a discontinuity. After the transition, A_C takes on the previous value, while the frequency of the self-oscillations remains equal to $4.36 \cdot 10^{-2}$ sec^{-1}.

NOTATION

ω, angular frequency; k, wave number; a, thermal diffusivity; T_T, thermostat temperature; T_C, transition temperature; T_0, the difference between the transition temperature and the thermostat temperature; T(x), the deviation of the temperature at point x from T_T; x, coordinate; t, real time; τ, normalized time; λ, thermal conductivity; ρ, density; u_1 and u_2, voltages; P, power; S, area; q, specific heat of transition; u_0, reference voltage; α, thermo-electromotive force coefficient; σ, Heaviside function; x_0, coordinate of the thermocouple; x_B, coordinate of the phase boundary (stationary component); $\tilde{x}_B$, the oscillating component of the phase boundary coordinate; ε, the small deviations of the parameters from their critical values; K, the controller gain coefficient; ξ, the dimensionless amplitude of the first harmonic; c_n and b_n, the Lyapunov coefficients; Θ_n and Ξ_n, the n-th harmonics of the temperatures of phases I and II, respectively; δ, thickness of the sample; x_n, the n-th harmonic of $\tilde{x}_B$; A, the generalized gain coefficient; B, the stationary temperature gradient; V(x) and W(x), the spatial parts of the first harmonic of the temperatures in phases I and II; $\varkappa$, φ_j, ψ_j, ℓ_j, μ_j, and D are auxiliary parameters.

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GENERALIZED STEFAN PROBLEM

A. A. Gukhman, A. A. Zaitsev, and B. P. Kamovnikov

UDC 621.56/57

A generalized Stefan problem is considered in which volume heat release during the freezing-out of bound moisture is taken into account. It is shown that the appearance of additional criteria does not prevent obtaining a self-similar solution.

A whole series of problems associated with a change in the aggregate state of a material (freezing, drying, heating, sublimation, and similar problems) can be solved in terms of Stefan model approximations. In accordance with this model the phase separation boundary moves from the periphery into the depth of an object depending on withdrawal of heat from its surface (or the addition of heat to it). It is assumed here that the liberation or absorption of heating during a phase change takes place in an infinitely thin region of the material, namely, on a moving "front" (the phase separation boundary).

Experimental verification of the "frontal" theory yields satisfactory results in those cases involving moisture found in a free state. The situation deteriorates substantially when it becomes necessary to take the effect of bound moisture into account. We consider

Moscow Institute of Chemical Technology. All-Union Polytechnical Correspondence Institute, Moscow. Moscow Institute of Applied Biotechnology.

Translated from Inzhenerno-fizicheskii Zhurnal, Vol. 62, No. 2, pp. 317-324, February, 1992. Original article submitted September 9, 1991.

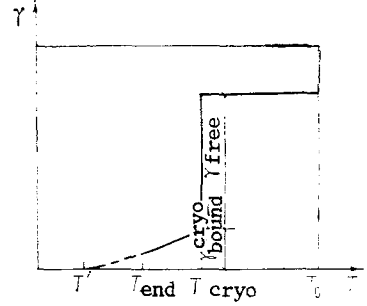

Fig. 1. Dependence of volumetric moisture content of material on the temperature (T_{end}, end temperature of process of freezing, determined by industrial regulation).

this question in more detail using as an example the freezing of moist bodies.

As is well known [1-3], in the process of freezing one can distinguish three characteristic periods: a preliminary cooling of the product, a natural chilling (formation and development of a zone of crystallization), and a terminal freezing. The cooling period continues up to the time that the surface of the object attains a cryoscopic temperature. In the second period the temperature in the chilled zone decreases and in this zone a freezing-out of the remaining moisture takes place. This period terminates when the crystallization "front" reaches the thermal center of the object. In the last stage of the process (up to the time that the material attains a mean-volume temperature level determined through industrial regulation) there is a continuation of the process of freezing-out of the bound moisture.

