of stress applied,
- ii/ continuous and smooth transition between linear and non-linear regions.

The requirement i/appears when the material exhibits the same behavior under compression and tension, or for torsion in opposite signs. In such cases all even terms of / 2.8 / must be omitted. To discuss the requirement ii/we shall consider the one-step history / 2.2 /. Integration by parts of / 2.8 / yield

$$\mathcal{E}_{c}(t, G_{o}) = J(t)G_{o} + K_{1}(t)G_{o}' + K_{2}(t, t)G_{o}'^{2} + \cdots / 2.9 /$$

and after dividing by G_{o}

$$J_{c}(t,G_{o}) = J(t) + K_{1}(t) \frac{G_{o}^{i}}{G_{o}} + K_{2}(t,t) \frac{G_{o}^{i2}}{G_{o}} + \cdots / 2.10 /$$

According to the published experimental results /4,6,8/ the $J_c = J_c(G_o)$ relationships for one-step history present the continuous and smooth transition between the linear and non-linear regions. Eq. / 2.10 / assures the continuity of $J_c = J_c(G_o)$ function, but due to the presence of the second term K_4 ther is a discontinuity of the first derivative for $G_o = G^*$. Thus, the second term of / 2.8 / should be also omitted.

3. Test procedure

To provide an example of the application of the approximate description proposed in the previous section creeptests were performed on a commercial polyvinyl chloride. Different loading programs of uniaxial tensile stress at the room temperature / 20° C / were applied to flat specimens / length: 23 cm, cross section: 0,385 cm² /. The specimens were cut from one sheet, 0,2 cm thick. To reduce eventual self-stresses specimens were annealed at 55° C for 6 hrs. The scatter of creep and recovery data / Fig. 4 / was small and did not affect the mathematical description. Axial elongation was measured by means of Amsler extensometer; two exchangeable dial gauges / accuracy of readings 0,01 mm and 0,001 mm / permitted to measure the axial strain with the accuracy of 1,25 · 10⁻⁶ and 1,67 · 10⁻⁷ respectively. Because of the small magnitude of observed strains /£max = 0,03 / the Cauchy strain measure was assumed and the nominal stress was calculated. Small initial prestressing 6° = 22 kg cm² was applied in order to eliminate small flexure of the specimen due to unsymmetric position of the extensometer.

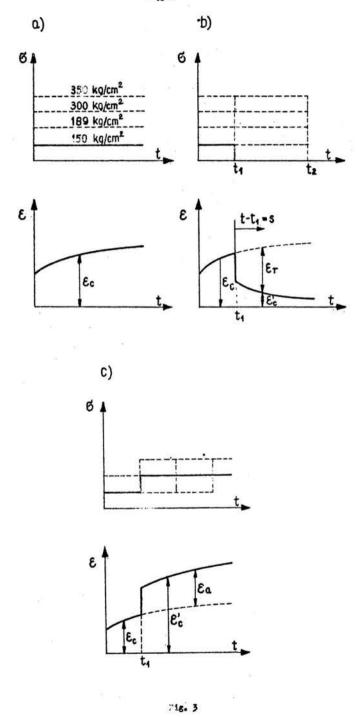
Three different loading programs consisting of oneand two-step histories were applied / Fig. 3/

I
$$T \le 0$$
, $G(T) = 0$; $T > 0$, $G(T) = G_0$; / 3.1 a /

II $T \le 0$, $G(T) = 0$; $0 < T \le t_4$, $G(T) = G_0$; $T > t_4$, $G(T) = 0$ /

III $T \le 0$, $G(T) = 0$; $0 < T \le t_4$, $G(T) = G_0^4$; $T > t_4$, $G(T) = G_0^2$ / 3.1 c /

In the I - program, the stresses $G_0 = 150 \text{ kg cm}^{-2}$, $G_0 = 189 \text{ kg cm}^{-2}$, $G_0 = 300 \text{ kg cm}^{-2}$, and $G_0 = 350 \text{ kg cm}^{-2}$ respectively were applied during 360 min. The creep and recovery program II with four values of stress, identical to those of I program, acting during 30 min. or 180 min. provided data to analyse the stress history effects. In the III-program, the following stress histories were used: $G_0 = 150 \text{ kg cm}^{-2}$, $G_0 = 300 \text{ kg cm}^{-2}$, and $G_0 = 300 \text{ k$



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From the geometry of Fig. 3 it follows that

