REVIEWS

RELAXATION METHODS IN INVESTIGATIONS OF TRANSPORT PROCESSES

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In spite of the large variety of existing transport processes they all have one fundamental property in common: these processes strive to neutralize external actions that take a system out of the equilibrium state. Any physical system has some inertia, related to internal processes specific to the system, which are activated with breakdown of equilibrium and which are characterized by their own internal variables. The approach of the system to equilibrium, which results in the observed transport processes, can be viewed as a collection of relaxation processes for these internal variables.

The internal variables (relaxation parameters) can include macroscopic parameters (temperature of some subsystem, concentrations of different substances, coordinates of chemical reactions, order parameters, etc.), as well as variables describing microscopic state of separate structural elements: molecules, atoms, ions, etc. Microstructural relaxation parameters usually include, for example, the energies of rotational and vibrational degrees of freedom of atoms and molecules.

If the time scales of the external actions are of the same order of magnitude as the relaxation times, then the internal nonequilibrium processes can have a significant effect on the macroscopic energy, mass, and momentum transport processes both between the surrounding medium and the system and within the system itself.

Since in many practically important situations the evolution of internal variables cannot be directly observed, it is important to develop a theory of relaxation processes that gives a phenomenological relation between the characteristics of the external perturbation and the response of the system and the internal processes occurring in the system.

There are two basic approaches to the description of relaxation phenomena and processes. One of them, based on the use of the corresponding kinetics equations [1, 2], requires a detailed knowledge of the processes at the molecular level and for this reason its application to systems of different classes is closely related to the development of methods specific to each class. The second approach is based on the methods of thermodynamics of irreversible processes, which are fundamentally identical for different types of systems [1, 3–10] and, therefore, has a great degree of generality. The first thermodynamic method was applied to problems of acoustic relaxation by Mandel'shtam and Leontovich [7]. The method was further developed by meixner et al. [3, 5, 6, 11–13]. The present state of the problem is described in [1, 4–10], which also contain a large number of references to examples of applications of the method to specific problems.

In this review we shall limit our analysis only to the relaxation formalism of nonequilibrium thermodynamics, and we shall briefly analyze its relation to the methods of the theory of dynamic systems [14]. For a thermodynamic interpretation of the results obtained with the help of this theory, it is entirely sufficient to examine the theory as a convenient approximate method for describing macroscopic processes corresponding to the thermodynamic behavior of the system [15].

As far as the kinetic theory of molecular relaxation processes is concerned, the present state of the problem is described in [1]. In addition, the foundations of the kinetics and thermodynamic theories of molecular relaxation in gases and liquids, as well as data on relaxation of the translational, rotational, and vibrational degree of freedom, on relaxation with dissociation, ionization, electronic excitation, and chemical transformations in gaseous mixtures are presented in [1, 2, 7, 16–25].

Kinetic methods permit finding the relaxation times, which can then be used in the thermodynamic theory.

Specific examples of the application of the relaxation formalism to thermodynamics of irreversible processes encompass a very wide range of problems from different branches of mechanics, physics, and chemistry. A very lary number of papers is devoted to problems of acoustical relaxation: powerful modern methods for studying the structure and properties of substances in different aggregate states are based on the study of the characteristics of propagation and absorption of ultrasound [1, 7, 8]. We also call attention to [9, 10, 26-31], which are concerned with the study of dislocation processes, thermo-, magneto-, and visco-elastic relaxation, as well as relaxation due to first- and second-order phase transitions in solids (including polymers, structural, and other materials with a considerably nonuni-form structure, etc.).

Many attempts at using the thermodynamic formalism to construct models in control theory, economics, biology, and sociology have been made in recent years (see, for example, [32-35]).

Aside from a brief exposition of the thermodynamic theory of relaxation processes, we shall examine various applications of these methods to problems of physicochemical mechanics and transport processes in single-phase and dispersed media. For the problems indicated, molecular relaxation processes, dissocation processes, phase transformations, as well as chemical reactions and fluctuation processes are of greatest interest. There are a large number of papers and monographs concerning these problems so that it is impossible to describe all of them in detail in a brief review. For this reason, we shall restrict out attention to a schematic description of the important approaches to the solution of these problems, examining as examples the work involving a purely thermodynamic approach. In addition, we shall point out the papers in which the effect of relaxation processes on the behavior of different systems is examined to make it easier for the interested reader to penetrate this complex and rapidly developing field.