In the solution of accurately formulated boundary-value problems of this kind the application of various forms of generalized analysis (in particular, the theory of similarity) often proves to be useful. However, from the standpoint of this method, it is usually only the first and second periods that are considered. The third period is either generally ignored [4, 5] or use is made of simplified models [1, 2] or empirical formulations. In this regard, at the second stage the effect of freezing-out the remaining mositure and the associated volume heat release are not infrequently neglected. Moreover, in the directed calculations for effecting a thermal balance it is assumed that the energy consumption of the freezing machinery is completely determined by expenditures of energy in the proper chilling stage and that similar expenditures in the final freezing period are immaterial. As for the very structure of the criterial relationships, it proves to be different for each stage of the freezing process. In this connection it is expedient to obtain a single criterial system for all the periods of this process, taking into account volume heat release owing to the freezing-out of bound moisture.

The general volumetric moisture content of a material (mass of moisture per unit volume) can be represented as a sum of two terms corresponding, respectively, to the volumetric content of the free moisture and of the bound moisture:

$$\gamma = \gamma_{free} + \gamma_{bound}. \tag{1}$$

Here the second term accounts only for the portion of the moisture (comparatively weakly bound with the material) which can take part in phase transitions. Hydrated water is generally excluded from consideration since it does not take part in phase transitions. We assume that crystallization of the free moisture occurs at the moving "front" and that of the bound moisture occurs within the volume of the frozen material depending on the lowering of its temperature below the cryoscopic point.

Without considering the mechanism of interaction of weakly bound water with the material, we use only the well-known fact that with a lowering of the temperature the quantity γ_{bound} decreases monotonically. We now attempt to approximate this behavior by the most rational method. Since in what follows we are presumed to apply a generalized analysis to obtain the most universal form of the solution, containing possibly the fewest number of criteria, the approximating function must also include a minimum number of parameters. This requirement calls for a monomial power term of the form

$$\gamma_{bound} = A\,(T - T')^{\beta}. \tag{2}$$

Here A, T', and β are macroscopic parameters of the frozen material.

It should be noted that this in no way signifies that function (2) approximates the sought-for behavior over the whole interval $T' - T_{cryo}$. Of practical interest is the significantly narrower range of variation of the temperatures (on the order of 15-25 K) close to T_{cryo}. Beyond the limits of this temperature range the temperature curve can go in a completely arbitrary way. To emphasize this situation, in the figure its course in the corresponding region is indicated by a dashed curve. It is known that the bound moisture can be observed even for very low temperatures (below T'). Therefore, the value T' must be regarded as a conditional characteristic not directly connected with the limiting temperature of the remaining moisture present.

The local constituent of the volumetric density of the thermal flow corresponding to liberation of heat owing to the freezing-out of bound moisture can now be represented in the following way:

$$dQ_V = r_{bound} \frac{\partial \gamma_{bound}}{\partial \tau} = r_{bound} \frac{d\gamma_{bound}}{dT} \frac{\partial T}{\partial \tau}. \tag{3}$$

Taking relation (2) into account, we can rewrite Eq. (3) in the form

$$dQ_V = \beta A r_{bound} (T - T')^{\beta-1} \frac{\partial T}{\partial \tau}, \tag{4}$$

and the Fourier equation for the frozen material in the form

$$\frac{\partial T}{\partial \tau} = a_1 \nabla_2 T - \frac{\beta A r_{bound}}{c_1 \rho_1} (T - T')^{\beta-1} \frac{\partial T}{\partial \tau}. \tag{5}$$

The minus sign before the second term on the right side of Eq. (5) stipulates that crystallization is an exothermic process.

Similar considerations, to an equal degree, are also valid for the drying process (thermal, atmospheric, sublimational). Actually, this process can also be divided into three characteristic periods: heating of an object, formation and motion of a dehydrated zone, and completion of drying. Of course, instead of the heat of crystallization, here we must consider heat of vaporization and, during sublimational drying, the heat of sublimation.

