MODELING OF HEAT AND MASS TRANSFER IN CAPILLARY-POROUS MATERIALS

P. V. Akulich and N. N. Grinchik   UDC 66.047

A system of differential equations for heat and mass transfer in capillary-porous materials in a hydroscopic moisture-content field is derived with allowance for kinetics of moisture desorption, two-phase filtration, and liquid pressure determined by the action of surface forces. An analysis is made of the results of a computational experiment illustrating evolution of the moisture content, temperature, and vapor pressure fields against the material characteristics and kinetic coefficient.

Introduction. Heat and mass transfer in capillary-porous media has attracted the attention of many researchers owing to the considerable scientific and practical importance of these processes.

To describe moisture transfer in capillary-porous bodies, A. V. Luikov has introduced the unified potential of mass transfer $\theta_{m.t}$ by analogy with the thermal potential (temperature) [1, 2]. To construct a scale of the potential $\theta_{m.t}$ and determine the moisture content as a function of $\theta_{m.t}$, a potentiometric method has been developed, which consists in measuring the specific moisture content of an investigated capillary-porous body as a function of the moisture content of the reference body, both being in direct contact in the state of thermodynamical equilibrium. The dependences $u = f(\theta_{m.t})$ are rather different in form (concave, convex, flat, etc.).

In [3], it is shown with the aid of the method of measuring the equilibrium curves $u = f(u_{ref})$ (u_{ref} is the specific moisture content of the reference body) and the method of mercury porosimetry that the function of pore distribution with respect to capillary pressure is the governing factor of moisture distribution in a porous body or in a system of contacting porous bodies that are in the equilibrium state. An analysis of experimental studies of the mechanisms of mass transfer in the drying of the model [4] and real materials, including those with the use of radioactive tracers [5], made by these authors testifies to the decisive influence of capillary and surface forces on mass transfer and the drying rate.

In some works [6-9], the process of mass transfer in drying has been accounted for by driving forces determined by pressure gradients of the vapor and liquid (or capillary pressure) with use of structural characteristics of capillary-porous materials.

Mathematical Model. We will consider a nondeformable capillary-porous body. The investigated system consists of a skeleton of the capillary-porous body, liquid and moist air representing a binary vapor-gas mixture in the body capillaries.

The heat-transfer equation can be written in the form

$$[(1 - m) c_s \rho_s + a_v \rho_m c_m + u \rho_s c_{liq}] \frac{\partial T}{\partial \tau} = \text{div} (\lambda_{ef} \nabla T) - c_m \rho_m V_m \nabla T - c_{liq} \rho_{liq} V_{liq} \nabla T + LI . \qquad (1)$$

The first term on the right-hand side of the equation takes into account heat transfer by heat conduction, the second and third terms — heat transfer due to molar transfer of a liquid and a vapor-gas mixture, the fourth term — phase changes. It has been assumed that the vapor in capillaries is in thermal equilibrium with the liquid [6].

In [1], a system of equations is proposed for heat and mass transfer in capillary-porous media involving phase changes. However, the system is not closed since the introduced criterion of a phase transition in the source term is not determined. The system of equations obtained in [9] is closed with the aid of the sorption (desorption)

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isotherm and the hypothesis on an equilibrium regime (momentarily reaching the equilibrium state). However, this approach does not allow for the dynamics of a phase change (a nonequilibrium regime).

Let the sorption (desorption) isotherm $u_e = f(\varphi, T)$, where $\varphi = P_v/P_{sat}$, P_v is the partial vapor pressure (Pa), be known from experimental studies.

Note that when the pressure of the medium considerably differs from atmospheric pressure, it is necessary to know the isotherm as a function of the medium pressure $u_e = f(\varphi, T, P_m)$. Then the equation for vapor sorption (desorption) kinetics is

$$I = \frac{db}{d\tau} = \gamma \, (\varphi - \varphi_e) = \frac{\gamma}{P_{sat}} \, (P_v - P_e) \,, \tag{2}$$

where P_e is the equilibrium partial vapor pressure (Pa) determined from the isotherm.

