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Effect of surface diffusion on transfer processes in heterogeneous systems

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Abstract

The paper presents a review of publications concerned with the influence of surface diffusion of adsorbed molecules on the course of processes in heterogeneous systems. The influence of resonance (in particular, laser) radiation on mass transfer in channels (porous bodies) with allowance for surface diffusion of adsorbed molecules is discussed.

Keywords: Adsorption; Gas flow; Surface diffusion; Porous bodies; Resonance radiation

1. Introduction

The pressing need for the study of new methods to control the processes proceeding in heterogeneous media becomes more and more urgent with the development of a number of areas of new technologies associated with the necessity of acting on both individual atoms or molecules and the systems composed of them. Ascribed to these should be the technology of deposition of coatings in production of integral circuits for various electronic systems, the technology of production of nanoparticles and nanotubes, the technology of separation of gas mixtures (including isotopes) in using porous membranes with nanosized pores. The surface mobility of adsorbed atoms and molecules plays an important role in such processes as vapor condensation on surfaces [1,2] and growth of crystals [3]. In the presence of the concentration gradient of adsorbed molecules, their directed motion over a surface arises, which may substantially influence mass transfer in porous bodies [4–9]. It is known that the role of surface diffusion becomes especially great in nanoscale systems [10,11]. Thus, e.g., in nanoscale cylindrical channels (pores) surface diffusion of adsorbed molecules may make a considerable contribution to the total mass transfer through a channel with the relative contribution of surface diffusion to the total flux of molecules passing through the channel (pore) being increased with a decrease in its radius [12,13]. This effect is used for separation of gas mixtures, when they pass through membranes with nanosized pores [14]. Surface diffusion also exerts a substantial effect on the heterogeneous chemical reactions in porous bodies [15], on coalescence of nanosized particles [11], as well as on the processes of deposition of thin films in manufacturing the elements of microelectronics [16]. The formation of carbon nanotubes is also directly associated with the process of surface diffusion [17]. The review of the papers concerned with investigation of the mechanisms of the process of surface diffusion can be found in [10,18–21]. It should be noted that not only individual atoms and molecules can participate in the process of surface diffusion, but also rather large clusters [22,23].

2. Processes affecting surface diffusion of molecules

Since mass transfer over the surface by surface diffusion depends on the concentration of the molecules adsorbed on a surface, an essential factor that influences the surface flux seems to be the character of interaction of gas molecules with the surface. It should be noted that the description of surface processes can be made with a different degree

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Nomenclature

b	D_s/kT
D_{Kn}	coefficient of Knudsen diffusion
D_s	coefficient of surface diffusion
D_0	pre-exponential factor in equation for D_s
F	force acting on adsorbed molecules
h	Planck constant
I^-	density of a flux of gas molecules incident on a surface
I_s	density of a surface flux of molecules
I_d	density of a flux of molecules desorbing from the surface
J	radiation flux density
j_c	flux of molecules passing through a capillary
k	Boltzmann constant
L	length of a capillary
m	mass of a molecule
n	total number density of molecules in a gas phase
n'	number density of excited gas molecules
n_0	number density of unexcited gas molecules
n_a	number density of adsorbed molecules
P	gas pressure
Q_a	energy of adsorption
Q_s	activation energy of surface diffusion
r	radius of a capillary
T	temperature
t	time
v	average velocity of molecules
X	coordinate directed along the capillary axis
x	X/L

Greek symbols

α	sticking coefficient
γ	fraction of excited molecules
θ	surface coverage
σ_ν	cross-section for transition of molecules from one state to another
τ	adsorption time
τ_ν	relaxation time of excited gas molecules
ν	radiation frequency
ν_0	resonance frequency of gas molecules

