CREEP OF POLYMER MATERIALS IN STRUCTURAL ELEMENTS

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This paper is a survey of works published between 1964 and 1967, i.e., between the 2nd and 3nd All-Union Conferences on Theoretical and Applied Mechanics. It was presented by the author at the latter conference. Surveys of earlier works on creep of plastics [1] and on the mechanics of oriented fiberglass plastics were published in the Transactions of the 2nd All-Union Congress.

Creep in articles and structures made of polymers and polymer materials is, as a rule, more pronounced than in metals. It occurs under mechanical stresses at fairly low temperatures, sometimes close to room temperature. Hence, creep properties of polymers must be taken into consideration in calculations of long-term strength, stability, etc. of articles and structures made of such materials.

Creep is one of the mechanical relaxation phenomena. Consideration of the latter is limited in this survey to phenomenological aspects. In spite of its physical and chemical inhomogeneity, the material is considered to be a quasihomogeneous continuous medium. Only in special cases in which the properties of a composite material are investigated in terms of properties of its components, models of composite media will be examined. In such cases, one of the phases will be considered as distributed in the other in the form of separate inclusions.

Plastics, as regards their mechanical properties, are classified as isotropic or anisotropic materials. The different behavior of isotropic and anisotropic materials and articles made of these may, obviously, become apparent only when, for example, the strength of a material under complex stressing is considered or when a more or less complex structure is investigated as a whole. Under conditions of simple stressing, the behavior of an isotropic material does not differ phenomenologically from that of an anisotropic one. For this reason, we shall first consider the fundamental laws of creep under conditions of simple stressing. Incidentally, this aspect is also the most fully investigated.

Let us, first, consider the behavior of polymers under constant stress throughout the duration of a test.

As an example, the curves of short duration creep of nonplasticized polyvinyl chloride at temperature T = 19°C, taken from [3], are shown in Fig. 1. The nominal stress was maintained constant throughout individual tests. The several curves relate to different test stresses. These tests did not end in the destruction of the sample, but cold elongation did occur under high stress and after some time after load application. Creep tests in certain cases of brittle or highly elastic polymers did end in rupture of the test piece as the result of appearance and propagation of cracks. For such materials the creep curve begins to show from a certain point an increasing (instead of a decreasing, as at the beginning) rate of creep which continues up to the instant of the test piece destruction. The latter part of the creep curve for brittle plastics is comparatively short and bends upward only slightly prior to rupture. The region of increasing creep rate of elastomers, particularly of the heavily loaded variety [4], is very pronounced, i.e., the point of inflection of the creep curve appears long before rupture of the test piece and is followed by rapidly increasing deformation, with the latter sometimes exceeding several times the deformation at the point of inflection.

No attempts were apparently made to find a function which would adequately fit the entire creep curve, including its last section, for polymers, similar to that proposed by Endrade and used for de-

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scribing the creep of metals (see, e.g., [5], p. 171). However, this may not be necessary, since during the last stage of accelerated creep an intensive accumulation of damage (usually in the form of cracks) occurs in the material, which is entirely unacceptable in an industrial article or structure. Because of this, calculation methods are based on relationships defining the initial part of the creep curve along which the strain rate monotonically decreases with time. A few of these methods are presented below, together with relevant references. Materials to which these relationships were applied and, in the case of prolonged tests, the duration of experiments are listed.

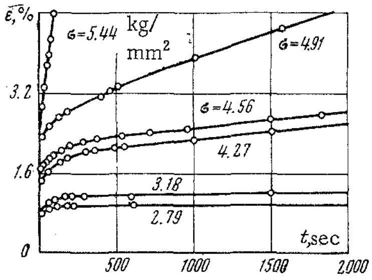

Fig. 1

Formulas Containing a Power Term. Sharma [6] (Penton), Obukhov [7] (Kapron, tests of up to 1000-h duration), Bongiovanni [8] (cellulose acetate, polymethyl methacrylate, polycarbonate, chlorinated polyester), Melent'ev [9] (polyethylene, polymethyl methacrylate, wood-laminate plastic, and others) had used for defining the dependence of strain ε on time t counted from the instant of load application an equation of the form

$$\varepsilon = \varepsilon_* t^n \tag{1}$$

(ε_* and n are, respectively, parameters of the curve and of the material).

Findley [10] (see also [1, 5]; a number of thermo- and reactoplastic, textolite, fiberglass plastics; tests up to and over 100,000-h duration), Sharma [11] (cellulose acetobutyrate; tests up to 100-h duration), Melent'ev [9] (materials as quoted above, exponent n in (2) was always assumed equal 0.25), and Smotrin and Chebanov [12] (textolite; tests up to 500-h duration) had used the relationship

$$\varepsilon = \varepsilon_0 + \varepsilon_c t^n \tag{2}$$

The strain defined by Eq. (2) is assumed to consist of two terms: one time-independent ε_0 (it will be called the momentary strain), and the second (the creep strain) increasing with time proportionally to t^n.

Melent'ev had used for describing the same experiments the three-term equation

$$\varepsilon = \varepsilon_0 + \varepsilon_c t^n + \varepsilon_v t \tag{3}$$

where the third term represents the strain increasing at a constant rate. Such strains are usually associated with viscous or viscoplastic behavior of matter. It should be noted, however, that the third term in Eq. (3) does not imply the presence of viscous or viscoplastic flow at constant nonzero stress proportional to t. For example, Plazek had established that the deformation of zinc polyphosphinate specimens, while increasing under load in proportion to t, disappeared completely after removal of load, although at first glance it could have been considered as viscous, i.e., these were elastic deformations.

Logarithmic Relationships. Khlopotov [14] (polyethylene) and Melent'ev used the relationship

$$\varepsilon = \varepsilon_0 + \varepsilon_c \lg t \tag{4}$$

Lemmens [15] (elastomere fibers) and Melent'ev [9] [who had assumed parameter τ in Eq. (5) to be equal zero] used the relationship

$$\varepsilon = \varepsilon_0 + \varepsilon_c \lg (t + \tau) + \varepsilon_v t \tag{5}$$

Ivanov and Sokolov [16] (organic glass test one year long) used the function

$$\varepsilon = \varepsilon_0 + \varepsilon_c \lg^2 (t + 1) \tag{6}$$

Exponential Functions. Bongiovanni [8] used the simplest exponential relationship corresponding to Burger's four-element rheological model (in the experiments mentioned above). This model has a single elastic recovery time or two relaxation times. Models with a limited number of elastic recovery or relaxation times are inadequate for describing the creep and relaxation of polymers, and are even less satisfactory for determining the dynamic behavior of real materials. To obtain a better correlation between theory and experimental results, the model is made more complex by the introduction of a large number of elements with a correspondingly large set of recovery and relaxation times. In this manner, a spectral method for the definition of mechanical relaxation behavior of polymers is obtained. Line, as well as

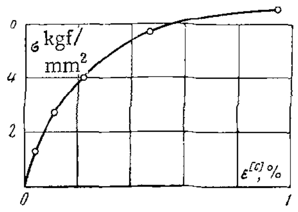

Fig. 2

continuous or band, spectra are widely used in polymer mechanics. One of such spectral models using the law of normal distribution yields the equation of the creep curve (Theocaris [17]) in the form

$$\lg \varepsilon = \lg Q_1 + \lg Q_2 \operatorname{erf}(h \lg t / k) \tag{7}$$

where k is the characteristic time and h is a parameter of the function of normal distribution of recovery time.

