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MATHEMATICAL MODELING OF THE PROCESS OF SUBLIMATION DRYING

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A complex physicomathematical analysis of vacuum sublimation drying has been given. Approaches to calculation and designing of sublimation dryers have been proposed.

Sublimation drying is the most complex nonstationary heat- and mass-exchange process. The kinetics and dynamics of sublimation drying depend on three interrelated processes: freezing of a material and its sublimation dehydration as well as vacuum desublimation (condensation) of a steam. In the strict sense, the identity of a dried material in technological batches can be attained under the same conditions of variation in the basic parameters in all these processes.

Mathematical modeling of sublimation drying involves: (a) calculation of the kinetics of sublimation drying, (b) calculation of the dryers, and (c) selection of the methods and conditions of optimization of sublimation dehydration.

In calculating the kinetics of sublimation drying, one uses two groups of methods: 1) empirical methods and (2) analytical ones.

Empirical Methods of Calculation of the Kinetics of Drying. The influence of the basic regime and technological parameters on the intensity of sublimation dehydration of a material has experimentally been studied using these methods on the basis of the physical sublimation-drying model [1–4]. An analysis of the results of investigations [1–4] on the kinetics enables us to generalize them in the form of the criterial dependence

$$\mathrm{Re}^{*} = \frac{J_{\mathrm{subl}} h}{\eta} = A\,\mathrm{Gu}^{n} K_1^{m} K_2^{d} K_3^{s}\,\mathrm{Le}^{i}\,. \tag{1}$$

Relation (1) is true, as a rule, for the temperature and pressure range and for the form and intensity of energy supply selected in the experiment.

The criterial dependence (1) was constructed in each case by statistical processing of experimental results. One drawback of empirical methods is their low accuracy in the case where dryers are used in calculations; the reasons for the low accuracy are as follows:

(1) no account is taken of the influence of the fraction of the radiative heat flux from sublimator walls; for this reason, the assumption of the participation of a certain convective component [1] allegedly intensifying the process of dehydration was made in obtaining criterial dependences of the type (1) for different products (the experiments of [3] have proved the fallacy of these opinions); 2) dependence (1) makes it impossible to obtain data on the kinetics of sublimation drying of uninvestigated materials and cannot be used in systems of monitoring of the process of sublimation drying and of control over it.

Analytical and Numerical Methods of Calculation of the Kinetics of Sublimation Drying. Calculation of a Dryer. The methods are based on mathematical models using heat- and mass-exchange and gasdynamic equations. Driving forces and mechanisms of the processes (a) and initial and boundary conditions (b) have been established for the mathematical models to compare theory and experiment.

A number of mathematical models have been considered in [2].

Figure 1 gives a physical model of transfer of heat and mass in sublimation dehydration in a batch dryer with conductive and radiative energy supplies. In accordance with Fig. 1, the most widespread for the process of sublima-

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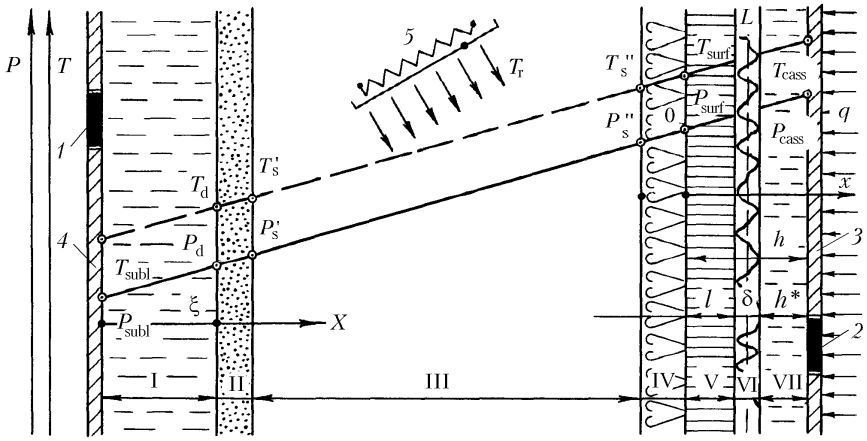

Fig. 1. Physical model of sublimation drying of a material in the dryer in the zones: I) desublimated ice; II) diffusion vapor layer; III) vapor-gas flow; IV) turbulent jet vapor flows; V) dry layer; VI) finely crystalline ice (sublimation zone); VII) ice; 1 and 2) heat-flux transducers; 3) cassette surface; 4) desublimator-wall surface; 5) radiator.

tion drying in the dryer are two dehydration models: 1) with a plane L phase boundary, when the sublimation zone is conventionally presented in the form of a plane [2, 5, 6]; 2) with a developed phase boundary (sublimation zone δ), where volume sublimation of frozen moisture from finely crystalline ice occurs [7].