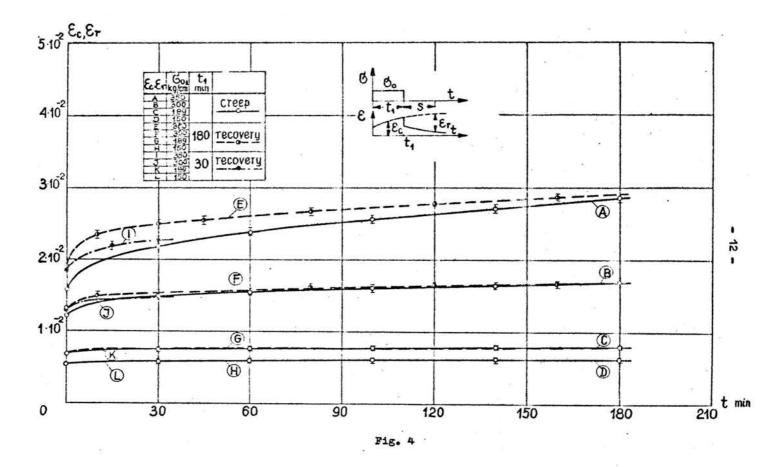
$$\mathcal{E}_{c}(t-t_{a}) = \mathcal{E}_{c}(t) - \mathcal{E}_{c}'(t-t_{a}),$$
 / 3.2 a / $\mathcal{E}_{c}(t-t_{a}) = \mathcal{E}_{c}'(t-t_{a}) - \mathcal{E}_{c}(t).$ / 3.2 b /

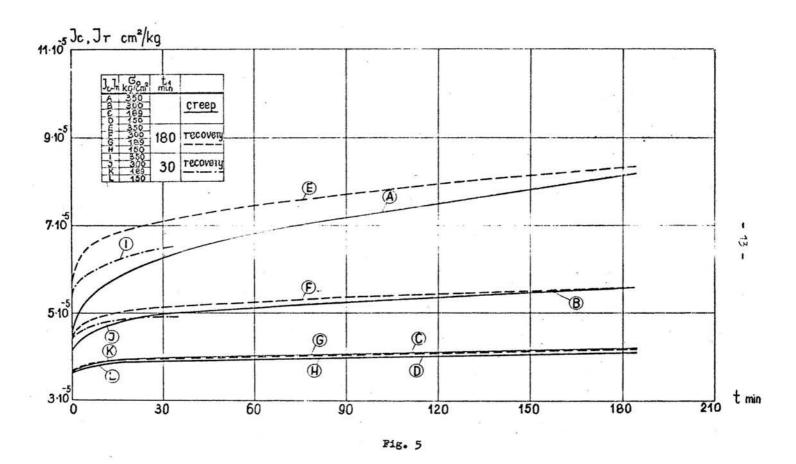
4. Test results and mathematical description

Ine creep and recovery response obtained from the test programs I and II are plotted in Fig. 4. All curves correspond to mean values of data of several tests, the scatter being also presented. The non-linear behavior of the material is evident from Fig. 4. It should be noted that creep and recovery strains are not identical for any value of stress the creep and recovery compliances presented in Fig. 5 show this effect clearly. Figs 4 and 5 show also the dependence of recovery response / recovery compliance / on the time lapse proceeding unloading. Thus the material exhibits nonlinear behavior with respect to both stress level and stress history. To determine the degree of non-linearity in function of stress applied, the relationships $J_c = J_c(G_o)$ are plotted in Fig. 6. From Fig. 6 it is seen that the departure from linearity becomes more pronounced for higher stress 60 . The dependences shown in Figs 6 and 7 are in qualitative agreement with the available data for other polymers /4.6.8 / The comparison of Jc and Jr is presented in Fig. 8.