A very good correlation between experimental and calculated curves is obtained with the use of the so-called fractional exponential functions in which the time t appears as t^k where k is a fraction. As an example, we write the function defining the elastic aftereffect in plastic (wood–laminate, fiberglass, and other plastics) in prolonged tests under stresses below a certain critical value, called by Yatsenko [18] the limit of long-term strength. It is of the form

$$\varepsilon = a + b \exp\left[-\alpha (t/t_1)^c\right] \tag{8}$$

where α and c are parameters of the material, a and b are stress functions, and t_1 the selected unit of time. Fractional exponential functions of a more complex form are at present used in the application of the theory of heredity to the solution of problems of strain, in polymer materials and their products. These functions are in the form of power series expansions, and will be considered below.

We shall also mention the equations of relaxation curves used by various authors for representing the behavior under conditions of constant strain $\varepsilon = \varepsilon_0 = \text{const}$.

Obukhov [7] had used the power relationship

$$\sigma = \frac{\sigma_0}{1 - [1 - (t^* / t)^n]^{1-n}} \tag{9}$$

Here n is a parameter of the material and σ_0 is the maximum stress reached at the instant $t = t^*$ of ending of loading applied at a constant rate $\varepsilon^\cdot$ until ε becomes equal to ε_0. Since $n \ll 1$, the author considers it reasonable to assume that $1 - n \approx 1$, which yields the equation

$$\sigma = \sigma_0 (t / t^*)^{-n} \tag{10}$$

Wegener [19] used a series expansion in powers of $\ln(1 + t)$ as the equation of the relaxation curve of polyamide-6. He has also noted the effect of the rate of relaxation $\varepsilon_0^\cdot$ at the beginning of the α test on the relaxation function [20] as

$$\sigma = \sum_{m,n} A_{mn} [\ln(1 + t)]^m (\varepsilon_0^\cdot)^n \tag{11}$$

where m varies from 0 to + 5, n from −6 to + 3, and the coefficients A_{mn} can be either positive or negative.

The equation used by Theocaris [17] is similar to Eq. (7). Slonimskii [21] used an equation containing an exponential function of fractional order of the form

$$\sigma(t) = \sigma_\infty + \sigma_r \exp(-at^k) \tag{12}$$

where σ_∞ and σ_r are parameters of the curve and a and k those of the material.

All of the above relationships for creep and relaxation curves describe more or less adequately the creep of polymer materials in a certain average time range attainable in investigations. Attempts at extrapolating these relationships to regions of very long or very short times t may yield estimates significantly different from actual characteristics of the material. Furthermore, the simple relationships cited above may not be satisfied in very wide intervals of time. Thus, for example, Tang [22] had described the creep of polyethylene by (2) for times of less than 1-min duration, and by (4) when t > 1 min.

For estimating the long-term strength of an article intended for a long service life, it is important to investigate the behavior of the material during considerable periods of time t which could be considered as tending to infinity in comparison with the time scale available for tests. From this point of view of particular interest are data provided by Findley who had shown that Eq. (2) holds for a considerable number of plastics tested in experiments of up to and over 100,000-h duration, i.e., the deformation for $t \to \infty$ increases proportionally to t^n. Smotrin and Chebanov [12] and also Yatsenko [18] had shown that at sufficiently high stresses greater than a certain critical σ_p, called in [12] the limit of proportionality and

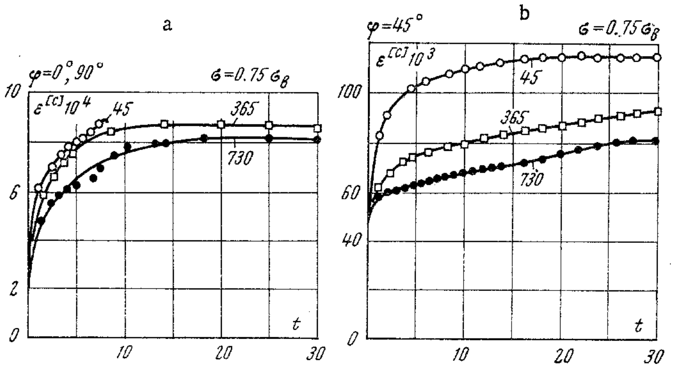

Fig. 3

in [18] the limit of long-term strength, the total deformation of the material contains a plastic component whose rate is independent of t, while being proportional to $\sigma - \sigma_p$. For certain reinforced plastics, such as fiberglass and wood–laminate plastics [18], subjected to not very high stresses, the strain ε approaches a certain limit when $t \rightarrow \infty$, i.e., the creep of such materials is limited.

The following singularities of creep and relaxation functions at $t \rightarrow 0$ should be noted. Equation (4) does not hold for very small t, since it results in an absurd estimate of strain for $t \rightarrow 0$. Hence, $t + \tau$, as in (5), or $t + 1$, as in (6) and (11), are to be taken as the arguments of the logarithmic terms of the creep or relaxation functions. Reverting to Fig. 1, we note that for small t the strain after load application increases very rapidly, so that at the beginning, the creep curve virtually merges with the axis of ordinates. Attempts at experimental determination of the actual rate of creep at σ = const and very short t (e.g., of the order of a hundredth of a second) end in failure, since either the creep rate is beyond the capacity of existing recording instruments, or the tensile test machine is incapable of producing a sufficiently high rate of loading; or, when the latter is very high, it is impossible to eliminate vibratory effects. Because of this, virtually all investigations of creep phenomena have come to the conclusion that for an actual material the creep function must necessarily have a weak (integrable) singularity, such that at σ = const and very small $t < \Delta$ (Δ is any arbitrary small positive number) the rate of creep $\dot{\varepsilon}$ infinitely grows, when Δ tends to zero. Such a singularity implies that τ in (5) must be equal to unity. Hence, the notable tendency of investigators to resort in the analysis of creep problems to creep and relaxation functions and also to use kernels of related integral equations with a weak singularity at $t = 0$.