In the first case of sublimation dehydration with thermoradiative energy supply [5, 6], the change in the temperature field for a dry product layer is described by the equation (Fig. 1, zone V)

$$\frac{\partial t_1}{\partial \tau} = a_1 \frac{\partial^2 t_1}{\partial x^2} + \frac{c_2 \rho_2}{c_1 \rho_1} \frac{dL}{d\tau} \frac{\partial t_1}{\partial x}, \quad 0 \le x \le L, \tag{2}$$

for a frozen layer (without energy supply), it is described by

$$t_2 = t_s'' . \tag{3}$$

With radiative energy supply we have $t_2 = t_s'' + \Delta t_s$, where Δt_s is the temperature jump on the sublimation surface (superheating of the frozen zone) due to the energy supply. For contact drying, Δt_s can be approximated by the dependence

$$\Delta t_s = q\delta / \lambda_{eff}, \tag{4}$$

where $\lambda_{eff} = f(P, q)$, and it is determined by the relations given in [2]. The initial conditions are

$$t_1 (x, 0) = t_s'', \quad L = 0; \tag{5}$$

$$\rho_2 r_{subl} \frac{dL}{d\tau} = C\left[\left(\frac{T_r}{100}\right)^4 - \left(\frac{T_s}{100}\right)^4\right], \quad x = 0. \tag{6}$$

The boundary conditions for $x = 0$ are

$$-\lambda_2 \frac{\partial t_1}{\partial x} = C\left[\left(\frac{T_r}{100}\right)^4 - \left(\frac{T_{surf}}{100}\right)^4\right], \quad x = 0. \tag{7}$$

For vacuum conditions, we have $\alpha_{conv}(T_{med} - T_{surf}) \approx 0$; when $x = L$ we obtain

$$-\lambda_2 \frac{\partial t_1}{\partial x} = \rho_2 r_{subl} \frac{dL}{d\tau}. \tag{8}$$

In [6], the problem posed has been solved numerically as applied to the sublimation drying of hydrogen-oxidizing bacteria, i.e., a product of microbiological synthesis. A satisfactory agreement between theory and experiment has been obtained with a total error of 1%. The optimum intensity of drying at a maximum permissible temperature T_{surf} of the product has been found. The algorithm can be used in a control system.

For the second case, the energy equation in the case of thermoradiative supply of heat for a thick layer ($h > 1$ mm, Fig. 1, zone V) is as follows:

$$\frac{\partial t_1}{\partial \tau} = a_1 \frac{\partial^2 t_1}{\partial x^2} - \frac{\Pi \rho_2 c_2}{(l+\delta)\, c_1\, \rho_1} \left(\frac{dl}{d\tau} + \frac{1}{2} \frac{d\delta}{d\tau} \right), \quad 0 \le x \le l + \delta. \tag{9}$$

The initial and boundary conditions are

$$t_1 (x, 0) = t_s'', \tag{10}$$

$$t_1 (h, \tau) = t_{cass} (\tau), \tag{11}$$

$$t_1 (0, \tau) = t_{surf} (\tau). \tag{12}$$

For a thin layer ($h \le 1$ mm), the general equation (when $\partial t_1 / \partial x = 0$) will be written as

$$\frac{\partial t_1}{\partial \tau} = \frac{\Pi \rho_2 r_{subl}}{h c_1 \rho_1} \left\{ \frac{dl}{d\tau} + \frac{1}{2} \frac{d\delta}{d\tau} \right\} + \frac{1}{W_0} \frac{d}{d\tau} \left[h^* (\tau)\, W^* (\tau) \right] + \frac{q_v (\tau)}{c_1 \rho_1} \tag{13}$$

with the following boundary conditions:

$$t_1 (0, \tau) = \tilde{\beta}\tau, \quad t_1 (h, \tau) = \tilde{\varepsilon}\tau. \tag{14}$$

The thermophysical characteristics c_1 and ρ_1 in (13) are taken for the region $l + \delta$, and $q_v(\tau)$ is the heat flux absorbed by the entire layer of the material. The problem of sublimation drying of melange has been solved under different assumptions in [7]. Equation (13) with boundary conditions (10) and (14) can be used for calculation of the kinetics of drying in designing continuous dryers.