We shall examine the simplest system whose behavior is determined by two dynamic variables: an external controlling action $x(t)$ and a response $y(t)$. In so doing, we shall assume that a natural internal process $\xi(t)$ occurs in the system.

In specific situations, the orders of magnitude of the characteristic times of the processes $y(t)$ and $\xi(t)$ are often different. The evolution of the system in the phase space of the dynamic variables can then be described by a system of autonomous differential equations containing a small parameter ε:

$$\varepsilon D_t y = f(x, y, \xi), \quad D_t \xi = h(x, y, \xi), \tag{1}$$

where D_t is the operator of differentiation with respect to time, while f and h are some functions, which in general are nonlinear.

Integration of the system (1) is usually a very complicated problem. For this reason, approximate and qualitative methods for studying the trajectories of dynamic systems in phase space are important and, in particular, the theory of bifurcations, whose foundations were developed by A. A. Andronov et al. [14]. For system (1), asymptotic expansions of the solutions in powers of ε with an degree of accuracy can be constructed. For a higher order system (when x, y, and ξ are collections of several quantities of a similar nature), such expansions are available only with accuracy up to ε; the problem of obtaining the next terms in the expansions has not yet been solved [36, 37]. The construction of these expansions is based on a theorem by A. N. Tikhonov [36-39], according to which, under certain conditions, the solution of the starting system (1) approaches with $\varepsilon \to 0$ the solution of the degenerate system

$$f(x, y, \xi) = 0, \quad D_t \xi = h(x, y, \xi), \tag{2}$$

in which the second equation describes the evolution of the "slow" variable ξ, while the first, "associated" equation is a consequence of the fact that there exists a "fast" variable y. To find an approximate solution of (1), y must be found as a function of x and ξ from the associated equation and the function obtained must be used in the second equation in (2).

The procedure for separating variables into fast and slow variables was widely used in solving problems of chemical kinetics and statistical mechanics [40-42] even before the formulation of A. N. Tikhonov's theorem. The method based on it will, evidently, be all the more accurate the larger the difference between the characteristic times. The law governing the variation of the controlling action $x(t)$ must in all cases be determined from several independent considerations.

The behavior of the system for which Eqs. (2) are approximately valid can be interpreted qualitatively as follows. The image point of the system in phase space rapidly moves along the line $\xi = \text{const}$ onto the surface $f(x, y, \xi) = 0$, correspoding to stationary states relative to fast motions, and then moves slowly along the surface. With a number of limitations on the forms of the functions f and h, the mathematical model based on (1) admits a direct thermodynamic interpretation, which permits using an effective formalism of classical and non-equilibrium thermodynamics for solving problems [32]. This is possible not only for problems of a physical nature, but also for problems based on purely formal analogies (for example, in control theory [32] and in economics [33, 34]).

The conditions for the autonomous systems (1) to be stationary, $f = 0$ and $h = 0$, determine in a particular case the thermodynamically equilibrium states of the system, for whose description it is sufficient to give only some of the thermodynamic quantities, since the remaining quantitities can be obtained from the equations of state. With deviations from equilibrium, the latter functional relations break down, but they can be formally restored by defining additional internal variables characterizing such a deviation. This approach was first used by Leontonovich [43] and it is presently used in solving a number of problems in nonequilibrium thermodynamics. Thus the dynamic variables y and x can be interpreted as coupled thermodynamic quantities, while ξ can be interpreted as additional internal parameters. The thermodynamic interpretation of states $f(x, y, \xi) = 0$ and $h(x, y, \xi) = 0$ is possible only if f and h form an involution [32]. The equations themselves can in general be viewed as generalized equations of state, which are equilibrium in the usual sense of the word, i.e., if $\xi = 0$.

We shall examine the linear response Δy of system (1) to an external perturbation Δx assuming that $\xi(t)$ is a slow process, while $y(t)$ is a fast process. Then, the system (2) is valid. From the first equation in (2) it follows that $y = y(x, \xi)$ and, in addition, x and ξ are viewed here as independent variables. We examine the second equation in (2) in the relaxation approximation, assuming that h depends on the affinity A of the relaxation process. Then, expanding h(A) near a stationary state in a Taylor series, we obtain the following dynamic equation for the internal variable:

$$D_t \xi = - L(A - A^{st}), \quad L = \left(\frac{\partial h}{\partial A} \right)^{st}. \tag{3}$$

Linearizing with respect to Δx, $\Delta \xi$, we obtain from (2) and (3)

$$\Delta y = c^\infty \Delta x + Q \Delta \xi, \quad D_t \Delta \xi = - (P \Delta x + R \Delta \xi), \tag{4}$$

where we introduced the notation

$$c^\infty = (\partial y / \partial x)_\xi, \quad Q = (\partial y / \partial \xi)_x, \quad P = L(\partial A / \partial x)_\xi, \quad R = L(\partial A / \partial \xi)_x. \tag{5}$$

Here the quantity $A - A^{st}$ plays the role of a thermodynamic force giving rise to the relaxation process and L is a phenomenological coefficient. The derivatives in (5) are calculated at the point of phase space corresponding to a stationary state of the system, while the quantities Δy, Δx, and $\Delta \xi$ describe the deviation from this state.

