

ON THE PROBLEM OF NONISOTHERMAL MASS TRANSFER IN POROUS MEDIA

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We propose a model of nonstationary processes of interrelated heat and mass transfer in porous media that takes into account the mutual influence of vapor and liquid pressures determined by the contribution of the capillary and surface forces and temperature on the interphase mass-transfer intensity, the thermocapillary flows, and the mechanical and dynamic equilibrium conditions of thin layers of liquids on curved interfaces. The results of the numerical solution of the system of equations of heat and mass transfer in capillary-porous bodies in discrete and continuous heating as well as with allowance for the effect of nonequilibrium of phase transformations have been analyzed.

Introduction. At present there is no single phenomenological approach to the description of heat and mass transfer processes in porous media. The discrepancies between the theories of sorption, filtering, and drying for describing one and the same interrelated heat and mass transfer in porous media are fundamental. The chief disadvantage of these theories is that they do not completely take into account the relationship between different mechanisms of moisture transfer and the influence of the surface phenomena.

To describe the drying processes, A. V. Luikov [1] proposed a system of differential equations of interrelated heat and mass transfer with allowance for the heat and moisture conduction:

$$j = D\rho_{\text{sol}} \left(\frac{\partial \tilde{u}}{\partial x} + \delta' \frac{\partial T}{\partial x} \right).$$

By virtue of the fact that the moisture content is not a transfer potential, the effect of heat and moisture conduction is no longer cross and can have a marked influence on the drying process. It is also important that the value of δ' depends, as a rule, on the process parameters.

The system of equations of interrelated heat and mass transfer proposed by A. V. Luikov played an important part in investigating the transfer processes in porous media at phase transformations. It should be noted, however, that its application does not permit estimating the role of each of the phases and surface phenomena in the resulting mass transfer in porous bodies in the presence of phase transitions. Therefore, in the present paper we propose a mathematical model that permits taking into account, in terms of the theory of multiphase filtering and with the aid of sorption isotherms and the Kelvin formula, the interphase mass-transfer intensity and the influence of the surface forces in describing the nonstationary heat and mass transfer in the case of both low- and high-intensity drying processes.

Mathematical Model. *Basic assumptions of the model.* In constructing the physical model, we will use the assumption of validity of the fulfillment of the hypothesis that in an elementary macrovolume of a porous body there is a local thermodynamic equilibrium, which can be substantiated as follows. In a porous medium, the boundary between two phases is broken down into many individual portions with a different curvature. The radius of curvature of each of them is close in order of magnitude to the characteristic size R of the pore channel. We consider capillary-porous media in which the transfer processes are strongly influenced by the capillary forces (according to [1], this holds for porous media with a capillary radius $R \leq 10^{-7}$ m). Imagine at some initial instant of time a hypothetical pore of

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cylindrical or spherical form whose walls are covered with a liquid film and the inner part of the which is filled with, e.g., clean air without impurities of this liquid's vapors. In the infinitely thin layer near the liquid surface, the concentration of vapors of this liquid, e.g., water, corresponds to some equilibrium value of C_e . As a result of the diffusion processes, in the course of time the vapor concentration in the inner volume of the pore will increase to the respective value of C_e on the film surface. The diffusion coefficient of vapors D_v of, e.g., water in the air, according to the data of [2, 3], is determined as

$$D_v = 0.216 \cdot 10^{-4} \left(\frac{T}{373} \right)^{1.8}.$$

Then for the relaxation time (or the equilibration time) we have the estimate $\tau_r \approx R^2/D_v \approx 5 \cdot 10^{-10}$ sec, i.e., τ_r is much shorter than the characteristic times of the heat and mass transfer processes.

In the heat and mass transfer theory, the thermal phase equilibrium hypothesis is widely used by practically all researchers [1, 9–18]. The relaxation time estimates, including those given above, show that the mass-transfer dynamic phase equilibrium hypothesis is also fairly well substantiated and can be used in a large number of situations. In this case, the nonequilibrium drying process should be understood as a quasi-equilibrium one where each local macrovolume of a porous body passes through a continuous series of instantaneous states of local thermodynamic equilibrium between phases. Nevertheless, for each particular case it is necessary to check the fulfillment of such assumptions since their fulfillment is associated with the time in which the process proceeds, the porous body structure, and the properties of the phases.

Note that the local thermodynamic equilibrium hypothesis is used in the theories of drying, sorption, and two-phase filtering in determining, respectively, the mass-transfer potentials and the heat-moisture conductivity [1, 4–8], the interphase mass-transfer intensity [9–12], and the Leverett function [13–18].

Under the conditions of dynamic liquid-vapor equilibrium, the phase potentials should be equal to

$$\mu_{\text{liq}}(p_{\text{liq}}, T) = \mu_v(p_v, T). \quad (1)$$

If $p_v = p_s$, where p_s is the saturated vapor pressure of volumetric liquid over a flat surface, then the liquid pressure in this case will be equal to the gas pressure over the flat surface and the equilibrium condition will take on the form

$$\mu_{\text{liq}}(p_s, T) = \mu_v(p_s, T). \quad (2)$$

The chemical potential of one mole of vapor at low pressures can be written as

$$\mu_v = \mu_0(T) + \tilde{R}T \ln \frac{p_v}{p_s}, \quad (3)$$

here $\mu_0(T)$ is the chemical potential of vapor in some standard state depending only on temperature. In our case, we assume that the phase temperatures are equal. Then from Eqs. (2) and (3) it follows that

$$\mu_{\text{liq}}(p_{\text{liq}}, T) - \mu_{\text{liq}}(p_s, T) = \tilde{R}T \ln \frac{p_v}{p_s}. \quad (4)$$

Taking into account that the liquid is slightly compressible and using the thermodynamic equality [19]

$$v'_{\text{liq}} = \left(\frac{\partial \mu_{\text{liq}}}{\partial p_{\text{liq}}} \right)_T, \quad (5)$$

we can expand the left-hand side of (4) into a Taylor series in the $p_{\text{liq}} - p_s$ variable. Then, restricting ourselves to the first expansion term, we have

$$v''_{\text{liq}}(p_{\text{liq}} - p_s) = \tilde{R}T \ln \frac{p_v}{p_s} \quad (6)$$

and, consequently, in the equilibrium state the liquid pressure can be determined by the equilibrium pressure of the vapor [20]

$$p_{\text{liq}} = p_s - \frac{\tilde{R}T}{v''_{\text{liq}}} \ln \frac{p_v}{p_s}. \quad (7)$$

Expression (7) is known as the Kelvin formula.

Thus, postulating only a change in the liquid pressure with changing vapor pressure at the interface boundary, we arrive at the following conclusion: knowing the partial vapor pressure for a real porous body with a macroscopic curvature and roughness, which are not known, as a rule, we can obtain the value of the liquid pressure. In a porous structure, it is composed of the capillary and disjoining pressures and is defined by the Kelvin formula, and, in this case, there is no need to introduce the effective radius of the pores and surface tension. As is known, the majority of natural porous media are fractal structures the description of whose inner surface on the basis of Euclidean geometry and the classical notions of integral and differential will no longer be correct. The introduction of the effective parameters — capillary radius and surface tension — is now not apparent and does not follow from the thermodynamics of the surface phenomena.

