DIFFUSION AND MASS TRANSFER IN MODERATELY CONCENTRATED DISPERSE SYSTEM WITH SMALL PECLET NUMBERS

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Nonsteady diffusion in a system with identical nonconducting spherical particles is considered, taking account of mass transfer with these particles.

Heat and mass transfer in heterogeneous two-phase or two-component systems forms the subject of a broad literature. Significant difficulties, which have yet to be overcome, arise even in studying the simplest problems on the dependence of the effective transfer coefficients (of heat conduction, diffusion) in a steady process on the corresponding coefficients for the phases or components, and also on the concentration and structural characteristics of the system (see, for example, the reviews [1-3]).

The situation is considerably complicated when analyzing nonsteady processes (in this case, the coefficients are not algebraic quantities but operators including differentiation and integration with respect to time [4]), and also when investigating combined processes of heat and mass at the interfaces between the phases or components. In the latter case, there also appear new physical effects — thermodiffusion and diffusional thermal conductivity of the heterogeneous system [5] — and the influence of crowding of the transfer processes on the coefficients of interphase or intercomponent transfer becomes significant.

In this work, the effects of the crowding are investigated, together with the effects of nonsteady conditions and interphase transfer, for a relatively simple diffusional process in a moderately concentrated system of identical spherical particles. The diffusion occurs only in the intervals between particles; the system is regarded as macroscopically homogeneous and steady (ε = const, α = const), and the Peclet number, characterizing the role of convection

This problem is of direct applied interest for diffusion processes in a cloud of droplets or solid particles in the presence of vaporization (sublimation), solution, condensation, or crystallization [7], the drying of disperse materials [8], sorption in granular media [9], the diffusional growth or solution of inhomogeneities in metals and other solid materials [10], mass transfer in a series of biological systems [11], etc.

The investigation is also performed on the basis of powerful methods averaging over the ensemble of particle configurations in the disperse phase, in combination with methods of self-consistent-field theory [12, 13]. These methods were briefly outlined in [14] in their application to processes of the transfer of a scalar characteristic (including heat and impurity mass); they were applied to specific problems, for example, in [4, 5].

Macroscopic Description of the Diffusion Process

According to the results of [12-14], the process of impurity transfer in the intervals between particles is described by the equation

$$\varepsilon \partial c_0/\partial t = -\nabla \mathbf{q} + h, \tag{1}$$

obtained by averaging over the ensemble; for the mean flux $\mathbf{q}$ and source function h in the given case, the following expressions are valid

$$\mathbf{q} = -D_0 \nabla c + D_0 n \int_{r=a} c_0^* \mathbf{n} d\mathbf{r},$$

$$h = -n \int_{r=a} \mathbf{q}^* \mathbf{n} d\mathbf{r}, \quad c = \varepsilon c_0 + \rho c_1. \tag{2}$$

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Here differentiation is performed with respect to the components of the radius vector $\mathbf{R} + \mathbf{r}$ and integration with respect to the surface of an isolated (sample) particle with its center at the point $\mathbf{R}$.

From general considerations of tensor dimensionality, the quantities in Eq. (2) may also be written in the form

$$\mathbf{q} = -D\nabla c_0, \; h = K(c_* - c_0) - Lc_0, \tag{3}$$

where D, K, and L are certain operators, in the general case.

Using Eq. (3), Eq. (1) is formally transformed to give

$$\varepsilon \partial c_0/\partial t = D\Delta c_0 + K(c_* - c_0) - Lc_0. \tag{4}$$

The quantity c_*, which is understood to be the saturated concentration of impurity at the particle surface (see below), depends on the time, in the general case.

The self-consistency condition of the theory, allowing D, K, and L to be determined in the final analysis, is obtained by equating Eqs. (2) and (3). To calculate the integrals in Eq. (2), a special problem regarding the perturbations introduced into the concentration field by the sample particle must be solved. Note that the "mean concentration in the disperse phase" c_1 at the point $\mathbf{R}$ is introduced formally in the given case (cf. [5]) and is the result of averaging the quantity c_0^* over the surface of the sample particle with its center at the given point.