Eliminating the quantity $\Delta \xi$ from (4), we obtain an operator relation between the response Δy and the pertrubation Δx, independent of $\Delta \xi$:

$$\Delta y = \hat{k} \Delta x, \quad \hat{k} = c^\infty - Q(R + D_t)^{-1} P. \tag{6}$$

The differential operator in (6) can be easily reduced to an integral operator, which permits writing down the solution of (6) in an explicit form [4].

The entire preceding discussion and the form in which relations (1)-(6) are written do not change if there are many variables y, x, and ξ. In this case, it is sufficient only to examine these variables as well as the quantity A as vectors, while L, c^∞, Q, P, R, and $\hat{k}$ must be viewed as matrices.

The matrix of phenomenological coefficients L is not necessarily symmetrical, i.e., its elements may not satisfy the Onsager reciprocity relations, since under stationary conditions far from equilibrium, the principle of local equilibrium may not be satisfied. However, as Glansdorf and Prigogine showed [44], for many important processes (for example, in chemical kinetics), this principle is still satisfied. Then, in studying relaxation to a stationary state of the system, it is possible to apply successfully the formalism of relaxation thermodynamics, devloped for states close to equilibrium.

Characteristics of a system describing its reaction to instantaneous changes of state and to slow displacements from the stationary or equilibrium state are quite common. In accordance with the separation of motions into fast and slow motions, the fast motions correspond to "frozen" internal processes (ξ = const), characterized by the quantity (or matrix) c^∞. On the other hand, it is evident from (3) that the condition $A = A^{st}$ corresponds to a stationary equilibrium ($A = 0$) state of the system, in which the response to an external perturbation corresponds to a slow stationary or reversible equilibrium process. In the last case, time does not enter explicity into the equation describing the evolution of the system and instead of (4) we have

$$y = y(x,\ A),\ \Delta y = c^+ \Delta x + Q_A \Delta A,\ c^+ = (\partial y/\partial x)_A,\ Q_A = (\partial y/\partial A)_x. \tag{7}$$

From (4)-(7), using the properties of Jacobians of the transformations or well-known relations between thermodynamic derivatives [2, 45], the generalized thermodynamic suscepti-bility

$$\hat{k} = (\partial\hat{y}/\partial x) = c^\infty + \frac{c^+ - c^\infty}{1 + \tau_x D_t} \tag{8}$$

can be expressed in terms of the characteristics of two limiting states of the system: the instantaneous (or frozen) c^∞ and stationary (or equilibrium) c^+ and the relaxation time τ_x.

The detailed procedure for obtaining the susceptibility in matrix form is described in [2, 4, 5] and, in addition, for relaxation to an equilibrium state, it is possible to use, as usual, the entropy and the thermodynamic potential of the system to calculate the matrix elements. In working with the operator D_t, it can be formally viewed as a number or as a parameter in the Laplace and Fourier transformations.

In [46], the variables were separated into fast and slow variables with the help of linear phenomenological Onsager equations

$$D_t y = -M_{yy}y - M_{y\xi}\xi,\ D_t\xi = -M_{\xi y}y - M_{\xi\xi}\xi \tag{9}$$

(this system can formally be obtained by linearizing (1)). If the ratio of the relaxation times $\tau_y/\tau\xi = \epsilon$, then $M_{y\xi}$ and M_{yy} are of the order of ϵ^{-1} relative to $M_{\xi\xi}$ and $M_{\xi y}$. It follows from here that in general the fast and slow variables are quite strongly coupled. This coupling can be greatly decreased with the help of special unimodular transformations of the quantities y, ξ.