In deriving the Kelvin formula, it was assumed that the vapor pressure over the interface differed from the liquid pressure only slightly. Indeed, only in this case will the change in the liquid pressure be insignificant and the Taylor expansion be valid. However, taking into account the fact that the liquid covering the porous body surface is also compressible only slightly (down to a few intermolecular distances) [21] and reasoning likewise, in some approximation we can also generalize formula (7) to the case where the liquid pressure considerably differs from the saturated vapor pressure and their difference $p_{\text{liq}} - p_s$ is not small.

The sorption isotherms (more precisely, their desorption branches) and expression (7) are used to construct curves of the radius-distribution of pores practically throughout the range of changes in their radii [11, 12, 22, 23].

Closing relations. Subsequently, we will need the dependence of the liquid pressure on the moisture content or saturation at various temperatures. In solving this problem, we will use the experimentally obtained sorption or desorption isotherms for a specific material. In practice, one usually determines the dependence of the equilibrium moisture content u_e on the relative humidity of vapors $\phi = p_v/p_s$ at a fixed temperature, i.e., $u_e = f(\phi, T)$. However, it is more convenient to first express the temperature dependence of saturated vapor pressure as

$$p_s = p_s(T) \quad (8)$$

and represent the sorption (desorption) isotherm in the form

$$u_e = f(p_v, T). \quad (9)$$

Hereinafter we assume that the total pressure of the mixture slightly influences the amount of equilibrium moisture, which is justified if the change in the total pressure of the mixture does not exceed a few atmospheres. Suppose that it is possible to determine analytically the vapor pressure in the moisture-content and temperature function with the aid of the inverse functions

$$p_v = F(u_e, T). \quad (10)$$

Then from relation (7) one can express the liquid-phase pressure depending on the moisture content and, which is particularly important, on the temperature:

$$p_{\text{liq}} = p_s + \frac{\tilde{R}T}{v''_{\text{liq}}} \ln \frac{F(u_e, T)}{p_s(T)}. \quad (11)$$

The experimental temperature dependences of the equilibrium moisture content for most materials are such that as the temperature increases, there is also an increase in the liquid pressure p_{liq} in accordance with (11). Provided the local equilibrium hypothesis is satisfied, this leads (as will be shown below by a number of examples) to the displacement of the liquid to the cold parts of the material, i.e., to the heat-moisture-conduction effect. This result is described in [24–29] and is of great importance in closing the moisture motion equations. Unlike the Kelvin formula, relation (11) takes into account the physicochemical properties of the liquid and the structural characteristics of the porous body and is needed to solve the liquid-phase transfer equation as a closing relation between the liquid pressure and the moisture content.

If the local thermodynamic equilibrium hypothesis is satisfied and the phases in the nonequilibrium process pass through a set of equilibrium instantaneous states, then the mass-transfer intensity for the one-component two-phase system is determined by the equation [9–12, 30]

$$I = \frac{du_e}{d\tau} = \left(\frac{\partial u_e}{\partial p_v} \right)_T \frac{\partial p_v}{\partial \tau} + \left(\frac{\partial u_e}{\partial T} \right)_{p_v} \frac{\partial T}{\partial \tau}. \quad (12)$$

This expression is widely used in the sorption theory.

Relation (12) holds if du_e is a perfect differential and does not depend on the path of integration. The wings of the sorption and desorption isotherms do not coincide and have a hysteresis. Therefore, (12) holds when the mass-transfer process proceeds in any one direction: the direction of sorption or desorption. In practice, the process proceeds most often in any one regime, and, therefore, with a correctly chosen isotherm branch relation (12) is quite feasible. This remark is also valid in determining the specific heat of sorption.

Let us make some transformations of the equality condition of chemical potentials (1) by the technique given in [19]. We differentiate it with respect to temperature and obtain

$$\frac{\partial \mu_{\text{liq}}}{\partial T} + \left(\frac{\partial \mu_{\text{liq}}}{\partial p_v} \right)_T \frac{\partial p_v}{\partial T} = \frac{\partial \mu_v}{\partial T} + \left(\frac{\partial \mu_v}{\partial p_v} \right)_T \frac{\partial p_v}{\partial T}. \quad (13)$$

Since

$$\begin{aligned} \left(\frac{\partial \mu_{\text{liq}}}{\partial T} \right)_{p_{\text{liq}}} &= -S_{\text{liq}}, & \left(\frac{\partial \mu_{\text{liq}}}{\partial p_{\text{liq}}} \right)_T &= v''_{\text{liq}}, \\ \left(\frac{\partial \mu_v}{\partial T} \right)_{p_v} &= -S_v, & \left(\frac{\partial \mu_{\text{liq}}}{\partial p_v} \right)_T &= v''_v, \end{aligned} \quad (14)$$

we have

$$\frac{\partial p_v}{\partial T} = \frac{S_{\text{liq}} - S_v}{v''_{\text{liq}} - v''_v} = \frac{r'_{\text{ev}}}{T(v''_{\text{liq}} - v''_v)}. \quad (15)$$

Since $v''_v \gg v'_{\text{liq}}$ and assuming $v''_v = \tilde{R}T/p_v$, on a mass-unit basis we have the Clapeyron–Clausius relation [19]

$$r_{\text{ev}} = \frac{\tilde{R}T^2}{v'_{\text{liq}}} \left(\frac{\partial \ln p_v}{\partial T} \right)_{u_e}. \quad (16)$$

Formula (16) holds in a wide range of pressures and temperatures only if the vapor can be considered as an ideal gas. Using (10), we can write expression (16) in the form of the formula

$$r_{\text{ev}} = \frac{\tilde{R}T^2}{v'_{\text{liq}}} \left(\frac{\partial \ln F(u_e, T)}{\partial T} \right)_{u_e}, \quad (17)$$

which was obtained by us in [24–29]. Most authors use (16) in combination with special methods of graphic differentiation that permit obtaining the value of r_{ev} only at some points, which leads to many difficulties in a continuous change in the parameters T and p_v . As is known, the methods of graphic differentiation of experimental functions can introduce large errors into calculations. Consequently, using the experimental dependences (8) and (9), one can also determine the specific heat of sorption and the mass-transfer intensity. It is also possible to determine, without additional experiments, the dependence of the liquid pressure on the moisture content and temperature.

The lower validity limit of (11), (17) is determined by the possibility of using (7). With pore sizes of the order of 10 Å, when the interaction between the pore walls and the gas molecules becomes appreciable, the gas is no longer ideal. The employment of formula (7) in this case gives a large error, and, therefore, pores of such an order determine the lower validity limit of the Kelvin equation [23].

The upper validity limit of (7) is determined by the complexity of obtaining sorption isotherms at $p_v/p_s \rightarrow 1$, which is due to the insufficient accuracy of measurement of the partial vapor pressure near p_s and the necessity of maintaining the sample temperature with exceptional accuracy. The above difficulties led many researchers [1, 4–8] to the statement that the macropores in the sorption process at $\phi \rightarrow 1$ are not filled. However, experiments [31] with special thermostating systems show that at $\phi \rightarrow 1$ the macropores, too, are filled; therefore, (7) is valid not only for desorption problems but for sorption problems as well.