In the case of the equilibrium sorption (desorption) conditions, $a = u_e \rho_s$; consequently,

$$I = \frac{db}{d\tau} = \frac{d \, [\rho_s u_e \, (\varphi, T)]}{d\tau} \,. \tag{3}$$

When the moisture content u of the body is higher than the maximum hygroscopic one, then the equilibrium vapor pressure approaches the saturated vapor pressure $P_e = P_{sat}$ and Eq. (2), in some approximation, can be written as

$$I = \frac{db}{d\tau} = \gamma \, (\varphi - 1) \,. \tag{4}$$

In this case, to determinate the kinetic coefficient, it is also necessary to carry out experimental studies.

To evaluate the specific heat of moisture evaporation of the material, we use the Clapeyron–Clausius equation

$$L = \frac{RT^2}{\nu_v} \frac{\partial \, (\ln P_v)}{\partial T} \,. \tag{5}$$

The vapor pressure P_v can be expressed from the equation of the desorption isotherm $P_v = F(u_e, T)$; then

$$L = \frac{RT^2}{\nu_v} \frac{\partial \, (\ln F \, (u_e, T))}{\partial T} \,. \tag{6}$$

This expression takes into account the amount of heat needed in drying to overcome the binding energy of the moisture with the material.

The continuity equations for a liquid and a vapor-gas mixture are written in the form

$$\frac{\partial \, (m \, \rho_{liq} \theta_{liq})}{\partial \tau} = - \nabla j_{liq} + I_1 \,. \tag{7}$$

$$\frac{\partial \, (m \, \rho_m \theta_m)}{\partial \tau} = - \nabla j_m + I_2 \,. \tag{8}$$

The motion of phases in a porous material under evaporation conditions is determined by the equations of two-phase filtration complicated by vapor diffusion in the gas phase:

$$V_{liq} = m\theta_{liq} v_{liq} = - \frac{k f_{liq} \, (\theta_{liq})}{\mu_{liq}} \nabla P_{liq} \,, \tag{9}$$

$$V_m = m\theta_m v_m = -\frac{k f_m(\theta_m)}{\mu_m} \nabla P_m,\tag{10}$$

where V_{liq}, V_m are the reduced phase velocities, m/sec.

For flows with an account of Eqs. (9) and (10), we can write

$$j_{liq} = \rho_{liq} V_{liq} = \rho_{liq} m\theta_{liq} v_{liq},\tag{11}$$

$$j_m = \rho_m V_m = \rho_m m\theta_m v_m.\tag{12}$$

Then Eqs. (7) and (8) are transformed into

$$\frac{\partial(m\rho_{liq}\theta_{liq})}{\partial\tau} = -\nabla(\rho_{liq}V_{liq}) + I_1.\tag{13}$$

$$\frac{\partial(m\rho_m\theta_m)}{\partial\tau} = -\nabla(\rho_m V_m) + I_2.\tag{14}$$

The equation of conservation is of the form

$$\frac{\partial(m\rho_v\theta_m)}{\partial\tau} = -\nabla(\rho_v V_m - D_v m\theta_m \rho_m \nabla(\rho_v/\rho_m) - \rho_v m\theta_m \chi\nabla T) + I_2,\tag{15}$$

where ρ_v is the reduced vapor density (vapor mass per unit volume of the mixture), kg/m^3.

The source terms are related by

$$I = I_1 = -I_2.\tag{16}$$

The equation of state of the ideal gas (vapor) is

$$P_v = \rho_v RT/v_v.\tag{17}$$

For saturations we can write

$$\theta_m + \theta_{liq} = 1.\tag{18}$$

Since, at the moist air pressure close to atmospheric pressure, the mass of vapor and air in capillaries is negligibly small compared to the liquid mass, it follows that

$$\omega \approx m\theta_{liq}\rho_{liq}, \quad u = \omega/\rho_s,\tag{19}$$

where ω is the volume concentration of moisture in the porous material, kg/m^3.