The formalism developed in [46] can be applied to the analysis of the behavior of temporal correlation functions of nonequilibrium macroscopic fluctuations, whose evolution is described by equations of the type (9) [47] and to the analysis of the Fokker-Planck equation. In analyzing Brownian motion, the separation into fast and slow variables can be realized because the change in the velocity of the Brownian particles occurs much more rapidly than the change in their positions. As a result, the solution of the Fokker-Planck equation is expressed as a series with respect to the eigenfunctions of the operator, in which the nonstationariness due to fast motions is neglected. The lowest eigenvalue of this operator corresponds to the equilibrium distribution function in velocity space.

We shall briefly point out some of the recently discussed generalizations of the theory of relaxation processes described above. In [48], the concept of internal variables is generally not used and the formal apparatus is constructed by analogy to the theory of electrical changes in the system that are linearly related to the parameters describing the state of the system, while the type of coupling is determined by the structure of the system.

In [49], the equations of nonlinear relaxation are obtained in the following form using the methods of statistical mechanics:

$$D_t\xi_i = -L_{ij}A_j - L_{ijk}A_jA_k. \tag{10}$$

In [50], a variational principle for linear irreversible processes is formulated. The solutions of the Euler-Lagrange equations of the variational problem describe both the characteristic relaxation and the "antirelaxation," which realizes the choice of dynamic system, perturbed by the random forces, and the most probable trajectory in phase space. The behavior of a Brusselator is examined as an example [15].

Finally, it is sometimes convenient to introduce some internal configuration space of the variably y [5, 51] and to assume that the variable ξ is a continuous function of y. For ex-

ample, the quantity $\xi(y)$ can be the particle density in a state characterized by a definite value of y, while y can be an angle determining the orientation of molecules, velocity of a Brownian particle, etc. The variable ξ satisfies the equation of continuity

$$\partial \xi / \partial t = -\nabla_y \vec{T}_\xi, \tag{11}$$

and, in addition, some phenomenological law must be indicated for the flux $\vec{T}_\xi$. Equation (11) naturally supplements the system of equations of conservation of mass, momentum, and energy. The expression for the entropy in this case contains an additional term, corresponding to diffusion in phase space. The application of the method in [5, 51] is illustrated for problems in chemical kinetics and Brownian motion.

In [11], the concept of internal variables is used to analyze and solve approximately the Louiville equation.

The dynamic equation (3) is often represented in the form

$$D_t \xi = -\frac{1}{\tau_{xy}} [\xi(t) - \xi_0], \tag{12}$$

where ξ_0 is the equilibrium value of the relaxation parameter. The relaxation time τ_{xy} is related to the affinity of the relaxation process [2, 5, 6]:

$$\tau_{xy} = -[L(\partial A/\partial \xi)_{xy}]^{-1}. \tag{13}$$

This expression and the known thermodynamic relations for A [2, 5, 6] permit using the entropy and thermodynamic potential in calculating the relaxation times.

All phenomenological methods for describing relaxation processes thus require an independent determination of either the phenomenological coefficients or relaxation times or the rate constants. A convenient practical technique for determining relaxation times from the inflection points and extrema on the experimental curves of different quantities as a function of ln t was proposed in [52].

Chemical relaxation and fluctuations are classical examples of processes whose description is based on the relaxation equation (12).

For small deviations of the reaction coordinate ξ from the equilibrium value ξ_0, when the relation $A(\xi_0)$ can be represented in the form $A = (\partial A/\partial \xi)(\xi - \xi_0)$, the reaction can be viewed as a relaxation process. In this case, the reaction rate satisfies Eq. (12). The quantity A is usually described by a relation expressing it both as a function of the chemical potentials of the substances and the stoichiometric coefficients, while the phenomenological coefficients are determined from the mass action law. For example, for a molecular reaction of the type $A \rightleftharpoons B$, occurring under isochoric–isothermal conditions [2], the relaxation time

$$\tau_{TV}^{-1} = kC_A \left(\frac{1}{C_A} + \frac{1}{C_B} \right), \tag{14}$$

where k is the reaction rate constant; C_A and C_B are concentrations.

The theory of chemical relaxation is described in detail in [2, 5, 6, 53, and 54]. In particular, in [54], for complex chemically reacting systems, methods for simplifying the initial problem, based on the analysis of a hierarchy of relaxation times and on separation of fast and slow processes, are proposed and some interesting recommendations are given for analyzing even more complicated biophysical and biochemical systems.

In the study of fluctuations in macroscopic systems, as well as in the analysis of relaxation processes, the same basic approaches are used: kinetic and thermodynamic. The thermodynamic theory of fluctuations in an equilibrium state is examined in detail in [5, 47]. Damping of fluctuations averaged over the equilibrium distribution satisfies linear equations of the type (9) or (12), which was checked experimentally. Based on the linear equations, it is easy to find the laws governing the changes in the temporal correlation functions [46, 47].

