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NON-STEADY-STATE SORPTION PROCESSES IN MICROHETEROGENEOUS DISPERSED SYSTEMS*

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The article formulates and solves the problem of asymptotic rapid mass transfer in microheterogeneous dispersed systems.

It is accepted practice to characterize steady-state transfer of a dissolved active component (AC) through a motionless layer of a liquid or solid dispersion by the diffusion coefficient D_*. This is formally determined as the material coefficient in Fick's first law $J = -D_*\nabla c_D$, where c_D is the local concentration of the AC in the dispersed system. However, experiments with some dispersed systems type molecular sieve [1, 2] or hard polymers [3] show that Fick's second law $\nabla\cdot(D_*\nabla c_D) = D_t c_D$ does not always provide an adequate description of the non-steady-state sorption processes in dispersed systems. An analogous conclusion was reached in the study of non-steady-state heat exchange in dispersed systems [4]. The deviations are of relaxational nature. We will henceforth characterize such processes by the material parameters of the dispersed system, viz., the relaxation time λ_S.

Deviations from the predictions of the classical theory and from the actual occurrence of sorption processes may be expected in dispersed systems for which the diffusion coefficient of the dispersed phase is lower than the diffusion coefficient of the continuous phase, but the sorptivity is concentrated in the dispersed phase. The deviations are particularly great upon sudden or very rapid (compared with λ_S) changes in the concentrations of the diffusing component in the dispersed system. Relaxation phenomena, noted in molecular sieves and other solid substances with polydispersed internal structure [1], may manifest themselves in liquid dispersed systems with analogous properties during rapidly occurring processes of mass transfer, e.g., in electrochemical measurements of the diffusion coefficients [5] or in their determination by methods of a free jet, wetted wall [6], or in industrial processes of contacting gases with suspensions.

The present work presents the asymptotic description of very rapid concentration processes in dispersed systems, i.e., it examines a special, asymptotic case of more general theories [1, 2]. It was found that such asymptotic models can be formulated independently of the internal geometric structure of the medium which in the models is represented only by the specific volume of the dispersed phase φ and its specific surface ψ. In view of the variety of physical situations, the transport model for different types of dispersed systems is presented in a more general form on the basis of the concepts of microheterogeneous dispersed systems.

Microheterogeneous Dispersed Systems. They have an internal structure whose microscopic scale is fairly large compared with the molecular dimensions. Yet it is still small compared with the typical macroscopic dimensions of test specimens [7]. Separate microscopically distinguishable details of dispersed systems may be viewed as homogeneous volumetric phases of a heterogeneous polyphase system. It is expedient to study the macroscopic behavior of dispersed systems as one entity by methods of the physics of the continuum. A similar dualism of the physics of the microheterogeneous continuum manifests itself particularly

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distinctly in the fact that an infinitesimal material element or material point of the dispersed system contains a sufficient number of elements of the microheterogeneous structure, e.g., particles of the dispersed phase, pores, etc. It is consequently expedient to introduce simultaneously magnitudes of the same type concerning the individual phases (concentration, diffusion flows, etc.) into the material point. We confine ourselves to the case of stable, statistically homogeneous and isotropic two-phase dispersed systems with a single continuous medium in which particles of a homogeneous material are scattered. We will deal with non-steady-state diffusion of the chemical component (AC) that is present in both phases of the microheterogeneous dispersed system. The course of non-steady-state diffusion under such conditions is decisively influenced by two fundamental processes:

1) the internal transfer of AC between the dispersed and the continuous phases characterized by instantaneous local volumetric intensity of the transition into the dispersed phase q within the material element of the dispersed system;

2) external transfer of AC between adjacent material elements of the dispersed system characterized by the instantaneous local diffusive flux J.

