Journal of Engineering Physics and Thermophysics, Vol. 71, No. 3, 1998

INTERNAL STRESSES, RHEOLOGY, AND STRUCTURE FORMATION IN DRYING OF A CONCENTRATED DISPERSE SYSTEM

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UDC 539.372:539.87

A mathematical model is suggested for the stress strain state and structure formation in drying of a concentrated disperse system. The interrelated process of deformation of the medium and motion of the liquid in a saturated region is described by equations of filtration consolidation. The rheological relationship of a saturated medium is determined on the basis of the micromechanics of the relative motion of the particles. A system of nonlinear integrodifferential equations that describe the process is obtained and numerical analysis of the problem is carried out under different drying conditions.

The stress-strain state (SSS) in drying of materials was investigated in [1-5]. It was assumed that the rheological properties of the medium were independent of the moisture content. The SSS in drying of a material with a different rheological behavior in the dry and saturated states was studied in [6]. A description of the behavior of disperse systems in a saturated region on the basis of the micromechanics of the approach of the particles was suggested in [7]. An analysis of internal stresses in drying with allowance for the filtration flux was made in [8].

Below, considering the micromechanics of the motion of the particles, we simulate the development of the SSS in drying of concentrated dispersions in their interrelation with the filtration flow of the dispersion medium in the pore space. We obtain a system of nonlinear differential equations and state and solve the problem of drying a free thin plate (layer). We carry out a numerical investigation and reveal the influence of the basic dimensionless parameters on the internal stresses and structure formation.

1. Mathematical Model. To describe the process of drying of deformed disperse systems in the zone of complete saturation, equations of filtration consolidation are used [9-11]:

$$\partial \overset{\mathrm{f}}{\sigma}_{ij} / \partial x_j - \partial p / \partial x_i = 0 , \tag{1.1}$$

$$\partial \theta / \partial t + \operatorname{div} \mathbf{q} = 0 , \tag{1.2}$$

$$\mathbf{q} = - f (| \nabla p | , \theta) (\nabla p / | \nabla p |) , \tag{1.3}$$

$$\sigma_{ij} = \overset{\mathrm{f}}{\sigma}_{ij} - p \delta_{ij} , \tag{1.4}$$

Equation (1.2) is written on the assumption that the matrix material and the liquid are incompressible, and, consequently, volumetric deformations of the medium occur only due to a change in the porosity.

To close the system of equations (1.1)-(1.3), rheological relations are needed. The presence of condensation contacts between the particles in the dry region allows one to use elasticity theory with account for free shrinkage [12]:

$$\sigma_{ij} = \frac{E}{1 + \nu} \left[\varepsilon_{ij} - \overset{0}{\varepsilon}_{ij} + \frac{\nu}{1 - 2\nu} (\varepsilon_{kk} - \overset{0}{\varepsilon}_{kk}) \delta_{ij} \right] . \tag{1.5}$$

Kazan State University. Russia. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 71, No. 3, pp. 417-423, May-June, 1998. Original article submitted July 9, 1996.

The mass balance of moisture on the evaporation surface gives an equation for determining the rate of evaporation-front motion V_n:

$$\rho m V_n = -(m/m_0)\, j_n + \rho q_n . \tag{1.6}$$

Since in the case considered volumetric deformations occur only due to changes in the porosity, then $dm/(1-m) = d\theta$.

Under conditions of intense drying the following approximate equation is valid [13]:

$$j_n = \frac{[D]\, p_s}{R_g T\, [l(t) + ([D]/D)\, \delta]}, \quad [D] = \frac{m_0 D}{\beta} . \tag{1.7}$$