On the other hand, the evolution of fluctuations in time can be viewed as the realization of some random process, which under certain conditions, can be described with the help of the Langevin or Fokker–Planck equations. Fluctuations have been studied in very many papers, in particular in [55-63], from this point of view.

A very important result of the theory of fluctuations is the well-known fluctuation-dissipation theorem, which establishes a relation between the generalized susceptibility $\hat{k}$ and the matrix of the spectral density of the correlation function [5, 47]. A large number of applications of the method of correlation functions to different problems is examined in [64]. Here, and in [65-68], some problems in fluctuation hydrodynamics are analyzed. A review of results obtained in investigations of fluctuations in dissipative and nonlinear systems in states far from equilibrium is given in the monographs [15, 44].

Fluctuations play a large role in self-organization processes in nonequilibrium systems, in which new dissipative structures can arise with the loss of stability of some state as a result of fluctuations. The stability of these new structures can be ensured by sufficiently rapid damping of fluctuations. The simplest criteria for determining the boundaries separating regions of stability for two different dissipative structures can be established with the help of the relaxation formalism [68]. Assuming that Einstein's equation is valid for the probabilities of nonequilibrium fluctuations [47]

$$W \sim \exp(\Delta S/k) \tag{15}$$

and using the relaxation equation following from (12) with $\xi \equiv S$ to describe the change in entropy

$$J_S = \frac{dS}{dt} = -\frac{\Delta S}{\tau}, \tag{16}$$

it is not difficult to obtain the relation

$$W \sim \exp\left(-\frac{J_S\tau}{k}\right), \tag{17}$$

from where it follows that the boundary indicated is determined by the condition $\tau_1 = \tau_2$. The conditions for the appearance of nuclei of a new phase, the boiling critical point, the critical point of gas glow in a tube, transition of laminar flow into turbulent flow, etc. were analyzed with the help of this simple criterion. Thus, for the critical value of the Reynolds number describing the onset of turbulent flow of a laminar boundary layer, the theory gives approximately $1.15 \cdot 10^5$, which is close to the experimental value $1.2 \cdot 10^5$. Many of the results obtained by applying the theory of random processes to the study of fluctuations are reviewed in [69].

We shall now discuss the problems of relaxation hydrodynamics of single-phase and dispersed media. For sufficiently high temperatures, pressures, and velocities, relaxation can strongly affect their properties. These effects can be related to incomplete chemical reactions in a flow, dissociation and ionization processes, deviations of the molecular and atomic distributions from the equilibrium distribution, and the presence of configuration effects in structured liquids and polymers. In dispersed media, relaxation processes due to nonequilibrium thermal and dynamic action of phases in phase transitions are also important. Extensive information on these problems, which have become especially important in recent years in connection with the demands for intensifying many technological processes, is collected in [70-79].

The construction of mathematical models of fluid flows in the presence of relaxation phenomena is based on the usual system of equations of conservation of mass, momentum, and energy [80]. Following the general method, this system is supplemented by equations of kinetics of relaxation processes (of the type (8) or (12)). In addition, it is assumed that the density, pressure, internal energy, and enthalpy of the fluid depend on the relaxation parameters ξ, i.e., generalized equations of state are available in which ξ enter as arguments. In the general case, relaxation processes can affect in an analogous manner the tensor of viscous stresses and the heat flow [5, 6]. In the most complete form, the concept of internal variables relative to hydrodynamics of relaxing media is examined in [81]. This approach is used in [81] for the simplest dispersed system: a dilute suspension of nonspherical particles.

Relaxation processes in suspensions, emulsions, gaseous suspensions, granular layers, porous media, and other dispersed systems occur, as a rule, due to the difference in temperatures and velocities of phases, as well as the presence of interphase mass transfer processes. The classification of mixtures when analyzing them using the methods of nonequilibrium thermodynamics is also based on an examination of the hierarchy of relaxation times [82]: mixtures

with strong and weak interaction between components or phases are separated. In so doing, mixtures of the first type can be successfully examined as a homogeneous continuous medium, while for mixtures of the second type it is necessary to use models of multivelocity and multitemperature interpenetrating continua [74-76]. Very often, especially in developing engineering techniques for analyzing processes in dispersed media, equations of hydrodynamics for coexisting continua are supplemented by phenomenological equations of hydrodynamic, thermal, and mass transfer interactions of phases of the same type of Stokes' or Newton's law. This makes it necessary to determine the frictional resistance coefficients and the heat and mass transfer coefficients independently. It is often convenient, on the other hand, to formulate these laws in terms of the relaxation formalism of nonequilibrium thermodynamics, whose limits of applicability to hydrodynamic problems are studied in [72], which also contains typical examples of relaxation processes in fluid flows.