Note again that knowledge of the sorption or desorption isotherms (depending on the process being investigated) is necessary not only for relation (11) but also for determining the interphase mass-transfer intensity and the specific heat of sorption. Therefore, reliable experimentally verified information on the dependence of the equilibrium moisture content on the vapor pressure and temperature is needed. There are many methods for finding sorption isotherms; however, the problem of obtaining them at vapor pressures close to the saturation vapor pressure in a wide temperature range remains. In this case, it is more correct to call sorption isotherms moisture-content (or water-yielding capacity) isotherms, and to obtain them at room temperature the method of contact standard porometry [31] can be recommended. Determining by the mercury porometry methods the dependence $u_e = f(p_{\text{liq}})$ for the standard and using the Kelvin relation, we can calculate the sorption isotherm. Then, recording the equilibrium moisture content of the sample being investigated, which is in contact with the standard, using the methods of [31], we can obtain for the sample the dependence $u_e = f(p_v/p_s)$ at $\phi \rightarrow 1$.

The difficulty of using (11), (17) is connected not only with the determination of the sorption isotherms, but also with the expression of the pressure in terms of inverse functionals in an analytical form convenient for calculations and analysis, since otherwise it is necessary to use methods of polynomial interpretation of the table function of two variables with the use of numerical methods.

Note that the justification of using the Kelvin equation to determine the capillary pressure of a liquid was verified in a number of works, e.g., [11, 12], and, in terms of the above-considered assumptions, causes no doubt.

Nonequilibrium effects. Consider a medium with double porosity, e.g., a silica gel fill through which warm air passes. In such a granular fill, in the spaces between particles, large pores are formed. In the grains themselves micropores dominate. The radius of the macropores is much larger than the size of the micropores. Due to the fact that the structure of the medium is nonuniform, in the dehydration process the moisture content in its elementary volumes is different, and the characteristic time of its change may turn out to be comparable to the settling time of phase permeabilities and capillary pressure. In this case, the process is not equilibrium and, therefore, it is expedient to operate with the effective moisture content. In so doing, it may be assumed that in the nonequilibrium process the relative phase permeability, desorption heat, and liquid pressure functions of the true moisture content correspond to the same functions in the equilibrium process but of some effective moisture content.

To obtain expressions relating the effective and true moisture contents, it is assumed that their difference only depends on the local rate of change in the moisture content and the characteristic relaxation time. As a result, for their relation the dimensional analysis gives an elementary kinetic relation. The basic ideas of taking into account the nonequilibrium effects for filtration problems were formulated in [14, 15].

Of great importance is also the account for the nonequilibrium of the transfer processes in the gas phase upon phase transformations in the porous body. The investigations of the mass-transfer kinetics of gases in capillaries in the presence of phase transformations, moving absorbed layers, and a deep evaporation zone in the porous body are described in [20]. The nonequilibrium effects have been analyzed on the basis of the solution of model kinetic equations describing the flow of a binary vapor-gas mixture in a cylindrical capillary in which evaporation of the fill occurs. A similar kinetic mass-transfer problem in gas filtration and in the presence of a deep evaporation zone was also considered for the model of a "dusty gas," i.e., when a high-porosity body is modeled by a system of randomly distributed spherical particles.

In intensive heat-and-mass transfer processes, the local thermodynamic equilibrium condition may not be fulfilled when the time of change in the thermodynamic parameters in physically small volumes is shorter than their relaxation time. In a rapid change in the vapor-gas mixture parameters in the porous body the vapor pressure in its pores can differ considerably from its equilibrium value with the body in the elementary volumes. In our case, it seems to be expedient to take into account the nonequilibrium state of the regime of heat and mass transfer on the basis of the phase transformation (sorption-desorption) kinetic equation, which can be given in the form

$$I = \frac{da}{d\tau} = \gamma(\varphi - \varphi_e) = \frac{\gamma}{p_s} (p_v - p_e), \quad (18)$$

where p_e is the equilibrium partial vapor pressure determined from the desorption isotherm.

When the moisture content of the body is higher than the maximum hygroscopic content, the equilibrium vapor pressure approaches the saturated vapor pressure $p_e = p_s$ and Eqs. (18) in some approximation will be written as

$$I = \frac{da}{d\tau} = \gamma(\varphi - 1). \quad (19)$$

Under equilibrium conditions, $a = u_e$ and, consequently, (18) takes on the form of (12).

Heat and Mass Transfer in the Drying Processes of Thermolabile Materials at Time-Periodic Absorption of Heat. To upgrade the quality of thermolabile materials, one uses in the process of their drying a time-discrete heat supply such as a periodic change in the temperature of the heat-transfer material (oscillating regime), heating with material circulation and intermediate cooling, intermittent irradiation, etc. Among the thermolabile materials are foods, raw cotton, paper, etc. In many works, the interrelated heat and mass transfer at a discrete heat supply is described by the A. V. Luikov system of equations where the temperature of the heat-transfer materials is a periodic time function. In most cases, however, the pure temperature problem is solved with allowance for the heating and cooling periodicity but ignoring the influence on the mass-transfer process.

Let us investigate the drying process of a cylindrical sample of K-120 cellulose paper, whose properties were determined in [32], of radius $R = 0.02$ m. At the initial instant of time at $\tau = 0$ the initial moisture content $\tilde{u}_0 = 0.06$, the temperature $T_0 = 293$ K, and the vapor pressure inside the pores determined by the desorption isotherm $p_{v0} = 1052$ Pa. The sample is placed in a vacuum chamber with a vapor pressure $p_{en} = 13.1$ Pa. Its surface is subjected to the action of a radiant flux from a radiator with a cyclically varying temperature. The total time of the drying process $Fo = 2.25$; the cycle time $Fo_c = 2 Fo/n$, $m = 1, 2, 3, \dots, n$. If the number m is odd, then the radiator temperature $T_{rad} = 420$ K, but if m is even, then $T_{rad} = 293$ K. We are to find the distributions of the vapor pressure fields, the moisture content, and the temperature for various instants of time. According to the data of [33], the vapor permeability is a constant, up to 10^3 Pa. In the calculations, we assume $D_v^* = K_v \rho_v / \eta_v = 5 \cdot 10^{-11}$ sec.

Direct measurements of the diffusion coefficients of a liquid are absent. Therefore, for tentative estimations, we use the vapor permeability $K_v \approx K_{liq}$; then $D_{liq}^* = K_{liq} \rho_{liq} / \eta_{liq} = 10^{-16}$ sec. We calculate the relative phase permeability of the liquid with some approximation by the S. V. Averiyarov formula

$$f_{liq}(\theta_{liq}) = [(\theta_{liq} - \theta_{liq}^*) / (1 - \theta_{liq}^*)]^{3.5}.$$