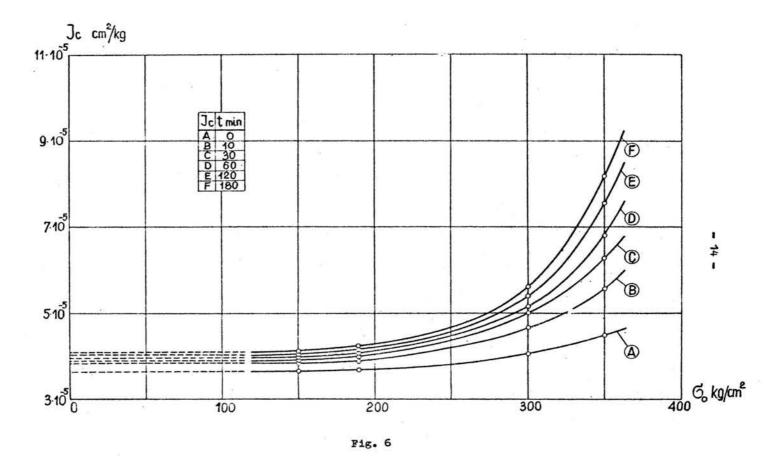
As a critical stress 6* we assume the value 6* = 150 kg cm⁻² / Fig.8 /. Experimental data indicate that for 6 > 150 kg cm⁻² non-linear effects become pronounced and cannot be ignored. Let us discuss first the one-step program. From / 2.8 / and / 5.1 a /, assuming previously mentioned requirements i/ and ii/ we have

$$\mathcal{E}_{c}(t,G_{o})=J(t)G_{o}+K_{3}(t,t,t)G_{o}^{13}+K_{5}(t,t,t,t,t)G_{o}^{15}+\cdots$$

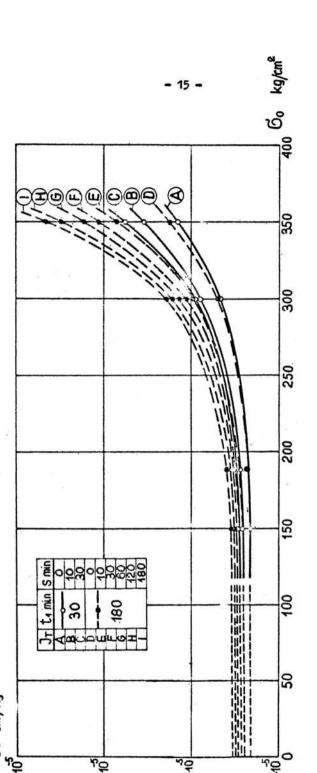




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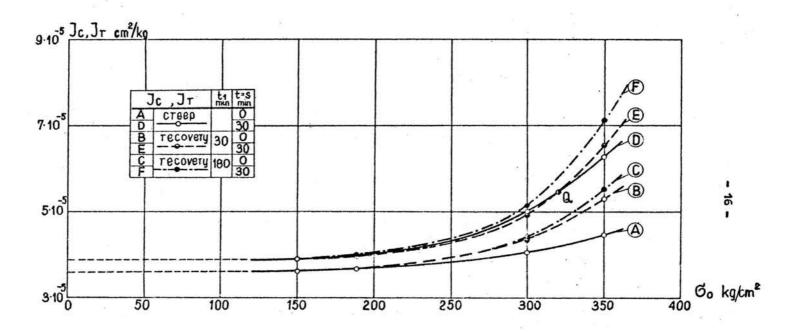


Fig. 8

and upon dividing by 60

$$J_c(t,G_o) = J(t) + K_3(t,t,t) \frac{{G_o^1}^3}{G_o} + K_5(t,t,t,t,t) \frac{{G_o^1}^5}{G_o} + \cdots$$

To establish, how many non-linear terms of / 4.2 / must be retained so as to describe test results, we shall first limit ourselves only to two terms J and K_3 . If these two terms suffice, than transforming / 4.2 / to the form

$$[J_c(t,G_o) - J(t)]G_o = K_3(t,t,t)G_o^{13},$$
 /4.3/

the experimental points, when plotted in the coordinates $y = [J_c(t, \delta_o) - J(t)] \delta_o$, $x = \delta_o^{13}$, should lie on the straight lines. The continuous lines in Fig. 9 show good agreement for t = 0, t = 30 min. and a significant discrepancy for t = 180 min. Including the third term of /4.2 / and transforming to the form

$$\frac{\left[\int_{C} (t, d_{0}) - J(t) \right] d_{0}}{d_{0}^{13}} = K_{3}(t, t, t) + K_{5}(t, t, t, t, t) d_{0}^{12} / 4.4 /$$

the same procedure can be applied. The dotted lines ... Fig. 9, now, show good agreement for the whole tested time interval. The position of the lines indicates that the third term K_5 should be included only for longer times / when the dotted lines are parallel to the horizontal axis it means that $K_5=0$ /. In other words, the mechanical behavior of tested material for one-step history can be described by the equation

$$\mathcal{E}_{c}(t,G_{o}) = J(t)G_{o} + K_{3}(t,t,t)G_{o}^{3} + K_{5}(t,t,t,t,t)G_{o}^{15}$$
 / 4.5 /