The continuum description of the process using Eqs. (1) and (4) and also Eq. (2) is meaningful in the case when the spatial scale A of the field c_0 is much larger than a, which is what is assumed blow. In this case, the concentration c_0 in the vicinity of the sample particle with its center at point R is described by the Taylor expansion

$$c_0(t, \; \mathbf{R} + \mathbf{r}) = C_0 + \mathbf{Er} + \mathbf{rMr} + \ldots \tag{5}$$

So as to work with algebraic quantities rather than operators, it is expedient to apply a Fourier transformation with respect to the time to all the relations. Then Eq. (4) is replaced by the relation

$$D'\Delta c_0 + G(C_* - c_0) = 0,$$

$$G = K' + L' + i\omega\varepsilon, \; C_* = c_* K'/G. \tag{6}$$

Here the prime is introduced to distinguish the parameters D', K', and L' from the operators D, K, and L; the previous symbols are retained to denote the Fourier transforms of the other quantities. It is clear that Eqs. (2) and (5) retain the same form after the transformation, while in Eq. (3) D, K, and L are replaced by D', K', and L'.

Note that any other linear integral transformation could have been used instead of the Fourier transformation — for example, a Laplace transformation.

From Eq. (6), the following relation exists between the coefficients of the expansion in Eq. (5)

$$\mathrm{Sp}\,\mathbf{M} = -(G/2D')(C_* - C_0). \tag{7}$$

Problem for the Sample Particle

In the general case of a concentrated disperse system, the quantities c_0^* and $\mathbf{q}^*$ appearing in the integrands in Eq. (2) are determined by formulating a problem regarding the impurity diffusion in a hypothetical inhomogeneous medium outside the sample particle, the effective diffusion coefficient in which depends in a special way on the distance to the particle center [12-14]. For a system of moderate concentration, this dependence may be neglected. Then the properties of the hypothetical medium coincide with the mean properties of the disperse system, and the well-known model of an "effective medium" usually used on an empirical basis is obtained [3].

Thus, the following problem is obtained for the Fourier transform of the concentration c_0^*

$$D'\Delta c_0^* + G(C_* - c_0^*) = 0; \; c_0^* \to c_0, \; r \to \infty;$$

$$-D'\mathbf{n}\nabla c_0^* = k(c_* - c_0^*), \; r = a. \tag{8}$$

It is natural to seek c_0^* in the form of the sum $c_0 + \varphi$; then, from Eq. (8), the following problem is obtained for φ

$$\Delta\varphi - s^2\varphi = 0;\ \varphi \to 0,\ r \to \infty;$$

$$\mathbf{n}\nabla(c_0 + \varphi) + \sigma(c_* - c_0 - \varphi) = 0,\ r = a. \tag{9}$$

Here and below, the following notation is used

$$s^2 = G/D',\ m^2 = K'/D',\ \sigma = k/D'. \tag{10}$$

Representing the desired solution of Eq. (9) in the form of a series in spherical functions, and transforming the expansion in Eq. (5) for the field c_0 to the same form, ordinary differential equations with boundary conditions for the coefficients of this series are obtained from Eq. (9).

Taking into account that the contribution to the integrals in Eq. (2) comes only from terms of this series with spherical functions of zero and first rank, the determination of the coefficients is limited solely to these functions. Then

$$\varphi = B_1\left(\frac{a}{r}\right)^{1/2} K_{1/2}(sr) + B_2\left(\frac{a}{r}\right)^{3/2} K_{3/2}(sr)\,\mathrm{Er} + \ldots, \tag{11}$$

where the Macdonald functions have been introduced, together with the coefficient

$$B_1 = (F_{1/2} + \sigma a K_{1/2})^{-1}\left\{\left[\sigma a\left(1 + \frac{m^2a^2}{6}\right) - \frac{m^2a^2}{3}\right](c_* - C_0) + \left(1 - \frac{m^2}{s^2}\right)\left(1 - \frac{\sigma a}{2}\right)\frac{s^2a^2}{3}\,C_0\right\},$$

$$B_2 = [F_{3/2} + (\sigma a - 1)K_{3/2}]^{-1}(1 - \sigma a),$$

$$F_j(sr) = -a\frac{d}{dr}\left[\left(\frac{a}{r}\right)^j K_j(sr)\right],\ \begin{Bmatrix} K_j \\ F_j \end{Bmatrix} = \begin{Bmatrix} K_j(sa) \\ F_j(sa) \end{Bmatrix}. \tag{12}$$