Now we transform the equations of conservation (13) and (14) to a more convenient form for solution. Substituting (9), (10), (17), (18), (19) into (13), (14) and neglecting small terms, we arrive, after some transformations, at

$$\frac{\partial(\rho_s u)}{\partial\tau} = \nabla\left(\frac{k\rho_{liq} f_{liq}(\theta_{liq})}{\mu_{liq}}\nabla P_{liq}\right) + I_1,\tag{20}$$

$$a_m \frac{\partial P_m}{\partial\tau} = \nabla\left(\frac{k\rho_m f_m(\theta_m)}{\mu_m}\nabla P_m\right) + I_2,\tag{21}$$

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$$\frac{\partial (a_v \rho_v)}{\partial \tau} = \nabla \left(\frac{k \rho_v f_m (\theta_m)}{\mu_m} \nabla P_m + D_v \rho_m a_v \nabla (\rho_v / \rho_m) + \rho_v a_v \chi \nabla T \right) + I_2, \tag{22}$$

where

$$a_m = \left(m - \frac{u \rho_s}{\rho_{liq}} \right) \frac{v_m}{RT}, \quad a_v = m - \frac{u \rho_s}{\rho_{liq}}, \tag{23}$$

$P_m = P_a + P_v$ is the vapor-gas mixture (moist air) pressure, Pa.

The second and third terms on the right-hand side of Eq. (22) indicate, respectively, the diffusional and thermodiffusional vapor transfer in air in capillaries (pores) of the body.

Equation (21) must be taken into consideration in the case of hydrodynamic (filtration) motion of the moist air, for instance, in filtration drying when a porous body is blown through by a drying agent. In the case where the content of dry air is insignificant compared to the amount of vapor, Eqs. (21) and (22) can be replaced by a single equation, such as Eq. (21) for the vapor pressure.

Instead of the functions $f_{liq}(\theta_{liq})$ and $f_m(\theta_m)$, we introduce the functions of relative permeability, expressed in terms of the moisture contents: $f'_{liq}(u)$ and $f'_m(u)$.

The liquid pressure is a function of the moisture content, temperature, and pressure of the mixture; therefore, the following relation is valid:

$$\nabla P_{liq} = \left(\frac{\partial P_{liq} (u, T, P_m)}{\partial u} \right)_{T, P_m} \nabla u + \left(\frac{\partial P_{liq} (u, T, P_m)}{\partial T} \right)_{u, P_m} \nabla T + \left(\frac{\partial P_{liq} (u, T, P_m)}{\partial P_m} \right)_{u, T} \nabla P_m. \tag{24}$$

Formulas (20), (21), and (22) can be written as

$$\rho_s \frac{\partial u}{\partial \tau} = \nabla \left(\frac{k \rho_{liq} f'_{liq} (u)}{\mu_{liq}} \left(\frac{\partial P_{liq}}{\partial u} \right)_{T, P_m} \nabla u + \frac{k \rho_{liq} f'_{liq} (u)}{\mu_{liq}} \left(\frac{\partial P_{liq}}{\partial T} \right)_{u, P_m} \nabla T + \right.$$

$$\left. + \frac{k \rho_{liq} f'_{liq} (u)}{\mu_{liq}} \left(\frac{\partial P_{liq}}{\partial P_m} \right)_{u, T} \nabla P_m + I_1 \right), \tag{25}$$

$$a_m \frac{\partial P_m}{\partial \tau} = \nabla \left(\frac{k \rho_m f'_m (u)}{\mu_m} \nabla P_m \right) + I_2, \tag{26}$$

$$\frac{\partial (a_v \rho_v)}{\partial \tau} = \nabla \left(\frac{k \rho_v f'_m (u)}{\mu_m} \nabla P_m \right) + \nabla (D_v \rho_m a_v \nabla (\rho_v / \rho_m) + \nabla (\rho_v a_v \chi \nabla T)) + I_2. \tag{27}$$