2. SSS and Structure Formation in Drying of a Disperse Layer. Let us consider the drying of an originally homogeneous concentrated disperse layer. We assume that the process of evaporation from the layer surface is independent of the coordinates x, y in a Cartesian coordinate system (x, y, z) whose plane (x, y) goes through the middle plane of the plate. Then, the moisture content and the SSS will depend only on the transverse coordinate z, and the following relations will be valid:

$$\begin{gathered} \sigma_{ij} \equiv 0 \quad (i \neq j), \quad \sigma_{xx} = \sigma_{yy} = \sigma(z), \quad \sigma_{zz} \equiv 0 , \\ \varepsilon_{ij} \equiv 0 \quad (i \neq j), \quad \varepsilon_{xx} = \varepsilon_{yy} = \varepsilon(z), \quad \varepsilon_{zz}(z) \neq 0 . \end{gathered} \tag{2.1}$$

The conditions of consistency of deformations [12] with account for Eq. (2.1) are reduced to the one condition $\partial^2\varepsilon/\partial z^2 = 0$, and the rheological relation (1.5) for the dry region takes the form

$$\sigma = E(\varepsilon - \varepsilon^0)/(1 - \nu) . \tag{2.2}$$

The shrinkage ε^0 of the material particle with the coordinate z is equal to deformation at the time $t(z)$ of passage of the evaporation front through it $\varepsilon^0 = \varepsilon\,(z^0 = z)$.

The motion of the particles in saturated disperse systems is determined by two main factors: the viscous flow of the dispersion medium in the gap between the particles and the forces of surface interaction [14]. In contrast to the forces of viscous resistance, the surface forces can both impede and promote approach of the particles, depending on the distance and physicochemical properties of the surfaces.

We will represent a macropoint by a set of spherical particles that are bound together by surface forces and whose dimensions are distributed according to a Gaussian law:

$$\varphi(r) = (2\pi \langle\sigma\rangle^2)^{-1/2} \exp\, [-(r - \langle r\rangle)^2/(2\langle\sigma\rangle^2] ,$$

Having separated out a pair of particles of radius r from such a set, we will write the equations of their relative motion without account for inertial forces [7]:

$$\begin{gathered} F_i^{e}(t) + F(h_i) + F^{v}(h_i, \partial h_i/\partial t) = 0 , \\ \langle S\rangle = \pi \langle r\rangle^2 ; \quad F_i^{e} = \langle S\rangle\, \sigma^{f}(t) , \quad i = x, y, z . \end{gathered} \tag{2.3}$$

On the basis of results obtained in [7] the rheological relations in the dry and saturated regions can be represented in the following form:

$$\sigma(z, t) = E(\eta)\,(1 - \nu)^{-1}\, [\varepsilon(t) - \varepsilon^0(z)] , \quad z^0 < z < h ; \tag{2.4}$$

$$\sigma^{\mathrm{f}}(t) = \frac{3\pi\mu r^2}{2h\langle S\rangle}\frac{\partial h}{\partial t} - \frac{\pi r G(h)}{\langle S\rangle}, \quad 0 < z < z^0; \tag{2.5}$$

$$\sigma^{\mathrm{f}}_{zz}(z, t) = \frac{3\pi\mu r^2}{2h_z\langle S\rangle}\frac{\partial h_z}{\partial t} - \frac{\pi r G(h_z)}{\langle S\rangle}, \quad 0 < z < z^0. \tag{2.6}$$

These equations involve both microparameters (h, r) and macroparameters $(\varepsilon, \varepsilon^0)$. The former characterize the state of the separate pair of particles and change within the limits of the macropoint, whereas the latter belong to the macropoint as a whole. Consequently, to close the system, in addition to the boundary condition on the edge of the free plate dried symmetrically on both sides

$$\int_0^h \sigma(z, t)\, dz = 0, \tag{2.7}$$

a relationship between the micro- and macroscopic values is needed. For small deformations ($\langle r\rangle >> \langle h\rangle$) we have

$$\varepsilon = (\langle h\rangle - \langle h_0\rangle)/\langle r\rangle, \quad \varepsilon_{zz} = (\langle h_z\rangle - \langle h_{0z}\rangle)/\langle r\rangle, \tag{2.8}$$

$$\langle h\rangle = \int_{r_-}^{r_+} h(r)\varphi(r)\, dr, \quad \langle h_z\rangle = \int_{r_-}^{r_+} h_z(r)\varphi(r)\, dr.$$

The sought quantities in the problem are the pore pressure $p(z, t)$, stress $\sigma(z, t)$, thickness of the interlayers $h(t, r)$, $h_z(z, t, r)$, and position of the evaporation front $z^0(t)$.

