RESEARCH IN THE FIELD OF PHENOMENOLOGICAL RHEOLOGY

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UDC 532.135

Rheology has been extensively developed in the past decades – this is the study of the deformation and flow of materials. Numerous problems of rheology are presently being published in the periodical literature of the USSR and abroad. Certain problems in the rheology of disperse and high-molecular systems have been generalized in previous surveys,* from which we see that Soviet scientists have contributed extensively to the fundamentals and development of rheology. We know that Shvedov founded the rheology of disperse and high-molecular systems in 1889 when he observed the relaxation of stresses in a gelatin sol and formulated the equation for viscoplastic flow.†

Numerous monographs and collections appeared abroad after the Second World War, devoted to the various branches of rheology. Some of these have been translated in Russian and published in USSR. In this survey we briefly outline some of the results achieved by foreign researchers in the field of phenomenological rheology, which have been published in the cited publications.

The term "rheology" was introduced at the suggestion of the American scientist E. Bingham at the Third International Symposium on the Theory of Plasticity (April, 1929). Even at that time it was felt necessary – within the general system of sciences – to separate that branch of knowledge devoted to problems of deformation and flow in a variety of materials which people encounter in their practical and scientific activity, i.e., manufacture, processing, and application. More detailed information as to the origin of the term "rheology," on the history of the establishment of the first society of rheology, on research into the processes of flow in materials – from the first efforts of Amenemhet (the inventor of the water clock in ancient Egypt) to the present day – can be found in the books of Scott-Blair [1]. These publications also contain a survey of the earliest research into the rheology of various materials, primarily those projects completed prior to the Second World War.

The Society of Rheology was organized in the USA in 1929, and its chairman for a long time was E. Bingham. For a number of years the society published the Journal of Rheology, and it regularly puts out The Rheology Leaflet, later changed to the Rheology Bulletin, which publishes abstracts in the field of rheology. At the initiative of Scott-Blair, the British Rheologist's Club was organized in England in 1940, and Professor G. I. Taylor served as its first chairman. This organization, also engaged in the regular publication of rheology bulletins, was subsequently renamed the British Society of Rheology. In conjunction with the English National Engineering Laboratory, this society publishes indexes of papers in the field of rheology in England (see, for example, [2]). A Rheology Group has been part of the National Science Research Center in France for about 10 years, and it publishes the special rheology journal Cahier du Groupe Francais d'Etudes de Rheologie. The Transactions of the Society of Rheology have been published in the USA since 1957. The journal Rheologica Acta has been published in the Federal Republic of Germany since 1958, i.e., after the Third International Rheological Congress.

Rheology‡ presently occupies a definite position in the general system of sciences, at the juncture of physics, chemistry, mechanics, and technology. Rheology owes its position at this juncture of the sciences

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*M. P. Volarovich, Kolloid. Zh., 16, No. 3 (1954); M. P. Volarovich, I. I. Lishtvan, and V. M. Naumovich, Inzh.-Fiz. Zh., 5, No. 2 (1962); M. P. Volarovich and N. I. Malinin, Inzh.-Fiz. Zh., 10, No. 6 (1966).

†See the article by M. P Volarovich and S. M. Levi [Kolloid. Zh., 18, No. 2 (1956)], devoted to the memory of F. N. Shvedov.

‡The definition of the word "rheology" can be found in an article by one of the authors of this paper in the Physics Encyclopedic Dictionary, Vol. 4 (1965), p. 435.

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to the fact that its limits have not yet been completely defined. The range of problems in rheology is quite broad and it deals with the problems of flow of deformation in the most varied of materials, beginning with Newtonian fluids and ending with the ideally elastic Hooke body. But, for example, the hydrodynamics of viscous fluids, which treat a liquid as a continuous medium, is also concerned with the flows of Newtonian fluids. The corresponding solutions of these problems are of interest both from the standpoint of rheology and hydromechanics, and it is impossible to establish a clear demarcation between rheology and hydrodynamics. There is also no clear boundary between rheology, on the one hand, and the theories of elasticity, plasticity, and creep, on the other hand. Unlike the cited sciences, hydrodynamics, and the theories of elasticity, plasticity, and creep, rheology devotes more attention to problems of interrelationships between the processes of deformation and flow and the structural features of various materials.

On the other hand, rheology is closely associated with that new branch of science – physicochemical mechanics – founded and developed in the works of Academician Rebinder and his school.* Physicochemical mechanics covers a broad range of subjects, in particular, the problems of structure formation in various materials in connection with the colloidal processes and chemical reactions taking place within them, makes extensive use of the methods of rheology and rheological apparatus.†

Contemporary rheology can be divided into three basic sections: phenomenological rheology, experimental rheology, and structural rheology. Structural rheology is occasionally referred to as microrheology; it establishes a relationship between the rheological properties and the structure of a material. A somewhat isolated phenomenon is the development of biorheology, which deals with research into the processes of flow in blood, protoplasm, synovial and hormonal fluids, etc., as well as the deformation of plant and animal tissues as this relates to the vital aspects of an organism. Biorheology – a branch of biophysics – employs essentially those methods as phenomenological, experimental, and structural rheology; however, the relationship between the properties of deformation and flow in biological tissues and fluids and the physiological processes of plant and animals governs the unique nature of this branch of science, situated at the juncture of mechanics, physics, chemistry, and biology.

This survey is devoted to problems of phenomenological macrorheology. Phenomenological rheology establishes the relationship between the stresses acting on a body, these stresses having been brought about by strains and their changes with time. The material being investigated is usually treated as a continuous medium which permanently occupies the space in which it is found. Examination of the problems of deformation and flow in products and structures of materials described by various phenomenological equations serves also as the subject matter of phenomenological rheology. The corresponding equations are frequently based for given materials on experimental research performed with the aid of a variety of rheology instrumentation. However, the problems of calculating the processes of deformation and flow are generally resolved in rheology by mathematical methods analogous to those of the theories of elasticity, plasticity, and hydrodynamics. This branch of rheology may therefore be referred to as theoretical, or mathematical, rheology.