Assuming that the liquid in capillaries is in equilibrium with the vapor-gas mixture, we can determine the liquid pressure using the pressure of vapor and liquid in the porous body by the Kelvin formula [10]:

$$P_{cap} = P_{liq} - P_m = \frac{RT}{v'_{liq}} \ln \frac{P_v}{P_{sat}} \tag{28}$$

or

$$P_{liq} = P_m + \frac{RT}{v'_{liq}} \ln \frac{P_v}{P_{sat}}. \tag{29}$$

In the state of capillary equilibrium, equality of capillary pressures in material pores takes place [3]. In a one-component, two-phase system "liquid–vapor," the capillary pressure is equal to

$$P_{cap} = P_{liq} - P_v .$$ (30)

The coefficient of gas or vapor permeability k is determined by the geometry of the porous structure of the body [11]:

$$k = \frac{m}{96} \int_{r_{min}}^{r_{max}} r^2 S(r) \, dr ,$$ (31)

where r_{min} and r_{max} are the smallest and largest radii of open pores; $S(r)$ is the differential function of pore size distribution in the interval $[r_{min}, r_{max}]$.

To determine the permeability coefficient, use is made of the equation of gas filtration through porous media [11]. Based on an analysis of filtration drying [12], the function $S(r)$ is replaced by the differential moisture (mass) content distribution with respect to the radii of straight-through filtration pores and a calculated formula for permeability is obtained. Here, a method to determine the permeability coefficient as a function of the moisture content of a porous body is given.

Example of the Numerical Solution and Discussion of the Results. We will consider the drying process of a single spherical particle in a gas flow with constant temperature T_{a1} and coefficients of heat transfer α and mass transfer β_1, β_2 (being either in an isothermal immovable or moving layer with similar effective coefficients). Assume that the pressure of dry air is constant; heat transfer by molar transfer of liquid and vapor as well as by thermal moisture diffusion can be neglected. Then the equations of heat and mass transfer for the one-dimensional case in the spherical coordinate system acquire the form

$$[(1 - m) c_s \rho_s + u c_{liq} \rho_s] \frac{\partial T}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \lambda_{ef} \frac{\partial T}{\partial r} \right) + LI ,$$ (32)

$$a_v^* \frac{\partial P_v}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{k \rho_v f_v'(u)}{\mu_v} \frac{\partial P_v}{\partial r} \right) - I ,$$ (33)

$$\rho_s \frac{\partial u}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{k \rho_{liq} f_{liq}'(u)}{\mu_{liq}} \left(\frac{\partial P_{liq}}{\partial u} \right)_T \frac{\partial u}{\partial r} \right) + I ,$$ (34)

where $a_v^* = a_v(\nu_v/RT)$.

The initial conditions are

$$\tau = 0 , \quad T(r, 0) = T_0 , \quad P_v(r, 0) = P_{v0} , \quad u(r, 0) = u_0 .$$ (35)

The boundary conditions are

$$\left. \frac{\partial P_v}{\partial r} \right|_{r=0} = 0 , \quad \left. \frac{\partial T}{\partial r} \right|_{r=0} = 0 , \quad \left. \frac{\partial u}{\partial r} \right|_{r=0} = 0 ;$$

$$\left. \lambda_{ef} \frac{\partial T}{\partial r} \right|_{r=R_p} = \alpha (T_{a1} - T) ;$$ (36)

$$\left. \frac{k \rho_v f_v'}{\mu_v} \frac{\partial P_v}{\partial r} \right|_{r=R_p} = \beta_1 (P_{a1} - P_v) ;$$