When considering problems of creep in structures made of polymer materials, it is important to know whether the material is linear. In creep tests of linear material, all components of strain, elastic, viscous (or viscoplastic), and of elastic aftereffects are proportional (under otherwise equal conditions) to the applied stress. A useful procedure for determining whether a material is linear is to plot the so-called isochronous curves $\sigma - \varepsilon$ derived, for example, from creep curves. A set of creep curves obtained for various σ makes it possible to find the values of ε (or creep strain $\varepsilon^{[c]}$) corresponding to each σ for the same values of t which, in this case, is considered to be a parameter. The points thus obtained represent, when plotted in the $\sigma-\varepsilon$ coordinates, the isochronous curve. An example of such curve in the $\sigma-\varepsilon^{[c]}$ coordinates is given in Fig. 2 for samples of isotropic fiberglass plastic 33-18°C cut at an angle of 45° to the direction of glass fibers and tested at 30°C with $\varepsilon^{[c]}$ calculated for 168 h. It is seen from this figure that at low stress the strain $\varepsilon^{[c]}$ is approximately proportional to the stress; a further increase of stress causes the strain to cease to be proportional to σ and to increase at a more rapid rate, which indicates the lowering of the strength of the material with increasing σ. This kind of nonlinearity which results in the decrease of the strength with increasing σ is typical of rigid and strong polymer materials, while another nonlinearity is typical of elastomers whose rigidity under increasing σ first decreases, then increases, and finally, falls again.

Of considerable interest and importance is the question whether the nonlinear properties of polymer materials are in evidence right from the beginning of load application (i.e., from $\sigma = 0$), or if there is a certain region of linear behavior. All investigators who have conducted experiments for clarifying this aspect favor the latter assertion. A large number of works is devoted abroad to the linear viscoelastic behavior of polymers (see, e.g., [23, 24]). From a number of Soviet works which mention the question of

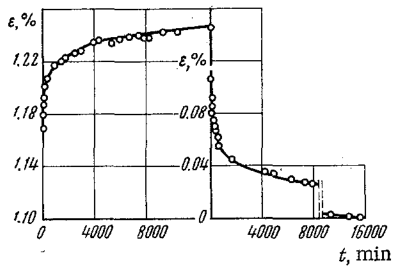

Fig. 4

the existence of a region of linear viscoelastic behavior of polymers and polymer materials, we would mention three dissertations.* Thus, Ozerov had noted that the epoxy compounds investigated by him had shown viscoelastic properties at a temperature of 295°K under stresses up to $\sigma = 0.8\sigma_b$, where σ_b is the tensile strength of the material at the given temperature, while at higher stresses its nonlinear properties became apparent. Under different conditions and in other polymers, the nonlinearity of the viscoelastic behavior is more pronounced.

In concluding the survey of creep in plastics under constant stress and other constant conditions, we would note that, as a rule, actual articles and structures are subject to varying stresses. In the case of materials with creep properties, this is not only due to variation of external forces, and may occur under constant load.

The behavior of materials under varying stresses is described by four, at present generally accepted, theories of creep: aging, flow, toughening, and heredity [5]. An important shortcoming of the first two of these is that their equations are not invariant with respect to the beginning of time count. The theories of aging and of flow can, however, provide satisfactory results, if the main load acting on the body varies only slightly with time and if the instant of its application is taken as the reference time. Thus, for example, stress–relaxation curves calculated by the theories of aging and flow from results of creep tests show a satisfactory correlation with data of direct experiments on relaxation (Minenkov: data on Kapron [25], polyethylene, and polypropylene [26]).

With certain limitations (less rigid than here described) the theory of toughening [7] also yields satisfactory results. However, the conventional toughening theory does not provide for recovery after the removal of load [5]. Hence, to obtain an acceptable correlation between theoretical estimates and experimental results on test pieces and articles, it is necessary to suitably modify these theories.

The theory most widely applied to plastics is that of heredity, which for varying stresses fits experimental data better than any of the theories mentioned above. The heredity and, in particular, the linear properties of materials may in certain cases be conveniently represented by mechanical models consisting of springs, viscous elements, etc. Models of linear media contain only linear elements, such as springs and dashpots. Nonlinear mechanical models comprising failing elements were proposed for weakening and thixotropic materials by Regel [27] and by Leonov [28].

The strain properties of a linear mechanical model are equivalent to a medium satisfying the linear differential equation

$$P\varepsilon = Q\sigma \tag{13}$$

where P and Q are differential operators of the form

$$\sum_{i=0}^{k} a_i \frac{d_i}{dt^i}$$

Equation (13) may be rewritten in the form containing Volterra's integral operator. The kernel of the obtained equation can be considered completely independently from the operator equation (13) or from the related rheological model. Only the Bolzmann–Volterra general principle of superposition, according to which the strain produced by the stress $\sigma_1 + \sigma_2$ is equal to the sum of strains produced by σ_1 and σ_2, is retained. It is also assumed that the properties of the medium are time-independent, and aging is excluded. This assumption is not exactly correct for plastics, since their properties vary with time. Data on creep of AVAM fiberglass plastic as function of its "age" (shown in the diagram in days) from the day of its

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*G. I. Bryzgalin, "Certain problems of creep of structural plastics," Candidate's Disseratation, SO AN SSSR, Novosibirsk (1964).

N. I. Malinin, "Analysis of the problem of creep and strength of plastics," Doctoral Dissertation, Institute of Problems of Mechanics of the AN SSSR, Moscow (1965).

V. I. Ozerov, "Investigation of anistropic viscoelasticity of oriented fiberglass plastics and of cylindrical shells in this material," Candidate's Dissertation, Institute of Mechanics of the AN UkrSSR, Kiev (1966).

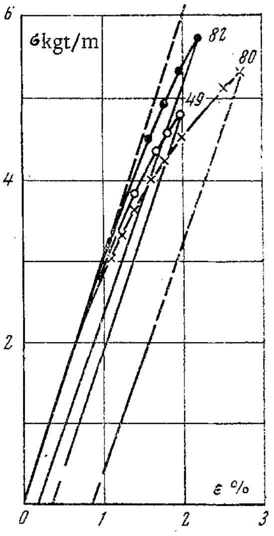

Fig. 5

manufacture are shown in Fig. 3 [29], where φ is the angle between the test piece center line and the direction of the glass fibers, and σ_b is the strength of test pieces determined on a tensile test machine. These diagrams indicate that the strength and rigidity of the material increase in the age range of 45-730 days. In certain cases, the strength and rigidity of polymer materials does, however, decrease with time, particularly when aging takes place in humid or corrosive atmosphere. Contemporary creep theories, primarily developed for concrete, can be recommended for stress calculations of articles of polymer materials, whenever it is essential to take into consideration the variation of properties of such materials in time. So far theories of this kind have not been applied in stress calculations of articles and structures made of polymer materials.