Subscripts

a	adsorption
c	capillary
d	desorption
Kn	Knudsen
min	minimum
s	surface

of detailing. In recent years, an increasingly greater attention of researchers is directed to the problems of interaction of gas molecules with a surface. The problem of scattering of gas particles by a surface was considered in the monographs [24–26]. The theories exist which detail both the surface relief, as well as the kinetics of the adsorption–desorption of molecules on the surface (for example, in describing the growth of crystals [3]), and in which the main emphasis is made on the interrelationship between different phases (gas–adsorbate–solid body) [27,28]. Approaches to the description of the gas–surface system, which take into account the interaction of a real gas with a phonon gas, have been developed in [29,30]. In a real system, both structural factors, which characterize the surface itself, and various nonequilibrium interphase processes will manifest themselves. Moreover, an exact quantitative description of the interaction of gas particles with a surface is difficult, since in a number of cases the real structure of the surface is unknown and, consequently, there is no exact enough expression for the potential of interaction of an incident gas particle with a condensed phase. The state of the surface is determined by the type of processing, purity, temperature; it can also change in the course of the process, since it may become covered by a layer of adsorbed molecules or, conversely, can be degassed [31]. Below, a brief qualitative picture of the interaction of gas molecules with a surface is given.

The gas molecule incident on the surface of a condensed phase may reflect from it elastically, having conserved its internal energy and the absolute value of the velocity of translational motion. In the case of inelastic interaction with the surface, the indicated parameters change upon collision with the surface. The particle that falls on the surface may also be adsorbed on it. In the majority of cases it is accompanied by accommodation of the gas particle (deactivation of the particle if prior to this it was in an excited state, as well as thermal accommodation). Within the framework of the Langmuir model, the density of the flux of molecules being adsorbed on the surface can be written as

$$J_a = I^- \alpha (1 - \theta), \tag{1}$$

where I^- at the Maxwellian gas velocity distribution function has the form

$$I^- = \frac{P}{(2\pi mkT)^{1/2}}. \tag{2}$$

In the Maxwellian diffuse-specular scheme of the scattering of gas molecules by a surface it is assumed that part of the molecules f is reflected diffusely and part $1 - f$ – specularly [31]. Often, it is not stipulated whether the first part consists of elastically reflected molecules or of the molecules being desorbed from the surface. From a more detailed analysis of transfer processes it is seen that such a division should be done (in particular, in the case of investigating the influence of resonance radiation on the transfer processes in heterogeneous systems). If the diffuse-specular scheme of interaction is interpreted as follows: the molecules reflected specularly are the elastically reflected ones, whereas the flux of the molecules scattered by the surface diffusely includes only desorbed molecules, then the value of f in the scheme indicated will coincide with the sticking coefficient α. The above-described scheme of scattering is applicable to a smooth surface. In the case of a rough surface, in [32] a scheme of backscattering is used, in which it is assumed that part of the molecules f is scattered diffusely and part $\beta = 1 - f$ – in the direction opposite to the motion of molecules. In [33], the relationship was established between the backscattering coefficient introduced in [32] and a concrete model of a rough surface. We note that in some cases the spatial distribution of the molecules being desorbed from the surface may differ from the cosine law. Thus, for example, when the molecules which formed in the process of recombination of atoms desorb from the surface, then, in some cases the scattering indicatrix of escaping molecules is extended in the direction of the normal to the surface [34]. In the field of resonance radiation, the distribution of the molecules being desorbed may also differ from the cosine law. This problem will be considered in more detail in what follows.

It should be noted that molecules may enter the adsorption layer from both the gas and condensed phases, then they can leave it by going into the gas phase or the condensed one. In [27,28], the theory of the kinetics of adsorption was developed with account for the noted processes of transfer in a gas and condensed phases. If we restrict the discussion to the case where the fluxes of molecules from an adsorbed layer into a condensed phase and back can be neglected, then in a steady state, in the assumptions adopted in Langmuir's theory of adsorption, for adsorbed molecules the continuity equation can be written in the form

$$-\nabla I_{\mathrm{s}} = I_{\mathrm{d}} - \alpha(1-\theta)I^{-}, \tag{3}$$

where I_{d} is expressed as [35]

$$I_{\mathrm{d}} = \frac{n_{\mathrm{a}}}{\tau}. \tag{4}$$

According to Karlov and Shaitan [36], the sticking coefficient α, which characterizes the probability of the gas particle that falls on the surface of not being elastically reflected from it, can be presented in the form