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$$\frac{k \rho_{liq} f'_{liq}}{\mu_{liq}} \left(\frac{\partial P_{liq}}{\partial u}\right)_T \frac{\partial u}{\partial r}\bigg|_{r=R_p} = \beta_2 \left(u_e - u\right).$$

The physical parameters of the model material were specified to be similar to those of bentonite. In conducting a numerical experiment, the parameters were varied with the purpose of exploring the regularities of internal heat and mass transfer. Knowing the desorption isotherms for the entire temperature range in the drying process, we can determine the closing relations. To describe the desorption isotherms of bentonite, we use the approximation formula [13]

$$u_e = u_{0.5}\,(T) \left(\frac{P_v}{P_{sat} - P_v}\right)^{1/n}, \tag{37}$$

where $u_{0.5}(T)$ is the equilibrium moisture content as a function of the temperature at $\varphi = P_v/P_{sat} = 0.5$. The index n is a constant value for the same material. After approximating the results from [14], we arrive at

$$u_{0.5}\,(T) = 0.023 \exp\left[-0.007\,(T - 293)\right], \quad n = 3. \tag{38}$$

Expression (5) for the specific heat of a phase transition of moisture with allowance for (37) can be written as

$$L = \frac{RT^2}{v_v} \left[\frac{15}{T} - \frac{(n-1)\,(u_{0.5}\,(T))^{n-1}}{u^n + u_{0.5}^n}\,\frac{\partial u_{0.5}}{\partial T}\right], \tag{39}$$

where $\partial u_{0.5}/\partial T = -0.007 u_{0.5}(T)$.

The saturated vapor pressure as a function of the temperature is determined by the approximate formula

$$P_{sat} = 10^5 \left(\frac{T}{373}\right)^{15}. \tag{40}$$

The derivative in (34) with regard for (29) and (37) can be written as

$$\left(\frac{\partial P_{liq}}{\partial u}\right)_T = \frac{RT}{v'_{liq}}\,\frac{1}{nu}\,\frac{u_{0.5}^{1/n} + 2u^{1/n}}{u_{0.5}^{1/n} + u^{1/n}}. \tag{41}$$

The liquid pressure was calculated by formulas (28), (30).

Calculations were made at the following constant parameters: $u_0 = 0.05$ kg/kg; $T_0 = 293$ K; $\rho_s = 1500$ kg/m^3; $m = 0.3$; $c_{liq} = 4.18$ kJ/(kg·K); $R/v_{liq} = 4.6 \cdot 10^5$ J/(m^3·K); $R/v_v = 461.6$ J/(kg·K); $\alpha = 30$ W/(m^2·K); $\beta_1 = 1 \cdot 10^{-5}$ kg/(Pa·m^2·sec); $\beta_2 = 1.5 \cdot 10^{-4}$ kg/(m^2·sec); $P_{at} = 2000$ Pa; $T_{at} = 353$ K; $R_p = 5 \cdot 10^{-3}$ m; $\mu_{liq} = 0.55 \cdot 10^{-3}$ Pa·sec; $\mu_v = 1.6 \cdot 10^{-5}$ Pa·sec.

It was assumed that a particle surface is exposed to a heat flux uniformly distributed over the sphere surface. The porosity of the material was assumed to be constant. In calculations, the dependence of the functions of relative permeability on the moisture content was taken into account by the formulas

$$f'_m\,(u) = \left(\frac{u_{max} - u}{u_{max} - u_{min}}\right)^\delta, \quad f'_{liq}\,(u) = \left(\frac{u - u_{min}}{u_{max} - u_{min}}\right)^\delta, \tag{42}$$

where $u_{max} = 0.3$ kg/kg; $u_{min} = 0.001$ kg/kg.