In connection with the above, of fundamental importance in rheology are the above-cited characteristics of the stressed–strained state of matter, i.e., stresses, strains, and their derivatives. In the general case these may be derivatives of n-th order with respect to time, where $n = 0, 1, 2, \ldots$, or it may be a fraction.‡ The definition of stress, as a tensor quantity, provided by Voigt** in the last century, has as yet undergone no change and exhibits no shortcomings which would serve as a basis for replacement, correction, or generalization of this macroscopic characteristic. As regards deformation, the Cauchy definition of deformation ε_{ij} in the form

$$\varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i}) \tag{1}$$

(u_i is the displacement of the point in the direction of the x_i coordinate line, with the comma denoting differentiation with respect to the corresponding space coordinate, so that $u_{i,j} = \partial u_i/\partial x_j$) is applicable only

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* P. A. Rebinder, Izv. Akad.Nauk. SSSR, Otd.Khim. Nauk, No. 11 (1957).

† See, for example, Physicochemical Mechanics of Soils, Clays, and Structural Materials [in Russian], Izd. Fan Uzbekskoi SSR, Tashkent (1966); Problems of Physicochemical Mechanics of Fibrous and Porous Disperse Structures and Materials [in Russian], Zinatne, Riga (1967).

‡ In connection with the use of fractional differentiation in the Liouville sense for the description of relaxation properties in high polymers, see: A. N. Gerasimov, Prikl. Matem. i Mekhan., 12, No. 3 (1948); G. L. Slonimskii, Dokl. Akad. Nauk SSSR, 140, No. 2 (1961).

**D. I. Kutilin, The Theory of Finite Deformations [in Russian], OGIZ Gostekhizdat (1947), p. 10.

to small deformations substantially smaller than unity. There is no proof that definition (1) is useful for finite (large) deformations of such materials, for example, as rubber whose elastic strains may reach a magnitude of 800-1000%. Reiner [3, 4] notes that any monotonically increasing function of the displacement may be treated as a measure of finite deformation, if it degenerates into the first part of formula (1) in the case of infinitely small deformations. Reiner provides absolutely no basis for this contention. In particular, he notes that even the Cauchy measure, in principle, can be used to determine finite deformations, although it exhibits certain drawbacks such as, for example, in the deformation of a ductile rod the magnitude of the deformation will be a function of that state of the rod which we regard as its initial state.

The theory of finite deformations has been developed more fundamentally abroad in the publications of Rivlin (see the articles in [4, Vol. 1], [5]), Lodge [6], Fredrickson [7], and of Green and Adkins [8].* These researchers define finite deformation with the relationship†

$$\varepsilon_{ik} = \frac{1}{2}(g_{ik} - a_{ik}), \tag{2}$$

where g_{ik} is the metric tensor of a material coordinate system associated with the particles of the material being deformed at the instant under consideration; a_{ik} is g_{ik} in the initial state, when the material has not yet been subjected to the effect of the stresses. Using this definition of finite deformation and its invariant characteristics, Rivlin derived the physical coupling equations of rather general form, from which he found the elastic potentials of Kuhn, Muni, et al. (see [9]) as special cases, these potentials having been proposed by the cited authors for rubber-like materials. The Rivlin closed system of equations, which he wrote in a system of material coordinates, and which, as is usual, contained equations of motion, continuity, boundary conditions, and nonlinear physical equations, makes it possible to formulate boundary-value problems for materials exhibiting great deformations. Certain of the problems were examined by Rivlin [4, Vol. 1]; here he investigated a number of nonlinear effects, such as the appearance of compressive stresses at the ends, in the twisting of round cylindrical objects, etc. We should point out that this kind of phenomenon is actually encountered in rubbers [9] and even in metals (the Poynting effect [3]), although at low deformations the magnitude of this effect (i.e., the corresponding stress or strain) is insignificant; it diminishes in approximate proportion to the square of the principal deformation.

Lodge [6] and Fredrickson [7] used definition (2) for finite deformation to investigate the processes of elastic aftereffect and viscous flow. The rheological equations which they derived provide an explanation, in particular, of such nonlinear phenomena as the Weissenberg effect (see Weissenberg [10], Freeman and Weissenberg [10], see also Pollett [11], Ward and Lord [11], Jobling and Roberts [4, Vol. 2], [11], Reiner [4, Vol. 1], [3], which arises in the appearance of normal stresses in a material subject to the deformation of pure shear. Problems from the theory of finite deformations are also covered in the Prager book [12].

Yet another means of explaining and mathematically describing the effects of tensor nonlinearity (for example, the Weissenberg effect) was suggested by Oldroyd [13]. Many physical laws employed in rheology, as indicated earlier, contain derivatives with respect to time, e.g., the derivatives of stresses or the derivatives of strains, with respect to time. An equation of rather general form, describing in particular the effects of viscous flow and elastic aftereffect, has the form

$$p'_{ik} + \lambda_1 \dot{p}'_{ik} = 2\eta_0 (e_{ik} + \lambda_2 \dot{e}_{ik}), \tag{3}$$

where $p'_{ik} = p_{ik} + p\delta_{ik}$ is the stress deviator; p_{ik} is the stress tensor; p is the hydrostatic pressure; δ_{ik} is the Kronecker delta; $\dot{e}_{ik}$ is the strain rate; λ_1, λ_2, and η_0 are the parameters of the material. The dots denote differentiation with respect to time. The rheological equation (3) does not differ from the equations for elastic sols (a Lesersich body) and relaxing gels (a Jeffries body) [3]. The magnitude of the derivative is a function of whether or not the reckoning system is fixed or whether the coordinate axes translate together with the fluid particle, or if they additionally are in rotation.

Oldroyd contends that the physical law associating stresses and strains should be independent of some arbitrary selection of a coordinate system. The only invariant coordinate system is the material (convection) coordinate system whose axes are associated with the particles of the medium being deformed. The

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*Academician L. I. Sedov [see L. I. Sedov, Introduction to the Mechanics of Continuous Media [in Russian], FM (1962)] contributed significantly to the development of the nonlinear mechanics of continuous media and the theory of finite deformations in the USSR.

