

PRESENTATION OF CERTAIN CONJUGATE PROBLEMS
OF HEAT AND MASS TRANSFER DURING PHASE AND
CHEMICAL TRANSFORMATIONS

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The problem of allowing for the kinetics of mass transfer in porous solids in the presentation of certain conjugate problems is considered.

It is well known that conjugate heat-transfer problems [1] involve the simultaneous solution of the heat-conduction equations for the solid and the liquid flowing around it (the velocity distribution in the liquid is determined by solving the corresponding hydrodynamic problem), i. e., the temperature or thermal flux at the solid/liquid interface is not specified in advance but determined from the solution of the problem. This approach is more correct for transient and high-intensity heat transfer [2]. It is a characteristic feature that in this case the solution depends on the properties of both the liquid and the solid around which it is flowing.

In a number of cases involving gas flow around a solid it is interesting to study heat transfer under conditions in which phase or chemical transformations are taking place on the solid surface (especially in sublimation or the combustion of the wall material), or in which some substance differing from the main gas flow passes through a porous surface (in the presence of deep evaporation zones in capillary-porous solids). Such problems are generally considered as conjugate with respect to heat transfer, i. e., the equations of the boundary layer in the gas and the heat-conduction equation in the solid are solved simultaneously [3-5]. However, the mass-transfer equation in the solid is usually absent, and the mass-transfer condition is specified at the gas/solid interface (for example, the condition of equilibrium or nonequilibrium evaporation).

In this article (which is based on investigations carried out in the Mathematical Transport Theory Laboratory, Institute of Heat and Mass Transfer, Academy of Sciences of the Belorussian SSR) we shall show that in certain cases, when formulating such conjugate problems for both polycrystalline and capillary-porous solids, it is essential to consider the mass-transfer equation and also to allow for the kinetics of transfer processes inside the solid.

I. During the high-temperature reaction of carbon on a solid surface the density of the sample was found to alter [6, 7]. The authors in question attributed this effect to the existence of a substantial solid-phase diffusion of atoms from the interior of the solid to its surface. The atoms from inside the solid tended to occupy the vacancies formed in the lattice at the surface as a result of the reaction, so that the vacancies in effect diffused into the interior.

A certain mathematical model was employed in [8] in an attempt to discover the conditions under which a substantial contribution might be made by the diffusive flow of atoms (vacancies) through the solid to the loosening of the structure in the presence of a surface reaction. The boundary condition for the vacancy diffusion equation allows for the fact that, in addition to ordinary evaporation, which corresponds to the complete removal of a surface atom from the crystal, incomplete evaporation processes have to be taken into consideration [9]. Atoms diffusing from the inside of the solid replace vacancies formed on the

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surface, and the velocity of the reaction front diminishes. The vacancy diffusion equation and boundary condition take the form

$$D_v \frac{d^2c}{dx^2} = \frac{c - c_0}{\tau_1}, \quad (1.1)$$

$$\omega_0(c - c_0) - v_0 = D_v \frac{dc}{dx} \Big|_{(x=0)}. \quad (1.2)$$

It follows from the solution to the diffusion equation (1.1) that cases may arise in which the velocity of the reaction front tends to zero, and the material loses mass from inside by the diffusion of the solid atoms to the surface. This arises, in particular, as a result of the fact that real crystals have paths of easy diffusion, associated with the development of networks of boundaries between various elements of their structure, including microcracks and pores, these defects being linked to the outer surface by way of microcapillaries [7, 10, 11]. We thus have to consider the boundary problem of the loosening of a solid, i. e., the change in its density with time. The expression for the density of the diffusive flow of atoms has to include a term allowing for the dependence of the mobility of the atoms on the vacancy concentration [9, 12]. The approximate solution given in [13] for the loosening of material yields qualitative agreement between the calculated surface density and experimental measurements [6].

Thus, a consideration of the vacancy kinetics within the framework of the model employed enables us to make a correct presentation of the problem both for the diffusion (mass transfer) equation and for the energy equation in the solid [14] in the presence of a surface reaction.