The source term in Eqs. (32)-(34) was determined by expression (2), as well as by the following formula:
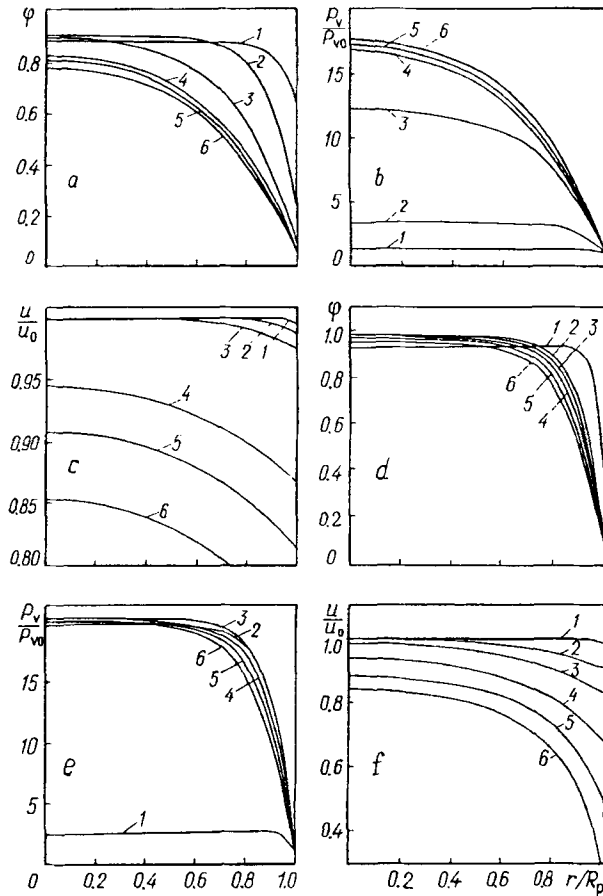
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Fig. 1. Relative humidity of the medium (a, d), vapor pressure (b, e) and moisture content (c, f) as a function of the particle radius for the nonequilibrium regime: a, b, c) $\gamma = 0.005$ kg/(m$^3 \cdot$sec); $\delta = 6$; $k = 1 \cdot 10^{-17}$ m^2; 1) $\tau = 2.58$, 2) 328, 3) 128, 4) 1230, 5) 1800, 6) 2730; d, e, f) $\gamma = 0.05$; 1) $\tau = 19.5$, 2) 324, 3) 587, 4) 1130, 5) 1660, 6) 2210.

$$I = \rho_s \frac{du_e}{d\tau} = \rho_s \left[\left(\frac{\partial u_e}{\partial P_v} \right)_T \frac{\partial P_v}{\partial \tau} + \left(\frac{du_e}{dT} \right)_{P_v} \frac{\partial T}{\partial \tau} \right] . \tag{43}$$

The latter expression indicates the equilibrium regime of moisture desorption.

A numerical solution of the system of equations was constructed using the iteration technique and applied programs for nonlinear parabolic equations [15].

Figure 1 shows the relative humidity of a vapor-air medium, vapor pressure, and moisture content as a function of the radial coordinate for the nonequilibrium regime of moisture desorption when the source term is calculated by formula (2) at $\gamma = 0.005$. These dependences have the form of monotonic parabolas, which, as far as moisture is removed, become flattened.