$$\alpha = 1 - \exp\left(-\frac{E_{\mathrm{c}}}{kT}\right), \tag{5}$$

where E_{c} is the specific energy of interaction of a gas molecule with the surface (critical energy of sticking) dependent on the relationship between the masses of the gas molecule and adsorbent atom and on the adsorption energy. Thus, for example, in the approximation which considers the collision of a molecule with the surface atom as collision of free particles upon fulfilment of the inequality $m_{\mathrm{g}}/M \ll 1$, where m_{g} is the mass of the incident gas molecule and M is the mass of the atom of the adsorbent lattice, the following expression can be written for E_{c} [36]:

$$E_{\mathrm{c}} \approx 4\frac{m_{\mathrm{g}}}{M}Q_{\mathrm{a}}. \tag{6}$$

It follows from Eqs. (5) and (6) that with a decrease in Q_{a} the value of the sticking coefficient will also decrease.

The time during which a gas particle resides on the surface (the time of adsorption) is a random quantity, but the average time of adsorption for a large number of molecules under constant external conditions has a definite value. The mean time of adsorption is determined by the energy (heat) of adsorption Q_{a} and by the surface temperature T as follows [4,35]:

$$\tau = \tau' \exp\left(\frac{Q_{\mathrm{a}}}{kT}\right), \tag{7}$$

where τ' is the quantity of the order of the period of the adsorbed molecule oscillation in the direction normal to the surface.

Adsorption of gases can be subdivided into physical and chemical. If during the adsorption the individualities of the adsorbed molecule and surface-forming particles are preserved, there is physical adsorption. But if a molecule during adsorption bonds chemically with the atoms (molecules) of the surface, then the adsorption is chemical (chemisorption) [35].

Adsorption is also subdivided into monomolecular and polymolecular. To realize monomolecular adsorption, it is required that the time of adsorption in the second and subsequent layers be much shorter than the time of adsorption in the first layer. If the molecules incident on the layer of the already adsorbed molecules experience a rather strong attraction, the time of their adsorption in the second and subsequent layers will no longer be negligibly short as against the time of adsorption in the first layer. In this case, polymolecular adsorption can take place. In [35], adsorption isotherms are given for both mono- and polymolecular adsorption.

At a sufficiently high external gas pressure, when the number of incident gas molecules sticking to the surface is higher than those evaporating, condensation of the given substance on the surface begins. It should be noted that the process of three-dimensional condensation is preceded by adsorption of molecules on the surface and two-dimensional condensation of adsorbed molecules. Here, a certain critical temperature corresponds to the molecular beam that bombards the surface [1,37]. Energetically, the solid body surface can be visualized as a set of adsorption centers in which the molecule being adsorbed attains a minimum of potential energy. For a homogeneous surface all adsorption centers are characterized by an identical energy; on an inhomogeneous surface the energy of different adsorption centers is not the same. In transition from one adsorption center to another, the adsorbed molecule must have an energy sufficient for overcoming the potential barrier between these centers, i.e., the movement of the molecule over the surface is usually of activation character. The mean time of the residence of the molecule at one adsorption center τ_0 is related to the activation energy needed for the transfer of the molecule as follows [4,35]:

$$\tau_0 = \tau_0' \exp\left(\frac{Q_s}{kT}\right), \tag{8}$$

where τ_0' is a constant close in the order of magnitude to τ'.

Generally, Q_s is lower than the adsorption energy Q_a. If Q_s is reasonably low, the transfer of the adsorbed molecules over the surface resembles their motion in a two-dimensional gas. In the case of an activated character of surface diffusion, the coefficient of surface diffusion D_s has the form [4,35]

$$D_s = D_{s0} \exp\left(-\frac{Q_s}{kT}\right). \tag{9}$$

In the case where the adsorbed molecules can be modeled by a two-dimensional gas, the coefficient of the surface diffusion is determined similarly to the three-dimensional case [4,35]:

$$D_s = \frac{1}{2}\lambda_s v_s. \tag{10}$$

Here, v_s is the average velocity of motion of particles over the surface and λ_s is the mean free path of particles.