The expression obtained for $C_1 = c_1(i\omega, R)$

$$C_1 = \left[1 + \left(1 - \frac{m^2}{s^2}\right)\frac{s^2a^2}{6}\right]C_0 - \frac{m^2a^2}{6}(c_* - C_0) + B_1K_{1/2}, \tag{13}$$

which allows the mean concentration c to be determined at any point R as the sum $\varepsilon C_0 + \rho C_1$ and then it is possible to find

$$\nabla c = \left\{1 + \rho\left[\frac{s^2a^2}{6} - \frac{\sigma a - (1 - \sigma a/2)\,s^2a^2/3}{F_{1/2} + \sigma a K_{1/2}}\,K_{1/2}\right]\right\}\mathrm{E}. \tag{14}$$

In calculating Eqs. (12)-(14), expressions for C_* and SpM are taken from Eqs. (6) and (7) and the definitions of Eq. (10) are used.

Using Eq. (14), and calculating the integrals in Eq. (2) using Eqs. (11) and (12), and then equating the resulting expressions for q and h with the corresponding representations in Eq. (3), the following system of nonlinear equations is obtained for the unknowns D', K', and L'

$$K' = \frac{3\rho k}{a}\left[1 + \frac{m^2a^2}{6} - \frac{\sigma a - (1 - \sigma a/2)\,m^2a^2/3}{F_{1/2} + \sigma a K_{1/2}}\,K_{1/2}\right],$$

$$L' = \rho k a(s^2 - m^2)\left[\frac{1}{2} + \frac{(1 - \sigma a/2)K_{1/2}}{F_{1/2} + \sigma a K_{2/2}}\right],$$

$$\frac{D'}{D_0} = 1 + \rho\left[-1 + \frac{s^2a^2}{6} - \frac{\sigma a - (1 - \sigma a/2)\,s^2a^2/3}{F_{1/2} + \sigma a K_{1/2}}\,K_{1/2} + \frac{(\sigma a - 1)K_{3/2}}{F_{3/2} + (\sigma a - 1)K_{3/2}}\right]. \tag{15}$$

As a result of solving this system, the quantities D', K', and L' (which, generally speaking, are complex) may in principle be expressed in the form of functions of the physical parameters and $i\omega$, which is completely determined by the relation in Eq. (6). Then, to obtain the diffusion equation in the form in Eq. (4), an inverse Fourier transformation must be used.

It is expedient to introduce, in addition to Eq. (10), the dimensionless time τ and the parameters

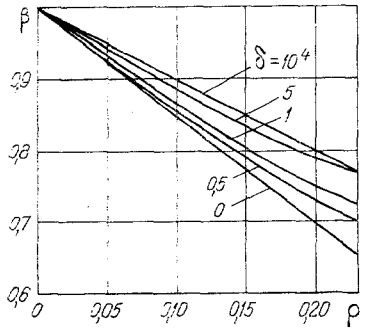

Fig. 1. The dependence of $\beta = D/D_0$ on ρ for various δ.

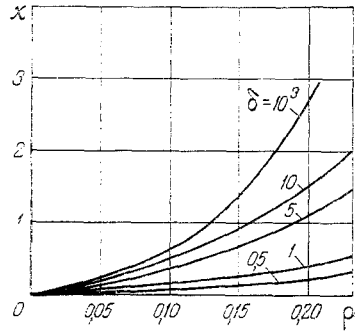

Fig. 2. The dependence of x on ρ at various δ.