†This definition of finite deformation is not the only one possible (see, for example, the Kutilin book cited earlier).

quantities $p'_{ik} = \mathscr{D}p'_{ik}/\mathscr{D}t$ and $e_{ik} = \mathscr{D}e_{ik}/\mathscr{D}t$ are thus convection derivatives associated with the moving and rotating system of coordinates which is simultaneously being subjected to strains in conjunction with the material. Let us consider the case of pure shear. Oldroyd derives a nonlinear relationship between the shear component of the stress tensor p_{xy} and the velocity gradient D. Oldroyd interprets this relationship as the law of non-Newtonian flow of a viscous fluid.

In establishing the relations between the stresses and strains, without which solution of the various working problems of mathematical rheology would be impossible, it is frequently useful to employ mechanical models. The latter are assemblies of various elements such as springs, ductile elements (hydraulic shock absorbers or dampers), dry-friction units, etc., whose individual points are displaced under the action of the forces applied to them. Some of the simplest models describing phenomena in actual media, i.e., elasticity, viscosity, plasticity, and combinations of these simplest of properties (viscoelasticity, viscoplasticity, and the behavior of an elastic–plastic material), has been covered in reviews by Reiner [3, 4, Vol. 1]. A particularly detailed study for the method of mechanical models is found in [14], where the chapters written by Persoz provide a description, in addition to the traditional, of such new models as well. These include, for example, the Képès model in which the plastic deformation is directly proportional to the applied stress. A model with a regulator is useful to describe phenomena such as dilatation (an elevation in the effective viscosity with an increase in the velocity gradient). The introduction of a limiter element makes possible the modeling of such phenomena as hardening under plastic deformation.*

The use of models with a limited number of elements usually results in laws of deformation in time (or in laws of variations in stress), which are poorly described by the curves observed for actual materials. This naturally results in a trend toward further complication of the mechanical models. Thus, for example, Alfrei [15] extensively uses the so-called model C to describe the linear properties of high polymers; this model is a series-connected combination of Hooke and Newtonian elements, in addition to a number of Kelvin elements. For C models use is frequently made of the concept of the distribution (spectrum) of the times of elastic aftereffect or, more exactly, the distribution of compliances with respect to the times of elastic aftereffect [15-17]. If the set of Kelvin elements is finite, we have a line spectrum. We also use the concepts of continuous spectra, when the models contain an infinite multiplicity of elements which make up these models. In addition to the generalized Kelvin model, the generalized Maxwell model [15-17] has also gained widespread acceptance, and it contains a finite or infinite number of Maxwell elements, connected in parallel.

The generalized Kelvin and Maxwell models correspond to linear equations describing the properties of the material. Similar constructions for nonlinear bodies were achieved by the Japanese authors Sawaragi and Tokumaru [18], Sawaragi, Taniguchi, and Furuichi [18], who in the cited references investigated models which, in addition to simple Hooke and Newtonian elements, also contain finite or infinite sets of three-element links; an individual link involves the parallel connection of a spring, shock absorber, and dry-friction unit. An assembly of such links is equivalent to the introduction of functions of compliance distribution with respect to the times of elastic aftereffect and the yield point. Sawaragi and Tokumaru [19] use a model of element groups connected in parallel. Each group is an infinite set of four-elements links connected in parallel, with each link made up of a series-connected Newtonian element and the three-element assembly referred to in the above-cited reference [18]. Sawaragi and Fukuda [19] use a model in the form of a parallel-connected set of groups, each of which is made up of an infinite number of links connected in parallel, with each link made up of a Hooke element connected in series with another Hooke element, with the last connected in parallel with the dry-friction unit. The Japanese models describe, in particular, such processes as the fatigue of elastomers, and their significant drawback is the excessive complexity.

The construction of a mechanical model for an actual material is the first step in the solution of the corresponding problems of phenomenological rheology, which deal with the various processes of deformation for such a material, as encountered in actual practice. In addition this step is entirely unnecessary; the physical (rheological) equations needed for the solution of the working problems can be compiled without the use of mechanical models. However, the construction of mechanical models is usually of some advantage; by analyzing a model we can prove, for example, the noncontradictory nature of the equations which we are using, and also the corresponding laws of thermodynamics, etc. A model made up of linear elements,

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*A model with destruction elements is proposed in the following paper: M. P. Volarovich, N. I. Gamayunov, and N. N. Sokolov, Kolloid. Zh., 28, No. 4 (1966).

i.e., of springs and shock absorbers, in terms of its mechanical properties is equivalent to a medium subject to a linear differential equation which, in operator form, is written as follows [3, 14, 20], Leaderman [4, Vol.2], Lee [5, 20]:

$$\sum_{i=0}^{i=m} a_i \frac{d^i}{dt^i} \varepsilon = \sum_{i=0}^{i=n} b_i \frac{d^i}{dt^i} \sigma, \tag{4}$$

where $a_0 = 1$, while a_i and b_i are the parameters of the material.†

Differential equations such as (4) can be rewritten to a form containing the integral Volterra operator. We can treat the kernel of the resulting linear Volterra integral equation as entirely independent of the operator equation (4) or independent of the corresponding rheological model. Here we retain only the general Boltzmann–Volterra rule of linear superposition according to which deformation resulting from the stresses $\Delta\sigma_1 + \Delta\sigma_2$ is equal to the sum of the deformations resulting from $\Delta\sigma_1$ and $\Delta\sigma_2$.

The methods to describe the rheological properties of linear viscoelastic materials with mechanical models, operator equations such as (4) and Volterra linear integral equations are the most general, and they are suitable for processes in which the stresses or strains vary arbitrarily. To determine the parameters of the model or the parameters of the operator equations, in particular, the integral equations, we need other methods of description, which may, perhaps, not distinguish themselves by their generality, but which are therefore more closely associated with a certain type of experiment, performed on an actual material. Among the latter we include those methods based on the determination of the creep function from experiments on creep, and on the determination of the relaxation functions from experiments on the relaxation of stresses, dynamic compliance I*, or the dynamic modulus E*. These last quantities are determined from experiments performed at variable stresses σ and strains ε, with σ and ε varying with time according to a sinusoidal law. With viscoelastic materials the deformation in the general case lags in phase from the applied stress.