The limiting saturation θ_{liq}^* corresponding to the value of the limiting moisture content $\tilde{u} = 0.04$ when the liquid is in the bound, stationary state was measured by the nuclear magnetic resonance (NMR) method. The self-diffusion coeffi-

cient was also measured by the NMR method [29]. Calculations were made for paper with a desorption isotherm of the form [32]

$$u_e = A \exp(-\alpha T) (\beta p_v)^{-\chi + \psi T}, \quad (20)$$

where $A = 367,935$, $\alpha = 0.033$, $\beta = 0.0076$, $\chi = 0.96$, and $\psi = 0.05$. In the cylindrical coordinates, the vapor, liquid, and energy transfer equations are of the form

$$m \frac{\partial (\rho_v \theta_v)}{\partial \tau} = D_v^* \frac{1}{r} \frac{\partial}{\partial r} \left(f_v(\theta_v) r \frac{\partial p_v}{\partial r} \right) + I, \quad (21)$$

$$m \frac{\partial (\rho_{liq} \theta_{liq})}{\partial \tau} = D_{liq}^* \frac{1}{r} \frac{\partial}{\partial r} \left(f_{liq}(\theta_{liq}) r \frac{\partial p_{liq}}{\partial r} \right) - I, \quad (22)$$

$$\left[(1-m) c_{p_{sol}} \rho_{sol} + m (\theta_v \rho_v c_{p_v} + \theta_{liq} \rho_{liq} c_{p_{liq}}) \right] \frac{\partial T}{\partial \tau} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \lambda_{eff} \frac{\partial T}{\partial r} \right) + r_{ev} I - m \left(p_{liq} - T \left(\frac{\partial p_{liq}}{\partial T} \right)_{\theta_{liq}} \right) \frac{\partial \theta_{liq}}{\partial \tau}. \quad (23)$$

Detailed derivation of (23) with allowance for the additional heat as a result of the change in the saturation of the porous medium with moisture was carried out in [29]. Determining p_v from (20) in explicit form and using the Clapeyron–Clausius and Kelvin formulas, we get expressions for $r_{ev}(u)$ and $p_{liq}(u)$:

$$r_{ev} = \frac{\tilde{R}T^2}{v_{liq}'} \left[\frac{\alpha - \psi \ln \beta}{-\chi + \psi T} + \frac{\ln(u/A) + \alpha T + \chi - \psi T}{(-\chi + \psi T)^2} \right], \quad (24)$$

$$p_{liq} = \frac{\tilde{R}Tu}{v_{liq}''} \left[\frac{\ln(u/A) + \alpha T}{-\chi + \psi T} - \ln(\beta \tilde{C}) - 15 \ln \left(\frac{T}{373} \right) \right], \quad (25)$$

where $\tilde{C} = 10^5$ Pa.

The saturated vapor pressure was calculated by the formula

$$p_s = \tilde{C} \left(\frac{T}{373} \right)^{15}. \quad (26)$$

The source term is determined by expression (12). The boundary conditions are

$$\tau = 0: T(r, 0) = T_0, \quad p_v(r, 0) = p_{v0}, \quad p_{liq}(r, 0) = p_{liq0};$$

$$r \equiv \frac{r}{R} = 0: \left. \frac{\partial p_v}{\partial r} \right|_{r=0} = \left. \frac{\partial p_{liq}}{\partial r} \right|_{r=0} = 0, \quad \left. \frac{\partial T}{\partial r} \right|_{r=0} = 0;$$

$$r = 1: p_v(1, \tau) = p_{en}, \quad p_{liq}(1, \tau) = p_s(T) + \frac{\tilde{R}T_{en}(\tau)}{v_{liq}''} \ln \left(\frac{p_{en}}{p_s(T)} \right),$$

$$\lambda_{eff} \left. \frac{\partial T}{\partial r} \right|_{r=1} = \sigma \varepsilon (T_{rad}^4(\tau) - T^4(1, \tau)), \quad (27)$$

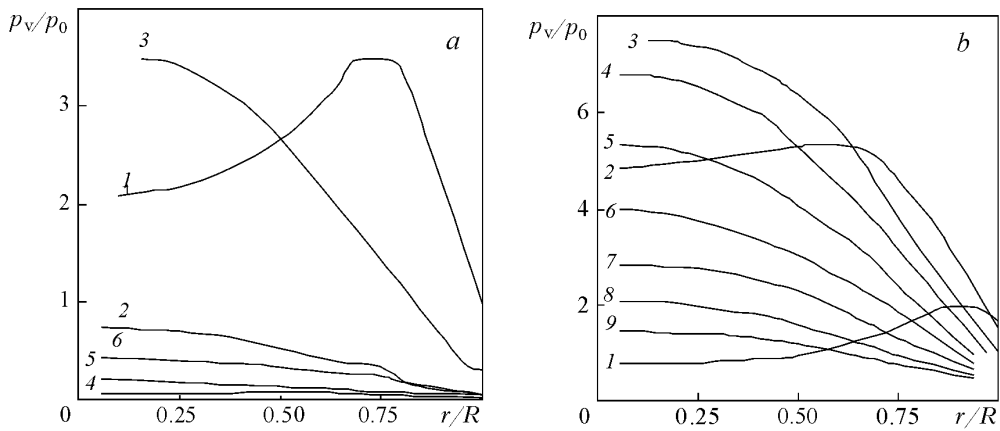


Fig. 1. Vapor pressure fields for various instants of time: a) discrete heat supply at $n = 10$; b) continuous heat supply. The curve number indicates the time for corresponding \tilde{k} .

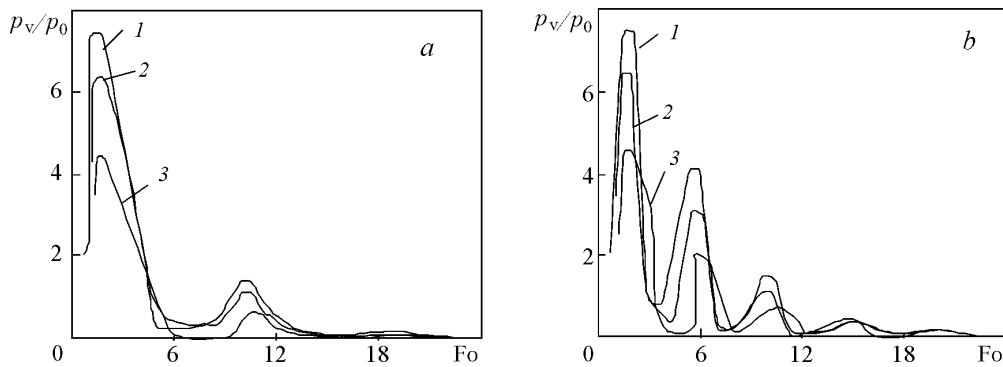


Fig. 2. Vapor pressure at three points of the sample as a function of time. Discrete heating: a) $n = 6$; b) 10.

where $\begin{cases} T_{\text{rad}} = 420 \text{ K} & \text{at } m = 2k + 1; \\ T_{\text{rad}} = 293 \text{ K} & \text{at } m = 2k, \quad k = 0, 1, \dots \end{cases}$

The system of equations (21)–(27) was solved by the method of successive approximations with iterations, and for the energy and vapor equations thereby we used numerical schemes in the Euler representation and for the flow equation — Lagrange variables [29].

Note that the equations of vapor and liquid filtration (21) and (22) are derived from the Navier–Stokes equations in considering the flow through a thin slotlike pore where the friction forces considerably exceed the inertial components. At the same time, in some cases it is necessary to take into account the convective component in the heat-conduction equation (23), the estimation method of whose contribution is described in [14]. In our case, its contribution is insignificant compared to the conductive component.

The calculation of the drying kinetics was made up to $\text{Fo} = 22.5$. We registered the values of the functions for nine instants of time ($\text{Fo} = 22.5\tilde{k}/9$, $\tilde{k} = 1, 2, \dots, 9$) in order to follow the evolution of the local values of the parameters.