$$x = K'\tau,\ y = L'\tau,\ z = D_0/D',\ p = i\omega\tau\varepsilon,\ \delta = ka/D_0,\ \tau = a^2/D_0 \tag{16}$$

in which Eq. (15) is written, after transformations using the well-known analytical expressions for the Macdonald functions, in the form

$$x = 3\rho\delta\left[1 + \frac{zx}{2} + \left(1 + \frac{zx}{6}\right) sa\right](1 + sa + z\delta)^{-1},$$

$$y = (\rho/2)\, z\delta\,(y + p)\,(3 + sa)\,(1 + sa + z\delta)^{-1},$$

$$\frac{1}{z} = 1 + \rho\left\{-1 + \left[1 + \frac{(sa)^2}{2} + \left(1 + \frac{(sa)^2}{6}\right) sa\right](1 + sa + z\delta)^{-1} - \right.$$

$$\left. - [3\,(1 + sa) + (sa)^2]\,[(2 + z\delta)\,(1 + sa) + (sa)^2]^{-1}\right\},$$

$$sa = [z\,(x + y + p)]^{1/2}. \tag{17}$$

The problem of describing the diffusional process evidently reduces to investigation of the system in Eq. (17).

Steady Process

In this case, D and K are algebraic quantities (the prime in the notation may be omitted), $p = 0$, $y = 0$, $sa = \sqrt{zx}$. Two equations are obtained from Eq. (17) for the unknowns x and z

$$x = 3\rho\delta\left[1 + \frac{zx}{2} + \left(1 + \frac{zx}{6}\right)\sqrt{zx}\right](1 + \sqrt{zx} + z\delta)^{-1},$$

$$\frac{1}{z} = 1 + \frac{x}{3\delta} - \rho\left[1 + \frac{3\,(1 + \sqrt{zx}) + zx}{(2 + z\delta)\,(1 + \sqrt{zx}) + zx}\right]. \tag{18}$$

It is of interest to investigate Eq. (18) in the limiting cases $\delta \ll 1$ and $\delta \gg 1$, when the mass transfer is limited by kinetic effects and diffusion, respectively. When $\delta \ll 1$, x and $\beta = 1/z$ may be sought in the form of power series in δ; it is evident from Eq. (18) that $x \sim \delta$, $\beta \sim 1$. The equations for the coefficients of these series are obtained in the standard manner from Eq. (18). Retaining only the first two terms, computations lead to the expression

$$x \approx 3\rho\,(1 - \delta)\,\delta,\ \beta \approx 1 - \frac{3\rho}{2} + \frac{\rho}{4}\,\frac{9\rho - 1}{1 - 3\rho/2}\,\delta. \tag{19}$$

When $\delta \gg 1$, $x \sim z \sim 1$. In this case, x and β are sought in the form of series in inverse powers of δ and, instead of Eq. (19), the expression obtained is

$$x \approx \varepsilon f\left\{1 - \frac{1}{\delta}\left[\varepsilon\,(1 + \sqrt{f})\,T - \frac{f}{3} + \rho\left(3 + \frac{f}{1 + \sqrt{f}}\right)\right]\right\},$$

$$\beta \approx \varepsilon\left\{1 - \frac{1}{\delta}\left[\rho\left(3 + \frac{f}{1 + \sqrt{f}}\right) - \frac{f}{3}\right]\right\}, \tag{20}$$

where f is the solution of the equation

$$f = 3\rho\left[1 + \frac{f}{2} + \left(1 + \frac{f}{6}\right)\sqrt{f}\right],$$

$$T = \left[1 - \frac{3}{2}\rho\left(1 + \frac{1}{\sqrt{f}} + \frac{\sqrt{f}}{2}\right)\right]^{-1}. \tag{21}$$

The quantities K and D are expressed in terms of x and $\beta = 1/z$ using Eq. (16). The dependences of x and β on ρ at different δ obtained numerically from Eq. (18) and also the limiting dependences in Eqs. (19) and (20) are shown in Figs. 1 and 2.

The effective diffusion coefficient thus depends on the intensity of the interphase mass transfer, which is a new property specific to heterogeneous disperse systems. With increase in δ from zero to infinity, it rises slowly at any $\rho \neq 0$ from the value $(1-3\rho/2)D_0$ in a system of neutral particles obtained earlier from the effective-medium method to the value εD_0 in a system of particles with a fixed concentration at their surfaces, which follows formally from the concept of perfectly identical topological properties of the phases of a heterogeneous system.