From the mathematical point of view all these phenomena are completely identical and lead to the generalized Stefan model. In addition, it whould be noted that here the Stefan problem is a generalized problem, not only and not so much because in its solution use is made of the apparatus of generalized analysis. Of much more importance is the fact that the necessity for taking into account the effect on the process of the presence of bound moisture requires formulation of the problem in its most general form. The classical Stefan problem corresponds to a particular case of it (for $\gamma_{bound} \equiv 0$).

We consider the process of freezing of an unbounded plate of moist material of thickness 2δ, which at time $\tau = 0$ is present in the medium with temperature T_m. The initial temperature of the plate is everywhere the same and equal to T_0 ($T_0 > T_m$). The coefficient of heat transfer from the surface of the plate to the cooling medium, as well as the thermophysical parameters of the material in both zones (frozen and unfrozen), are assumed to be constant. If we introduce the temperature difference

$$\vartheta = T - T_m, \tag{6}$$

then the generalized Stefan problem can be represented by the following system of equations and boundary conditions:

$$\frac{\partial \vartheta}{\partial \tau} = a_1 \frac{\partial^2 \vartheta}{\partial x^2} - \frac{\beta A r_{bound}}{c_1 \rho_1} (\vartheta - \vartheta')^{\beta-1} \frac{\partial \vartheta}{\partial \tau}; \tag{7}$$

$$\frac{\partial \vartheta}{\partial \tau} = a_2 \frac{\partial^2 \vartheta}{\partial x^2}; \tag{8}$$

$$\alpha \vartheta_{wall} - \begin{cases} -\lambda_1 \left(\dfrac{\partial \vartheta}{\partial x}\right), \\ -\lambda_2 \left(\dfrac{\partial \vartheta}{\partial x}\right), \end{cases} \quad \text{for } x = \pm \delta; \tag{9}$$

$$\frac{\partial\vartheta}{\partial x}=0 \text{ for } x=0; \tag{10}$$

$$\vartheta=\vartheta_0 \text{ for } \tau=0; \tag{11}$$

$$\lambda_1\left(\frac{\partial\vartheta}{\partial x}\right)_1-\lambda_2\left(\frac{\partial\vartheta}{\partial x}\right)_2=r\gamma_{\text{free}}\frac{\partial\xi}{\partial\tau};\ \vartheta=\vartheta_{\text{cryo}} \text{ for } x=\xi. \tag{12}$$

The system (7)-(12) differs from its analog for the classical Stefan model [6] by additional terms on the right side of Eq. (7), and also by the fact that in condition (12) on the moving boundary, instead of the total moisture content γ of the material, we have the quantity γ_{free} of the free moisture. To the stages of proper freezing and terminal freezing there corresponds a first alternate version of relation (9), and to the period of preliminary cooling there corresponds a second. Of course, for the cooling and terminal freezing stages one must confine oneself only to the corresponding part of Eqs. (7)-(12).

Thus, as an example of the application of generalized analysis the system of equations (7)-(12) completely encompasses all periods of the freezing process and makes it possible to obtain a criterial relation valid for all stages of this process. It should be noted, however, that for each of these periods we have a very rational system of similarity criteria and reference scale for the variables, obtainable from most general considerations through corresponding simplifications, some of which we consider below:

$$\frac{\vartheta_*}{\tau_*}=a_1\frac{\vartheta_*}{l_*^2}=\frac{\beta A r_{\text{bound}}\vartheta_*^{\beta}}{c_1\rho_1\tau_*};\ \vartheta_*=\vartheta'; \tag{13}$$

$$\frac{\vartheta_*}{\tau_*}=a_2\frac{\vartheta_*}{l_*^2}; \tag{14}$$

$$\alpha\vartheta_*=\lambda_1\frac{\vartheta_*}{l_*};\ l_*=\delta; \tag{15}$$

$$\vartheta_*=\vartheta_0; \tag{16}$$

$$\lambda_1\frac{\vartheta_*}{l_*}=\lambda_2\frac{\vartheta_*}{l_*}=r_{\text{free}}\frac{l_*}{\tau_*};\ \vartheta_*=\vartheta_{\text{cryo}}. \tag{17}$$