From Fig. 1a it is seen that initially the vapor pressure in the section $r/R = 0.75$ has a characteristic wavy projection (curve 1) and then decreases (curve 2). At some instant of time (curve 3) the vapor pressure reaches the maximum value of $3.5p_0$, and at $\text{Fo} = 10$ (curve 4) it is already close to the equilibrium value. However, at subsequent instants (curves 5, 6) the vapor pressure somewhat increases again, but the maximum pressure is lower than $p_{\text{max}} = 0.4p_0$ and then (curves 7, 8, 9 are not given) relaxation to the equilibrium pressure occurs. For comparison, Fig. 1b gives the vapor pressure fields at a continuous operation. It is seen that the vapor pressure increases wave-like (curves 1, 2, 3) to $7.5p_0$, which is twice as high as the maximum value of the vapor pressure (see Fig. 1a).

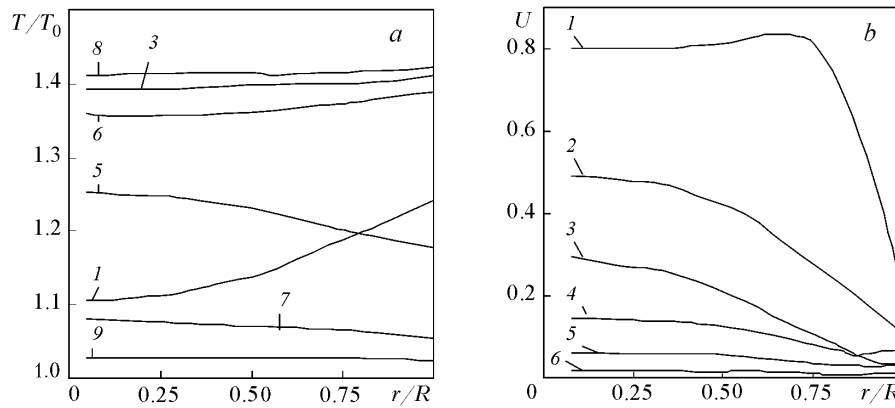


Fig. 3. Temperature (a) and moisture-content (b) fields in the sample cross section for various instants of time. Discrete heating at $n = 10$. The curve number indicates the time for the corresponding \tilde{k} .

It should be emphasized that comparison of the curves in Fig. 1a and b is made for the corresponding instants of time (curves 1–9); therefore, the pressure peaks should not necessarily fall exactly on them. Figure 2 shows the local vapor pressures as a function of time for three points at a discrete heat supply: 1 — center, 2 — middle, 3 — near-surface layer. From Fig. 2a it is seen ($n = 6$) that the maximum vapor pressure is $7.45p_0$ and is reached at time $Fo = 1.8$. Then the vapor pressure decreases to $\sim 0.3p_0$, but at $Fo = 10$ there arises the next peak of the maximum vapor pressure $1.3p_0$, and so on. Wave-like relaxation of the vapor pressure with decreasing amplitude occurs. Note that the number of pressure peaks is determined by the number of sample heating cycles and is half of the number of switches. Figure 2b gives the local vapor-pressure fields at three points as a function of time for $n = 10$.

Figure 3 shows the temperature and moisture-content fields at various instants of time at a discrete heat supply (curves 2, 4 (a) and 7, 8, 9 (b) are not shown). From their analysis it follows that the sample temperature changes cyclically, its periodic rise and fall occurs, and the temperature gradient in the sample thickness thereby is much lower than in a continuous heating.

Thus, note that the drying of thermolabile materials at a discrete heat supply is followed by the oscillation of not only the temperature but the vapor pressure as well, and the vapor pressure and moisture-content gradients are much smaller than at a continuous heat supply.

Nonisothermal Mass Transfer in Porous Media. Consider a soil which is a mixture of sand and peat with a desorption isotherm of the form

$$u_e = A \exp(-\alpha T) p_v - B \exp(\beta T). \quad (28)$$

At $\alpha = \text{const}$ and $\beta = \text{const}$ the coefficients A and B are, respectively, in the range $A = (1-3) \cdot 10^5$ and $B = 4-5$ and reflect the experimental data of the selected soils.

The soil contains an infinite cylinder on whose surface a constant temperature T_{en} is maintained. We are to find the time evolution of the moisture content and temperature fields. For the system of equations (21)–(23) the boundary conditions are of the following form:

$$\begin{aligned} \tau = 0: \quad & T(r, 0) = T_0, \quad p_v(r, 0) = p_{v0}, \quad p_{liq}(r, 0) = p_{liq0}; \\ r = \infty: \quad & \left. \frac{\partial T}{\partial r} \right|_{r=\infty} = 0, \quad \left. \frac{\partial p_v}{\partial r} \right|_{r=\infty} = 0, \quad \left. \frac{\partial p_{liq}}{\partial r} \right|_{r=\infty} = 0; \\ r = R: \quad & T(R, \tau) = T_{en}, \quad D_{liq}^* \left. \frac{\partial p_{liq}}{\partial r} \right|_{r=R} = D_v^* \left. \frac{\partial p_v}{\partial r} \right|_{r=R}. \end{aligned} \quad (29)$$

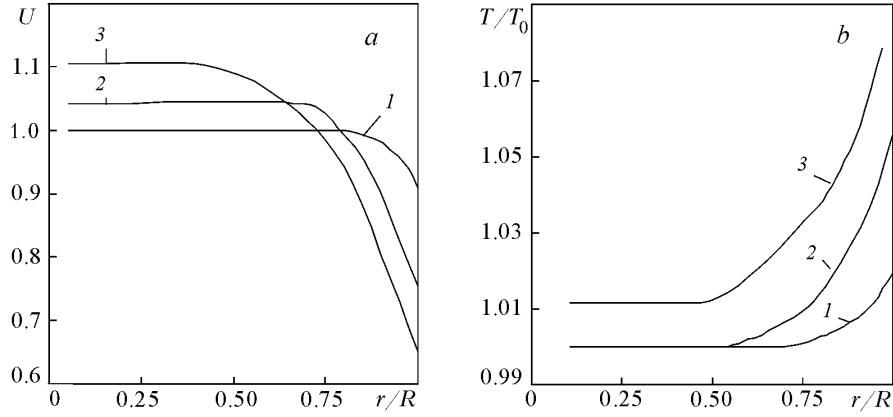


Fig. 4. Moisture-content (a) and temperature (b) field distribution for various instants of time. 1) $\tau = 1.5$; 2) 4.5; 3) 13.5 h.

Figure 4 shows the results of the calculations of the moisture content and temperature distribution for a soil with $D_{\text{liq}}^* = 0.5 \cdot 10^{-5}$ sec and $D_v^* = 0.5 \cdot 10^{-9}$ sec.

Because of the heat-moisture conduction phenomenon, there is an increase in the moisture content at "colder" points of the soil. The calculations have shown that the heat-moisture conduction intensity is strongly influenced by the temperature and radius of the heat source, the initial moisture content, and the thermophysical properties of the soil. The results obtained are in good agreement with the experimental data for an aggregated heavy-loamy soil [34].

The heat-moisture conductivity is usually determined experimentally from the analysis of the stationary moisture distribution in the stationary temperature field. The thus-obtained coefficient is difficult to use in calculations, since it is the result of the interrelated heat and mass transfer in porous media.