The overall content of AC in the dispersed system is characterized by the summary concentration c_D, the full content of AC in the continuous and in the dispersed phases:

$$c_D = \varphi c_S + (1-\varphi)\, c_F\,, \tag{1}$$

where c_S, c_F are the concentrations of AC in both phases. For isochoric systems the following local relationships apply

$$D_t c_D = -\nabla\cdot J + \varphi r_S + (1-\varphi)\, r_F, \tag{2}$$

$$\varphi D_t c_S = q + \varphi r_S, \tag{3}$$

where r_S, r_F are the volumetric production intensities due to the chemical reaction in individual volumetric phases. The operator of the convective derivative $D_t = (\partial_t + v\cdot\nabla)$ was introduced for a continuum moving at the local speed v, which is common to the continuous and the dispersed phases.

Mass transfer in the dispersed phase is fully determined by the concentration processes in the vicinity of the particles. The local instantaneous values of the magnitudes J, q, r_F, r_S can therefore be determined from the prehistory of the concentration field c_F and the velocity field v in the elementary circle circumscribing the material point. In accordance with such a notion it is expedient to unify the relationships (1)-(3) in the balance of AC for the continuous phase. After J, q, r_F are expressed by the corresponding definitions, the balance refers to the sole field c_F:

$$-\nabla\cdot J = (1-\varphi)\, D_t c_F + q - (1-\varphi)\, r_F. \tag{4}$$

The influence of the "internal" chemical reaction in the dispersed phase is already included in the expression for the intensity of the internal transfer q.

In the case of a two-phase dispersed system consisting of Fick continua with constant diffusion coefficients D_F, D_S and with constant equilibrium distribution coefficients H,

$$c_S = Hc_F \quad \text{(equilibrium)}, \tag{5}$$

corresponding to the functional of the memory [8] expressing the dependences of J, q, r_F on the prehistory of the field c_F; they are fully determined by the internal geometric structure of the dispersed system and the material constants D_S, D_F, H. We assume that even in the case of deviations from the internal concentration equilibrium, the external diffusive flux J can be expressed by Fick's first law with an effective diffusion coefficient that does not depend on the history of the concentration processes. It is also accepted that in a continuous medium the resistance to internal transfer of AC may be neglected. For the case of absence of chemical reactions ($r_S = r_F = 0$), transport models type (4) in connection with the dynamics of sorption processes in molecular sieves, bidisperse catalysts [1, 2], hard polymers [3], etc., were formulated and investigated. The assumption was used that the dispersed phase consists of identical particles with simple shape. We will deal with asymptotic problems of non-steady-state diffusion without a chemical reaction ($r_S = r_F = 0$) when it suffices to characterize the internal geometry of the dispersed system by two parameters: specific volume (volumetric fraction) φ and specific surface ψ of the dispersed phase.

Intensity of Internal Transfer of AC. The transition of AC into particles of the dispersed phase depends on the diffusion resistance of the surrounding continuous medium as well as on the resistance inside the particle. However, if $\alpha \ll 1$, where

$$\alpha = HD_S/D_F, \tag{6}$$

then the resistance of the continuous medium compared with the internal interphase transfer of AC may be neglected [9]. The concentration of AC on the surface of the particles is then determined by the equilibrium relationship (5), and the change in the mean concentration of AC in the particle, in accordance with the classical theory of non-steady-state diffusion [9], can be expressed by the corresponding Duhamel convolution integral in dependence on the concentration history in the surrounding continuous medium [2, 9]. For the geometrically similar particles in this integral, the relaxation time λ_S appears:

$$\lambda_S = R_S^2/D_S \tag{7}$$

as the only characteristic parameter. It is expedient to determine the equivalent particle radius R_S as the ratio of the volume to the surface of the particles, i.e.,

$$R_S = \varphi/\psi. \tag{8}$$

The convolution integrals for particles with different shapes have identical asymptotes of very slow and very fast processes [10, 11]. Asymptotic approximations of the convolution integrals for very fast and slow processes (comparable with λ_S) can be obtained by a formal method [11] or on the basis of physical intuition.