If we take as reference scales

$$\vartheta_*=\vartheta_0;\ l_*=\delta;\ \tau_*=\frac{\delta^2}{a_1}, \tag{18}$$

we obtain, from the ten scaling equations, seven criteria of similarity (four of their number being parametric):

$$\frac{a_1}{a_2};\ \frac{\lambda_1}{\lambda_2};\ \frac{\vartheta'}{\vartheta_0};\ \frac{\vartheta_{\text{cryo}}}{\vartheta_0};\ \text{Bi}\equiv\frac{\alpha\delta}{\lambda_1};$$
$$K_1\equiv\frac{\beta A r_{\text{bound}}\vartheta_0^{\beta-1}}{c_1\rho_1};\ \text{Ste}^*\equiv\frac{c_1\rho_1\vartheta_0}{r\gamma_{\text{bound}}}. \tag{19}$$

The corresponding generalized relationship may be put into the form

$$\vartheta_+=\frac{\vartheta}{\vartheta_0}=f\left(\frac{x}{\delta};\ \frac{a_1\tau}{\delta^2};\ \frac{a_1}{a_2};\ \frac{\lambda_1}{\lambda_2};\ \frac{\vartheta'}{\vartheta_0};\ \text{Bi};\ \frac{\vartheta_{\text{cryo}}}{\vartheta_0};\ K_1;\ \text{Ste}^*\right). \tag{20}$$

The relationship obtained differs from the solution of the classical Stefan problem in that here two additional criteria are involved in the number of arguments: K_1 and ϑ'/ϑ_0. Their characteristic feature is that neither one of them includes parametric values of time or extension. But this means that their appearance does not prevent our obtaining self-similar solutions of corresponding degenerate problems [7-9].

The simplest self-similar problem in the classical case corresponds to the Lamé–Clapeyron approximations [6] for a semibounded body. Its natural generalization may be presented in the form

$$\frac{\partial\vartheta}{\partial\tau} = a_1\frac{\partial^2\vartheta}{\partial x^2} - \frac{\beta A r_{\text{bound}}}{c_1\rho_1}(\vartheta - \vartheta')^{\beta-1}\frac{\partial\vartheta}{\partial\tau}; \tag{21}$$

$$\vartheta = \vartheta_{\text{cryo}} \text{ for } x > \xi; \tag{22}$$

$$\vartheta = 0 \text{ for } x = 0; \tag{23}$$

$$\frac{\partial\vartheta}{\partial x} = 0 \text{ for } x \to \infty; \tag{34}$$

$$\vartheta = \vartheta_{\text{cryo}} \text{ for } \tau = 0; \tag{25}$$

$$\lambda_1\frac{\partial\vartheta}{\partial x} = r\gamma_{\text{free}}\frac{d\xi}{\partial\tau}; \ \vartheta = \vartheta_{\text{cryo}} \text{ for } x = \xi, \tag{26}$$

where $\nu = T - T_{\text{wall}}$. Just as in the classical case, we consider boundary conditions of the first kind, and we assume the temperature of the nonfrozen zone to be constant and equal to the initial cryoscopic temperature. The system of equations of the scaling relationships becomes simpler in comparison with relations (13)-(18) and can be rewritten in the form

$$\frac{\vartheta_*}{\tau_*} = a_1\frac{\vartheta_*}{l_*^2} = \frac{\beta A r_{\text{bound}}\vartheta_*^{\beta}}{c_1\rho_1\tau_*}; \ \vartheta_* = \vartheta' = \vartheta_{\text{cryo}}; \ \lambda_1\frac{\vartheta_*}{l_*} = r\gamma_{\text{free}}\frac{l_*}{\tau_*}. \tag{27}$$

There are five equations of scaling relations in the three transformed variables. But if from these relations we eliminate those which lead to the appearance of criteria not involving ℓ_* and τ_*, there then remains only one equation of constraint between the characteristic scales ℓ_* and τ_*. It is known [7] that in this case the problem has a self-similar (similar) solution

$$\vartheta_+ = \frac{\vartheta}{\vartheta_{\text{cryo}}} = f\left(\frac{a_1\tau}{x^2}; \ \frac{\vartheta'}{\vartheta_{\text{cryo}}}; \ K_1; \ \text{Ste}^*\right). \tag{28}$$