Consider now a porous medium, e.g., a flat sample of K-120 insulating paper, with a small initial moisture content $u_0 = 65.73 \text{ kg/m}^3$ at $T = 293 \text{ K}$. In this case, according to the sorption isotherm (20), the intraporous pressure $p_{v0} = 1052 \text{ Pa}$. The sample is encased in a vapor- and moisture-proof sheath. We are to find the stationary moisture distribution in the sample thickness in a known stationary temperature field.

Let the temperature field vary as the linear dependence

$$T(x) = T_1 + \xi x,$$

where $\xi = (T_2 - T_1)/l$.

At low moistures the moisture transfer in the form of a liquid can be ignored [29]. In this case, in the stationary state under insulation conditions it may be assumed that $p_v = \text{const}$ throughout the sample thickness. To determine the unknown vapor pressure, we use the fact that the total amount of moisture before the beginning and at the end of the experiment is a constant, i.e., the mass balance equation is of the form

$$\int_0^l u_0 dx = \int_0^l A \exp(-\alpha T(x)) (\beta p_v)^{-\chi + \psi T(x)} dx, \quad (30)$$

where $A = 367,935$, $\alpha = 0.0033$, $\beta = 0.0076$, $\chi = 0.96$, and $\psi = 0.005$ are constants in the sorption isotherm (20).

Introducing the designation $b = \beta p_v$ and using the substitution $b^x \exp(x \ln b)$ for a sample with thickness $l = 1 \text{ m}$, we obtain

$$A \exp(-\alpha T_1) b^{-\chi + \psi T_1} \int_0^l \exp(-\alpha \xi x) \exp(x \ln b^{\psi \xi}) dx = u_0, \quad (31)$$

and after integration

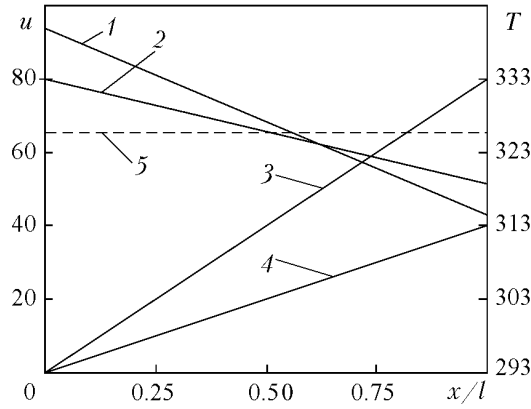


Fig. 5. Stationary sample-thickness distribution of moisture (1, 2) and temperature (3, 4) at low humidity under stationary conditions: 1, 3) $\Delta T = 40$ K; 2, 4) 20; 5) u_0 .

$$\frac{A \exp(-\alpha T_1) b^{-\chi + \psi T_1} \left\{ \exp[\xi(-\alpha + \psi \ln b)] - 1 \right\}}{\xi(-\alpha + \psi \ln b)} = u_0. \quad (32)$$

We are to determine the constant from the solution of the following equation:

$$A \exp(-\alpha T_1) b^{-\chi + \psi T_1} [b^{\xi \psi} \exp(-\alpha \xi) - 1] = u_0 \xi(-\alpha + \psi \ln b). \quad (33)$$

For the sample of thickness $l = 1$ and $\Delta T = T_2 - T_1 = 40$ K, the transcendental equation is of the form

$$23.25b^{0.5} (0.267b^{0.2} - 1) = -86.77 + 13.48 \ln b. \quad (34)$$

Having solved (34), we obtain $b = 16.25$ and $p_v = 2144$ Pa. Substituting the found pressure value into the sorption isotherm equation, we obtain

$$u(x) = A \exp(-\alpha T(x)) (2144\beta)^{-\chi + \psi T(x)}. \quad (35)$$

Figure 5 shows the moisture (curve 1) and temperature (curve 3) distributions as a function of the x/l coordinate. The stationary moisture distribution depends on not only the initial moisture and temperature but the temperature gradient as well. At the same thickness l and $\Delta T = 20$ K the transcendental equation (33) is of the form

$$23.25b^{0.5} (0.516b^{0.1} - 1) = -43.85 + 6.74 \ln b. \quad (36)$$

The root of Eq. (36) is $b = 11.7$, which corresponds to $p_v = 1539.3$ Pa. Under these conditions, moisture (curve 2) and temperature (curve 4) distributions take place. It is seen from Fig. 5 that as a result of the vapor recondensation the moisture content is higher in the "cold" parts of the sample. Usually the steady moisture distribution in the stationary temperature field is found from experimental studies. Knowing the moisture distribution in the temperature field under stationary conditions, the heat-moisture conductivity coefficient $\delta' = (\Delta \tilde{u} / \Delta T)_{\tilde{u}}$ is calculated and used in the general heat and mass transfer equations. The difficulty of modeling with such an approach is due to the fact that δ' strongly depends on the process parameters.

Investigation of the Heat and Mass Transfer with Allowance for the Nonequilibrium Effect. Consider the drying process of a spherical bentonite particle in a gas flow with a constant temperature T_{en} and heat-transfer α^* and mass-transfer coefficients β_1^* and β_2^* (or in an isothermal stationary or moving layer with analogous effective coefficients). It is assumed that the dry air pressure is constant and the heat transfer due to the molar liquid and vapor transfer can be neglected. Then the heat and mass transfer equations in the spherical coordinate system will be written as follows [35, 36]:

$$\left[(1-m) c_{p_{\text{sol}}} \rho_{\text{sol}} + \tilde{u} c_{p_{\text{liq}}} \rho_{\text{sol}} \right] \frac{\partial T}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \lambda_{\text{eff}} \frac{\partial T}{\partial r} \right) + r_{\text{ev}} I, \quad (37)$$

$$a_v^* \frac{\partial p_v}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{K \rho_v \dot{f}_v(\tilde{u})}{\eta_v} \frac{\partial p_v}{\partial r} \right) - I, \quad (38)$$

$$\rho_{\text{sol}} \frac{\partial \tilde{u}}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{K \rho_{\text{liq}} \dot{f}_{\text{liq}}(\tilde{u})}{\eta_{\text{liq}}} \left(\frac{\partial p_{\text{liq}}}{\partial \tilde{u}} \right)_T \frac{\partial \tilde{u}}{\partial r} \right) + I, \quad (39)$$

where

$$a_v^* = a_v \frac{v_v''}{RT}.$$

The initial conditions are

$$\tau = 0 : T(r, 0) = T_0, \quad p_v(r, 0) = p_{v0}, \quad \tilde{u}(r, 0) = \tilde{u}_0. \quad (40)$$

The boundary conditions are

$$\begin{aligned} \left. \frac{\partial p_v}{\partial r} \right|_{r=0} &= 0, \quad \left. \frac{\partial T}{\partial r} \right|_{r=0} = 0, \quad \left. \frac{\partial \tilde{u}}{\partial r} \right|_{r=0} = 0, \\ \lambda_{\text{eff}} \left. \frac{\partial T}{\partial r} \right|_{r=R} &= \alpha^* (T_{\text{en}} - T), \\ \frac{K \rho_v \dot{f}_v}{\eta_v} \left. \frac{\partial p_v}{\partial r} \right|_{r=R} &= \beta_1^* (p_{\text{en}} - p_v), \\ \frac{K \rho_{\text{liq}} \dot{f}_{\text{liq}}}{\eta_{\text{liq}}} \left(\frac{\partial p_{\text{liq}}}{\partial \tilde{u}} \right)_T \left. \frac{\partial \tilde{u}}{\partial r} \right|_{r=R} &= \beta_2^* (\tilde{u}_e - \tilde{u}). \end{aligned} \quad (41)$$

In the numerical experiment, we varied the parameters in order to study the laws of internal heat and mass transfer. The closing relations can be determined, knowing the desorption isotherms for the whole temperature range in the drying process. To describe the bentonite desorption isotherms, one uses the approximation formula [37]

$$\tilde{u}_e = \tilde{u}_{0.5}(T) \left(\frac{p_v}{p_s - p_v} \right)^{1/\omega}, \quad (42)$$

where $\tilde{u}_{0.5}(T)$ is the temperature dependence of the equilibrium moisture content at $\varphi = p_v/p_s = 0.5$. The index ω is constant for one and the same material.