According to the Bolzmann–Volterra principle of linear superposition the contribution of the σ-stresses, prevailing in the time interval θ preceding the instant of time t, to the strain at instant t is

$$d\varepsilon^{[c]} = \sigma(\theta) J(t-\theta)\, d\theta \tag{14}$$

where J is a smoothly decreasing function of its argument – a function of "memory" or the kernel of creep. Integrating this relationship from 0 to t (we assume that up to the instant t = 0 the body was free of stresses) and taking into account the elastic strain, which according to the hypothesis of linearity of properties is related to stress by the Hooke's law, we obtain

$$\varepsilon(t) = \frac{1}{E}\left[\sigma + E\int_0^t J(t-\theta)\,\sigma(\theta)\, d\theta\right] \tag{15}$$

This linear integral equation, when considered in relation to the creep test, shows that the total strain consists of two parts: one time-independent, the other – a function of time. If such separation of strain could be obtained with adequate accuracy from a creep curve, the modulus of elasticity E and the creep kernel J(t) would be, by the same token, also determined. Unfortunately, such separation is not possible, since, owing to the previously mentioned singularity of creep curves at t = 0, this cannot be made objectively. Hence, the modulus of elasticity E is determine by some special experiments. It can, for example, be derived by the frequency method (in which case it is unavoidably a function of circular frequency) or, as in [3], from the diagram of rapid load removal. In principle, it is not always necessary to separate the elastic strain from that of creep, i.e., it is possible to consider the total strain as due to creep. This results in the degeneration of Volterra's integral equation of the first kind (15) into an integral of the kind of Stieltjes convolution.

The integral equation (15) can be written with the strain ε as the integrand. This results in another function – the relaxation kernel K(t) which is the resolvent of the creep kernel – appearing in the integral. The properties of kernels J(t) and K(t) at $t \to \infty$ and $t \to 0$ are of considerable interest. The tendency of J(t) and K(t) to vanish when $t \to \infty$ is equivalent to the condition of complete reversibility. Here, obviously, we mean not the thermodynamic reversibility, but the total recovery of strain on removal of load. Rigid and strong plastics, as well as three-dimensional ones, behave in exactely this manner under not too high stresses, as can be seen from Fig. 4 from the work of Zambakhidze and Rabinovich [30]. The curve shown in this diagram relates to the "uniformly strong" SVAM fiberglass plastic under tension of ~ 32 kg/mm^2 in the direction of one of the glass fibers. Its recovery curve at T = 70°C and $\varphi = 0$ is also shown there. It is clear from this figure that the recovery is total.

Singularities at $t \to 0$ of the form $\dot{\varepsilon} = \infty$ on the creep curve and of the form $\dot{\sigma} = -\infty$ imply that the kernels must have a weak (integrable) singularity. Fractional exponential functions in the form of power series expansions for describing creep and relaxation were mentioned above. Below we shall write the expressions for the kernels of two functions with the view to their application to solving practical problems by operational-symbolic methods. For the Rabotnov kernel (see [5], p. 128), we have

$$\vartheta_\alpha(\beta, t-\tau) = \sum_{n=0}^{\infty} \frac{\beta^n (t-\tau)^{n(1+\alpha)-\alpha}}{\Gamma[(n+1)(1+\alpha)]} \tag{16}$$

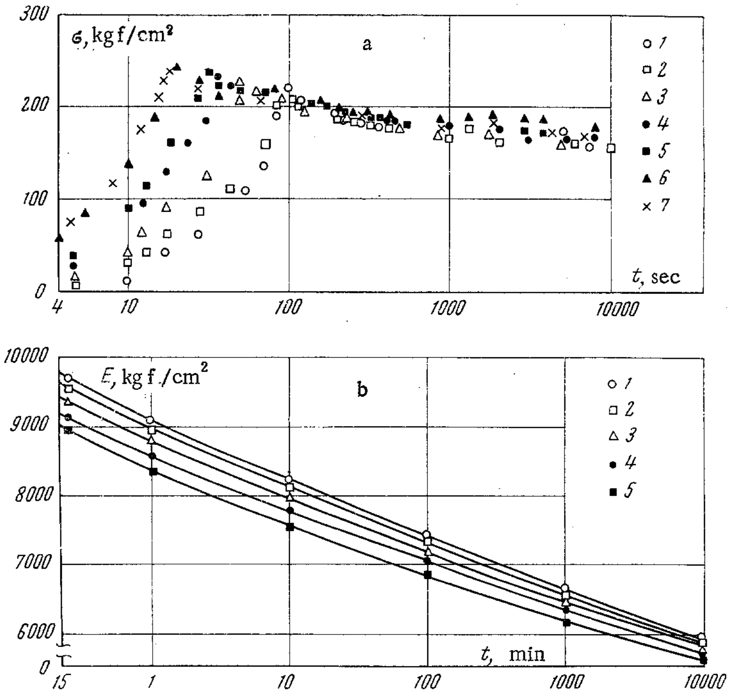

Fig. 6

The second kernel, considered in [31-33], is the resolvent of the Rzhanitsin kernel, which is a fractional exponential function. It is of the form

$$E_\alpha[\beta, \varkappa, t-\tau] = e^{-\beta t} \vartheta_\alpha(\varkappa, t-\theta) \tag{17}$$

It was shown in the previously mentioned dissertations that the Rabotnov kernel (16) defines with sufficiently high accuracy the creep of polymers and polymer materials. Furthermore, Ozerov, who in his dissertation had dealt with various loading programs including those in which stress and strain were varied in jumps, confirmed once again the validity of the Bolzmann–Volterra principle of superposition for polymer materials (epoxy resins, fiberglass plastics) in the region of linear behavior of these.

A method of computer calculation yielding a reasonably accurate determination of the Rabotnov kernel parameter from experimental data was suggested in [34].

The nonlinear viscoelastic behavior of polymers at fairly high stresses has been already noted above. The nonlinear relationships of viscoelasticity of polymers can be derived, if one assumes that the inherited strain not only contains the contribution defined by the right-hand side of Eq. (14), but also depends on terms which determine the effect of stresses acting at various instants of time preceding the one under consideration. This assumption leads to the nonlinear Frechet integral equation written for nonaging materials with heredity properties in the form

$$\varepsilon(t) = \sum_{i=1}^{k} \int_0^t \ldots \int_0^t \sigma(\theta_1)\ldots\sigma(\theta_i)\, J_i(t-\theta_1, \ldots, t-\theta_i)\, d\theta_1 \ldots d\theta_i \tag{18}$$

where kernels J_i can have strong singularities, for example, in the form of Dirac δ-functions; these strong singularities determine momentary strains.