If, following tradition [6], we introduce the variable

$$\zeta_+ = \sqrt{x^2/(a_1\tau)} = 1/\sqrt{\text{Fo}_x}, \tag{29}$$

then the fundamental equation of the problem in the partial derivatives of Eq. (21) turns out to be equivalent to the ordinary differential equation

$$\vartheta_+'' + \frac{1}{2}\zeta_+\left[1 + K_1\left(\vartheta_+ - \frac{\vartheta'}{\vartheta_{\text{cryo}}}\right)^{\beta-1}\right]\vartheta_+' = 0. \tag{30}$$

The boundary value problem corresponding to Eq. (30) can be solved numerically. But if we restrict ourselves to a linear approximation for the amount of moisture, putting $\beta = 1$ in expression (2), we can then obtain a solution in analytic form. We have

$$\vartheta_+'' + \frac{K_2}{2}\zeta_+\vartheta_+' = 0; \tag{31}$$

$$\vartheta_+ = 1 \text{ for } \zeta_+ > \xi_+; \tag{32}$$

$$\vartheta_+ = 0 \text{ for } \zeta_+ = 0; \tag{33}$$

$$\vartheta_+' = \frac{\xi_+}{2\text{Ste}^*}; \ \vartheta_+ = 1 \text{ for } \zeta_+ = \xi_+, \tag{34}$$

where $K_1 = 1 + Ar_{\text{bound}}/(c_1\rho_1)$. It follows directly from this [6] that

$$\vartheta_+ = \operatorname{erf}\frac{\zeta_+}{2} \bigg/ \operatorname{erf}\frac{\xi_+}{2}, \tag{35}$$

where the quantity ξ_+ is determined from the relation

$$\frac{\exp(-\xi_+^2/4)}{\operatorname{erf}\xi_+/2} = \frac{\xi_+\sqrt{\pi}}{2\text{Ste}^*}. \tag{36}$$

We can make solution (35), (36) completely identical to that of the classical case [6]. To do this it is sufficient to introduce the concept of volumetric heat capacity of the frozen material

$$c'_{\text{eff}} = c_1\rho_1 + Ar_{\text{bound}}. \tag{37}$$

In addition, a fallout from the equations of problem (31)-(34) is the criterion K_2 since, by the same token, the two scaling relations equations

$$\frac{\vartheta_*}{\tau_*} = a_1, \quad \frac{\vartheta_*}{l_*^2} = \frac{Ar_{\text{bound}}\,\vartheta_*}{c_1\rho_1\tau_*} \tag{38}$$

may be replaced by the single equation

$$\lambda_1\frac{\vartheta_*}{l_*^2} = \vartheta_* c'_{\text{eff}}\,'\tau_*. \tag{39}$$

Of course, all these considerations remain valid even for the more complete formulation of the self-similar generalized Stefan problem for a semibounded body, a classical analog for which was considered in [6].

A solution of the non-self-similar problem (20) also simplifies somewhat if we limit ourselves to a linear approximation and introduce an effective thermal heat capacity of the material. In this case the criteria K_1 and ϑ'/ϑ_0 drop out and expression (20) takes on the form

$$\vartheta_+ = f\left(\frac{x}{\delta};\ \frac{\lambda_1\tau}{c'_{\text{eff}}\delta^2};\ \frac{\lambda_1}{\lambda_2};\ \frac{c'_{\text{eff}}}{c_2\rho_2};\ \frac{\vartheta_{\text{cryo}}}{\vartheta_0};\ \frac{\alpha\delta}{\lambda_1};\ \frac{c'_{\text{eff}}\vartheta_0}{r\gamma_{\text{free}}}\right). \tag{40}$$

It is necessary to note that this type of representation has meaning only for the stages of proper freezing and terminal freezing. As for the period of preliminary cooling of an object, it is here that one applies the ordinary criterial relation

$$\vartheta_+ = f\left(\frac{x}{\delta};\ \frac{a_2\tau}{\delta^2};\ \frac{\alpha\delta}{\lambda_2}\right), \tag{41}$$

in which for the parameters one takes the values corresponding to the frozen state.