When the changes in concentration on the surface of the particles of the dispersed phase are sufficiently slow, it may be expected that the equilibrium inside the particles has time to stabilize. Conversely, in very fast processes, even substantial changes in concentration on the surface of the particles will manifest themselves only in a very thin surface layer of the particle. In this case, the particles of the dispersed phase may be regarded as a semi-infinite space bounded by a plane interphase boundary with surface ψ in the unit volume of dispersion [11, 12].

For the volumetric intensity of internal mass transfer of AC between phases in the approximation of asymptotically slow concentration processes, we may postulate the relationship

$$q = HD_t c_F \quad \text{(equilibrium asymptote)} \tag{9a}$$

and for asymptotically fast processes

$$q = H\lambda_S^{-1/2} D_t^{1/2} c_F \quad \text{(penetration asymptote)} \tag{9b}$$

where the complex

$$D_t^{1/2} c = \pi^{-1/2} D_t \left(\int_0^t (t-s)^{-1/2} \{c(s) - c(0)\}\, ds \right) \tag{10}$$

is the convective modification of the semidifferential operator [11, 12, 13].

Local Diffusive Flux of AC. For the type of dispersed system under examination, it can be shown by the methods of statistical physics [7] that Fick's first law remains valid under steady-state conditions of the process or if local internal equilibrium is maintained between the continuous and the dispersed phases:

$$J = -D_* \nabla c_F. \tag{11}$$

The effective diffusion coefficient D_* depends on the detailed internal geometry of the dispersed system. In case $\alpha < 1$, the upper estimate, according to [7], is given by the ratio

$$D_* = D_F \frac{2 - 2\varphi + \alpha(1 + 2\varphi)}{2 + \varphi + \alpha(1 - \varphi)}. \tag{12}$$

In the case of asymptotically fast processes, with $\alpha \ll 1$, the penetration of AC into the particles of the dispersed phase may be neglected. From the point of view of the total diffusive flux, the dispersed phase behaves like impermeable ballast material. In accordance with (12), this position is met by the corresponding asymptote for $\alpha = 0$, whereas the mean equilibrium concentration for the corresponding steady-state process with $c_S = 0$ is equal to $(1 - \varphi) c_F$:

$$J = -(1 - \varphi) D_0 \nabla c_F, \tag{13}$$

$$D_0 = D_F (1 - \varphi)/(1 + \varphi/2). \tag{14}$$

Asymptotic Transport Models. If the local values of c_F change so little with time that for equalizing the concentration field in the particle the internal diffusion suffices,

$$|D_t \ln c_F| \ll \lambda_S^{-1}, \quad t \in \langle \theta - \lambda_S;\ \theta \rangle, \tag{15}$$

then we consider such a process asymptotically slow [11]. For such processes, the substitution of the corresponding determining relationships (9a), (11) into (4) leads to the quasiequilibrium model of diffusion:

$$D_* \nabla^2 c_F = H_* D_t c_F. \tag{16}$$

The parameter

$$H_* = 1 - \varphi + \varphi H \tag{17}$$

is, according to (1), (5), the proportionality factor between c_F and c_D in the state of internal equilibrium:

$$c_D = H_* c_F \quad \text{(equilibrium)}. \tag{18}$$

If at the instant $t = 0$ the external influence manages to disturb the internal equilibrium of the concentration of the dispersed system ($c_F = c_F(0)$, $c_S = Hc_F(0)$, $t < 0$) and if the total time of the process θ ($0 < t < \theta$) is very small compared with λ_S:

$$\theta \ll \lambda_S, \tag{19}$$

then we consider such processes asymptotically short. Periodic processes whose time interval θ is very short compared with the relaxation time λ_S are considered asymptotically fast [11]. For asymptotically fast processes, the substitution of the corresponding asymptotic relationships (9b), (13) into (4) leads to a quasipenetration model of diffusion

$$D_0 \nabla^2 c_F = D_t c_F + b \lambda_S^{-1/2} D_t^{1/2} c_F, \tag{20}$$

where the parameter

$$b = \varphi H / (1 - \varphi) \tag{21}$$

yields the ratio of the sorptivities of the continuous and the dispersed phases.