II. Now let us consider some questions relating to the transfer kinetics of moisture in vapor form inside a capillary-porous solid in the presence of a deep evaporation zone.

1. As already noted, in certain cases surface diffusion has a considerable influence on transfer processes in porous solids. Experimental investigations into the flow of gases in capillaries under free-molecular conditions reveal a marked difference between the measured conductivity and that calculated by the Knudsen formula. One of the reasons for this difference is surface diffusion [15, 16]. The free-molecular flow of a vapor (with due allowance for surface diffusion) in a cylindrical capillary of radius R bounded on one side ($X = 0$) by the plane surface of the evaporating liquid and connected to a reservoir filled with vapor of the same liquid on the other ($X = L$), may be given an approximate analytical description by means of the following integrodifferential equation for the surface density of the adsorbed molecules n_{ad} as a function of the dimensionless coordinate $x = X/L$ [11]:

$$\frac{1}{EL^2} \frac{d^2 n_{ad}}{dx^2} = n_{ad}(x) - \int_0^1 n_{ad}(\xi) [K_1(|x - \xi|) + \alpha K_1(x + \xi)] d\xi - N_1 \tau K(x) - N_2 \tau [K(1 - x) - \alpha K(1 + x)], \quad (2.1)$$

where τ is the adsorption time; $E = (1/\tau D_S)$, $l = L/R$, α is the specular reflection coefficient of the liquid surface,

$$K(x) = \frac{l^2 x^2 + 2}{2(l^2 x^2 + 4)^{1/2}} - \frac{l}{2} x, \quad K_1(x) = -\frac{dK(x)}{dx}; \quad (2.2)$$

$$N_1 = (1 - \alpha) \frac{p_1}{(2\pi mkT)^{1/2}}, \quad N_2$$

are, respectively, the flows of molecules diffusely emitted by unit area of the liquid meniscus and molecules passing through unit area of the open end from the reservoir.

The boundary conditions have the form [11, 17]

$$\frac{dn_{ad}}{dx} = LV\bar{E}(n_{ead} - n_{ad}); \quad n_{ead} = N_2 \tau, \quad (2.3)$$

$$n_{ad}(0) = n_0. \quad (2.4)$$

It follows from Eq. (2.1) that, on the one hand, the greater the dimensionless parameter $G_L = 1/EL^2$, the more significant is the role of surface diffusion in the distribution of n_{ad} over the surface of the capillary. On the other hand, if $G_L \ll 1$ Eq. (2.1) has a small parameter attached to the leading derivative.

Hence, for arbitrarily small values of G_L regions (boundary layers) usually exist at the ends of the capillary in which it is essential to allow for the differential term in (2.1). In order to obtain an approximate analytical solution for the problem (2.1)-(2.4) the kernel was replaced by the exponential $(1/2) \exp \{-lx\}$ [18].

The resulting flow of molecules at the exit from the capillary equals the sum of the surface diffusion flow $N_S(1)$ and the flow of molecules $N_0(1)$ escaping through the open end of the capillary:

$$N(1) = N_s(1) + N_0(1), \quad (2.5)$$

where

$$N_s(1) = 2\pi R J_s(1) = \frac{2\pi R}{L} D_s \frac{dn_{ad}}{dx}, \quad (2.6)$$

$$N_0(1) = \pi R^2 \{N_1 K_2(1) - N_2 [1 - \alpha K_2(2)]\} + \frac{2\pi R^2 n_{0l}}{\tau} \int_0^1 n_{ad}(x) [K(1-x) + \alpha K(1+x)] dx, \quad (2.7)$$

$$\frac{dK_2(x)}{dx} = -2lK(x).$$

Here we must remember that in (2.5) for $G_R = (1/ER^2) \ll 1$ $N_S \ll N_0$ and for $G_R \gg 1$ $N_S \gg N_0$, i. e., G_R characterizes the ratio of the conductivities due to surface and Knudsen diffusion. It should be noted that, when the gas flow passes around the capillary-porous solid as a whole, the vapor pressure of the evaporating liquid p_2 at the surface of the solid is an unknown quantity entering into N_2 . Thus we have to solve the diffusion equation in the boundary layer and Eq. (2.1) simultaneously.