For thermophysical characteristics of materials in calculations we can use:

(1) the effective values λ_{eff}, c_{eff}, and ρ_{eff} for the known porosity Π or the radius of the pores of a dry skeleton;

(2) determination of λ_1, c_1, and ρ_1 from the kinetic curves and the thermograms of drying of a material, as has been indicated in [5, 6].

The quasistationary level of pressures in a sublimation dryer (Fig. 1) must be ensured by the efficient operation of condensers (desublimators). The process of desublimation is active when the temperature (pressure) of a cooled desublimator wall is lower than the saturation temperature (pressure) of a steam, i.e., $P_{subl} < P_s'$ at $P_s' \approx 1$ mmHg.

When $P > 1$ mmHg we should take $P = P_{cr}$, where P_{cr} is the pressure corresponding to the critical temperature at which ice is formed [8, 9]:

$$P_{cr} = 1.0465 \cdot 10^{47} \exp (-27760 T_{cr}). \tag{15}$$

For desublimators with plane desublimation surfaces, a mathematical formulation of the problem has the form (Fig. 1, zone I)

$$\frac{\partial T_{\text{ice}}}{\partial \tau} = a_{\text{ice}} \frac{\partial^2 T_{\text{ice}}}{\partial x^2}, \quad 0 \le x \le \xi ; \tag{16}$$

$$T_{\text{ice}}(\xi, \tau) = T_{\text{d}}(\xi) ; \tag{17}$$

$$\lambda_{\text{ice}} \left. \frac{\partial T_{\text{ice}}(x, \tau)}{\partial x} \right|_{x=\xi-0} = \rho_{\text{ice}} r_{\text{d}} \frac{d\xi}{d\tau} ; \tag{18}$$

$$T(0, 0) = T_{\text{subl}} ; \tag{19}$$

$$\rho_{\text{ice}} \frac{d\xi}{d\tau} = \frac{\overline{\alpha}\,[P_{\text{s}}' - P_{\text{d}}(\xi, \tau)]}{\sqrt{2\pi R T_{\text{d}}(\xi, \tau)}} . \tag{20}$$

In accordance with [2], the quantity $P_{\text{d}}(\xi, \tau)$ in Eq. (20) for the range of residual pressures 600–13.33 N/m^2 can be described by the dependence

$$P_{\text{d}}(\xi, \tau) = A T_{\text{d}}^n(\xi, \tau) , \tag{21}$$

where $A = 10^{-53.56}$ and $n = 23.13$.

The value of λ_{ice} in (19) is determined for desublimated ice as follows [2, 10]:

$$\lambda_{\text{ice}} = \frac{1100}{T_{\text{ice}}} . \tag{22}$$

It follows from dependence (22) that the thermal conductivity of desublimated ice on the desublimation surface having a temperature of 200 K is equal to 5.5 W/(m·K), i.e., is 1.72 times larger than the thermal conductivity of atmospheric ice.

The condensation coefficient $\overline{\alpha}$ in Eq. (20) in the residual-pressure range 6.67–266 N/m^2 is determined by the relation [9, 11]

$$\overline{\alpha} = 0.03 + 0.01255 \cdot 10^{-2} P_{\text{s}}' . \tag{23}$$

It has been shown experimentally that the ice temperature can be described by the dependence

$$T_{\text{ice}}(x, \tau) = T_{\text{w}} + (T_{\text{d}} - T_{\text{w}}) \left(\frac{x}{\xi} \right)^n . \tag{24}$$

For the selected linear temperature distribution in the ice layer [2]

$$T_{\text{ice}}(x, \tau) = T_{\text{w}} + (T_{\text{d}} - T_{\text{w}}) \frac{x}{\xi} \tag{25}$$

the duration of the process of desublimation is

$$\tau = 0.109 \cdot \frac{\rho_{\text{ice}} r_{\text{d}}}{\lambda_{\text{ice}}} \xi^2 . \tag{26}$$

By analogy with the condensation of a liquid, the effective coefficient of heat transfer in formation of an ice layer can be determined in the form [11]

$$\alpha_{\text{eff}} = B\,(\lambda_{\text{ice}}\,\rho_{\text{ice}} r_{\text{d}}/\tau)^{1/2}, \quad B = \text{const}. \tag{27}$$

For maintaining a constant vacuum in the process of sublimation dehydration the relation

$$J_{\text{subl}}(\tau) = J_{\text{d}}(\tau) \tag{28}$$

is true for the dryer.