Rich experience has now been accumulated on solving a wide range of problems of relaxation hydrodynamics of single- and multiphase media. The basic directions of the investigations can be formulated as follows [70-101]:

1) relaxing flows in nozzles, channels, and pipes;

2) relaxing flow around bodies with different shapes;

3) propagation of weak perturbations with small and finite amplitude (including sound waves) in a relaxing medium;

4) shock waves and other surfaces of discontinuity in the presence of relaxation processes.

Many papers are concerned with studying directly the effect of relaxation processes on transport of matter, energy, and momentum [102-131]. Diffusion, thermal, and chemical relaxation accompanying liquid—vapor phase transitions are examined [102-107]. Dynamic equations of state [6, 89, 108, 109], nonequilibrium effects accompanying percolation [110-112], problems of describing heat conduction and diffusion with relaxation with the help of different generalizations of the usual transport equations, in particular, hyperbolic equations [114-131], are also studied. Here we shall consider in greater detail only investigations in which the relaxational formalism described above is used in some way.

The results obtained in relaxational thermodynamics up to 1975, an exact method based on numerical integration of equations of gas dynamics and chemical kinetics, and an approximate method for solving the problems of flows in the channels of jet engines, etc. are discussed in [70]. The last method is actually based on the scheme examined for separating fast and slow variables: up to some section of the channel, the flow is assumed to be an equilibrium or almost an equilibrium flow, and then it is assumed that the external (from the side of the channel) actions vary so rapidly that the relaxation processes appear to be frozen (ξ = const). In [73], based on an analysis of the spectra of relaxation times, a criterion is proposed for determining the "stagnation point" of the flow and specific calculations are performed for chemical, vibrational, and rotational relaxation processes. In [71], numerical methods for integrating the equations of relaxational gas-dynamics are examined in detail, and a general scheme is discussed for describing relaxation processes and finding relaxation times. In addition, the relaxation formalism is used in this paper to describe phase transitions in rarefied dispersed flows in turbine machines.

Recently, many important problems of relaxational gas- and hydrodynamics were discussed at the sixth All-Union Conference on Rarefied Gas Dynamics (Novosibirsk, 1979). Supersonic flow past bodies, shock waves, flow in channels in the presence of relaxation, and other relaxational phenomena were examined, as well as problems of using the apparatus of kinetic theory of gases for constructing a complete system of equations of relaxational gas dynamics [83].

The propagation of perturbations in two-phase media is investigated in [84-99]. Thermal and hydrodynamic interaction of phases in gaseous suspensions containing solid particles [86-88, 96-99] as well as phase transitions in mixtures containing evaporating drops [84, 89-92, 95] are most often examined at the phenomenological level by introducing appropriate relaxation times. In addition, either dynamic or thermal relaxation is neglected to make it possible to use either a single-velocity or single-temperature model of two-phase mixtures [91, 98, 99]. It should be noted that the use of quasistationary relations to describe interphase interactions leads to considerable errors for high-frequency perturbations. To eliminate

these disadvantages, it is necessary to solve special nonstationary problems of interphase exchange, which greatly complicates the analysis. The problem of obtaining a single evolutionary equation for waves with small, but finite amplitude has been studied in recent years [74, 90, 96, 98, 99]. Nonlinear waves in gas—liquid mixtures were also studied in [92] and, in addition, interphase mass transfer was examined in a relaxation approximation. Transport processes on the interface between two immiscible media, taking into account relaxation effects, were studied in [104] using linear thermodynamics. In particular, it was demonstrated that their presence narrows the possible relations between the thermodynamic forces and flows, which decreases the number of boundary conditions. The relaxation formalism is used in [108] to derive a dynamic equation describing adiabatic compression of gas with solid particles. This equation gives the adiabat for compression of a pure gas for rapid processes and the adiabat for compression of an equilibrium mixture for slow processes.