In [38], based on consideration of the model of a two-dimensional gas for adsorbed molecules and use of the solution of kinetic equations by the Chapmen–Enskog method, expressions are obtained for the coefficients of surface diffusion and thermodiffusion for an adsorbed phase. The results obtained are compared with the available expressions for a three-dimensional gas. In [39], also a molecular-kinetic approach to the process of surface diffusion is used which makes it possible to correctly account for the interaction of the diffusing particles with both one another and the substrate.

In the general case, where an external force acts on adsorbed molecules (for example, when adsorbed molecules can be represented as ions [40] and they experience the action of the electric field), the density of the surface flux of molecules can be presented in the form

$$I_s = -D_s \frac{\mathrm{d}n_a}{\mathrm{d}X} + n_a bF, \tag{11}$$

where the first term on the right-hand side describes the diffusion component and the second term corresponds to the flux component which is due to the action of the external force.

It should be noted that generally the coefficient of surface diffusion depends on the surface coverage by adsorbed molecules [6,10]. To describe the indicated dependence in the region of change in the surface coverage $0.1 < \theta < 0.6$, the following expression is often used [6]:

$$D_s = D_s|_{\theta \to 0} \frac{1}{1-\theta}. \tag{12}$$

In the region of sufficiently high values of the surface coverage θ in [41] a modified expression for D_s, which does not lead to infinity when $\theta \to 1$, was obtained.

It should be noted that surface diffusion can be influenced by the surface curvature. Thus, in [42] the problem of the influence of surface curvature on surface diffusion in application to carbon nanotubes is investigated. The dependence of the surface flux on surface curvature has been revealed. The dependence of surface diffusion on the size of spherical nanoparticles was discussed in [11].

3. Influence of resonance radiation on surface processes

It is known that resonance (in particular, laser) radiation exerts its influence on the processes of transfer in both a gas phase and heterogeneous systems [43–85]. Often, the surface processes turn out to be the means by which the coupling of the phenomena of transfer with different kinds of external effects on the system occurs. Thus, for example, the resonance (in particular, laser) radiation, which changes the character of interaction of gas particles with a solid surface, may lead to a change in the permeability of porous bodies and individual capillaries relative to the gas passing through them [46–52]. In the case, where resonance radiation acts on the system that was equilibrium prior to the effect of radiation, a resulting mass flux can be realized in the latter system [53–69]. The influence of radiation on the flow of gases in capillary-porous bodies in the absence of sources and sinks of molecules on the walls of pores may be realized both by means of the process of surface diffusion and through a radiation-affected change in the trajectory of motion of gas molecules in capillaries.

With the aid of resonance radiation one can also influence the processes of deposition of a substance from a gas phase. Resonance radiation influences the processes of adsorption–desorption, evaporation-condensation, and migration of molecules over the condensed phase surface [70–85]. For the case of vibrationally excited molecules the following expression for the sticking coefficient is given in [72]:

$$\alpha = 1 - \exp\left(-\frac{E_c}{E_g}\right), \tag{13}$$

where E_g is the gas molecule energy involving the excitation energy.

Expressions for the time of adsorption and coefficient of surface diffusion in the field of resonance radiation can be presented in the first approximation in the form

$$\tau = \tau' \exp\left\{\frac{Q_a}{kT}(1 + \Delta_1)\right\}, \tag{14}$$

$$D_s = D_{s0} \exp\left\{-\frac{Q_s}{kT}(1 + \Delta_2)\right\}, \tag{15}$$

$$\Delta_1 = \frac{\Delta Q_\mathrm{a}}{Q_\mathrm{a}} - \frac{\Delta T}{T}, \quad \Delta_2 = \frac{\Delta Q_\mathrm{s}}{Q_\mathrm{s}} - \frac{\Delta T}{T}, \tag{16}$$

where ΔQ_a and ΔQ_s describe, respectively, changes in the energy of adsorption and the activation energy of surface diffusion under the action of radiation. Their signs are determined by the character of radiation effect. In representing Eqs. (14)–(16) it was assumed that $\frac{\Delta Q_\mathrm{a}}{Q_\mathrm{a}}$, $\frac{\Delta Q_\mathrm{s}}{Q_\mathrm{s}}$, $\frac{\Delta T}{T} \ll 1$. The quantity ΔT describes a change in the surface temperature because of the heating of the system by radiation, including also such a kind of heating as deactivation of preliminarily excited molecules.