The quantities I* and E* are treated as complex; this treatment is associated with the representation of σ and ε and, consequently, of I* and E* in the form of vector diagrams on a complex plane as is frequently done in the case of harmonic oscillations. Completely to define the viscoelactic properties, we must know of the compliance $I(t) = \varepsilon(t)/\sigma$ in the creep experiment with constant σ with time t ranging from zero to ∞ or with the relaxation modulus $E(t) = \sigma(t)/\varepsilon$ in the experiment on relaxation at constant deformation for the same range of time t from zero to ∞, or with the complex compliance as a function of the angular frequency ω (for all possible variations in frequency from $\omega = 0$ to $\omega = \infty$) $I^*(\omega) = \varepsilon(t)/\sigma(t)$, or for the complex modulus $E^*(\omega) = \sigma(t)/\varepsilon(t)$ at frequencies in the interval $[0, \infty)$.

As was mentioned earlier, in the determination of $I^*(\omega)$ and $E^*(\omega)$, ε and σ are sinusoidal functions of t. These functions are complex quantities and contain real and imaginary parts. To determine the viscoelastic properties of a material, it is sufficient to know one of these four functions of ω (moreover, we must have the constants which determine the elasticity of a material and its ductility, if elastic and viscous strains arise in the deformation of the material). For example, if we know the imaginary part of the complex modulus of elasticity over the entire range of frequencies from 0 to ∞, from these data we can also determine the real part of E* as a function of ω [16].

Each of the above-enumerated methods is convenient for the solution of a specific range of problems. For greater universality of the methods of phenomenological rheology in the case of a linear viscoelastic material, here we must have at our disposal methods for the transition from one method of describing the properties of a material to another method. The most complete review of such methods is found in [16]. The figure shows a diagram of the methods to describe the linear viscoelastic properties of materials and the methods of changing from one to the other, this diagram having been borrowed from the cited monograph (see also, Gross [11]). Many of the methods for the transition from one description method for the properties of a linear viscoelastic material to another are associated with extensive mathematical difficulties. In part these difficulties have not yet been overcome; the arrows in the diagram indicating the transition from one description (indicated by a rectangle) to another therefore do not connect all of the rectangles.

In certain cases, for the transition from one description to another, we use approximate methods (see [16], Leaderman [4, Vol.2]), e.g., to calculate the relaxation spectrum from the function E(t) of the relaxation

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†Chapter III of Yu.N. Rabotnov's monograph [Creep of Structural Elements [in Russian], Nauka (1966)] is devoted to the behavior of linear viscoelastic media. A large portion of this book is devoted to the creep of metals at high temperatures, and to an examination of the corresponding phenomenological problems.

modulus. In certain cases, existing exact methods of transition are inconvenient for practical calculations; despite the existence of some exact solution, we therefore also use approximate calculations. For example, we know of an exact solution for the problem of determining the relaxation spectrum from dynamic data (from the complex modulus of elasticity, from its real and imaginary part as functions of the angular velocity), as derived by Gross. However, for practical calculations this is an inconvenient solution, since for the calculations of the above-enumerated dynamic functions they must be specified in the form of analytical functions. In actual practice, we therefore proceed in the following fashion: from the experimental data we usually derive the curves for the dynamic modulus and for the modulus of the losses as a function of ω, with the analytical equations describing these functions, as a rule, unknown. Here it is more convenient to use approximate methods (see, for example, Leaderman [4, Vol. 2]).*

The above-enumerated theories assume that the medium exhibits constant properties that are independent of time and that the effects of aging, i.e., the time variations of the mechanical properties of the material, can be neglected. For many materials (in particular, for concretes, certain types of polymer materials, and disperse systems) aging is significant. Certain researchers therefore attempted to generalize the theories of viscoelasticity for media whose properties are a function of their age.† One such attempt is made by Bismuth and Saunier in [21, part 2], who introduced a term containing time in explicit form into an operator equation such as (4).

Most of the materials encountered by researchers in the field of rheology are nonlinear in nature. Many materials exhibit a linear elastic, viscous, or viscoelastic behavior in the region of low stresses, but behave as nonlinear materials at high stresses. A certain boundary exists in this case to separate the regions of linear and nonlinear behavior. The linear viscoelastic materials are referred to by Leaderman [22]‡ as materials subject to the linear Boltzmann–Volterra principle of superposition, and their deformation properties are described by linear Volterra integral equations with different kernels. It is Lee's opinion [5, 20] that linear and nonlinear materials are most easily distinguished in the following manner. If similar forces $P_i = C_i P(t)$ acts on completely identical items at similar points (the shape of the item is irrelevant), and these forces are oriented identically with respect to the axes of this item, the deformations of the linear material vary with time t in proportion to the forces P_i, while for an object made of a nonlinear material the deformations will not be proportional to P_i.

It is sometimes incorrectly assumed that a linear material exhibits a linear $\sigma - \varepsilon$ diagram. This is an entirely unacceptable criterion with regard to viscoelastic materials. The very simplest linear material described by the mechanical Maxwell model will exhibit a nonlinear diagram (see, for example, Alfrei [15]).

Reiner [3] distinguishes physical and tensor nonlinearity. Physical nonlinearity is evidenced by the fact that scalar quantities characterizing stresses, deformations, and rates of deformation (for example, their invariance), are associated with others by nonlinear relationships which can be investigated, for example, in the case of uniform deformation patterns (simple tension, simple shear, etc.). For materials exhibiting nonlinear viscoelastic properties (such materials include, in particular, many of the high polymers) the creep strains for specified values of t and for various values of σ, for example, grow in proportion to σ^n, where $n > 1$ (Staverman and Schwarzl [23]), or in proportion to $\mathrm{sh}(\sigma/\sigma_m)$, where n and σ_m are the parameters of the material (Findley [24]), etc.**

Consideration of the simultaneous elasticity and viscosity effects in solving problems of rheology for a physically nonlinear material generally involves considerable mathematical difficulties. Fortunately, for most cases of importance under practical conditions, one of the components of deformation – elasticity, viscosity, plasticity, or the deformation of elastic aftereffect – usually predominantes over the others.††

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*In the USSR, a number of investigations devoted to the methods of describing linear viscoelastic properties of materials and to find methods of making the transition from one method to another, have been performed by Shermergor, Prikl. Mekhan. i Tekh. Fiz., No. 1 (1960); Fizika i Metallovedenie, 9, No 2 (1960).