Integrating Eqs. (2.4) and (2.5) over the plate thickness with allowance for the condition on the edge (2.7), we obtain

$$\left[\frac{3\pi\mu r^2}{2h(t, r)\langle S\rangle}\frac{\partial h(t, r)}{\partial t} - \frac{\pi r G(h)}{\langle S\rangle}\right] z^0 -$$

$$- \int_0^{z^0} p(z, t)\, dz + \int_{z^0}^{1} E(\eta)(1 - \nu)^{-1}[\varepsilon(t) - \varepsilon^0(z)]\, dz = 0, \tag{2.9}$$

and from Eq. (2.6), with allowance for Eq. (1.4) and $\sigma_{zz} \equiv 0$ (see Eq. (2.1)), we have

$$\frac{3\pi\mu r^2}{2h_z\langle S\rangle}\frac{\partial h_z}{\partial t} - \frac{\pi r G(h_z)}{\langle S\rangle} - p(z, t) = 0. \tag{2.10}$$

We carry out further transformations: Eqs. (2.9) and (2.10) are multiplied by $r^{-2}h(r)\varphi(r)dr$ and $r^{-2}h_z(z, r)\varphi(r)dr$, respectively, and integrated over r. Taking account of Eq. (2.8), we obtain

$$d\varepsilon/dt = \left[\int_0^{z^0} p(z, t)\, dz - \int_{z^0}^{h} E(\eta)(1 - \nu)^{-1}[\varepsilon(t) - \varepsilon^0(z)]\, dz\right]\frac{2\langle S\rangle \nu_2(t)}{3\pi\langle r\rangle\mu z^0(t)} + \frac{3\nu_1(t)}{3\langle r\rangle\mu}, \tag{2.11}$$

$$\partial\varepsilon_{zz}/\partial t = \frac{2\langle S\rangle p(z, t) u_2(z, t)}{3\pi\langle r\rangle\mu} + \frac{2u_1(z, t)}{3\langle r\rangle\mu}, \tag{2.12}$$

$$u_1(z,t)=\int\limits_{r_*}^{r_+} r^{-1} h_z g(h_z)\varphi(r)\,dr\,,\quad u_2(z,t)=\int\limits_{r_*}^{r_+} r^{-2} h_z\varphi(r)\,dr\,,$$

$$v_1(t)=\int\limits_{r_*}^{r_+} r^{-1} hg(h)\varphi(r)\,dr\,,\quad v_2(t)=\int\limits_{r_*}^{r_+} r^{-2} h\varphi(r)\,dr\,.$$

For the considered case of drying a thin disperse layer, Eqs. (1.2) and (1.6), with account for Eqs. (2.12) and (1.7), take the form

$$\frac{\partial}{\partial z}\left[K_f(\theta)\frac{\partial p}{\partial z}\right]-\frac{2\langle S\rangle u_2(z,t)}{3\pi\langle r\rangle\mu}p(z,t)=\frac{2u_1(z,t)}{3\mu\langle r\rangle}+2\frac{d\varepsilon}{dt}, \tag{2.13}$$

$$\rho m\frac{dz^0}{dt}=-\frac{m}{m_0}\frac{[D]p_s}{R_gT(h-z^0+\delta[D]/D)}+\rho\left[-K_f(\theta)\frac{\partial p}{\partial z}\right]_{z=z^0}. \tag{2.14}$$