The nonlinear integral equations of the form of (18) were used by Ward [35, 36] for defining creep in uniaxial tension of polypropylene fibers, while Findley [37] had used these for describing the creep of nonplasticized polyvinyl chloride under conditions of complex stressing. Ward retained in the Frechet expansion single and triple integrals, and Findley used single, double and triple integrals. Control experiments carried out by these authors had shown a good correlation between theoretical estimates and experimental data.

Unfortunately Eqs. (18) are complex and their use for solving practical problems is difficult, hence, the understandable efforts of several investigators to find simple relationships for describing the heredity

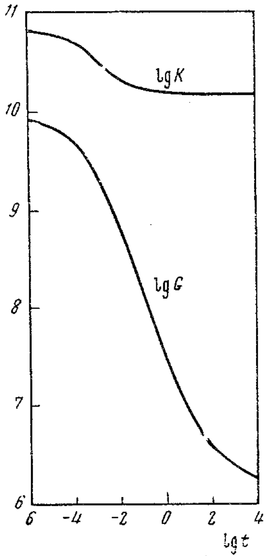

Fig. 7

properties of polymers. The writer [3] has made the following attempt. Let us assume that the dependence of the inherited strain on stress $\sigma(\theta)$ is not linear, as in (14), but is of the more general form

$$d\varepsilon^{[c]} = J\,[\sigma(\theta),\, t - \theta]\, d\theta \tag{19}$$

If the elastic strain is a linear function of stress, the integration of this equation yields

$$\varepsilon(t) = \frac{\sigma}{E} + \int_0^t J\,[\sigma(\theta),\, t - \theta]\, d\theta \tag{20}$$

The linearity of the dependence of the elastic strain $\varepsilon^{[e]}$ on stress was checked for nonplasticized polyvinyl chloride. As an example, the curves related to loading and unloading at a temperature T = 15°C are shown in Fig. 5. The loading rates for test pieces Nos. 49, 80, and 82 were, respectively, $\sigma^{\cdot} = 8.6 \times 10^{-3}$ kg/mm^2 sec, 3.86×10^{-3} kg/mm^2 sec, and 6.6 kg/mm^2 sec.

The application of load in these tests was fast with an even faster unloading (0.03 sec), which was considered to be virtually instantaneous. The unloading was represented on the diagram by a straight line, and the Young modulus was determined by the tangent of its angle of inclination.

Experimental checks had confirmed the nonlinear principle of superposition defined by Eq. (20) and have enabled us to distinguish more or less clearly three subregions in the range of stresses from $\sigma = 0$ to $\sigma = \sigma_b$ (ultimate tensile stress). In the linear subregion the material behaves as a viscoelastic one. In the subregion of weak nonlinearity, function $J(\sigma, t - \theta)$ can be represented as the product of two functions: one, a function of only σ, and the other of only $t - \theta$. Finally, at high stresses we have the subregion of strong nonlinearity where such separation is not possible. The latter aspect can be considered together with the effect of the acting stress on the relaxation time of the material, as was done by Rabinovich (see e.g., [2, 30]).

For an anisotropic body, Eq. (20) was rewritten by Bulgakov [38, 39] in the generalized form

$$\varepsilon_{ij}(t) = \varepsilon_{ij}^{[e]}(t) + \int_0^t Q_{ij}\,[t - \theta,\, \sigma_{\alpha\beta}(\theta),\, T(\theta)]\, d\theta \qquad (i, j, \alpha, \beta = 1, 2, 3) \tag{21}$$

which takes into account possible temperature effects.

He had carried out tests at constant temperature on tubular test pieces of organic glass and polyester resin subjected to tension, compression, and torque, as well as to combined compression and torque. These had shown that the nonlinear principle of superposition defined by (21) is satisfied with reasonable accuracy at T = const.

Yet another equation for describing the heredity properties of polymer materials in the region of strong nonlinearity was proposed in 1951 by Rozovskii. It is based on the nonlinear integral equation of the Volterra kind of the form

$$\varepsilon(t) = \frac{\sigma}{E} + \int_0^t J\,(\sigma,\, t - \theta)\, \sigma(\theta)\, d\theta \tag{22}$$

Here, unlike in Eqs. (20) and (21), the stress σ in the argument of the nonlinear kernel J is considered as a function of $t - \theta$. Equation (22) was used in [40] for comparing the results of experiments on creep in a condition of periodic loading with those under static load. The relationship derived by the author for the determination of creep properties of a material in the presence of periodic stress component on the basis of static tests was experimentally checked (by Buyanov, Kasyuk, and Panshin) on CO-120 organic glass under conditions which prevented heat generation in the sample itself. This check had confirmed the possibility of using Eq. (22) for determining the heredity properties of materials, provided the temperature of the sample, either caused by external or internal heating owing to dissipation of mechanical energy, remains

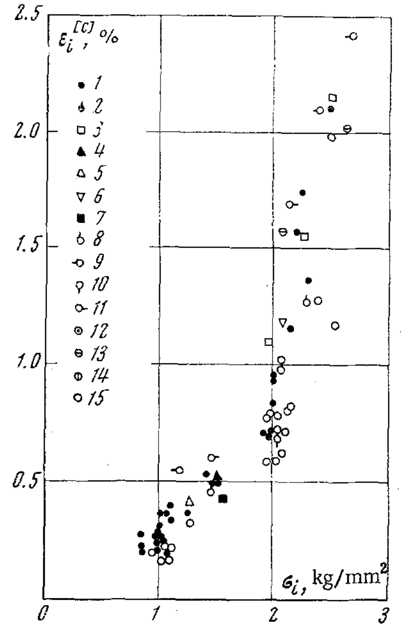

Fig. 9

[see, e.g., Eq. (15)], except that time normalized with respect to temperature is substituted everywhere for actual time. Moreover, a term which takes into account temperature-induced stresses is introduced in the physical relationships. The concept described in [46, 47] is supported by Il'yushin and Ogibalov [48].

The formula of Williams, Landel, and Ferry (see, e.g., [23]) is generally used for the determination of the dependence of coefficient a_T on temperature. Recently this formula was experimentally confirmed, although the basic concept of its authors was criticized.

It should be noted that the concept of simple thermorheological behavior and also relationships of the form of the Williams–Landel–Ferry formula are valid for amorphous polymers only. A number of crystalline polymers can be listed for which this concept is completely invalid. The greater complexity of the temperature-dependence of relaxation of crystalline polymers is caused by considerably greater sensitivity of their structure to temperature and other effects and the effect of the structure on the properties of such materials is much more pronounced than in the case of amorphous polymers.

When considering the viscoelastic behavior of polymers and polymer materials in variable and nonstationary temperature fields, it can also be assumed that the parameters and functions appearing in the hereditary relationships depend on temperature. Equation (21), which also contains the temperature, can be applied to a wider range of materials than the thermorheologically simple ones.