†See, for example, N. Kh. Arutyunyan's book: Certain Problems in Creep Theory [in Russian], Gostekhteoretizdat (1952), as well as the above-cited Rabotnov monograph.

‡The cited articles make up the report of the Committee on Terminology of the USA Society of Rheology.

**A survey of Soviet and foreign papers devoted, in particular, to an examination of the theories of nonlinear viscoelasticity can be found in the Malinin report published as The Transactions of the Second All-Union Congress on Theoretical and Applied Mechanics, No. 3, Mechanics of Solids [in Russian] (Moscow, 1964), Nauka (1966).

††See M. P. Volarovich, the footnote at the beginning of the article.

In calculating the flow processes it is therefore sufficient to account only for that one of these components, neglecting all the others. Thus, in examining the laws of flow in disperse systems, solutions, and melts of high polymers we usually neglect the elastic strains, as well as the deformations of elastic aftereffects as small in comparison with the irreversible effects – viscosity or plasticity strains. The effect of physical nonlinearity for viscous and plastic materials becomes evident in the fact that the flow diagram – the strain rate as a function of the acting stress – is a curve. In investigating viscosity our attention is basically devoted to the viscous behavior of the material in the case of shear strains, while the flow diagram (the consistency curve [1, 3]) is the curve showing the shear rate $\dot{\gamma}$ as the shear stress τ. In phenomenological and structural rheology, the effective viscosity is of great significance, and this quantity is defined by the equation*

$$\tau = \eta_{\mathrm{eff}}\dot{\gamma}, \tag{5}$$

with the quantity η_{eff} in the general case being a function of the shearing stresses τ. For Newtonian fluids η_{eff} = const. This is the way liquid low-molecular substances which contain no suspended macroscopic particles behave when the shear rates range from $\dot{\gamma} = 0$ to the high values of $\dot{\gamma}$ at which turbulent flow begins. At low concentrations for the solid-phase particles in the liquid, a disperse system continues to behave as a true Newtonian fluid (see, for example, the well-known work of Einstein). However, if the concentration arises above some critical level, we find effects of anomalous viscosity; here the effective viscosity is a function of the shearing stresses τ.† For various disperse systems, under a variety of conditions, the effective viscosity η_{eff} may diminish or increase with a rise in τ. Scott-Blair, in his monographs [1], analyzes in detail the factors leading to a change in η_{eff}; these are closely associated with the change in the structure of the systems during the flow process. In this connection, the effects of anomalous viscosity are of great interest for structural rheology; some of these effects have been described by Richardson [25].

The effect of raising the effective viscosity with an increase in the shear rate in the case of disperse systems and high polymers is encountered quite rarely in practice. Conversely, we very frequently find the inverse phenomenon in which the effective viscosity is reduced with a rise in τ. If the solid-phase concentrations in suspensions and in colloidal solutions or in polymer solutions are not too great, and if the temperatures are not too low (this latter circumstance is particularly important in examining flows of polymer melts) the system begins to flow at negligible stresses of $\tau \to 0$. Consequently, the system is a non-Newtonian fluid: the limit shearing stress is virtually equal to zero, while η_{eff} is a variable quantity. For solution of the problems of phenomenological rheology it is important to know, for this case, the law governing the relationship between the stresses τ and the strain rate $\dot{\gamma}$. This law must satisfy the obvious but contradictory requirement of, first of all, corresponding to the experimental data with a sufficiently high degree of accuracy, and secondly, it must be as simple as possible, since the range of problems capable of solution with contemporary mathematics and cybernetics facilities is markedly reduced as the rheological law is made increasingly complicated.

To solve the problems of flows for melts and solutions of polymers, rather extensive use is made of the Ostwald–De Vale "power law" [26-29]

$$\tau = K\dot{\gamma}^{n}, \tag{6}$$

where K and n are the parameters of the material; the Prandtl rheological equation [26, 28, 29]

$$\tau = A\,\mathrm{Arsh}\,\frac{\dot{\gamma}}{C} \tag{7}$$

(A and C are parameters) and the Eyring–Powell equation [26-29], whose left-hand member, as in (6) and (7), is τ, and its right-hand member is the sum of the right-hand members of Eqs. (6) and (7).

As the temperature is reduced, or as the solid-phase concentration in disperse systems or solutions of high polymers is increased, the effects of anomalous viscosity become increasingly evident.‡ Under certain conditions, the effective viscosity η_{eff} for certain critical stresses varies so rapidly** and in such a

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*See P. A. Rebinder and N. V. Mikhailov, Kolloid. Zh., 17, No. 2 (1955).

†The effects of anomalous viscosity for concentrated disperse systems are discussed by the authors in a survey article [Inzh.-Fiz. Zh., 10, No. 6 (1966)].

‡See the authors' survey [Inzh.-Fiz. Zh., 10, No. 6 (1966)].

**P. A. Rebinder describes the experiments in which, in the shear of clay structures, a drop in effective viscosity by a factor of 10^7 and more was observed (P. A. Rebinder, Physicomechanical Mechanics [in Russian], Znanie (1958); also see his comments to the translation of this book [4, Vol. 1]).