Thus, we have the system of equations (2.9), (2.10), (2.13), and (2.14) for determining the functions $p(z, t)$, $z^0(t)$, $h(t, r)$, and $h_z(z, t, r)$ with allowance for Eqs. (2.8) and (2.11) under the following boundary and initial conditions:

$$z=0:\ \ \partial p/\partial z=0\,; \tag{2.15}$$

$$z=z^0:\ \ p(z^0)=p_c(\varepsilon)\,; \tag{2.16}$$

$$t=0:\ \ h=h_z=h_0(r)\,,\ \ \varepsilon=\varepsilon_{zz}=0\,,\ \ z^0=h\,, \tag{2.17}$$

where h_0 is the initial distance between the particles, determined from the condition $F_c = \pi rG(h_0)$. For the permeability of the medium k we use the Kozeni formula [13] $k = m^3/(5S_0(1-m)^2)$, $S_0 = 3(1-m)/r$.

The shrinkage $\varepsilon^0(z)$ is equal to the deformation $\varepsilon(t)$ at the time $t(z)$ when the evaporation front is at the position z. Consequently, the shrinkage is variable over the plate thickness and is determined in the course of solution, $\varepsilon^0(z) = \varepsilon(z^0 = z)$. To determine the dependence of the capillary pressure p_c on the deformation ε, we should take into consideration the model of the structure of the disperse medium. For the model selected, i.e., an ensemble of spherical particles, the pressure p_c can be approximately represented as

$$p_c(\varepsilon)=\frac{-2\alpha}{\langle r\rangle(\varepsilon+\langle h_0\rangle/\langle r\rangle)}.$$

To investigate the problem numerically, we represent the system of nonlinear integrodifferential equations (2.9), (2.10), (2.13), and (2.14) with the initial and boundary conditions (2.15)-(2.17) in dimensionless form:

$$\frac{\partial}{\partial\zeta}\left[k_0^0\frac{\partial P(\zeta,\tau)}{\partial\zeta}\right]-\frac{2U_2(\zeta,\tau)P(\zeta,\tau)}{3\kappa}=\frac{2}{3\kappa}\gamma U_1(\zeta,\tau)+$$

$$+2\left[\frac{V_2}{1-\zeta^0}\left(\int\limits_{\zeta^0}^{1}P(\zeta,\tau)\,d\zeta-\psi\int\limits_0^{\zeta^0}\psi_0(e(\tau)-e^0(\zeta))\,d\zeta\right)+\gamma V_1\right]; \tag{2.18}$$

$$m(\theta)\frac{d\zeta^0}{d\tau} = \frac{m(\zeta = \zeta^0)}{m_0}\frac{QJ(0)}{\zeta^0 D/(|D|\delta_0) + 1} - \kappa k_0^0 \frac{\partial P}{\partial \zeta} \quad (\zeta = \zeta^0), \tag{2.19}$$

$$\left[\frac{3R^2}{2H}\frac{\partial H}{\partial \tau} - R\gamma g(H)\right](1 - \zeta^0) - \int_{\zeta^0}^{1} P d\zeta + \psi \int_0^{\zeta^0} \psi_0 \,[e(\tau) - e^0(\zeta)]\, d\zeta = 0, \tag{2.20}$$

$$\left[\frac{3R^2}{2H_z}\frac{\partial H_z}{\partial \tau} - R\gamma g(H_z)\right] - P(\zeta, \tau) = 0 \tag{2.21}$$

with the boundary and initial conditions

$$\zeta = 1: \ \partial P/\partial \zeta = 0; \tag{2.22}$$

$$\zeta = \zeta^0: \ P(\zeta^0) = P_c(e) = -\delta_1/(e + \delta_1); \tag{2.23}$$

$$\tau = 0: \ H = H_z = H_0(R), \ e = e_{zz} = 0. \tag{2.24}$$

The effective and total stresses are determined from relations (2.4), (2.5), and (1.4):

$$\Sigma^f(\tau) = \frac{3R^2}{2H}\frac{\partial H}{\partial \tau} - R\gamma g(H), \ \zeta^0 < \zeta < 1;$$

$$\Sigma(\zeta, \tau) = \Sigma^f(\tau) - P(\zeta, \tau), \ \zeta^0 < \zeta < 1;$$

$$\Sigma(\zeta, \tau) = \psi\psi_0\,[e(\tau) - e^0(\zeta)], \ 0 < \zeta < \zeta^0;$$

$$\Sigma_{zz}^f(\zeta, \tau) = P(\zeta, \tau), \ \Sigma_{zz} = 0, \ \zeta^0 < \zeta < 1.$$