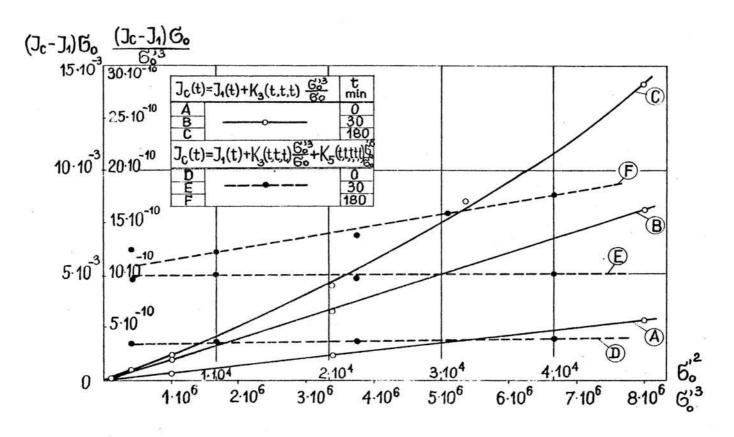


Fig. 9

Following from the assumed constitutive equation

$$\begin{split} \mathcal{E}(t) = \int_{0}^{t} J(t-T) \dot{S}(T) dT + \iiint_{000}^{t+t} K_{3}(t-T_{4}, t-T_{2}, t-T_{3}) \dot{S}(T_{4}) \dot{S}(T_{2}) \dot{S}(T_{3}) dT_{4} dT_{2} dT_{3} + \\ + \iiint_{00000}^{t} K_{5}(t-T_{4}, \dots t-T_{5}) \dot{S}(T_{4}) \dots \dot{S}(T_{5}) dT_{4} \dots dT_{5} \\ / 4.6 / \end{split}$$

Let us now consider the II - loading program. From / 4.6 / and / 3.1 b / we obtain for $t > t_4$

$$\begin{aligned} & \mathcal{E}_{c}^{'}(t, \mathcal{G}_{o}, t_{a}) = \left[J(t) - J(t - t_{a}) \right] \mathcal{G}_{o} + \left[K_{3}(t, t, t) - 3 K_{3}(t, t, t - t_{a}) + \right. \\ & + 3 K_{3}(t, t - t_{a}, t - t_{a}) - K_{3}(t - t_{a}, t - t_{a}, t - t_{a}) \right] \mathcal{G}_{o}^{13} + \\ & + \left[K_{5}(t, t, t, t, t) - 5 K_{5}(t, t, t, t, t - t_{a}) + 10 K_{5}(t, t, t, t, t - t_{a}, t - t_{a}) + \right. \\ & - 10 K_{5}(t, t, t - t_{a}, t - t_{a}, t - t_{a}) + 5 K_{5}(t, t - t_{a}, t - t_{a}, t - t_{a}, t - t_{a}) + \\ & + K_{5}(t - t_{a}, t - t_{a}, t - t_{a}, t - t_{a}, t - t_{a}) \right] \mathcal{G}_{o}^{15}. \end{aligned}$$

The recovery strain & according to / 3.2 a / has the form

$$\mathcal{E}_{\tau}(t, 6_{0}, t_{1}) = J(t - t_{1}) 6_{0} + [3 K_{3}(t, t, t - t_{1}) + \\
- 3 K_{3}(t, t - t_{1}, t - t_{1}) + K_{3}(t - t_{1}, t - t_{1}, t - t_{1})] 6_{0}^{13} + \\
+ [5 K_{5}(t, t, t, t, t - t_{1}) - 10 K_{5}(t, t, t, t - t_{1}, t - t_{1}) + \\
+ 10 K_{5}(t, t, t - t_{1}, t - t_{1}, t - t_{1}) - 5 K_{5}(t, t - t_{1}, t - t_{1}, t - t_{1}) + \\
+ K_{5}(t - t_{1}, t - t_{1}, t - t_{1}, t - t_{1}, t - t_{1})] 6_{0}^{15} . \qquad / 4.8 /$$

Denoting $t-t_1 = S$, equation / 4.8 / with the help of / / 4.1 / can be written as follows