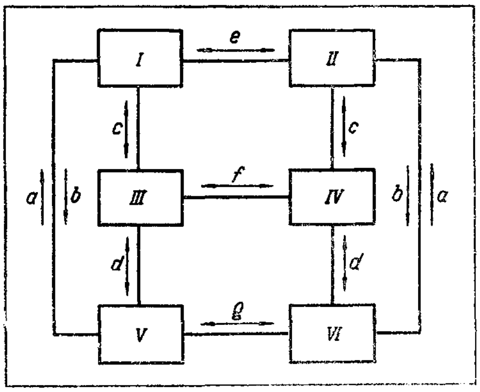

Fig. 1. Diagram illustrating the methods of describing linear viscoelastic properties and transitions from one to the other; I) the complex compliance function $I^*(\omega)$; II) the complex modulus function $E^*(\omega)$; III) the creep function $\psi(t)$; IV) the relaxation function $\psi(t)$; V) the distribution function for the aftereffect times; VI) the distribution function for the relaxation times; a) the Stieltjes integral; b) an algebraic inversion formula in complex variables; c) the Fourier transform; d) the Laplace transform; e) algebraic equations; f) Volterra intergral equations; g) integral transformations.

narrow range of values for τ that for a phenomenological description of the processes of the flow of such media it is more convenient to use the nonanalytical relationships than relationships such as (6) and (7), and those relationships similar to these, for which η_{eff} is a smooth function of the stresses τ. The value of the nonanalytical functions in rheology has been stressed in the surveys by Prager ([4, Vol. 1], W. Prager [30]).

As an example of a nonanalytical relationship used extensively in contemporary rheology we can cite the Bingham flow law (see [1, 3, 30-32], a Bingham body is a degenerate Shvedov body, see [3]), which in the one-dimensional case is written mathematically in the form†

$$\begin{aligned} \dot{\gamma} &= 0 \qquad (\tau \leqslant \theta), \\ \tau &= \theta + \eta_{\text{pl}}\dot{\gamma} \ (\tau > \theta), \end{aligned} \tag{8}$$

where θ is the yield point of the system (the limit shearing stress); η_{pl} is the plastic viscosity. The Bingham flow law provides a good description of the flow of various disperse systems such as, for example, paints ([3, 31, 32], Bantoft [13]), food products ([1, 3], Steiner [13]), peat [1, 3], and similar materials.

From among the nonanalytical relationships which make provision for the rise in the viscosity resistance with an increase in the velocity gradient, let us recall the Casson equation

$$\begin{aligned} \dot{\gamma} &= 0 \qquad (\tau \leqslant K_0^2), \\ \sqrt{\tau} &= K_0 + K_1\sqrt{\dot{\gamma}} \quad (\tau > K_0^2) \end{aligned} \tag{9}$$

(K_0 and K_1 are the parameters of the material), which finds application to describe the flow of typography inks (Casson [13], Bantoft [13]), and of blood (Merril, Margetts, Cokelet, Gilliland [21, part 4], Copley [33]).

As the concentration of the solids in the disperse system increases, the yield point θ of the system (or the parameter K_0 in (9)) usually increases more rapidly with the concentration c than the plastic viscosity η_{pl} (as in the case of the parameter K_1 in (9)). Thus, with high c the contribution of the second term to the overall magnitude of τ in (8) and (9) is frequently negligibly small in comparison with the yield point θ. Relationships such as (8) and (9) in this case degenerate into relationships of ideal plasticity; Van Iterson [34] stressed the possibility of using the methods of the theory of plasticity to describe the processes of flow in concentration disperse systems (soils, bricks, ceramics, etc.). These possibilities have been adequately exploited for disperse systems, and the theory of plasticity has been developed primarily for processes of deformation and metal flow. The contemporary theory of metal plasticity (primarily the mathematical theory) has been the subject of extensive literature. To familiarize the reader with the plastic properties of metals and of certain nonmetal materials, we can recommend the basic work of Nadai [35], in which many of the aspects of rheology have been covered.

Plasticity theory makes no provision for the relationship between plastic deformations and time, since this relationship is insubstantial for most metals, as well as for many nonmetallic crystalline and amorphous solids at low temperatures. However, at high temperatures, the effect of time on metal deformation becomes quite substantial. Under these conditions, the metal will creep with the passage of time under the action of stresses. Creep theory has been developed to describe the creep of metals: the theory of aging, of flow, of heredity (the above-mentioned descriptions with the aid of mechanical models, operator equations, and Volterra integral equations – these are all particular examples of the application of the concepts from

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† The general case of a three-dimensional stressed state is discussed in the Volarovich and Gutkin article [Zh. Tekh. Fiz., 16, No. 3 (1946)]. Generalization to the three-dimensional case of relationship (7) leads to the system of equations derived by Hencky and by Il'yushin.

the theory of heredity) and the theory of hardening ([35, Vol. 2], [36]).* While the best results for polymer materials are given by the theory of heredity from among the above-named theories, for metals the most satisfactory results are found for the equations from the theory of hardening, based on the utilization of concepts to the effect that a structural change takes place in the metal during the process of metal creep, leading to the hardening of the material and to a reduction in the creep rate [35, 36]. In simplest form, the equation of state from the theory of hardening associates the rate of deformation with the stress and with the deformation that has been accumulated.

The tensor nonlinearity (as well as the geometric nonlinearity) becomes evident in the fact that the principal axes of the stress tensor do not coincide with the principal axes of the strain tensor, or of the deformation rate [3]. This noncoincidence is made evident in the above-cited effects of Poynting and Weissenberg (see also Markovitz [21, part 1], Reiner [21, part 1], Ginn, Metzner [21, part 2]) and in other phenomena, in particular, the effect of the expansion in the jet diameter on discharge of a solution or of a polymer melt from a tube (see Shertzer, Metzner [21, part 2]). Some authors refer to this last phenomenon as the Barus effect, using the name of the researcher who first described the phenomenon during the nineties of the last century (see Lodge [33]).

The geometric and tensor nonlinear effects were found in the most varied of materials:† metals [3], polymer systems, and air ([3], Reiner [21, part 1]). On the basis of available data we are, as yet, unable to express an opinion as to whether or not these effects in such diverse systems are the result of a single factor or whether various mechanisms are operative in different cases. In a number of cases, the geometric and tensor nonlinear effects can be predicted on the basis of physically nonlinear relationships which make use of the concepts of finite deformations different from the Cauchy deformations (see, for example, [8]). The Poynting effect in metals was satisfactorily explained by Reiner [3] on the basis of the tensor nonlinear elasticity law which he proposed in conjunction with Hanin, and this law has the form

$$\sigma_{ij} = K_0\delta_{ij} + K_1\varepsilon_{ij} + \sum_{\alpha=1}^{3} K_2\varepsilon_{i\alpha}\varepsilon_{\alpha j}, \tag{10}$$

where σ_{ij} is the component of the stress tensor, and δ_{ij} is the Kronecker delta. In the simplest K_0, K_1, and K_2 are constants for nonlinear materials and they may be scalar functions of the deformation invariants. The right-hand part of (10) is a second-degree polynominal with respect to the component of the strain tensor; Reiner and Hanin demonstrated (see [3]) that relationships such as (10) for physically, geometrically, and tensor nonlinear materials – as complex as you please – contain no terms higher than the second degree for ε_{ij}.