As a result of the experimental data approximation, we obtained

$$\tilde{u}_{0.5}(T) = 0.023 \exp[-0.007(T - 293)], \quad \omega = 3. \quad (43)$$

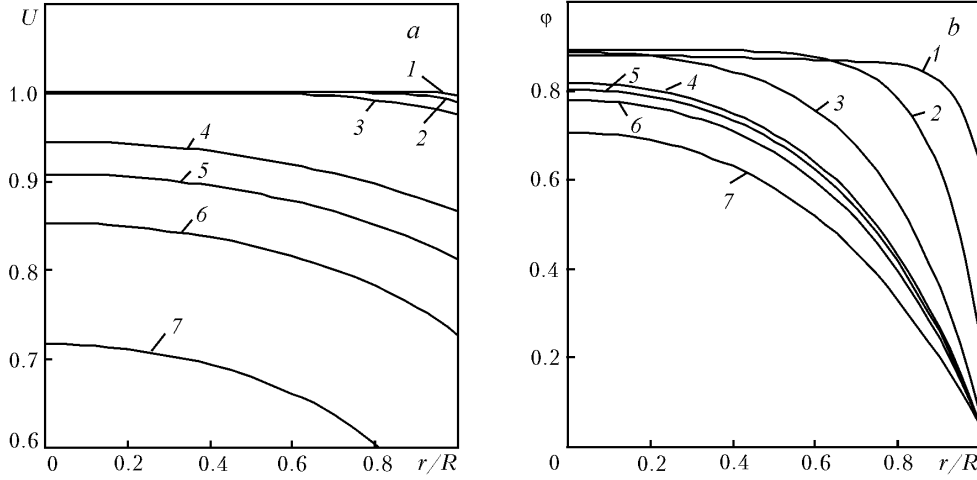


Fig. 6. Moisture content (a) and relative humidity of the medium (b) as a function of the radial coordinate r/R for nonequilibrium conditions ($\gamma = 0.005$ kg/(m³·sec); $\delta = 6$; $K = 1 \cdot 10^{-17}$ m²): 1) $\tau = 2.58$; 2) 32.8; 3) 128; 4) 1230; 5) 1800; 6) 2730; 7) 5600 sec.

Expression (17) for the specific heat of the moisture phase transition in view of (42) will be written as

$$r_{ev} = \frac{\tilde{R}T^2}{v'_v} \left[\frac{15}{T} - \frac{(\omega - 1) (\tilde{u}_{0.5}(T))^{\omega-1} \frac{\partial \tilde{u}_{0.5}}{\partial T}}{\tilde{u}^\omega + \tilde{u}_{0.5}^\omega} \right], \quad (44)$$

where $\frac{\partial \tilde{u}_{0.5}}{\partial T} = -0.007 \tilde{u}_{0.5}(T)$.

The saturated vapor pressure as a function of temperature is determined by the approximated formula (26). The derivative in expression (39) in view of (6) and (42) is of the form

$$\left(\frac{\partial p_{liq}}{\partial \tilde{u}} \right)_T = \frac{\tilde{R}T}{v''_{liq}} \frac{1}{\omega \tilde{u}} \frac{\tilde{u}_{0.5}^{1/\omega} + 2\tilde{u}^{1/\omega}}{\tilde{u}_{0.5}^{1/\omega} + \tilde{u}^{1/\omega}}. \quad (45)$$

The liquid pressure was calculated by formula (7).

The calculations were made at the following constant parameters: $\tilde{u}_0 = 0.05$ kg/kg, $T_0 = 293$ K; $\rho_{sol} = 1500$ kg/m³; $m = 0.3$; $c_{p_{liq}} = 4180$ J/(kg·K); $\tilde{R}/v'_{liq} = 4.6 \cdot 10^5$ J/(m³·K); $\tilde{R}/v'_v = 461.6$ J/(kg·K); $\alpha^* = 30$ W/(m²·K); $\beta_1^* = 1 \cdot 10^{-5}$ kg/(m²·sec·Pa); $\beta_2^* = 1.5 \cdot 10^{-4}$ kg/(m²·sec); $p_{en} = 2000$ Pa; $T_{en} = 353$ K; $R = 5 \cdot 10^{-3}$ m; $\eta_{liq} = 0.55 \cdot 10^{-3}$ Pa·sec; $\eta_v = 1.6 \cdot 10^{-5}$ Pa·sec.

It was assumed that the particle surface is subjected to the action of a thermal flow uniformly distributed on the sphere surface. The material porosity was taken to be constant. In the calculations, we took into account the moisture-content dependences of the relative permeability by the formulas

$$f_v^\circ(\tilde{u}) = \left(\frac{\tilde{u}_{max} - \tilde{u}}{\tilde{u}_{max} - \tilde{u}_{min}} \right)^\delta, \quad f_{liq}^\circ(\tilde{u}) = \left(\frac{\tilde{u} - \tilde{u}_{min}}{\tilde{u}_{max} - \tilde{u}_{min}} \right)^\delta, \quad (46)$$

where $\tilde{u}_{max} = 0.3$ kg/kg; $\tilde{u}_{min} = 0.001$ kg/kg.

The source term in Eqs. (37)–(39) was determined by expressions (18) and (12).

The numerical solution of the system of equations (37)–(39) was found by the iteration method with the use of the programs for nonlinear parabolic equations.

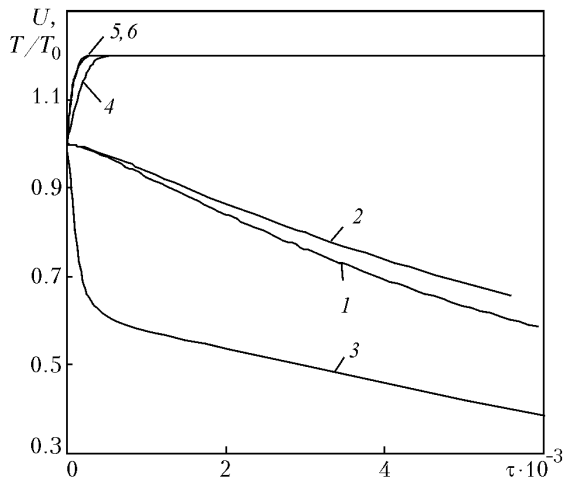


Fig. 7. Curves of material drying U (1–3) and heating T/T_0 (4–6) ($K = 1 \cdot 10^{-17} \text{ m}^2$; $\delta = 6$): 1, 6, $\gamma = 0.01 \text{ kg}/(\text{m}^3 \cdot \text{sec})$; 2, 5) 0.005; 3, 4) equilibrium conditions. τ , sec.