The following dimensionless variables and parameters are introduced:

$$P = p/(|p_c(0)|), \ \tau = t\,|p_c(0)|/\mu, \ \zeta = 1 - z/h_{pl}, \ R = r\langle r\rangle,$$

$$e = \varepsilon/|\varepsilon_+|, \ H = h/\langle r\rangle, \ H_z = h_z/\langle r\rangle, \ \Sigma = \sigma/|p_c(0)|,$$

$$J = i/i_{0*}, \ g(H) = G(h)/G_+, \ g_z(H_z) = G(h_z)/G_+, \ \delta_0 = \delta/h_{pl}, \ \psi_0 = E/E_0,$$

$$\kappa = k_0/h^2, \ \gamma = G_+/\langle r\rangle\,|p_c(0)|, \ \psi = E_0\,|\varepsilon_+|/(1 - \nu)\,|p_c(0)|, \ \delta_1 = \langle h_0\rangle/\langle r\rangle\,|\varepsilon_+|,$$

$$V_2 = \int_{R_*(\tau)}^{R_+} R^{-2} H\Phi(R)\,dR, \ V_1 = \int_{R_*(\tau)}^{R_+} R^{-1} Hg(H)\,\Phi(R)\,dR,$$

$$U_2 = \int_{R_*(\tau)}^{R_+} R^{-2} H_z\Phi(R)\,dR, \ U_1 = \int_{R_*(\tau)}^{R_+} R^{-1} H_z g(H_z)\,\Phi(R)\,dR,$$

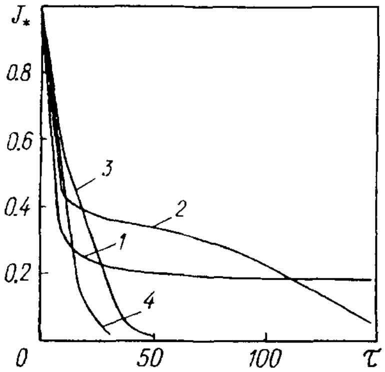


Fig. 1. Dependence of the critical vapor flux J_* on the time τ: 1) $\kappa = 10^{-4}$; 2) 10^{-3}; 3) $5\cdot 10^{-3}$; 4) 10^{-2}.

$$Q = i_{0*}\,\mu/h\,|p_c(0)|\,\rho\,,\quad k_0^0 = \frac{k\,(\zeta = \zeta^0)}{k_0}\,,\quad \Phi\,(R) = \varphi\,(r)\,\langle r\rangle\,.$$

The second boundary condition for the pressure does not always have the form of Eq. (2.23). Two regimes are possible depending on the intensity of vapor removal. Calculations showed that if J is smaller than a certain value, then the pressure $P(1, \tau)$ on the surface turns out to be greater than the capillary pressure $P_c(e)$, which corresponds to incompletely developed menisci. In this case (the first regime) the evaporation surface does not recede and equality is established between the filtration inflow of the liquid and the vapor flow. The boundary condition for the pressure in this regime follows from Eq. (2.19) at $\zeta^0 = 0 = \text{const}$ and has the form

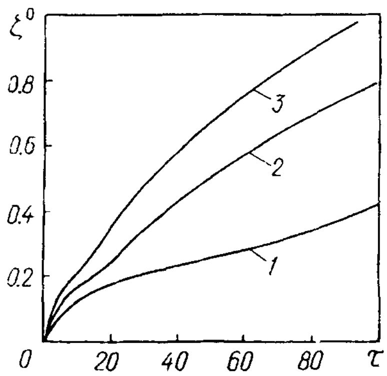
$$\frac{\partial P}{\partial \zeta}\,(\zeta = 0) = \frac{m\,(\zeta = 0)}{m_0}\,\frac{QJ\,(0)}{\kappa k_0^0}\,. \tag{2.25}$$