The nonisothermal case was considered in [19].

2. Many processes of chemical technology involve gas transport in capillary-porous solids, in which phase transformations occur at the sides of the pores. A description of such processes is especially important for engineer's calculations of porous evaporative cooling and calculations of the time required for drying wet materials. Usually when using the model of capillary tubes the transport of moisture vapor is calculated by means of the Knudsen formula for a capillary of infinite length (in the case of a rarefied medium) or from the mutual diffusion equations for a binary vapor-air mixture, with a correction for the Stefan flux (diffusive region of mass transfer), but without allowing for the evaporation from the cells of the capillary or deviations from isothermal conditions [20]. An attempt was made in [21, 22] to give a detailed description of the transport of vapor for the case of free molecular flow in a narrow capillary of length L , allowing for evaporation not only from the meniscus of the liquid, but also from the side of the capillary in the presence of a temperature gradient along the walls of the latter.

The number of molecules evaporating from unit surface per unit time is

$$j(\sigma, T) = \sigma \frac{p_e(T)}{(2\pi mkT)^{1/2}},$$

where p_e is the saturated vapor pressure, σ is the evaporation (condensation) coefficient. Let us assume that the flow of molecules escaping from unit surface contains a proportion σ of evaporating molecules and a proportion $(1 - \sigma)$ diffusely reflected [23]. Then for the flow of molecules escaping from unit lateral surface of the capillary $I(x)$ we obtain the following equation (the flow is referred to the quantity $j(1, T_0) = j_0$; $x = X/L$):

$$I(x) = \sigma \frac{p_e(T)}{p_e(T_0)} \left(\frac{T_0}{T}\right)^{1/2} + (1 - \sigma) \left[\int_0^1 I(\xi) K_1(|x_1 - \xi|) d\xi + N_0 K(x) \right] \quad (2.8)$$

Here the first term describes the evaporating molecules, the second describes those incident upon unit area of the lateral surface (the integral term) and bottom ($N_0 K(x)$), subsequently being reflected (for simplicity we assume that the external medium is a vacuum):

$$N_0 = \sigma_0 + 2l(1 - \sigma_0) \int_0^1 I(\xi) K(\xi) d\xi. \quad (2.9)$$

If the surface temperature of the capillary varies in accordance with the law $T(x) = T_0(1 + Ax)$ and $|A| \ll 1$, the pressure p_e may be written as follows:

$$p_e(x) = B \exp \left\{ -\frac{Q}{kT(x)} \right\} = B \exp \left\{ -\frac{Q}{kT_0} (1 - Ax) \right\}. \quad (2.10)$$

Substituting (2.10) into (2.8) and once again replacing the kernels K and K_1 by exponentials, we obtain an approximate analytical solution for Eq. (2.8). In particular, for the flow of molecules at the outlet from the capillary

$$N = j_0 \pi R^2 \left[N_0 K_2(1) + 2l \int_0^1 I(x) K(1-x) dx \right]$$

in the particular case $\sigma = 1$ and $l \gg s = (QA)/(kT_0)$ we have

$$N = j_0 \pi R^2 \left(1 - \frac{s}{l} \right) \exp \{s\} \approx j_0 \pi R^2 \exp \{s\},$$

i. e., N tends to a value independent of l . In the isothermal case ($s = 0$) we obtain $N = j_0 \pi R^2$. A similar result was explained qualitatively in [24].