Problems of creep in plastics under conditions of unidirectional stress were considered so far. Under conditions of complex stresses, the behavior of polymer materials depends to a considerable extent on whether these are isotropic or anisotropic. Let us first consider the properties of isotropic materials.

In the case of isotropic materials with linear-heredity properties the system of physical equations relating the components of stress–strain tensors is divided into two tensor equations: one for spherical tensors, the second for deviators. For a body with linear properties, the relation between the σ_{ij} and the ε_{ij} components can be written in the form

$$\varepsilon_{ij}(t) = \delta_{ij}\left[B_0\sigma_{kk}(t)] + \int_0^t B_1(t-\theta)\,\sigma_{kk}(\theta)\,d\theta\right] + J_0\sigma_{ij}(t) + \int_0^t J_1(t-\theta)\,\sigma_{ij}(\theta)\,d\theta \tag{24}$$

This is a Volterra integral equation of the first kind. Summation is carried out with respect to the double subscripts. For a weakly nonlinear body, Eq. (24) was generalized by Findley, and, in accordance with Leaderman's nonlinear integral equation, written as follows:

$$\varepsilon_{ij}(t) = \delta_{ij}\left\{B_0 f\,[\sigma_{kk}(t)] + \int_0^t B_1(t-\theta)\,f_1[\sigma_{kk}(\theta)]\,d\theta\right\} + J_0 F\,[\sigma_{ij}(t)] + \int_0^t J_1(t-\theta)\,F\,[\sigma_{ij}(\theta)]\,d\theta, \ldots \tag{25}$$

where f and F are nonlinear functions of σ_{kk} and σ_{ij}, such that at low stress f and F degenerate into σ_{kk} and σ_{ij}, respectively.

The majority of authors, when solving practical problems, take into consideration hereditary properties only in relation to shear strain. Polymer materials under volume tension (compression) behave as elastic bodies and are sometimes considered as being incompressible. In the majority of cases, the inaccuracies in the definition of bulk properties of a material (the adjective "bulk" is used here to denote the properties of material relative to the strain resulting from volume tension or compression) do not appreciably affect the results of calculations of the pattern of the stress–strain state. There are, however, problems in which an injudicious choice of the law defining bulk properties substantially affects the results. One of such is the plane problem of deformation of a hollow viscoelastic cylinder with an outer elastic shell and an inner ablative surface under internal pressure. This problem was considered by

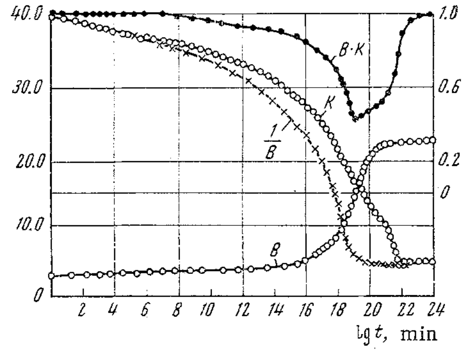

Fig. 8

constant. If the periodic component of stress has a sufficiently high frequency and a not very small amplitude, the self-heating of the material induced by mechanical losses must be taken into account [41]. The phenomenon of vibration-induced heat generation in polymer materials was investigated by Barenblatt et al. [41], who had noted that this did sometimes increase the rate of creep by 2/10–3/10. As the main cause of creep acceleration under vibratory conditions, the authors of [41] considered the effects of vibratory heating which results in lowering of mechanical properties of polymers and the related increase of the rate of $\varepsilon^\cdot$ for a given σ, in accordance with the known temperature-dependence of the relaxation properties of polymer materials. Maksimov and Urzhumtsev [42] found that small vibrations accelerate the creep rate not only by increasing the temperature of material, but also in consequence of some other effects unconnected with those of temperature.

When dealing in a previous section with stress relaxation, attention was drawn to the effect of the loading program for the initial stage during which the stress rises from $\varepsilon = 0$ to $\varepsilon = \varepsilon_0$. In addition to the data presented there, we should note the work of Sharma [43], who had investigated the stress relaxation of penton in the nonlinear region, and for the curves defining this phenomenon at various rates of strain $\varepsilon_0^\cdot$ during the loading stage proposed the relaxation function of the form

$$\Psi(t, \varepsilon_0^\cdot) = \Phi(t)(\varepsilon_0^\cdot)^n \qquad (23)$$

where time t is counted from the instant at which the strain reaches a specified value.

It should be noted here that the effect of the rate of $\varepsilon_0^\cdot$ on the pattern of consequent creep and relaxation curves is in itself not surprising. Since various rates of $\varepsilon_0^\cdot$ obtain at various loading programs, the laws of stress variation with time will also vary in the course of an experiment on relaxation. However, owing to the indicated above property of hereditary materials, according to which the creep and relaxation kernels tend to vanish when their argument $t - \theta$ tends to infinity, the material, after extended periods of time, must have a tendency to "forget" gradually the initial singularities of the loading program. For example, the relaxation curves obtained for various $\varepsilon_0^\cdot$ should in the course of time merge into a single curve by virtue of the laws of linear and nonlinear heredity. Koltunov [44], referring to his not entirely convincing data, suggests the introduction of $\varepsilon^\cdot$ in addition to ε and σ into his nonlinear integral equation of the Rabotnov kind (see [5], p. 209).

The effect of initial stages of the loading program on the relaxation curve pattern, based on data of Sharman and Haas [43], is shown in Fig. 6. Initial sections of relaxation curves obtained during tests at five different rates of $\varepsilon_0^\cdot$ are shown in Fig. 6a, where points 1-7 relate to $10^5 \times \varepsilon_0^\cdot = 23$, 27, 41, 56, 66, 108, and 115, respectively. At the beginning the increase of σ is consistent with the rate of $\varepsilon_0^\cdot$, then follows the relaxation process. Results of a more prolonged experiment are shown in Fig. 6b, where points 1-5 relate to $10^4 \times \varepsilon_0^\cdot = 25$, 20, 15, 10, and 5, respectively. The ratio of rates of $\varepsilon_0^\cdot$ in the various tests was approximately the same as in Fig. 6a. According to conventional theories of heredity, the differences in the initial stages of tests on various specimens should have vanished from their memories after 10,000 min, and the curves should have merged. However, it was not so, as shown by the approximately 5% difference in the value of σ on curves corresponding to the lowest and highest values of $\varepsilon_0^\cdot$, respectively.

As regards the temperature-dependence of creep and relaxation properties of polymers and polymer materials, the following should be noted. The majority of investigators consider polymers and polymer based materials as thermorheologically simple bodies. The term "thermorheologically simple body" was introduced in 1952 by Schwartzl and Staverman [45]. The strain of such body under a given stress and at temperature varying from T_0 to T is the same as at temperature T_0, but the rate of creep strain development is altered by a factor a_T.