In the case of intense removal of the vapor flux J the pressure on the layer surface $P(1, \tau)$ becomes equal to the capillary pressure P_c, and the evaporation front recedes, which corresponds to the second regime of drying. We denote by J_* the magnitude of the vapor flux that separates the two regimes indicated and call it critical. In order to determine J_*, it is sufficient to find the pressure field $P(\zeta, \tau)$ in the plate that satisfies, in addition to the boundary condition (2.8) at the plate center, the two boundary conditions (2.23) and (2.25) at $\zeta^0 = 0$. Here, one of them is used to recover the pressure, and the other to determine J_*.

By analyzing numerically problem (2.18)-(2.24), we revealed the basic dimensionless parameters (κ, ψ, γ) that influence its solution. The first parameter is the ratio of the viscous resistance of the matrix due to the liquid motion in the gap between the particles to the filtration resistance; the second parameter is the ratio of the elasticity modulus of the dry region to the capillary pressure, and the third parameter is the ratio of the specific surface force to the capillary pressure. The dependence of the elasticity modulus E on the concentration η was taken to be linear, $E = E_0(1 + \eta)$, and, consequently, $\psi_0 = 1 + \eta$.

Figure 1 presents the dependences of the critical vapor flux J_* on the time τ for different values of the parameter κ. In the initial period of drying the magnitude of the critical flow decreases sharply irrespective of the value of the parameter κ. As is seen from Fig. 1, for small values of κ (curves 1, 2) a time segment during which the quantity J_* changes little is typical. As the parameter κ increases, there is no flat portion in the graph (curves 3, 4). The presence of a "plateau" in the graphs of $J_*(\tau)$ for small values of κ is explained by the fact that initially deformations e develop in plan in disperse media. At the same time, in the transverse direction ζ the compression is small, and one can even observe extension at the center of the plate. Then, inflow of the liquid is formed due to penetration of transverse deformations e_{zz} into the plate, which maintains an almost constant magnitude of the flux J_*.

As a result of a numerical investigation of the regime of drying with formation of a dry zone, we obtained time dependences of the position of the evaporation front ζ^0 (Fig. 2) for different values of the parameter κ.



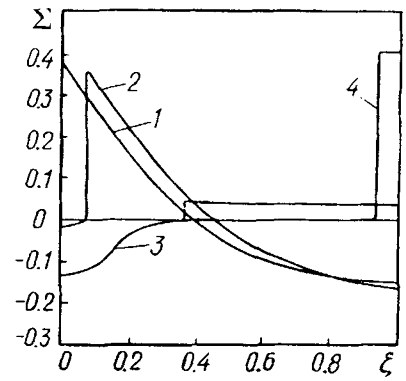


Fig. 2. Influence of the parameter κ on the time dependence of the evaporation-front position ζ^0: 1) $\kappa = 10^{-3}$; 2) $5 \cdot 10^{-3}$; 3) 10^{-2}.

Fig. 3. Distribution of the stresses Σ over the plate thickness ζ ($\psi = 3$; $\kappa = 10^{-2}$): 1) $\zeta^0 = 0$; 2) 0.08; 3) 0.37; 4) 0.95.

Attention should be paid to the fact that a change in the parameter ψ has no effect on the dependence $\zeta^0(\tau)$. We also obtained distributions of stresses over the plate thickness (Fig. 3) for different positions of the evaporation front. Curve 1 (Fig. 3) corresponds to the initial time of drying, with the evaporation front on the layer surface. In this period the saturation region has both a region of extension and a region of compression (curves 1, 2). As the evaporation front recedes farther into the layer, the stress in the saturation region becomes constant due to equalization of the liquid pressure over the plate thickness (curves 3, 4, Fig. 3). Usually, in the process of drying, the dry region is in a state of compression, and the level of compressive stresses grows with increase in the parameter ψ. On completion of drying, the structure of the dried medium will be inhomogeneous over the plate thickness, i.e., the medium is hardened from the periphery to the center.