To explain the effects of tensor nonlinearity in fluids, Reiner [3] proposes the use of (10) in which ε_{ij} denotes not only the deformation of the elastic material, but the rates of deformation as well. However, this type of relationship leads to a different distribution for the normal stresses (for example, in the clearance between two disks, of which one is fixed, while the second rotates) than is encountered in the experiments associated with the Weissenberg effect described in the above-cited projects in which rotation equipment was used.

De Witt, and then Gieseckus (cited in [3]) explained the Weissenberg effect and similar nonlinear effects on the basis of their proposed relationships which made provision for the stress tensor as a function not only of the symmetrical portion of the velocity gradient (i.e., of the strain rate), but also as a function of its nonsymmetrical part (i.e., the velocity of rotation). The relationships proposed by De Witt and Gieseckus are generalizations, for a ductile material, of the physical equations of the moment theory of elasticity developed by the Kossero brothers in the tenth century.

Phenomenological rheology is not limited to the determination of the laws relating stresses and deformations for the materials being investigated. In and of themselves, these laws are of limited value. But they are completely necessary to examine the various problems of deformation and flow in the continuous media which they describe. The solution of the boundary-value problems for deformation and flow of rheological media form the subject matter for one of the most imporant branches of rheology which, in analogy with the mathematical theory of elasticity and the mathematical theory of plasticity, may be referred to as

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*See the Rabotnov monograph entitled: Creep of Structural Elements [in Russian], Nauka (1966).

†One of the authors of this survey found the Poynting effect in a solid polymer – polymethyl methacrylate [Mekhan. Polimerov, No. 5 (1966).

mathematical rheology. Because of the limited scope of this survey, we have no opportunity here to undertake a detailed study of the contemporary methods of mathematical rheology. We will therefore limit ourselves to an enumeration of the basic literature containing such information.

Methods for the solution of some of the simplest problems of linear viscoelasticity (for example, problems of beams) can be found in the Alfrei monograph [15], as well as in the article by Baltrukonis and Vaishnav [21, part 2]. An analysis of the methods for the solution of quasistatic problems of linear viscoelasticity, when the stresses which arise in a deformed material, due to inertial forces, are negligibly small in comparison with the stresses due to external static forces, as well as an analysis of dynamic problems, is found in the book by Blend [20] and the survey article by Lee [5, 20]. The book by Nowacki [37] is devoted to these same problems.

The main difficulty in solving the boundary-value problems of viscoelasticity is associated with the fact that the physical equations describing the behavior of a material include time. In the West the generally accepted methods of solving the problems of linear viscoelasticity are based on the application of such integral transformations as the Laplace transform, for example, to the overall system of equations describing the state of a material. With the aid of the integral transformation the problem reduces to the corresponding elasticity problem and the inversion of the elasticity solution yields the time variation in the picture of the stressed–strained state. Operator methods have been developed in the USSR for the solution of similar problems, and these are based on the use of the so-called Volterra principle, according to which the elasticity solution is suitable even for the corresponding viscoelasticity problem; it is only in the final result that the constant elasticity parameters are replaced by viscoelasticity integral operators.*

Some problems with regard to the propagation of viscoelasticity waves are covered in the books by Kol'skii [38] and Davis [39],† as well as in the articles by Valanis [21, part 2], Arnold, Lee, and Panarelli [21, part 2].

The development of the theory of viscoelasticity was motivated in great part by the extensive use of plastics as structural materials. A number of interesting problems arose in connection with the development of solid-propellant rocket engines (see, for example, [40], Williams [41], Klosner and Sadre [21, part 2], and Moghe and Hsiao [21, part 2]).

There is no need here to touch upon the problems of the viscous flow of Newtonian fluids, since these problems, as noted earlier, are the subject matter of hydrodynamics. The mathematical rheology of non-Newtonian fluids was developed primarily in connection with the progress in the technology of processing various rheological media: polymer materials and disperse systems. In the article by Gore and McKelveys [4, Vol. 3] and in [26, 27, 29, 42] we find discussions of the problems of flow for certain non-Newtonian fluids (primarily of the Ostwald–DeVale medium, subject to the "power law" (6) of flow). A survey of the solutions for the problems of flow of non-Newtonian fluids in viscosimeters of various design is given by Oka [4, Vol. 3]. A number of problems in nonlinear rheology are covered in the Fredrickson [7] and Wilkinson [28] monographs, as well as in the article by Gerrard and Philippoff [21, part 2].

In [43] problems concerning the flow of viscoplastic materials are discussed. In recent years the theory of flow in these materials and problems arising from it have been developed intensively in the USSR.‡

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*See the Rabotnov monograph entitled: Creep of Structural Elements [in Russian], Nauka (1966); see also, A. R. Rzhanitsyn, The Theory of Creep [in Russian], Stroiizdat, Moscow (1968).

†See also Kh. A. Rakhmatulin and Yu. A. Dem'yanov, Strength under Intensive Short-Term Loads [in Russian], FM (1961).