Moreland and Lee [46] and later Sternberg and Gurtin [47] proposed to consider problems of viscoelasticity in variable and nonstationary temperature fields of thermorheologically simple bodies in the following manner. In the general system of equations defining the problem, only the equations of physics are associated with temperature. The latter are written in the form corresponding to constant temperature

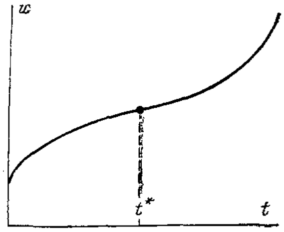

Fig. 10

Huag et al [49], who had used the law of bulk relaxation derived from experimental data obtained by Merwin, Aldrich, and Seki with the use of temperature–time superposition, as described in [23], pp. 467-468.

Data on the frequency-dependence of polyisobutylene bulk modulus at T = 25°C, converted to the bulk relaxation modulus by the approximate formula of Ninomi and Ferry ([23], pp. 93), are shown in Fig. 7 in log K–log t coordinates (K is the bulk modulus in dyne/cm^2). It is seen that the variation of the relaxation modulus K(t) in the range of log t from -5 to +2 is of the order of 0.85 tenths (t is in sec). The dependence of the relaxation modulus G on time is, also, shown for comparison. The analysis had shown that in this problem the results of stress field calculations depend on the extent to which the bulk creep of the material is taken into account.

Sharma and McCarty [50] had measured directly the bulk creep of the "Hycar" elastomer (cross-linked copolymer of butadene and acrylic acid) highly filled with inert and live fillers. The bulk creep and relaxation moduli were investigated by Theocaris [51]. His data on these moduli for the C-100-O-8 cold-setting epoxy resin are shown in Fig. 8. To extend the time range he had also used the principle of temperature–time superposition.

The temperature of 298°K is normalized by

$$B^* = T/298\, B(t)\cdot 10^{-9}\ \mathrm{dyn}\times\mathrm{cm}^2;\ K^* = \frac{258}{T}\, K(t)\cdot 10^{11}\ \mathrm{dyn}^{-1}\times\mathrm{cm}^2$$

The coefficients at B and K represent temperature corrections which take into account the increase of the equivalent modulus of the polymer with increasing temperature, in accordance with the kinetic theory of elasticity of rubber.

The product of the relaxation modulus and pliability is also shown in Fig. 8. According to the formula given by Bronskii [52], this product must be equal to unity. It is, however, seen that when creep is sufficiently prominent, it differs considerably from unity.

Findley [10] had experimentally checked the possibility of using Eqs. (24) and (25) for defining the creep of isotropic glass-like polymers (annealed strong polyvinyl chloride tested at room temperature). It was established that Eq. (24) and, even better, Eq. (25) in which functions f and F depend on their arguments according to the hyperbolic sine law, define the inherited component of total strain with adequate accuracy. Nonlinear integral equations of the form of (18) with single, double, and triple integrals retained were used in [37] for describing the nonlinear hereditary properties of isotropic polymers under complex stress and complicated loading program.

This equation with respect to tensor components ε_{ij} and σ_{ij} is quadratic, since third power terms are expressed by quadratic terms in accordance with the Cayley–Hamilton identity. Scalar functions, of which there are 12 or 9 depending on whether the stress state is three-dimensional or plane, contain the first, second, and third invariants of the stress tensor.

In concluding this section, it seems expedient to consider the results of experiments on creep [53] carried out on tubular test pieces of polymethyl metacrilate plastic under combined tension and torque:

points	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*
$\tau/\sigma =$	0	0.2	0.3	0.34	0.4	0.5	0.62	0.8

points	*9*	*10*	*11*	*12*	*13*	*14*	*15*
$\tau/\sigma =$	0.9	0.94	1.0	1.07	1.8	2.0	$-\infty$

These results are shown in Fig. 9 in coordinates of creep strain $\varepsilon_i^{[c]}$ and σ_i. The test, which lasted 5 h, was at room temperature and under simple loading. The ratio of tangential to tensile stress J/σ was varied from 0 to ∞. It can be seen from Fig. 9 that in spite of some scatter the experimental points lie in a fairly narrow band. All points obtained at constant τ/σ ratio lie in an even narrower band, which shows that the position of a particular point in this diagram depends not only on the given σ_i, but also on the τ/σ ratio. Thus, under specific conditions σ_i defines ε_i in the first approximation only. It seems that the relationship between scalar invariants of the stress and strain tensors is affected not only by the second variant of tensor σ_{ij}, but also by the first and third invariants. Unfortunately, this problems remains at present virtually unexplored.

Certain data cited in the same paper prove that the relationship between deviators of stress and strain tensors is linear with respect to these tensors only in the first approximation, while nonlinearity may become prominent at considerable strains of the order of 100%.

A series of experiments on creep of polymer materials under complex stressing in simple and complicated loading programs was carried out at the Institute of Polymer Mechanics [54, 55]. The obtained data are interpreted on the basis of Malmeister's theory, a variant of the plastic slip theory.

It should be noted that from the point of view of maximum utilization of the strength of material the use of isotropic plastics in highly stressed articles and structures is inadvisable. Technologists, designers, and engineers have at their disposal various means for increasing the resistance of polymer materials in the direction of maximum stress, sometimes at the expense of weakening the material in another direction where stresses may be much lower.

Generally anisotropic materials have, also, anisotropic creep properties. Unfortunately there have been recently only a few works dealing with the investigation of anisotropic polymers on the basis of phenomenological theories. As an example of experimental work in this field, the paper by Smotrin and Chebanov [12], who had investigated creep of testone test pieces cut under various angles to the base material, may be quoted.

Attention should be drawn here to two theoretical papers devoted to the determination of the stress–strain relationship at considerable deformations. Ultimate strain can be determined by various means of which the most theoretically sound is the determination of the stress tensor in terms of the metric tensor of a moving system of Lagrangian coordinates related to mass points by equation [56]

$$\varepsilon_{ij}(t) = {}^1\!/_2\,[g_{ij}(t) - g_{ij}(0)] \tag{26}$$

where $g_{ij}(t)$ are the metric tensors of this system at instant of time t, and $g_{ij}(0)$ are the tensors at $t = 0$ [$g_{ij}(0)$ is usually taken as the unit tensor]. The strain is thus uniquely defined by the metric tensor $g_{ij}(0)$.

The most general expression for the relationship between the stress and strain tensors for a medium with time-independent hereditary properties was proposed by Pipkin [57] in the form

$$\sigma_{ij}(t) = \frac{\partial X_i(t)}{\partial x_p}\,\frac{\partial X_j(t)}{\partial x_q}\,S^{pq}\,[g_{rs}(t-\theta)\,\big|_{\theta=0}^{\infty}] \tag{27}$$

where S^{pq} is the stress tensor at a given point of the material in the system of Cartesian coordinates X_i and x_i attached to the material. The expression in brackets implies that S^{pq} is a functional of g_{rs}. The functional is subsequently expanded into a Frechet series, and various possible forms of presentation of the obtained relationship are analyzed.