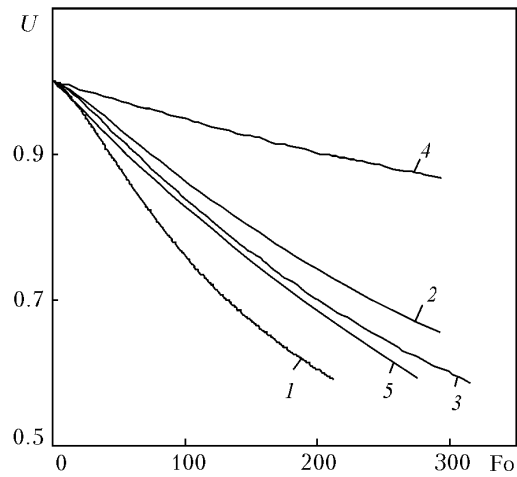


Fig. 8. Curves of material drying: 1) $\gamma = 0.05 \text{ kg}/(\text{m}^3 \cdot \text{sec})$; 2) 0.005; 3) 0.01 (for 1–3 $K = 1 \cdot 10^{-17} \text{ m}^2$; $\delta = 6$); 4) $\gamma = 0.005$ ($K = 1 \cdot 10^{-18} \text{ m}^2$; $\delta = 6$); 5) $\gamma = 0.005$ ($K = 1 \cdot 10^{-18} \text{ m}^2$; $\delta = 4$).

Figure 6 shows the moisture content and relative humidity of a vapor-air medium as a function of the radial coordinate of the particle for the nonequilibrium regime of moisture desorption where the source term was calculated by formula (18) at $\gamma = 0.005$. They have a monotonic parabolic form and equalize with time as the moisture is removed.

Figure 7 shows the kinetic curves of the material drying and heating for nonequilibrium conditions with a different value of the kinetic coefficient and equilibrium conditions, for which the source term was calculated by expression (12). All the other parameters remained unaltered. From Fig. 7 it is seen that the chosen material is warmed up fairly quickly and the subsequent drying process proceeds at a practically constant temperature. Apparently, with increasing coefficient γ the drying intensity increases. As would be expected, in the warming-up period the moisture content of the material changes only slightly, and on the kinetic curves of drying this period is observed, which is more clearly seen in Fig. 8. The equilibrium regime leads in the warming-up period to a sharp decrease in the moisture content, which is due to the decrease in its equilibrium value with increasing temperature. Therefore, the equilibrium regime of moisture desorption (determination of the source term by formula (12)) appears to be not always good, as in this case, and can obviously be used for slow heat and mass transfer processes.

For the nonequilibrium regime of moisture desorption, an increase in the kinetic coefficient leads to an increase in the relative humidity of the medium and in the vapor pressure inside the particle and to an increase in the vapor pressure and moisture-content gradients in the near-surface zone of the particle. As a result, the drying process is speeded up. The drying kinetic curves given in Fig. 8 are indicative of a retardation of the heat and mass transfer process with decreasing permeability coefficient K and increasing exponent δ determining the relative permeability function. This is due to the increase in the vapor and liquid transfer resistance.

CONCLUSIONS

As a result, it may be concluded that the heat and mass transfer in a porous medium cannot be adequately described by the diffusion equations without taking into account the physics and thermodynamics of the surface phenomena.

The proposed physical model of the nonstationary processes of interrelated heat and mass transfer of a single fluid takes into account the mutual influence of the vapor and liquid pressures determined by the contribution of the capillary and surface forces and temperature on the interphase mass-transfer intensity and the mechanical and dynamic equilibrium conditions of thin layers of liquids on curved interfaces.

The system of differential equations formulated on the basis of the proposed physical model, which relies on the mass and energy conservation laws for each phase, the salient points of the classical theory of two-phase filtration, the Kelvin and Clapeyron–Clausius formulas, and the family of sorption isotherms permits describing, from a single point of view, the drying, sorption, and filtration processes in incomplete saturation.

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NOTATION

A and B , constants in the sorption isotherm; a , amount of sorbed (desorbed) moisture in the nonequilibrium process, kg/m^3 ; C , concentration, kg/kg ; c_p , specific heat, $\text{J}/(\text{kg}\cdot\text{K})$; D , diffusion coefficient, m^2/sec ; $D^* = K\rho/\eta$; $f(\theta)$ and $f^0(\tilde{u})$, relative phase permeabilities, respectively, in the saturation and moisture-content functions; I , source term, $\text{kg}/(\text{m}^3\cdot\text{sec})$; j , moisture flow, $\text{kg}/(\text{m}^2\cdot\text{sec})$; K , permeability coefficient of the porous body, m^2 ; l , linear dimension, m ; m , porosity of the material; n , number of switches; p , pressure, Pa ; R , radius of pores, particle (of the sample), m ; \tilde{R} , universal gas constant, $\text{J}/(\text{mole}\cdot\text{K})$; r , radial coordinate, m ; r'_{ev} and r_{ev} , specific heat of evaporation of bound moisture, J/mole and J/kg ; S , entropy, $\text{J}/(\text{mole}\cdot\text{kg})$; T , temperature, K ; T_1 and T_2 , temperatures on boundary surfaces, K ; u and \tilde{u} , moisture content, kg/m^3 and kg/kg ; $U = u/u_0 = \tilde{u}/\tilde{u}_0$; x , current coordinate, m ; α , β , δ , χ , ξ , ψ , ω , constants; α^* , heat-transfer coefficient, $\text{W}/(\text{m}^2\cdot\text{K})$; β_1^* and β_2^* , mass-transfer coefficients assigned, respectively, to the vapor pressure and the moisture content, $\text{kg}/(\text{m}^2\cdot\text{sec}\cdot\text{Pa})$ and $\text{kg}/(\text{m}^2\cdot\text{sec})$; γ , kinetic desorption coefficient, $\text{kg}/(\text{m}^3\cdot\text{sec})$; θ , saturation of porous material; θ^* , limiting saturation; δ' , heat and moisture conductivity coefficients, $1/\text{K}$; ϵ , emissivity of material; λ , heat-conductivity coefficient, $\text{W}/(\text{m}\cdot\text{K})$; η , coefficient of dynamic viscosity, $\text{Pa}\cdot\text{sec}$; μ , chemical potential, J/mole ; v' , molecular mass, kg/mole ; v'' , molar volume, m^3/mole ; ρ , density, kg/m^3 ; σ , Stefan–Boltzmann constant; τ , time, sec ; ϕ , relative vapor pressure; $\text{Fo} = D\tau/R^2$, Fourier number. Subscripts: 0, parameters of the initial state; c, cycle; e, equilibrium; eff, effective parameters; en, environment parameters; ev, evaporation; liq, liquid; max, maximum; min, minimum; r, relaxation parameters; rad, radiation; s, parameters of the saturated-state phase; sol, solid body parameters; v, vapor.

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