The effective coefficient of interphase transfer referred to unit volume of mixture is found to differ from the value which would be obtained with simple summation of the contributions from all the particles in unit volume, under the assumption that the kinetic and diffusion coefficients are k and D_0, respectively. Taking into account that the quantity k should not depend on the crowding, this effect is described by introducing the new diffusion coefficient D_e strictly specific to the mass-transfer process. Equating the total flux of n particles having the previous value of k and $D = D_e$ with the value $K(c_* - c_0)$ gives

$$\frac{ka/D_0}{1 + ka/D_e} = \frac{x}{3\rho}, \quad \frac{D_e}{D_0} = \left(\frac{3\rho}{x} - \frac{1}{\delta}\right)^{-1}. \tag{22}$$

In particular, it follows from Eqs. (19) and (20) that

$$D_e \approx D_0,\ \delta \ll 1;\ D_e \approx (\varepsilon f/3\rho)D_0,\ \delta \gg 1. \tag{23}$$

At small ρ, it follows from Eq. (21) that $f \approx 3\rho(1 + \sqrt{3\rho})$, i.e., $D_e/D_0 \approx 1 + \sqrt{3\rho}$. Thus, the influence of crowdedness on the effective values of the diffusion coefficient determined from the rate velocity of longitudinal "transitional" diffusion in the system and from the rate of interphase mass transfer is found to be completely different. The dependence of D_e/D_0 on ρ at various δ is shown in Fig. 3. As in the case of large Peclet numbers [6], the crowdedness of the process intensifies the interphase transfer.

Nonsteady Process

For simplicity, only the limiting cases $\delta \to 0$ and $\delta \to \infty$ will be considered. It may be shown that Eq. (4) is differential only at sufficiently small frequencies. It is therefore assumed that $|p| \ll 1$.

In the case when the mass transfer is limited by kinetic effects ($\delta \to 0$), it follows from Eq. (17) that $x \approx y \approx 0$, $sa \approx \sqrt{zp}$. In this case, retaining terms of order up to p, Eq. (16) and the third relation in Eq. (17) lead to the result

$$\beta = \frac{D'}{D_0} \approx 1 - \frac{3\rho}{2} + \frac{3\rho\varepsilon}{4(1 - 3\rho/2)} i\omega\tau, \quad \tau = \frac{a^2}{D_0}. \tag{24}$$

This relation corresponds to the following relaxational equation, corresponding to the first relation in Eq. (3)

$$\left[1 - \frac{3\rho}{2} + \frac{3\rho\varepsilon}{4(1 - 3\rho/2)}\tau\frac{\partial}{\partial t}\right]\nabla c_0 \approx -\frac{q}{D_\theta}. \tag{25}$$

Substituting Eq. (24) into Eq. (6) and performing in an inverse Fourier transformation, simple computations lead, within the limits of the adopted accuracy, to the result

$$\varepsilon\left[1 - \frac{3\rho\varepsilon}{4(1 - 3\rho/2)^2}\tau\frac{\partial}{\partial t}\right]\frac{\partial c_0}{\partial t} \approx \left(1 - \frac{3\rho}{2}\right)D_0\Delta c_0. \tag{26}$$

This equation is obviously of elliptical type.

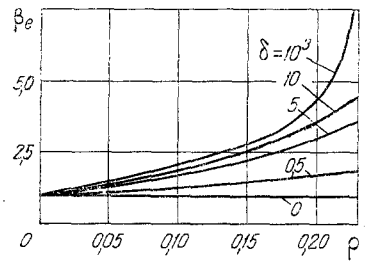

Fig. 3

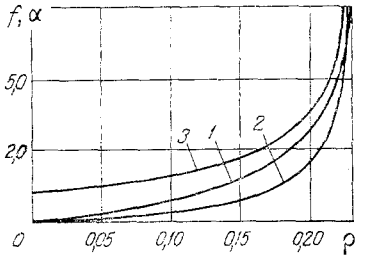

Fig. 4

Fig. 3. Dependence of $\beta_e = D_e/D_0$ on ρ at various δ.