According to the assumption, the applicability of the quasipenetration model is limited by the very short contact time (or very fast periodic processes) $\theta \ll \lambda_S$. In the range of changes of θ, we can distinguish the following two asymptotic cases: the self-similar braked regime

$$\theta \ll \lambda_0 \tag{22a}$$

and the self-similar penetration regime

$$\lambda_0 \ll \theta \ll \lambda_S, \quad b \gg 1, \tag{22b}$$

where

$$\lambda_0 = \lambda_S / b^2 = (1 - \varphi)^2 (\psi H)^{-2} D_S^{-1}. \tag{23}$$

In the self-similar braked regime (22a), the sorptivity of the dispersed phase does not play any role. The model (20) changes into the well-known form of writing Fick's second law for systems with a ballast dispersed phase: $D_0 \nabla^2 c_F = D_t c_F$.

In the self-similar penetration regime (22b), the sorptivity of the dispersed phase by far exceeds the sorptivity of the continuous phase. The model (20) assumes the asymptotic form

$$(\lambda_0^{1/2} D_0) \nabla^2 c_F = D_t^{1/2} c_F. \tag{24}$$

Penetration Experiment. The parameters (D_*, H_*) and (D_0, λ_0) may be viewed as structurally determined material constants. However, it would be more correct to view them as phenomenological coefficients of the asymptotic transport models (11), (17) or (13), (20), which have to be determined by experiments under the corresponding kinetic conditions.

Let us examine a penetration experiment of unidimensional non-steady-state sorption of AC into a motionless semi-infinite space [9] bounded by the plane surface $x = 0$. Before the experiment begins ($t \leq 0$), the dispersed system is in the state of internal concentration equilibrium that is characterized, e.g., by zero concentration of AC in both phases of the dispersed system. In consequence of external action, i.e., contacting of the dispersed system at the instant $t = 0$ with the homogeneous phase containing AC, its concentration on the surface in the continuous phase changes jumplike from zero to $c_F = c_1$. The layer of dispersion, $0 \leq x \leq h$, is so diluted that for the contact time θ taken into consideration, the penetration of AC does not manifest itself by appreciable changes in concentration at points far from the surface, i.e.,

$$h^2 H_*/D_* \equiv \theta_D \gg \theta. \tag{25}$$

The total amount of AC passing during the time of contact into the dispersed system through unit surface is

$$M(\theta) = \int_0^\theta |J|_{x=0}|\, dt. \tag{26}$$

The boundary conditions for the concentration field $c_F = c(t, x)$ can be written in the form

$$c_F = 0 \quad (x > 0,\ t \leq 0), \tag{27a}$$

$$c_F \to 0 \quad (x \to \infty,\ t \geq 0), \tag{27b}$$

$$c_F = c_1 \quad (x = 0,\ t > 0). \tag{27c}$$

With the conditions as specified, the quasilinear model changes into the linear integrodifferential equation

$$D \partial_{xx}^2 c = \partial_t c + \lambda_o^{-1/2} \partial_t^{1/2} c, \tag{28}$$

which, with the boundary conditions (27a, b, c), can be solved, e.g., by ordinary methods of integral transformations [13, 14]. The Laplace transform of the function $M = M_0(\theta)$ has the form

$$\overline{M}_o(p) \equiv \int_0^\infty \exp(-p\theta)\, M_o(\theta)\, d\theta = (1-\varphi)\, c_1 D_o^{1/2} p^{-1/2} [1 + (\lambda_o p)^{-1/2}]^{1/2}. \tag{29}$$

The inverse transformation leads to a series that converges for every finite θ:

$$M = M_o(\theta) = (1-\varphi)\, c_1 D_o^{1/2} (4\theta/\pi)^{1/2} A(\theta/\lambda_o); \quad \theta \ll \lambda_S, \tag{30}$$

where

$$A(T) = \frac{1}{2} \pi^{1/2} \sum_{k=1}^{\infty} \binom{1/2}{k} T^{k/2} / \Gamma\left(\frac{3}{2} + \frac{k}{2}\right) \approx 1 + 0.4431 T^{1/2} - \cdots; \quad \theta \ll \lambda_o \tag{31}$$

and $\Gamma(x)$ is Euler's second integral.