We will briefly consider the literature data on the influence of resonance laser radiation on the processes of adsorption–desorption [70–85]. In [74], a decrease in the number of adsorbed (condensed) gas molecules on their vibrational excitation was revealed. This phenomenon may be due to both the increase in the probability of reflection of excited molecules and also to the local release of heat in deactivation of excited particles on the surface leading to a more intense evaporation of particles. The mechanisms of the laser radiation-induced desorption of particles from the adsorbent surface were considered in [49,70,72,77–79,81–85].

It should be also noted that on resonance excitation of the molecules, they can be passed into the region of predissociation [72]. On collision with the surface, such excited molecules can dissociate into atoms. However, the energies of adsorption and activation of surface diffusion of molecules and of their constituting atoms can be different. Correspondingly, the quantities α, τ, and D_s will also differ. It should be noted that in the field of resonance radiation the indicatrix of the scattering of gas particles by a surface may also change. In [51], an assumption was made that the radiation-induced change in the angular distribution of the particles scattered by the surface (the indicatrix is extended in the direction of the normal to the surface) can serve as one of the reasons for the experimentally observed sharp decrease in the flux of gas molecules passing through capillaries in the field of resonance radiation [46]. In [86], the problems of nonequilibrium desorption of gas particles from the surface are considered. The nonequilibrium desorption is understood to be the desorption of particles when the distribution function of the escaping particles is not described by the Maxwellian distribution function with a temperature equal to the surface temperature. It is noted that usually for nonequilibrium desorption the cosine law is not satisfied and the angular distribution of desorbing particles is proportional to $\cos^\mu\Theta$, where Θ is the angle between the direction of motion of a particle and a normal to the surface; according to [86], the value of the parameter μ lies within the range 3–5. In [85], a similar scattering indicatrix of particles evaporating on exposure to radiation was considered experimentally. It is also shown that evaporating particles are subdivided into high-energy ones, which escape at small angles relative to the normal to the surface, and low-energy ones, the angular distribution of which corresponds to the cosine law, whereas the mechanism underlying the formation of particles can be considered a quasi-equilibrium thermal one.

4. Free-molecular gas flow in long channels with account for surface diffusion

First, we consider a simple model of free-molecular gas flow in a long cylindrical channel with additive account for the transfer of molecules in a gas phase and by surface diffusion under isothermal conditions. A flux of molecules passing through the cross-section of the cylindrical channel (capillary) of radius r in a free-molecular regime of gas flow on the assumption of diffuse scattering of molecules by the channel wall and with neglect of the surface mobility of adsorbed molecules has the form [87]

$$j_\mathrm{c} = -\pi r^2 \cdot \frac{2}{3} r v \frac{\mathrm{d}n}{\mathrm{d}X} = -\pi r^2 D_\mathrm{Kn} \frac{\mathrm{d}n}{\mathrm{d}X}. \tag{17}$$

The experimentally observed excess of the measured value of the flux of molecules over the value resulting from Eq. (17) is usually attributable either to the influence of specular reflections of molecules from the channel wall, which in the Maxwellian scheme of the scattering of molecules by the channel wall leads to the factor $(2-f)/f$ in the expression for D_Kn, or to the transfer of adsorbed molecules by surface diffusion. Here, in (17) the additional term $-2\pi r D_\mathrm{s} \frac{\mathrm{d}n_\mathrm{a}}{\mathrm{d}X}$ is introduced. The second approach to the explanation of the experimentally observed increase in the flux of molecules passing through the cylindrical channel, as compared to Eq. (17), was employed in [12,13]. It was assumed in these works that all the molecules leaving the surface are distributed in conformity with the cosine law (i.e., diffuse scattering of molecules by the wall is realized and the parameter $f = 1$).