There can be several approaches to solution of problems of desublimation in the dryer with account for (28):

(a) determination of the desublimation intensity J_{d} with the known change in the nonstationary field in the ice layer and comparison to J_{subl} (relation (28));

(b) determination of the temperature field in the ice layer according to the known approximate dependence of the intensity $J_{\text{d}}(\tau)$ by solution of the problem on the kinetics of drying.

The vacuum main from the sublimator to the condenser is calculated from the well-known gasdynamic dependences [12]. The above physical models and mathematical formulations of the problems are basic for calculation of the overall dimensions of a dryer, determination of the dimensions of a desublimator, and selection of a vacuum pump.

The second period of sublimation drying is the least investigated theoretically and experimentally. Construction of the dependence for the drying rate by differentiation of the drying curves for this period of sublimation drying has an extremely low accuracy [1]. The calorimetric method of direct measurement enables us to obtain the curve of the rate of drying of materials on dryers of any type. The most characteristic curves of drying rate for the second period of dehydration of capillary-porous and colloidal materials have been obtained in [13]. As has been indicated in [14], the S-shaped form of the drying-rate curve is the most characteristic for the second period of drying. In this particular case, as our experiments have shown, the dependence of the drying rate

$$\frac{dW}{d\tau} = f(\text{Le}) = f\left(\frac{W}{W_{\text{f}}}\right)^{n} \tag{29}$$

is legitimate for this period. However, the peculiarity of the mechanism of sublimation dehydration from the critical point K_1 and K_2 (Fig. 2) on the drying-rate curve (presence of the local phase of ice in the material and of the dry and moist zones in the capillary-porous structure) is determined by a more complex dependence of the drying rate in the form

$$\frac{dW}{d\tau} = a + b\tau - e\tau^2\Big|_{\tau_{K_1}}^{\tau_{K_2}} - d\exp(-f\tau)\Big|_{\tau_{K_2}}^{\tau_{K_1}}. \tag{30}$$

Thus, for sublimation drying of brewer's yeast we have

$$\frac{dW}{d\tau} = \omega_1\left[0.19 + 4.56\tau - 0.5\tau^2\right]\Big|_{\tau_{K_1}}^{\tau_{K_2}} + \omega_2 \cdot 194.95\exp(-0.48\tau)\Big|_{\tau_{K_2}}^{\tau_{K_3}}. \tag{31}$$

In relations (29) and (30), the time from τ_{K_1} to τ_{K_2} is the transient period of drying (when local zones of finely crystalline ice exist in the material). The interval from τ_{K_2} to τ_{K_3} is the period of removal of bound moisture. Monitoring of these periods is of great practical importance for developing energy-saving technologies of drying and for obtaining a high-quality product.

Optimization of Sublimation Drying of Materials in Batch Dryers. To optimize the process of sublimation drying one has used Eq. (1), as a rule. However, the complex interrelation of the parameters in it makes this problem rather difficult. We have tested the method of optimization of sublimation drying for a generalized parameter — heat