By the method of asymptotic expansion, the inverse transformation of expression (29) can be determined in the region $\theta/\lambda_0 \to \infty$ in the form (30), where

$$A(T) = \frac{1}{2} \pi^{1/2} \sum_{k=0}^{\infty} \binom{1/2}{k} T^{1/4-k/2} / \Gamma\left(\frac{7}{4} - \frac{k}{2}\right) \approx T^{1/4} (0.9643 - 0.4889 T^{-1/2} - \ldots); \quad \theta \gg \lambda_o. \tag{32}$$

The shape of the function $A(T)$ in the range of medium values of θ is shown in Fig. 1.

The quasiequilibrium model (16), which with the given conditions changes into the parabolic equation

$$(D_*/H_*)\, \partial_{xx}^2 c = \partial_t c, \tag{33}$$

leads to the ratio for accumulation of AC

$$M = M_*(\theta) = c_1 (H_* D_*)^{1/2} (4\theta/\pi)^{1/2}; \quad \lambda_S \ll \theta \ll \theta_D. \tag{34}$$

The solutions found for (30) and (34) reveal possible errors in evaluating the effectiveness of the diffusion coefficient with dispersed systems in case the experimental data, obtained for very brief contact, are processed in accordance with the classical theory from relationship (34). We will examine the case $\varphi \ll 1$, $\alpha \ll 1$, when the relationships $D_0 \approx D* \approx D_F$ remain valid, and we will assume that the dependence $M(\theta)$ was measured in the range of contact time $\theta \leq \lambda_0$. If we erroneously use relationship (34) instead of (30), we obtain, in evaluating the diffusion coefficient D_0, the erroneous value

$$(D_o)_{\text{error}} = \frac{M_o^2(\theta)}{c_1 H_*} \frac{\pi}{4\theta} \approx \frac{1 + 0.4 (\theta/\lambda_o)^{1/2}}{H_*} (D_o)_{\text{acct}}, \tag{35}$$

which in the case of $H_* \gg 1$ may be as much as one order of magnitude smaller than the accurate value $D_0 \approx D_F$. Such relaxation effects are encountered in measurements of the diffusion coefficient in dispersed systems by electrochemical [5] and sorption [6] methods in the range of contact times comparable with λ_S. According to data of [1, 14], the range of λ_S is very broad: $\lambda_S \epsilon (10^{-2}$ sec; 10^5 sec), and it consequently encompasses sorption processes in suspensions running down a wetted wall or in free jets [6].

For the experimental determination of the parameters D_0, λ_0, the self-similar regimes for fast concen-

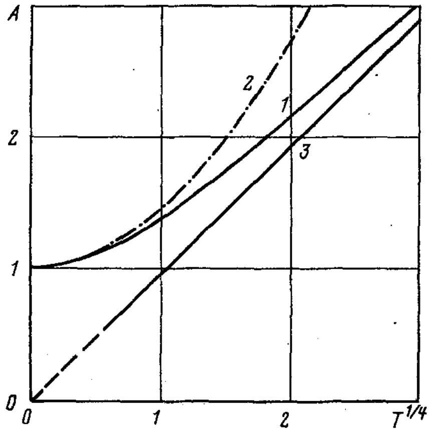

Fig. 1. Shape of the function A(T): 1) exact dependence; 2) asymptote for T → 0 according to (31); 3) asymptote for T → ∞ according to (32).