The main advantage of relations of the type (40) is that, at the expense of a change of reference scale τ_*, it becomes possible to use well-known results from the theory of heat conduction. For the initial and final stages this solution of the problem relating to the temperature field of a homogeneous body tends toward equilibrium, where in the second case use of the concept of an effective volumetric heat capacity is mandatory. This method makes it possible to apply results of numerical solutions of the classical Stefan problem for the stage of proper freezing. Essentially, this portion of the parametric criteria is involved only in the limiting inequalities and, therefore, cannot turn out to have an influence on the specific form of a function at this or another stage of a process. In particular, this allows us, on the basis of known solutions, to determine the instant that the regular regime of cooling commences, the degeneration of a particular criterion, and similar items. The value of solutions (including analytic solutions), valid for individual periods of a process, diminishes somewhat as a result of the fact that each preceding stage yields, as initial conditions for the succeeding stage, a rather involved expression for the temperature distribution. Nevertheless, use of solutions of this kind can prove to be very useful.

The application of numerical methods to obtain the most complete relation of the type (40) involves no special difficulties. The situation is somewhat more complex with a generalization of the experimental data. Representation of experimental data in generalized form on the basis of a Stefan model makes it possible to compare quantitative characteristics of miscellaneous processes (for example, sublimational drying and freezing). However, not infrequently, in this connection, specific simplifying prerequisites are applied, appropriateness for which must be proven in each individual case.

A whole series of solutions has been obtained on the basis of the so-called quasi-stationary approximation. It is assumed that in the equation of heat conduction the derivative of the temperature with respect to the time can be neglected if the heat of transition is large in comparison with the heat accumulated by the material, i.e.,

$$r \gg c_1\delta T, \tag{42}$$

and, more exactly,

$$r\gamma \gg c_1\rho_1\delta T, \tag{43}$$

where δT is a characteristic temperature difference. For the generalized Stefan problem condition (43) is obviously insufficient. Here the factor accompanying the derivative with respect to the time is the quantity $c_{eff}' \equiv c_1\rho_1 + Ar_{bound}$; therefore, discarding the corresponding term in the Fourier equation is only possible providing the influence of the bound moisture is negligibly weak. In other words, in this case the classical model of Stefan must be considered, but its generalized modification and quasi-stationary approximation in connection with a strong formulation of the problem are generally incompatible.

Of course, this does not mean that it is impossible to take into account the effect of the remaining moisture in the framework of some approximate quasi-stationary model. Such a model can be constructed, for example, on the basis of corresponding balance relationships. However, its consideration would take us beyond the bounds of the present paper.

NOTATION

a, thermal diffusivity; c, heat capacity; Q_V, local component of volume density of thermal flow; r, heat of phase transition; r_{bound}, heat of phase transition of bound moisture; T, temperature; T_{cryo}, cryoscopic temperature; T_0, initial temperature; T_m, temperature of surrounding medium; T_{wall}, wall temperature; x, coordinate; α, heat transfer coefficient; γ, total volume moisture content; γ_{free} and γ_{bound}, total content of free and bound moisture, respectively; δ, plate thickness; ϑ, temperature difference; λ, thermal conductivity; ξ, coordinate of moving "front"; ρ, density; τ, time; $Bi \equiv \alpha\delta/\lambda$, Biot number; $Fo \equiv a\tau/\delta^2$, Fourier number; $Ste^* \equiv c_1\rho_1\delta T/r\gamma_{free}$ and $Ste^{**} \equiv c_{eff}'\delta T/\gamma_{free}$, Stefan number modifications. Indices: *, reference scales of corresponding quantities; +, dimensionless quantities; 1 and 2, parameters of frozen and nonfrozen zones, respectively.

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