Let us determine the dimensionless resultant flux of molecules on the lateral surface of the capillary:

$$J(x) = I(x) - I^-(x) = \sigma [\exp(sx) - I^-(x)], \quad (2.11)$$

where I^- is the flux of molecules falling on unit lateral surface, which is determined from the equation

$$I^-(x) = \int_0^1 I(\xi) K_1(|x - \xi|) d\xi + N_0 K(x).$$

The behavior of the function $J(x)$ is determined by the dimensionless parameters s and l . In particular, for $l = s$ and $l < 1$, $J(0) > 0$ and $J(1) > 0$, i. e., evaporation occurs along the whole length of the capillary, while for $l > 1$, $J(0) < 0$, $J(1) > 0$, i. e., for an increasing value of x , a transition from condensation to evaporation takes place. For $l = -s$ the vapor migration mechanism will be different from $l = s$. Thus the direction of the temperature gradient has a considerable effect on evaporation and condensation processes in the capillary.

We note that the problem here considered is easily generalized to the case in which the pressure of the external medium differs from zero (i. e., when it is not a vacuum). Yet another term outside the integral then appears in Eq. (2.8). Once again we now have to consider an equation of type (2.8) and the system of equations of the boundary layer above the capillary-porous solid at the same time.

3. Apart from the model of capillary tubes, the pseudogas model [25, 26] is often employed in order to describe transfer processes in porous solids. In this model the porous solid-gas system is considered as a binary mixture of gases, the molecules of one of these being stationary and having a size and weight much greater than those of the real molecules.

An attempt was made in [27] to describe the flow of gas in a highly dispersed porous solid in the intermediate pressure range, with phase transformations at the pore walls, the ratio of the thickness of the porous layer to the range being assumed arbitrary, i. e., in the case under consideration diffusion theory was inapplicable (the problem of the flow of vapor through a dry layer of porous solid in which evaporation only took place from the bottom of the layer was considered in [21]).

The probability that a molecule will traverse a distance x without any collisions and will then collide with another molecule in a distance dx is

$$W = \exp \left\{ -\frac{x}{\lambda} \right\} \frac{dx}{\lambda}, \quad \frac{1}{\lambda} = \frac{1}{\lambda_1} + \frac{1}{\lambda_2}, \quad (2.12)$$

where λ_1 is the free path for the case of a molecule-molecule collision, i. e., the ordinary hydrodynamic free path; λ_2 is the free path for a molecule-"stationary molecule" collision. It may be shown that

$$\lambda_2 = \frac{2}{3} \frac{\Pi}{1 - \Pi} d.$$

Let Φ_1 and Φ_2 be the number of molecules appearing in unit volume per unit time as a result of intermolecular collisions and the collisions of molecules with solid particles (including the sublimation of the actual porous solid). Allowing for (2.12) and the assumptions made when deriving the Peierls integral equation [28], we obtain the following equation for the functions Φ_1 and Φ_2 :

$$\begin{aligned} \Phi_1(x) = & 2\tilde{\Phi}(0) E_2\left(\frac{x}{\lambda}\right) \frac{1}{\lambda_1} + \frac{1}{2} \int_0^L \Phi_1(x') E_1\left(\frac{|x-x'|}{\lambda}\right) \frac{dx'}{\lambda_1} + \\ & + \frac{1}{2} \int_0^L \Phi_2(x') E_1\left(\frac{|x-x'|}{\lambda}\right) \frac{dx'}{\lambda_1} + 2\tilde{\Phi}(L) E_2\left(\frac{L-x}{\lambda}\right) \frac{1}{\lambda_1}, \end{aligned} \quad (2.13)$$

$$\begin{aligned} \Phi_2(x) = & \frac{\sigma Z \beta p_e(T)}{(2\pi mkT)^{1/2}} + 2(1-\sigma) \tilde{\Phi}(0) E_2\left(\frac{x}{\lambda}\right) \frac{1}{\lambda_2} + \frac{1}{2} (1-\sigma) \int_0^L \Phi_2(x') E_1\left(\frac{|x-x'|}{\lambda}\right) \frac{dx'}{\lambda_2} - \frac{1}{2} (1-\sigma) \times \\ & \times \int_0^L \Phi_1(x') E_1\left(\frac{|x-x'|}{\lambda}\right) \frac{dx'}{\lambda_2} + 2(1-\sigma) \tilde{\Phi}(L) E_2\left(\frac{L-x}{\lambda}\right) \frac{1}{\lambda_2}, \end{aligned} \quad (2.14)$$

where Z is the number of "stationary molecules" in unit volume, β is the surface area of a "stationary molecule", σ is the evaporation coefficient, $\Phi = \Phi_1 + \Phi_2$ is the total number of particles appearing in unit volume, $\tilde{\Phi}(0)$, $\tilde{\Phi}(L)$ are the flows of molecules from the substrate and from the external medium, respectively.