tration processes $0 \ll \lambda_S$ are suitable. For the self-similar braking of the regime, the following asymptotic relationship is valid in accordance with (30) and (31):

$$M \approx c_1(1-\varphi) D_0^{1/2} 2\pi^{-1/2}\theta^{1/2}; \quad \theta \ll \lambda_0, \tag{36a}$$

whereas according to (30), the self-similar penetration regime is described by the asymptotic relationship

$$M \approx c_1(1-\varphi) D_0^{1/2} \lambda_0^{-1/4}\theta^{3/4}/\Gamma(7/4); \quad \lambda_0 \ll \theta \ll \lambda_S. \tag{36b}$$

In the coordinates log M−log θ these regimes can be easily found, and the evaluations of the corresponding parameters obtained according to (36a, b). From published data on absorption curves [1] for molecular sieves, the self-similar penetration regime can be identified as the region of the convex initial course of the dependence in the coordinates $M-\theta^{1/2}$.

Technically interesting could be the application of the relationship (36b) for describing steady-state sorption of gases in a turbulently moving suspension according to Higby's penetration theory [15]. If the mean dwelling time of an element of the suspension at the surface ϑ corresponds to the inequalities $\lambda_0 \ll \vartheta \ll \lambda_S$, then the non-steady-state diffusion into the element can be described by (36b). The mean intensity of the flow of the AC through unit surface of the suspension J, flowing turbulently, can be expressed in accordance with (36b) by the relationship

$$J = M(\vartheta)/\vartheta \approx c_1(1-\varphi) D_0^{1/2} \lambda_0^{-1/4}\vartheta^{-1/4}. \tag{37}$$

The empirical parameter ϑ depends on the hydrodynamics of the stream of suspension.

NOTATION

AC, diffusing component; b, ratio of the sorptivities of the dispersed and continuous phases; c, concentration of the diffusing component of the AC; D, diffusion coefficient of the AC; H, equilibrium distribution factor of the AC between the dispersed and continuous phases; J, diffusive flux of AC in the dispersed system; M, accumulation of the AC in the dispersed system per unit contact surface; q, volumetric intensity of internal exchange of AC; r, volumetric rates of the reaction; t, time; x, distance from the contact surface; Γ, Euler's second integral; θ, time of contact; λ, diffusion time of the relaxation of the dispersed system; φ, specific volume of the dispersed phase during dispersion (volumetric fraction); ψ, outer specific surface of the dispersed phase. Subscripts: D, dispersed system as a continuum; *, slow, quasiequilibrium processes, $\theta \gg \lambda_S$; o, fast, quasipenetration processes, $\theta \ll \lambda_S$; F, continuous phase as a continuum; S, dispersed phase as a continuum.

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GAS FLOW IN A CAPILLARY WITH AN EXTERNAL DISTURBANCE VARYING THE COEFFICIENT OF MOLECULAR ADHESION

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The effect of change in the molecular adhesion coefficient on mass transfer in a capillary is studied for the free-molecular gas flow regime.

In recent years the interaction of laser radiation with material, leading to selective occurrence of many physicochemical reactions, has attracted ever greater interest of scientific investigators [1-3]. Among such reactions is a wide class of processes which occur on the boundary dividing two media. This class includes physical and chemical adsorption, heterogeneous catalysis, evaporation from the surface of solids and liquids, and diffusion in porous bodies. As was noted in [1], a correct understanding of the mechanisms by which they occur is extremely important for laser control of processes occurring on the surface of solids. Using the molecular-kinetic approach, the present study will consider certain questions related to the action of an external disturbance (in particular, laser radiation) on mass transfer in a capillary in the free-molecular gas flow regime.

As was noted in [1, 2] there are two possible variants of laser action on heterogeneous processes: 1) the radiation acts directly on the phase boundary and the molecules adsorbed thereon (the beam is incident on the surface) ; 2) the radiation acts upon the gas near the surface (the beam is parallel to the surface).

In the first case, aside from such possibilities as desorption stimulated by radiation and surface mobility of adsorbed molecules, one must also consider the possibility of direct radiation action on the adsorbent (producing changes in its catalytic properties, heating, etc.). Since clarification of all possible details of laser action on the material is quite difficult, in the first approximation we should select some parameters which

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