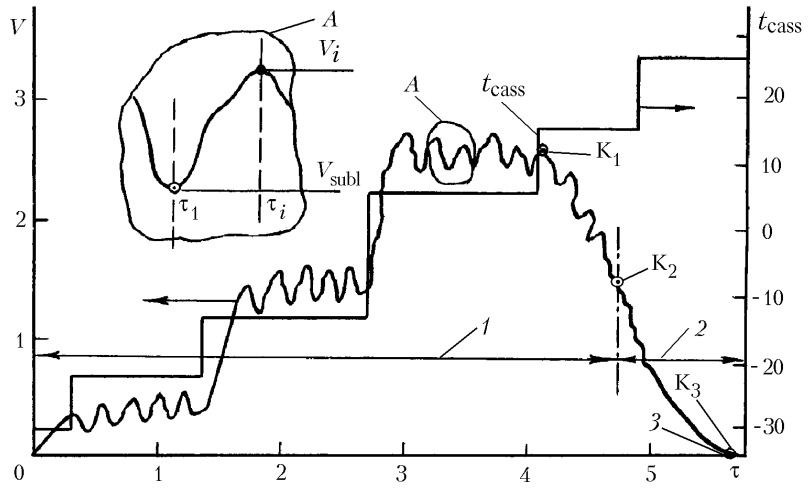

Fig. 2. Kinetics of drying of brewer's yeast (in the signal of a heat-flux transducer–time coordinates) in cyclic change in the cassette temperature [A) region of a portion of the drying curve]: 1) first period of drying; 2) second period; 3) end of drying. V, mV; τ, h; t_{cass}, ^{o}C.

of phase transition (crystallization, melting, sublimation, and desublimation heats). Figure 1 shows the arrangement of heat-flux transducers for monitoring of the heats of phase transition [13], whereas Fig. 2 gives the curve of the rate of sublimation drying of brewer's yeast in the signal of a heat-flux transducer–time coordinates at an elevated surface temperature T_{cass} of the cassette base and its cyclic change. The oscillations of the drying-rate curve (Fig. 2) in the first period are caused by the successive local action of the processes of crystallization and sublimation on the frozen product; in the second period, they are caused by the action of the processes of melting and sublimation. To determine optimization conditions for the process of sublimation dehydration in the first period we formulate the problem in generalized form (Fig. 1, zone V [15, 16])

$$\frac{\partial \bar{T}_1}{\partial \mathrm{Fo}} = \frac{\partial^2 \bar{T}_1}{\partial X^2} - \pi_1 \frac{d\bar{L}}{d\mathrm{Fo}} \frac{\partial \bar{T}_1}{\partial X}, \quad 0 \le X \le \bar{L}. \tag{32}$$

For the frozen layer (zone VII) we have

$$\bar{T}_2 = \frac{t - t_s''}{t_{cass} - t_s''}. \tag{33}$$

The boundary conditions are

$$\frac{\partial \bar{T}_2}{\partial X} = -\pi_2 \frac{\partial \bar{T}_1}{\partial X} - Q_{subl}(\theta) + Q_{cryst}(\theta), \quad X = \bar{L}; \tag{34}$$

$$\frac{\lambda_1 \partial \bar{T}_1}{L(t_{cass} - t_s'')\,\partial X} = 0, \quad X = 0. \tag{35}$$

The initial conditions are

$$\bar{T}_1(X, 0) = \bar{T}_{cass}(\theta), \quad X = 1; \tag{36}$$

$$\bar{T}_1(X, 0) = \bar{T}_2(X, 0) = 0\,, \quad 0 \le X \le 1\,. \tag{37}$$

In crystallization and sublimation of unfrozen moisture, we have

$$\bar{T}_2(1, 0) = 1\,, \quad \bar{T}_2 < \bar{T}_{\text{eu}}(\omega)\,. \tag{38}$$

The temperature determined from (32)–(37) can be written in the general functional form

$$\bar{T} = f[\text{Fo}, \pi_1, \pi_2, Q_{\text{subl}}(\theta)\,, \; Q_{\text{cryst}}(\theta), \bar{T}_{\text{cass}}]\,. \tag{39}$$

The quantity $\bar{T}_{\text{eu}}$ for preparations having a eutectic zone depends on the percentage of unfrozen moisture ω. According to (38), we have crystallization and sublimation at $T_2(x, \theta) \le T_{\text{eu}}$ and crystallization and beginning of melting (unfreezing) at $T_2(x, \theta) \ge T_{\text{eu}}$. In the first period of sublimation drying, the relation

$$\tilde{a} \le \frac{Q_{\text{subl}}}{Q_{\text{cryst}}} \le \tilde{b} \tag{40}$$

is true.

In each interval of the cyclogram (Fig. 2), we have

$$Q_{\text{subl}} = kV_{\text{subl}}(\tau_i - \tau_0) \tag{41}$$

for the sublimation period and

$$Q_{\text{cryst}} = k\int_{\tau_0}^{\tau_i} V_{\text{cryst}}(\tau)\, d\tau \tag{42}$$

for the crystallization period.