Equation (8) for the generalized susceptibility permits introducing the operator representation for thermodynamic functions of nonequilibrium states in the relaxation approximation [45]. In this manner it is possible to determine the index of the adiabat $\hat{n}$, effective heat capacity of the process $\hat{c}_z$, velocity of sound $\hat{a}$, etc. We have

$$\hat{n} = n^{(\infty)} + \frac{n^{(0)} - n^{(\infty)}}{1 + \tau_V D_t}, \quad \hat{c}_z = c_z^{(\infty)} + \frac{c_z^{(0)} - c_z^{(\infty)}}{1 + \tau_T D_t},$$

$$\hat{a}^2 = \hat{n}\frac{p}{\rho} = (a^{(\infty)})^2 + \frac{(a^{(0)})^2 - (a^{(\infty)})^2}{1 + \tau_V D_t}, \tag{18}$$

where τ_V, τ_T are relaxation times for constant volume and temperature, respectively; p, ρ are the pressure and density, while the indices (∞) and (0) indicate instantaneous and equilibrium change of state.

If the operator D_t is replaced in the expression for $\hat{a}^2$ by its eigenvalue, then a relation is obtained which was first derived by Meixner in the theory of acoustic relaxation [6]. In [96], the two-phase medium was examined as a system with N internal relaxation processes. In this case, the square of the velocity of sound is expressed as follows:

$$a^2 = (a^{(\infty)})^2 + \sum_i^N \frac{a_{i-1}^2 - a_i^2}{1 + \omega^2 \tau_i^2}, \tag{19}$$

where a_{i-1}, a_i are the velocities of sound in the case when $i - 1$ or i relaxation processes are frozen. Specific calculations of the velocity of sound were performed for a vapor-drop system taking into account rapid relaxation and relaxation of the temperature of the vapor and of the drop to the saturation temperature. Analogous calculations were performed for systems of bubbles in liquids.

The operator form of the equation of the adiabatic process with index $\hat{n}$ from (18) or equations for change of volume with deformation, in which the presence of relaxation is accounted for with the help of the operator of the volume modulus of elasticity $\hat{\varkappa}$, introduced in analogy to [108], permits writing the dynamic equation of compression in the following form:

$$\frac{d^2 p}{dt^2} V + \frac{dp}{dt}\left(\frac{dV}{dt} + \frac{V}{\tau_V}\right) + \varkappa^\circ\left(\frac{\tau_V}{\tau_p}\frac{d^2 V}{dt^2} + \frac{1}{\tau_V}\frac{dV}{dt}\right) = 0. \tag{20}$$

At constant volume, this equation describes relaxation of pressure:

$$\frac{p(t) - p_{\text{fin}}}{p_{\text{in}} - p_{\text{fin}}} = \exp\left(-\frac{t}{\tau_V}\right). \tag{21}$$

This relation is confirmed by experimental data on pressure working of multiphase mixtures and oil [109].

The results of investigations of the effect of relaxation on percolation of viscoelastic polymer media and oil are presented in [110, 111]. The theoretical investigation of percolation with relaxation is based on the assumption that the permeability k_ω relaxes according to a law of the type (12). A theory of percolation, based on the relaxation formalism of nonequilibrium thermodynamics, was proposed in [112]. Using an operator representation of the type (8) for quantities inversely proportional to the permeability, the dynamic form of Darcy's law can be written as follows:

$$-\nabla p = \hat{k}_\Phi^{-1} J, \tag{22}$$

where J is the velocity of percolation and ∇p is the pressure gradient. Using the expression for $\hat{k}_\Phi^{-1}$ we obtain from (22)

$$-\nabla p = \left(a - \sum_i \frac{\mu_i}{\tau_i} \right) J + \sum_i \int_0^t \frac{\mu_i}{\tau_i^2} \exp\left[-\frac{t-u}{\tau_i} \right] J(u)\, du, \tag{23}$$

where a and μ_i are phenomenological coefficients. If there is only one relaxation process, then it is easy to obtain from (23) the equation proposed in [111].

The possibility of including relaxation in transport of momentum and energy in viscoelastic media was first proposed by Maxwell [114]. Later, such ideas were examined for the process of heat conduction with finite velocity [115-117].

If we introduce the generalized Fourier law

$$\vec{q}(t) = -\int_0^\infty \lambda_\theta(\theta)\, \nabla T(t-\theta)\, d\theta, \tag{24}$$

and define, in analogy to [114, 115], the function $\lambda_\theta(\theta)$ in the form

$$\lambda_\theta(\theta) = \frac{\lambda}{\tau} \exp\left(-\frac{t}{\tau} \right), \tag{25}$$

then we obtain the following expression from (24) and (25) for the heat flux density, taking into account the finiteness of the velocity of propagation of heat:

$$\vec{q} = -\lambda \nabla T - \tau \frac{\partial \vec{q}}{\partial t}, \tag{26}$$

where λ is the usual coefficient of thermal conductivity.