The author wishes to express his gratitude to Professor A. V. Kosterin for discussion of the results of this work and valuable comments.

The work was carried out under financial support from the Russian Fund for Fundamental Research (project No. 93-013-17300).

NOTATION

$[D]$, D, effective and molecular coefficients of diffusion; E, E_0, Young's modulus and its value at $\eta = 0$; e, e_{zz}, dimensionless deformations; F_i^e, the force determined by the effective macrostresses; F^ν, F, the force of the viscous resistance and of the surface interaction between the particles; F_c, capillary forces at the initial time of drying; G, G_+, specific energy of the surface interaction of plane particles and its maximum value; g, g_z, dimensionless specific energies of the surface interaction of the particles; h_{pl}, layer thickness of the disperse medium; h_i, shortest distance between particles in the direction of the coordinate axes; $h = h_x = h_y$, h_z, H, H_z, current distances between particles and their dimensionless values; h_0, H_0, initial thickness of the liquid interlayers between the particles; $\langle h\rangle$, $\langle h_z\rangle$, $\langle h_0\rangle$ mean distances between particles at the macropoint; $\langle h_0\rangle$, mean initial distances between particles and its dimensionless value; j, j_n, j_{0*}, mass vapor flux, its component normal to the evaporation surface, and its critical value at the initial instant of time; J, J_*, dimensionless vapor flux and its critical value; k, k_0, permeability and its initial value; k_0^0, dimensionless parameter; K_f, coefficient of filtration; l, depth of the dry zone; m_0, m, initial and current porosities; P_c, P, dimensionless capillary and current pressures of the liquid; p_c, p, capillary and current pressures of the liquid; p_s, pressure of the saturated vapor; Q, dimensionless constant; q, q_n, filtration-rate vector and its component normal to the evaporation surface; R_g, universal gas constant; R, dimensionless radius of the particles; r_-, r_+, R_+, minimum and maximum radii of the particles and their dimensionless values; R_*, dimensionless critical radius of the particle; $\langle r\rangle$, r, mean and current radii of the particles; S_0, specific surface of the particles; $\langle S\rangle$, cross-sectional area of a particle of mean radius; T, temperature; t, time; u_1, u_2, U_1, U_2, v_1, v_2, V_1, V_2, notation in formulas (2.11), (2.12), (2.18); V_n, component of the evaporation front velocity vector normal to the evaporation surface; x, y, z, axes of a Cartesian coordinate system; z^0, coordinate of the evaporation front; α, surface tension coefficient of the dispersion medium; β, tortuosity coefficient; δ_{ij}, Kronecker delta; δ, thickness of the diffusion boundary layer on the outer surface of the specimen; ε_{ij}, ε_{ij}^0, components of the deformation and shrinkage tensors; ε^0, shrinkage of the material elements of the medium in the (x, y) plane; ε, ε_{zz}, normal component of the deformation tensor along the x (or y) and z axes; γ, δ_0, δ_1, κ, ψ, ψ_0, dimensionless parameters; ζ, dimensionless coordinate; ζ^0, dimensionless coordinate of the evaporation front; η, concentration of rigid contacts at the macropoint; $\theta = \varepsilon_{kk}$, first invariant of the macrodeformations; μ, viscosity of the dispersion medium; ν, Poisson coefficient; ρ, liquid density; σ_{ij}, σ_{ij}^f, total and effective stresses in the porous matrix; σ, σ_{zz}, normal component of the stress tensor along the x (or y) and z axes; $\langle r\rangle^2$, dispersion; τ, time; φ, Φ, dimensional and dimensionless distribution functions.

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