The flow of molecules escaping from the porous solid is determined as follows:

$$N = 2\tilde{\Phi}(0) E_3\left(\frac{L}{\lambda}\right) + \frac{1}{2} \int_0^L \Phi(x') E_2\left(\frac{L-x'}{\lambda}\right) dx' + \frac{\sigma p_e(T_L)}{(2\pi mkT_L)^{1/2}} (1-\Pi). \quad (2.15)$$

On approximating the kernels in Eqs. (2.13) and (2.14) by exponentials we may obtain an approximate solution for this system of equations. In particular, if:

a) $\lambda_1 \gg \lambda_2$, i. e., the free-molecular mode of flow, and $\tilde{\Phi}(L) = 0$. In this limiting case $\Phi_1 = 0$, and

$$\Phi_2(x_1) = a_1 \exp\{cx_1\} + a_2 \exp\{-cx_1\} + \frac{1}{s^2 - c^2} \cdot \frac{\sigma p_e(T_0)}{(2\pi mkT_0)^{1/2}} - (s^2 - 4l^2) Z\beta \exp\{sx_1\}, \quad (2.16)$$

and

$$c^2 = 4\sigma l^2, \quad l = \frac{L}{\lambda}, \quad x_1 = \frac{x}{L}, \quad c \neq \pm s = \frac{Q\Delta T}{kT_0^2},$$

Q is the heat of vaporization, the coefficients a_1 and a_2 are from the system of algebraic equations given in [27].

For $\sigma = 1$ $\Phi_2 = \sigma Z \beta p_e / (2\pi mkT)^{1/2}$. In this case, on allowing for the exponential approximations of the functions E_2 and E_3 , for the flow (2.15) we have

$$\begin{aligned} N = & \frac{\Pi p_e(T_0)}{(2\pi mkT_0)^{1/2}} \exp\left\{-\frac{3}{2}l\right\} + \frac{1}{4} \frac{Z\beta \lambda_2 p_e(T_0)}{(2\pi mkT_0)^{1/2}} \times \\ & \times \frac{\exp\left\{-\frac{3}{2}l\right\} \left[\exp\left(s + \frac{3}{2}l\right) - 1 \right]}{1 + \frac{2}{3} \frac{s}{l}} + (1-\Pi) \frac{p_e(T_L)}{(2\pi mkT_L)^{1/2}}. \end{aligned} \quad (2.17)$$

From (2.17) for the isothermal case $s = 0$ and on allowing for $Z\beta\lambda_2 = 4\Pi$ we easily obtain

$$N = \frac{p_e}{(2\pi mkT)^{1/2}},$$

i. e., we have the same result as for the capillary.

b) $s = 0$ and evaporation only takes place from the bottom of the dry layer. The resultant flow of molecules at the boundary of the dry layer with the external medium then equals

$$N = \frac{4\Pi\sigma}{(2\pi mkT)^{1/2}} \cdot \frac{p_e - p(L)}{4\lambda + 3L\sigma\Pi}. \quad (2.18)$$

It should be noted that on considering (2.18) as $(\lambda/L) \rightarrow \infty$ we obtain an expression for the flow of molecules corresponding to evaporation from a free surface, while as $(\lambda/L) \rightarrow 0$ we have the Fick law for a diffusive flow.

III. In conclusion, let us assess the influence of the kinetics of the transfer process in certain of the cases considered in Sec. II on the motion of the evaporation front inside the porous solid.