Fig. 4. Dependence of the coefficient in Eq. (28) on ρ: 1) f; 2) α; 3) $1 + \alpha + \alpha'$. The value of f is determined only when $\rho \leqslant 0.231$, which is associated with the use of the approximation of a moderately concentrated disperse system.

Thus, it follows from Eq. (25) that the gradient of the mean concentration relaxes to its steady value, corresponding to the given mean impurity flux q, and not vice versa, as is fairly often assumed for processes of heat and mass transfer in relaxing media, and leads not to Eq. (26) but to the hyperbolic equation of heat conduction or diffusion (see [15, 16] and also [17, 18], where analogous conclusions are reached from general thermodynamic considerations).

Note that Eqs. (25) and (26) are only applicable to sufficiently slow processes of characteristic frequency $\omega \ll 1/\tau$. Further refinement of the results for small p leads to the appearance in Eq. (24) of a term proportional to $(i\omega)^{3/2}$; in this case, Eqs. (25) and (26) become integrodifferential, as shown for the example of the problem of heat transfer in a disperse system in [4].

The situation when the mass transfer is limited by diffusion ($\delta \to \infty$) is now considered. It follows from the third relation in Eq. (17) that the dispersion D' is absent in the given case, i.e., $D' = \varepsilon D_0$. Assuming that $|p| \ll \varepsilon f \sim \rho$

$$x \approx \varepsilon (f + \alpha i\omega\tau), \quad y \approx \varepsilon\alpha' i\omega\tau,$$

$$\alpha = \frac{3\rho}{2\sqrt{f}}\left(1 + \frac{f}{6}\right)(1+\alpha')T, \quad \alpha' = \frac{\rho}{2}\left[\frac{1}{3+\sqrt{f}} - \frac{\rho}{2}\right]^{-1}, \tag{27}$$

where f, as before, is determined from Eq. (21).

In this case, Eq. (4) takes the form

$$(1+\alpha+\alpha')\frac{\partial c_0}{\partial t} \approx D_0 \Delta c_0 + f\frac{c_* - c_0}{\tau} + \alpha\frac{dc_*}{dt}. \tag{28}$$

Thus, with a fixed impurity concentration, there is no relaxation of the mean-concentration gradient, in general, at the surface of the disperse-phase particles, but the dispersion of the quantities K' and L' leads to new effects, in particular, it is as if the volume of space between the particles which is accessible to the diffusing impurity is increased; the rate of change of the surface concentration also begins to play a role. The coefficients of Eq. (28) are shown in Fig. 4.

Equation (28) is only valid for processes of characteristic frequency satisfying the condition $\omega \ll \rho/\tau$. Taking the next terms in the expansions of Eq. (27) in powers of $i\omega$ again leads, as is readily shown, to an elliptical diffusion equation.

NOTATION

A, spatial scale of the concentration field; a, particle radius; B_i, coefficients in Eq. (11); c_0, c_1, c, mean concentrations in the intervals between particles, in the particles, and in the mixture (the last two quantities are formally introduced); c_*, concentration at the particle surface; D, D', operator of the diffusion coefficient and its Fourier transform; D_0, the molecular diffusion coefficient of the impurity; D_e, effective diffusion coefficient de-

termining the rate of mass transfer; C_*, G, quantities defined in Eq. (6); f, root of Eq. (21); h, source function; k, kinetic coefficient; K, L and K', L', operators in Eq. (3) and their Fourier transforms; C, **E**, **M**, coefficients of the expansion in Eq. (5); n, numerical concentration of the particles; $p = i\omega\tau\varepsilon$; **q**, mean impurity flux; **R**, **r**, radius vectors; m, s, quantities introduced in Eq. (10); x, y, z, parameters introduced in Eq. (16); α, α', coefficients in Eq. (27); $\beta = 1/z$; $\delta = k\alpha/D_0$; $\varepsilon = 1-\rho$; ρ, volume concentration of particles in the system; $\sigma = k/D_0$; $\tau = a^2/D_0$; φ, perturbation of the field c_0 close to the sample particle; ω, frequency (parameter of the Fourier transformation); an asterisk denotes quantities pertaining to the sample particle.

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