Figure 2 presents kinetic curves of material drying and heating for two regimes, namely, nonequilibrium with different kinetic coefficients and equilibrium when the source term is calculated by expression (43). The other parameters remain unchanged. As is seen, the chosen material is heated rather quickly and then the drying proceeds practically at a constant temperature. Apparently, with an increase in the coefficient γ, the drying rate will increase. As could be expected, during heating the moisture content of the material changes insignificantly and the kinetics curves of drying show a heating period, which is more distinctly seen in Fig. 3. The equilibrium regime leads to a drastic decrease in the moisture content during the heating period, which is caused by a decrease in the equilibrium moisture content with an increase in temperature. Therefore, the equilibrium regime of moisture desorption
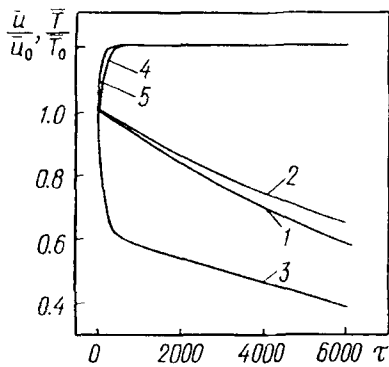
Fig. 2. Curves of drying (1, 2, 3) and heating (4, 5) of the material ($k = 1 \cdot 10^{-17}$ m^2; $\delta = 6$): 1) $\gamma = 0.01$ kg/(m^3·sec); 2, 5) 0.005; 3, 4) equilibrium regime.
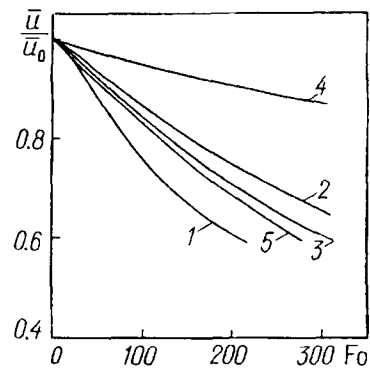
Fig. 3. Drying curves of the material: 1) $\gamma = 0.05$ kg/(m^3·sec); 2) 0.005, 3) 0.01 (for 1-3) $k = 1 \cdot 10^{-17}$ m^2, $\delta = 6$); 4) $k = 1 \cdot 10^{-18}$ m^2, $\gamma = 0.005$, $\delta = 6$; 5) $k = 1 \cdot 10^{-18}$, $\gamma = 0.005$, $\delta = 4$.

(determination of the source term by formula (43)) is not always valid, e.g., in the present case, and it may apparently be adopted for rather slow processes of heat and mass transfer.

For the nonequilibrium regime of moisture desorption, an increase in the kinetic coefficient results in an increase in the relative humidity of the medium and the vapor pressure inside a particle, amd also in the vapor-pressure gradients and moisture content in the near-surface zone of a particle for the considered time interval (Fig. 1d, e, and f) and, as a consequence, speeds up the drying process. The kinetic curves of drying in Fig. 3 are indicative of a slowing down of the heat and mass transfer process with a decrease in the permeability coefficient k and an increase in the exponent δ determining the relative function of permeability. This is related with an increase in resistance to vapor and liquid transfer.

This model can be used provided the structural characteristics of the porous material are known and slightly change in the investigated moisture-content range.

NOTATION

a, amount of the desorbed moisture, kg/m^3; c, heat capacity, J/(kg·K); D, diffusion coefficient, m^2/sec; f, relative permeability; L, specific heat of desorption, J/kg; m, material porosity; P, pressure, Pa; R, gas constant, J/(kmole·K); R_p, particle radius, m; $S(r)$, differential function of pore radius distribution; T, temperature, K; u, $\bar{u}$, local and mean moisture content of the material, respectively, kg/kg; v, phase velocity, m/sec; V, reduced phase velocity, m/sec; α, heat-transfer coefficient, W/(m^2·K); β_1, β_2, mass-transfer coefficients based on vapor pressure and moisture content, respectively, kg/(Pa·m^2·sec) and kg/(m^2·sec); γ, kinetic sorption (desorption) coefficient, kg/(m^3·sec); θ, saturation of the porous material; χ, thermal diffusion coefficient of vapor, m^2/(sec·K); λ_{ef}, effective thermal conductivity, W/(m·K); μ, coefficient of dynamic viscosity, Pa·sec; ν, molar mass, kg/kmole; ν_{liq}, molar volume of the liquid, m^3/kmole; ρ, density, kg/m^3; τ, time, sec; φ, relative humidity of the vapor-gas mixture; Fo, Fourier number. Subscripts: a, air; at, environment; e, equilibrium; ref, reference body; liq, liquid; m, vapor-air mixture; s, solid; sat, saturated; v, vapor.

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