In a real case there may probably occur both specular reflections of gas particles from a sufficiently smooth surface of the channel and the processes of adsorption and surface diffusion. Taking into account the fact that only the molecules adsorbed on the channel walls may participate in the process of surface diffusion, assuming the additivity of the volumetric and surface fluxes and satisfaction of the Henry law for adsorption, the value of the flux of molecules passing through the channel in a stationary case can be written in the form

$$j_\mathrm{c} = -\pi r^2 \left(\psi \cdot \frac{2}{3} r v + \frac{D_\mathrm{s} \alpha v \tau}{2r}\right) \frac{\mathrm{d}n}{\mathrm{d}X}, \tag{18}$$

where for diffuse-specular scheme of the scattering of molecules $\psi = \frac{2-f}{f}$.

As noted above, on the assumption that the molecules sticking to the surface are scattered diffusely, while those which do not stick are reflected specularly, the value of f will be equal to the sticking coefficient α. Here, from Eq. (18) for the criterion G, which represents the ratio of the surface flux to the volumetric one, we obtain the relation

$$G = \frac{3\tau D_s \alpha^2}{4r^2(2-\alpha)}. \tag{19}$$

From Eq. (19) it is seen that the value of G is increased with a decrease in r and an increase in α. It follows from Eq. (18) that at $f = \alpha$ the value of $j_c(\alpha)$ will be characterized by a minimum at $\alpha = \alpha_{\min}$. Here, the value of $\alpha_{\min}$ is equal to

$$\alpha_{\min} = 2r\left(\frac{2}{3D_s\tau}\right)^{1/2}. \tag{20}$$

It should be noted that in [32] for rough surfaces the scheme of backscattering was used in which it is assumed that part of molecules f is scattered diffusely and part $\beta = 1 - f$ – in the direction opposite to the molecule motion. Here, the quantity ψ in Eq. (18) has the form $\psi = f/(2-f)$ [32]. Since the value of ψ in the backscattering scheme at $f < 1$ is less than unity (in contrast to the scheme of diffuse-specular scattering), the situation may be realized, where an increase in the flux of molecules passing through the capillary due to the surface diffusion of molecules will be compensated by backscattering of molecules. This will occur when the following equality is realized:

$$\frac{1-\beta}{1+\beta} + \frac{3D_s\alpha\tau}{4r^2} = 1, \tag{21}$$

from which one may find the values of the parameters that correspond to the condition indicated.

It is worth noting that in the case of a rough surface the surface diffusion will manifest itself not only in the second term of Eq. (18), but also via its influence on the quantity $\psi = f/(2-f)$ [59]. This is due to the fact that at the characteristic length of the surface diffusion $l_s = (D_s\tau)^{1/2}$ larger than the size of roughness the molecules may pass over the roughness facets by means of surface diffusion, which will lead to a higher coefficient of diffuse scattering [68]. This effect will depend substantially on temperature, since according to Eqs. (7) and (9), the dependence of l_s on temperature has the form

$$l_s = (D_{s0}\tau')^{1/2} \exp\left(\frac{Q_a - Q_s}{2kT}\right). \tag{22}$$

The dependence on temperature of a flux of molecules passing through a cross-section of a long cylindrical channel under the assumptions made and subject to Eqs. (5), (7), (9) and (18) can be presented as

$$j_c = -\pi r^2 \left(\frac{8kT}{\pi m}\right)^{1/2} \left\{\psi \cdot \frac{2}{3} r + \frac{D_{s0}\tau'}{2r} \exp\left(\frac{Q_a - Q_s}{kT}\right) \times \left[1 - \exp\left(-\frac{E_c}{kT}\right)\right]\right\} \frac{dn}{dX}. \tag{23}$$

It should be noted that apart from the factors considered above, the flow of gases in narrow channels can also be affected by the potential field of the walls of the channels [88,89].

5. Influence of resonance radiation on free-molecular gas flow through long channels (capillaries) with account for surface diffusion

Since, as noted above, the values of α, τ, and D_s may change under the action of resonance laser radiation [72], it follows from Eq. (18) that this effect can be employed to alter the flux of molecules passing through a capillary when a pressure drop along its length is realized, as well as to initiate mass transfer in an initially equilibrium system (Fig. 1).