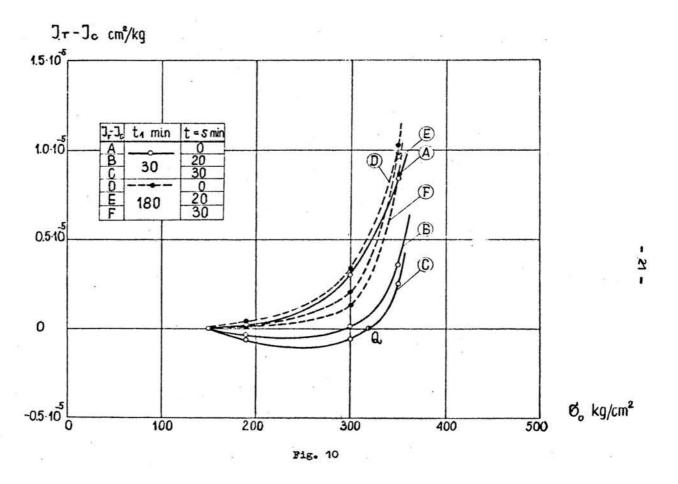
$$\mathcal{E}_{\tau}(s, G_0, t_1) - \mathcal{E}_{c}(s, G_0) = [3 K_3(s+t_1, s+t_1, s) +$$

- 3
$$K_3$$
 (s+t₄,s,s)] $6_0^{13} + [K_5(s+t_4,s+t_5,s+t_4,s+t_5,$

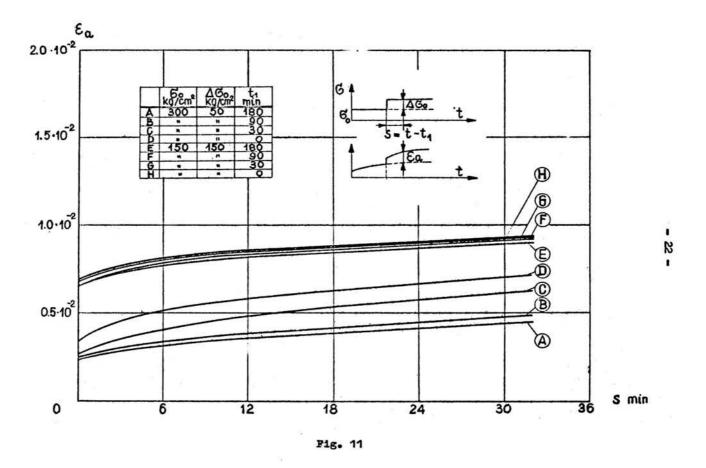
Dividing by 6, we finally arrive at

$$J_{\tau}-J_{c} = [3K_{3}(s+t_{4},s+t_{4},s)-3K_{3}(s+t_{4},s,s)] \frac{G_{o}^{13}}{G_{o}} +$$

Eq. / 4.10/ implies that the representation / 2.8 / can describe the difference between the creep and recovery compliances, shown in Fig. 3. The linear term does not affect this relation. From /4.10 / it is also seen that when only one non-linear term of / 2.8 / is retained, the difference $J_{\tau} - J_{c}$ must be one-signed function for arbitrary value of S and From Fig. 10 it is seen that the difference $J_{\tau} - J_{c}$ changes sign after short time t_{4} / point Q /. In consequence at least two non-linear terms are necessary in order to describe



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qualitatively the experimental data.

The data of additional creep test will not be confronted with functional description and only qualitative analyse will be presented.

The additional strain response shown in Fig. 11 exhibits similar effect to that observed on polyethylene by Lifshitz and Kolsky /9/. With the increase of time of loading before further load is added, the material becomes more har $t_A = 180 \text{ min. lies below the}$ dened. The curve A for curves B . C or D obtained for $t_4 = 90, 30, and$ O min. for the same initial and additional loads. This effect is more pronounced for higher values of the initial load. It should be noted that qualitatively opposite effect is observed in the recovery tests. Fig. 4 shows that with the increase of time interval preceeding unloading, the hardening of the material decreases. Similar conclusion can be made when analysing test results of Ward and Onat /4/.

The presented analysis of test results demonstrates the posibility of description of the mechanical behavior of tested polyvinyl chloride by the representation / 2.8 /. The question as how the proposed description simplifies the mathematical expressions as compared to / 2.1 / for other materials, will depend on the material response and assumed accuracy. The proposed approximation assures, however, the continuous and smooth description in cases when linear and non-linear regions are observed.

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