‡The following books are devoted to the hydrodynamics of viscoplastic fluids in the petroleum industry: R. I. Shishchenko, The Hydraulics of Clay Solutions [in Russian], Aznefteizdat (1951); A. Kh. Mirzadzhanzade, Problems in the Hydrodynamics of Viscoplastic and Viscous Fluids in the Petroleum Industry [in Russian], Aznefteizdat (1959). Certain problems in the flow of viscoplastic peat systems are covered in the book by N. N. Kulakov [Introduction to the Physics of Peat [in Russian], Gosénergoizdat (1947)]. A number of books by Soviet authors, devoted to a theoretical examination of the problems in the flow of viscoplastic materials, is cited in the Volarovich survey [Kolloid. Zh., 16, No. 3 (1954)]. In recent years, in the area of the theory of the flow of viscoplastic materials successful work is being done by Gutkin [Kolloid Zh., 17, No. 6 (1955); 19, No. 1 (1957); 23, No. 3 (1961); 24, No. 1 (1962)], by Myasnikov and Mosolov [Prikl. Mekhan. i Tekh. Fiz., Nos. 2 and 5 (1961); No. 4 (1962); Prikl. Matem. i Mekhan., 29, No. 3 (1965); 30, No. 4 (1966); 31, No. 3 (1967)]. Extensive research into the rheological properties (in particular, the viscoplastic properties) of clay suspensions has been carried out by Academician Ovcharenko, and his co-workers (see F. D. Ovcharenko, N. N. Kruglitskii, S. P. Nichiporenko, and É. G. Agabal'yants, Mountain Leather in Drilling [in Russian], Tekhnika, Kiev (1968), and others).

The problem of the propagation of a stress wave in an elastic–viscoplastic material whose mechanical model is a combination of Hooke, Newtonian, and St. Venant elements connected in parallel is examined in the article by Cristesku and Predeleanu [21, part 3].

Extensive literature [44-50]† is devoted to the solutions of problems from the mathematical theory of plasticity; unfortunately, the use of these solutions is limited virtually in its entirety to metallic systems.

A solution is given in the Phillips and Tsan Sun Chang article [21, part 1] for the problem of the distribution of stresses and strains in a hardening medium in an underground explosion (for example, a nuclear explosion).

In the above-cited articles on the hydrodynamics of non-Newtonian fluids it is primarily laminar flows with which we are concerned. Engineers frequently deal with the turbulent flow of non-Newtonian fluids in actual practice, and for these, for example, in flow through tubes, the relationships between flow rate and pressure difference will differ from the analogous relationships in the case of a laminar regime. A special chapter in [29] is devoted to the problems of turbulent flow for a laminar fluid. As is well known, the existence of a laminar or turbulent flow regime depends on the Reynolds number Re for the given flow; for Re < Re* (Re* is the critical Reynolds number) the flow regime is laminar; for Re > Re* the regime is turbulent. The magnitude of the Re number also determines the frictional losses for the flow of a fluid in a given channel.

For anomalous fluids the Reynolds number cannot be determined in the usual fashion, i.e., as in the case for viscous fluids, since the effective viscosity of the anomalous fluid is a variable quantity that is a function of the velocity gradient. Determinations of the Reynolds numbers and other similar criteria are given in [29] for various anomalous fluids subject to the "power law" of Ostwald–DeVale flow, subject to the Bingham law, etc.‡

In connection with the consideration of the problem of critical Reynolds numbers, it is appropriate at this point to refer to the two effects observed in the flow of viscous and viscoelastic fluids. The so-called Toms effect (see Toms [10], Oldroyd [10], and Fabula [21, part 3]) involves the fact that insignificant additions of high-molecular substances to the viscous fluid (for example, to water) leads to an extremely great increase in the critical Reynolds number Re*. Thus, for example, Kelley and Brodnyan [21, part 2] observed an increase in Re* from 2000 to ≈7000 in this case. Simultaneously with the increase in Re* there is a pronounced drop in the loss of dynamic head. No satisfactory physical explanation for the Toms effect has apparently yet been found.**

The second effect (some researchers refer to it as the effect of "hard turbulence") shows up in the discharge of polymer melts from tubes, nozzles, dies, etc.; it is occasionally observed in the extrusion of polymers. At high flow velocities, the surfaces of the products occasionally show signs of rippling, waviness, and sharkskin defects, etc. (see Benbow, Brown, and Howells [30]). This effect is associated with the fact that the polymer melt exhibits substantial elasticity relative to the shearing strains, and in the case of high flow velocities, when the potential energy of deformation is rather great, self-oscillations arise within the material, leading to the appearance of surface defects.††

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†See also the books by the Soviet authors: V. V. Sokolovskii, The Theory of Plasticity [in Russian], GTTI (1952); L. M. Kachanov, The Fundamentals of the Theory of Plasticity [in Russian], GTTI (1956); D. D. Ivlev, The Theory of Ideal Plasticity [in Russian], Nauka (1966).

‡In the above-cited monograph, Shishchenko determines the generalized Reynolds number from the same formula as for a viscous fluid; however, in the place of ordinary viscosity, use is made of the effective viscosity in this relationship.

**In the USSR the Toms effect has been studied persistently by Barenblatt and his co-workers [see, for example, Prikl. Mekhan. i Tekh. Fiz., No. 3 (1965); No. 5 (1965)], as well as in the articles by Él'perin and Smol'skii, with their co-workers [see Inzh.-Fiz. Zh., No. 8 (1964); 10, No. 2 (1966); Izv. AN BSSR, Ser. Fiz.-Tekh. Nauk, Nos. 2 and 3 (1965)]. A patent was granted to Él'perin for a method of reducing drag in tubes and channels (USSR Patent No. 169955, effective as of December 6, 1954).

††This effect was studied by Vinogradov, Malkin, and Leonov [Dokl. Akad. Nauk SSSR, 151, No. 2 (1963); Kolloid–Z. u. Z. Polymere, 191, No. 1 (1963)]. They demonstrated that the appearance of the oscillations associated with the effect of "hard turbulence" is governed by the magnitude of the singular criterion which the authors refer to as the elastic Reynolds number and which represents the measure of the ratio between the viscosity forces and the elasticity forces.

A number of specific problems in the hydrodynamics of non-Newtonian fluids arises in connection with the problems of their boundary layer.*

In conclusion of this survey, we must cite the Ziegler book [51]. This book contains an interesting attempt to provide a specific completed formulation to the phenomenological approach to the problems of rheology. In particular, it contains a unique discussion of the principles of the thermodynamics of irreversible processes, attempts to validate these, in addition to a systematic demonstration of the importance of applying these principles to the description of media with dissipation.

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