It is well known that in automodel Stefan problems of the phenomenological theory of transfer in both solid [13] and porous [29, 30] materials the velocity of the phase transition v is proportional to $1/\sqrt{t}$. On the other hand, in the case of evaporation from the free surface of the solid into a vacuum when there is no resistance to mass transfer the evaporation rate [9, 31] is

$$v_0 \approx a \exp \left\{ -\frac{Q}{kT_0} \right\}. \quad (3.1)$$

Hence when the evaporation zone penetrates deeply into capillary-porous solids [20] we must study the influence of the transfer kinetics of the vapor passing through the dry layer on the velocity of the phase-transition front.

First let us consider this problem for a single microcapillary of radius R and length $L(t)$ experiencing the free-molecular mode of flow. We shall consider that the time required for the molecule to penetrate through a distance $L(t_1 = L/a)$ is much less than the characteristic time for the progressive motion of the evaporation front ($t_2 = L/v_0$). For the law of motion of the meniscus in a single capillary we may then write the following equation:

$$\pi R^2 n \frac{dL}{dt} = \pi R^2 n v_0 - 2\pi R \sigma \int_0^{L(t)} I(x) K(x) dx, \quad (3.2)$$

where $K(x)$ is the probability that a molecule from the wall will fall on the bottom of the capillary (2.2), and n is the number of particles in unit volume of the solid.

The right-hand side of (3.2) represents the resultant flow of molecules from the evaporating surface. The function $I(x)$ characterizing the flow of particles escaping from unit lateral surface of the capillary is determined from (2.8). Let evaporation take place solely from the bottom of the capillary, while the walls reflect the incident molecules diffusely. Solving (2.8) for $\sigma_0 = 1$, $\sigma = 0$ and using the exponential approximation for the function (2.2), and then substituting the solution into (3.2), we obtain

$$v \equiv \frac{dL}{dt} = \frac{v_0}{1 + \frac{L}{2R}}. \quad (3.3)$$

The solution of Eq. (3.3) takes the form

$$L(t) = 2R \left[\sqrt{1 + \frac{v_0 t}{R}} - 1 \right]. \quad (3.4)$$

We see from (3.3) and (3.4) that for fairly long times ($L \gg R$) $v \sim 1/\sqrt{t}$. On the other hand, for $L = 0$ we obtain $v = v_0$.

Now let us consider the problem of the law of motion of the evaporation front in a porous solid simulated by a "dust-laden gas." On evaporation into a vacuum ($p_L = 0$) in the case of the free-molecular mode of flow, we have the following from (2.18):

$$\left(\sigma = 1; \quad \lambda = \frac{2}{3} \frac{\Pi}{1-\Pi} d \right) N = \Pi n v_0 / \left[1 + \frac{9}{8} (1-\Pi) \frac{L}{d} \right].$$

For the time dependence of L we then have the equation

$$\frac{dL}{dt} = v_0 / \left[1 + \frac{9}{8} (1-\Pi) \frac{L}{d} \right], \quad (3.5)$$

whence

$$L = \frac{\sqrt{\frac{9}{4} v_0 t d (1-\Pi) + d^2} - d}{\frac{9}{8} (1-\Pi)}. \quad (3.6)$$

It follows from (3.5) and (3.6) that for long times ($L \gg d$)

$$v \approx v_0 / \frac{3}{2} \sqrt{\frac{v_0 t}{d} (1-\Pi)}.$$

Thus mass transfer in the capillary has a substantial effect on the evaporation of the liquid, i. e., the thermal problem should be considered using the new $v(t)$.

NOTATION

c , vacancy concentration; c_0 , equilibrium vacancy concentration; τ_1 , relaxation time; D_v , self-diffusion coefficient of the vacancies; w_0 , vacancy mobility [9]; Π , porosity of the solid; d , diameter of a "stationary molecule"; α , velocity of sound; k , Boltzmann's constant; Q , heat of vaporization, $E_k(x) = \int_0^x \mu^{k-2} \exp(-x/\mu) d\mu$; $\tilde{\psi}(0) = p_e(T_0)/(2\pi mkT_0)^{1/2}$.

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