The parameter determining the degree of crystallization and sublimation on each ith interval of the cyclogram is as follows:

$$E_i = \frac{Q_{\text{cryst}} - Q_{\text{subl}}}{Q_{\text{cryst}}} = 1 - \frac{V_{\text{subl}}(\tau_i - \tau_0)}{\int_{\tau_0}^{\tau_i} V_{\text{cryst}}(\tau)\, d\tau}\,. \tag{43}$$

The quantities E_i, in developing the optimum cyclogram, have specific values for each product on condition that its high quality is preserved. An analogous parameter can also be introduced for the second period of drying (Fig. 2). For evaluation of the quality of the drying process in the dryer, we obtain (with condition (28))

$$E = \frac{k_{\text{subl}}}{k_{\text{d}}} \frac{\int_{\tau_0}^{\tau_i} V_{\text{subl}}(\tau)\, d\tau}{\int_{\tau_0}^{\tau_i} V_{\text{d}}(\tau)\, d\tau}\,. \tag{44}$$

The maximum value of E_2 characterizes the optimum operation of the dryer.

The basic promising directions in mathematical modeling of the process of sublimation drying with the aim of organizing energy- and resource-saving technologies include:

(1) formulation and solution of conjugate problems of sublimation drying and desublimation, selection of the optimum vacuum and refrigerating equipment, evaluation of the overall dimensions of a dryer, and development of programs for calculation of the type-dimension series of dryers for 15, 25, 50, and 100 kg of evaporated moisture;

(2) development of methods of solution of conjugate nonlinear problems of the vacuum desublimation of a steam for different designs of heat exchangers with a possible regeneration of their heat-exchange surfaces;

(3) creation of algorithms of control of the process of sublimation drying with the use of the optimum number of control parameters.

NOTATION

A, a, b, $\tilde{a}$, $\tilde{b}$, C, d, i, m, n, e, s, $\tilde{\beta}$, and $\tilde{\varepsilon}$, constant coefficients; a_1, thermal diffusivity, m^2/sec; c, heat capacity, J/(kg·K); J, intensity of phase transition, kg/(sec·m^2); k, conversion factor of the heat-flux transducer, W/(m^2·mV); h and l, thicknesses of the product layer and the dry layer, m; L, coordinate of the sublimation surface, m; P, pressure, N/m^2; Q, heat, J; q, specific thermal load, W/m^2; R, universal gas constant; r, latent heat of phase transition, J/kg; T and t, temperature (t, °C and T, K); T, dimensionless temperature; W, moisture content, kg/kg of moisture; V_d, V_{subl}, and V_{cryst}, electric signals of the heat-flux transducers in the processes of desublimation, sublimation, and crystallization, mV; X, x, ξ, coordinates, m; α, heat-transducer coefficient, W/(m^2·K); $\bar{\alpha}$, condensation coefficient; δ, sublimation-zone thickness, m; η, coefficient of kinematic viscosity of a steam, m^2/sec; λ, thermal conductivity, W/(m·K); Π, porosity; τ, time, sec; ω, humidity, %; ρ, density, kg/m^3; $Fo = a_1\tau/l$; $Gu = (T_{subl} - T_{mat})/T_{subl}$; $K_1 = (P_1 - P_{cond})/P_{mat}$; $K_2 = T_r/T_{mat}$; $K_3 = l/h$; $Le = W/W_f$; $\theta = \tau/\tau_f$; $T_1 = (t_1 - t_s'')/(t_{cass} - t_s'')$; $T_2 = (t_2 - t_s'')/(T_{cass} - t_s'')$; $T_{cass} = t(\theta)/(t_{cass} - t_s'')$; $X = x/h$; $\pi_1 = \lambda_2/(c_1\rho_1 h)$; $Q_{subl}(\theta) - Q_{cryst}(\theta) = r_{subl}J_{subl}(\theta)\delta - J_{cryst}(\theta)\delta/(t_{cass} - t_s'')\lambda_1$; $\pi_2 = \lambda_1/\lambda_2$; $T_{eu} = [t_{eu}(\omega) - t_s'']/(t_{cass} - t_s'')$. Subscripts and superscripts: d, desublimation; r, radiation; f, final; cass, cassette; cond, condensation; v, volume; conv, convective; cr, critical; cryst, crystallization; ice, ice; mat, material; surf, surface; subl, sublimation; w, wall; eu, eutectic; eff, effective; med, medium; 0, initial; s, saturation; 1, dry layer; 2, frozen layer; i, running value; ′, in the desublimator; ″, sublimator; ∗, zone VII.

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