Similar constructions are developed by Herrmann [58] with the use of a variant of the potential theory.

In conclusion we note certain practical investigations devoted to the development of fundamental principals of the theory of viscoelasticity and of solving boundary value problems.

An attempt at a strict axiomatic construction of a linear theory of viscoelasticity was made by Gurtin and Sternberg ([59] and other works). They have investigated such aspects as the existence and uniqueness of solution, analysis of certain properties of boundary value problems, generalization of known theorems and principles of the theory of elasticity (Beatty's theorem, the prinicple of St. Venant, etc.), and the possibility of extending the methods of Galerkin, Papkovich, and others to problems of viscoelasticity.

Theoretical problems of viscoelasticity differ from those of elasticity in that physical relationships in the former contain time, so that the stress–strain relationship appears there either as an operator equation with d(·)/dt as the operator, or as a Volterra equation of the kind described in the foregoing. To solve the stated boundary value problem of the theory of viscoelasticity the difficulties related to the time-dependence of the deformation process must be overcome. Many problems are solved by using the Laplace transformation (see, e.g., [60]) which reduces it to the so-called conjugate problem of elasticity in the mapping space. The solution of such problems depends on the transformation parameter p. The solution is obtained by carrying out the inversion of transformation. In certain cases, solution of the conjugate problem of elasticity is very involved, and the inversion of the Laplace transformation impossible. In such cases, numerical methods, usually with the aid of a computer, are resorted to (see, e.g., [61]) or various approximate methods are used [62].

Morland [63] solved the problem of rolling contact of the linear theory of viscoelasticity by means of the Fourier transformation.

In certain problems (e.g., those containing mixed boundary conditions or related to an ablating surface), the conjugate problem of elasticity remains undefined and the method of integral transformation cannot be used.

Problems of linear viscoelasticity can be analyzed by the method of Volterra based on the following. Differentiation (when operational equations of physics are used) and integration (if these equations are integral equations of Volterra) with respect to time are interchangeable with similar operations with respect to space coordinates. Hence, problems of viscoelasticity can be solved as in the theory of elasticity, always bearing in mind that the parameters in the equations of physics are not merely constants but time-dependent differential or integral operators. The integration of differential equations in partial derivatives with respect to space coordinates yields differential, integral, or integro-differential equations with t as the variable. Integration of these equations provides the solution of the problem. Examples of this approach appear in the papers of Shinozuka [64] and of Badran [65]. Owing to the great complexity of these equations, their integration is carried out by numerical methods on a computer.

Rabotnov [5] and later Rozovskii with his collaborators had developed a method based on the principle of Volterra, according to which the solution of a problem of elasticity, in which viscoelastic operators have been substituted for elasticity constants, are also valid for viscoelastic materials. The interpretation of such solutions presents certain difficulties, owing to the necessity to carry out algebraic and other mathematical operations on the operators. Rabotnov and Rozovskii with their collaborators [16] had developed a fairly detailed algebra for operators with fractional exponential kernels. A number of asymptotic formulas aimed at the simplification of calculation of Э_{α}-functions for a considerable range of t, is proposed. Tabulation of Э_{α}-functions has now been completed at the Mechanical Engineering Institute. An attempt is being made by Gromov [33] to formulate an algebra of integral operators based on kernels (16).

The methods described here have been used by numerous researchers in the USSR and abroad for analyzing many problems of the theory of linear viscoelasticity.

Particular attention is at present being paid by researchers to the analysis of properties of composite materials in terms of properties of their constituent phases. Several problems of this kind were considered by Skudra.* He had investigated the behavior of reinforced polymer materials (e. g., fiberglass) in terms of the filler and binder properties, concentration of the binder, and the orientation of reinforcing elements. The reinforcing elements were assumed by the author to be of rectangular cross section and the stresses in these and in the binder to be uniformly distributed. Creep of the reinforcing elements was taken into account and the binder and filler were considered to be normal linear bodies.

Wang Fou Fu [66, 67], considering a similar problem, assumed that the reinforcing filaments were distributed in the cross section at nodal points of a regular doubly periodic mesh. He took into consideration the effects of stress concentration in the binder in the gaps between filaments, and described creep by the Volterra integral equation with Rabotnov kernels.

Bolotin [68] had introduced the concept of so-called "diffusion" into the theory of reinforced media, according to which the higher strength of reinforcing elements is not concentrated in any specific parts of the material but is uniformly "diffused" throughout its volume. Using variational principles, he had established that the medium must conform to the fundamental relationships of the Voigt–Kosser momentary stress. Extension of this concept to viscoelasticity by using differential operators was proposed.

Teregulov [69, 70] investigated creep in plates and shells of viscoelastic material using the principle of Volterra and of Rabotnov operators. He established [70] that under an external load exceeding a certain critical value, determined by the long-term moduli of the plate, the deflection of the plate with some deviation from a perfect form increases with time, approximately as shown in Fig. 10, where t^* is the point of inflection. In accordance with Shesterikov's definition of the critical state, the time corresponding to the point of inflection is suggested as the critical time t_*. The instant of time at which the deflection exceeds its initial value by a factor k is proposed in [71] as the definition of critical time. The initial deflection allowance and the amount of permitted deflection are taken into consideration in the selection of k.

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*A. M. Skudra, Doctoral Dissertation, Institute of Problems of Mechanics, AN SSSR (1967).

Huang and Lee [72] solve the known problem of strain in a viscoelastic cylinder enclosed in an elastic shell under internal pressure, taking into consideration the ultimate strain and using Eq. (26) to determine the strain. A similar analysis was made by Badran [65] who used the logarithmic representation of the ultimate strain, as suggested by Hankey.

Il'yushin and Ogibalov [73] had attempted to formulate a general linear tensor theory of viscoelasticity based on relationships of the type of (18) in which only single and triple integrals with only linear tensor terms were retained in the right-hand side. Moskvitin [74], when considering the same theory, assumed the physically nonlinear terms to be proportional to a certain small parameter, and suggests solving the boundary value problem by the method of successive approximations.

Pobedrya [75] had proved by means of functional analysis a number of theorems for linear viscoelasticity defined by Eq. (18), and in [76] proposed a method for constructing the resolvents of kernels of this type of functionals.

A further case of nonlinear viscoelastic behavior of thermorheologically simple bodies induced by vibratory heating was considered by Barenblatt [77].

Unfortunately, the formulation of new theories is not always supported by experimental data which would confirm or refute theoretical deductions.

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