A method is proposed in [118] for determining the relaxation time of the heat flux and the functions describing the relaxation of this flow and of the internal energy for media with thermal memory.

The equation of heat transfer in a substance for which the law (26) is valid is a hyperbolic equation:

$$\frac{\partial T}{\partial t} + \tau \frac{\partial^2 T}{\partial t^2} = a \nabla^2 T. \tag{27}$$

Using instead of (26) the more general law (24) leads to an integrodifferential heat-conduction equation [117].

In [119], the effect of relaxation on heat transport is included with the help of linear phenomenological relations

$$\vec{q} = -\frac{L_{qq}}{T^2} \nabla T - \sum_i L_{qi}\vec{\xi}_i, \quad \rho\vec{\xi}_i = -\frac{L_{qq}}{T^2} \nabla T - \sum_k L_{ih}\vec{\xi}_h. \tag{28}$$

For a single relaxation process, it follows from here that Eq. (26) is replaced by

$$\rho \frac{\partial \vec{q}}{\partial t} + L_1\vec{q} = -\frac{\rho L_{qq}}{T} \nabla T - \frac{(L_1 L_{qq} - L_{q1}^2)}{T^2} \nabla T. \tag{29}$$

This expression leads to a third-order differential equation of heat conduction:

$$\frac{\rho}{L_1} \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \left(\frac{L_{qq}}{T^2 L_1} \frac{\partial}{\partial t} + \frac{L_1 P_{qq} - L_{q1}^2}{L_1 \rho T^2} \right) \nabla^2 T. \tag{30}$$

If the internal variable ξ changes sign under time reversal, then Onsager's relations in (28) must be replaced by the corresponding Casimir relations, i.e., the sign of the first term on the right side of the equation for $\rho\xi_i$ must be changed. This changes the sign in front of $L_{q_1}^2$ in (29) and for $L_{qq} = 0$ (29) transforms into (26) and, in addition (30), in this case, changes into the hyperbolic equation (27). If ξ is even, then $L_{qq} = 0$ only under the

condition that $L_{q_1} = 0$ or $\nabla T = 0$, since otherwise the requirement that the entropy-generating function be positive-definite is not satisfied. It is thus evident that only odd internal relaxation variables can be responsible for the finite velocity of propagation of heat. Two- and multitemperature systems are also examined in [119], and higher-order heat-conduction equations in solids with relaxation are obtained in [120].

If the heat capacity is represented in the form (18) when formulating the equation for conversation of heat in a relaxing or nonequilibrium dispersed medium, then we obtain the generalized heat-conduction equation [45]

$$\frac{\partial^2 T}{\partial t^2} c_p^{(\infty)'} \tau_T + c_p^{(0)'} \frac{\partial T}{\partial t} = \lambda \left(1 + \tau_T \frac{\partial}{\partial t} \right) \nabla^2 T. \tag{31}$$

An analogous heat-conduction equation in a granular medium, but with different coefficients, was obtained in [121]. The derivation was based on a special representation of the temperature fields in the continuous phase and in particles. It was shown in [122] that Eq. (31) can in principle be used to model heat conduction in biological tissues, in particular, in a contracting muscle. It was proposed in [123] that (27) be used for fluidized bed.

If the operator $(1 + \tau_T D_t)^{-1}$ in the expression for the heat capacity (18) is expanded in a series in powers of $\tau_T D_t$, then we obtain an equation containing an infinite chain of increasingly higher-order time derivatives of the temperature [124]. In the linear approximation with respect to $\tau_T D_t$, this equation is a second-order equation and it is hyperbolic or elliptic depending on the ratio of the heat capacities $c_p^{(\infty)'}/c_p^{(0)'}$. In dispersed media, the first approximation usually leads to an elliptic equation [124, 125].

Operator representations based on (8) were introduced in [126] for the chemical potential and coefficient of diffusion

$$\hat{D} = D^{(\infty)} + \frac{D^{(0)} - D^{(\infty)}}{1 + \tau D_t}. \tag{32}$$

From here it is easy to obtain a general equation of diffusion in a form analogous to the form of Eq. (31), Replacing D_t in (32) by the eigenvalues i_ω and separating the real and imaginary parts, a dispersion relation is obtained for the diffusion coefficient. This was done for a fluidized bed in [127]. A hyperbolic diffusion equation was obtained in [128–130] using different methods. It was applied in [131] to modeling of transport of calcium ions in heat muscle.

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