Eq. (18) is written for the case, where the values of α, τ, and D_s not change along the capillary. This may be valid for a capillary-porous body transparent for the incident radiation at a sufficiently slight absorption of radiation in the gas phase. Radiation can also act not on the porous body, but only on the gas phase (the beam is parallel to the porous body surface [72]). With the effect thus arranged, part of the molecules entering the capillary from the side of irradiation zone will be in an excited state. The part of the excited molecules can be found from the system

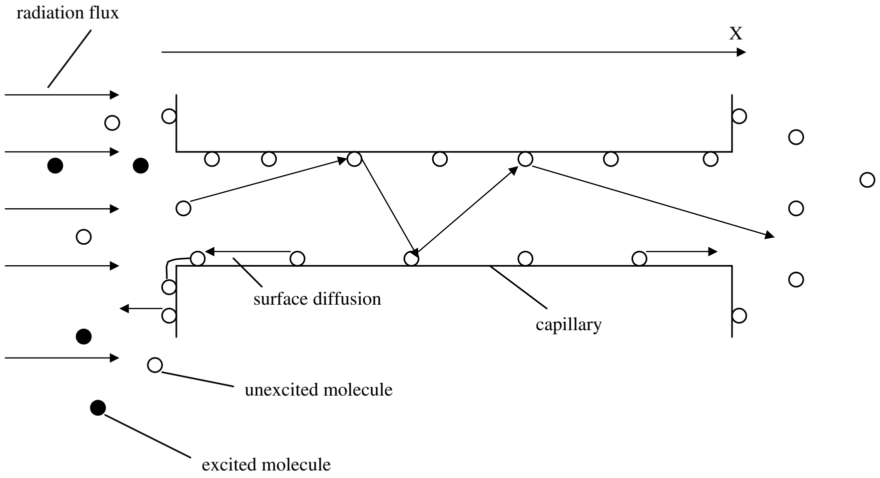

Fig. 1. Scheme of radiation-induced drift of molecules in capillaries.

of kinetic equations for transitions of molecules from the ground state into an excited one and back. For simplicity we will consider a two-level system. In this case, the above-indicated equations have the form [48]

$$\frac{\mathrm{d}n'}{\mathrm{d}t}=\frac{J\sigma_v}{hv}(n_0-n')-\frac{n'}{\tau_v}, \tag{24}$$

$$\frac{\mathrm{d}n_0}{\mathrm{d}t}=\frac{J\sigma_v}{hv}(n'-n_0)+\frac{n'}{\tau_v}, \tag{25}$$

The quantity γ, which characterizes the fraction of excited molecules, is defined as $\gamma=\frac{n'}{n}=\frac{n'}{n_0+n'}$. In a steady-state γ can be presented as

$$\gamma_i=\frac{(J\sigma_v/hv)\tau_v}{1+(2J\sigma_v/hv)\tau_v}. \tag{26}$$

The mean value of the sticking coefficient $\bar{\alpha}$ can be written in the form

$$\bar{\alpha}=\alpha_1-\gamma(\alpha_1-\alpha_2), \tag{27}$$

where α_1 and α_2 are the sticking coefficients of unexcited and excited molecules, respectively.

When a surface is exposed to radiation, then in addition to such possibilities as radiation-induced desorption and surface mobility of adsorbed molecules one should also take into account the influence of radiation directly on an adsorbent (change in its absorptive properties, heating, etc.). Since it is difficult to reveal all the possible details of the process of the action of radiation on a substance, in the first approximation one should define the parameters that in some way take into account the overall effect. Thus, in the exposure scheme noted one of such parameters can, in all probability, be the mean time of the gas particle residence on the surface (adsorption time). In the case, where radiation acts on the gas phase, the influence of radiation on the transfer processes is realized via the difference in the interaction with the surface of excited and unexcited molecules that, in particular, manifests itself in the difference between their sticking coefficients.

The above-described distinction between the influence of radiation on heterogeneous processes is, of course, conventional. When radiation falls on the surface, it can, naturally, excite also molecules in a gas phase. On the other hand, in the second scheme of exposure to radiation heating the surface may occur because of the deactivation of radiation-excited gas particles on the surface and, as a consequence, there may occur a change in the time of adsorption and in the surface diffusion coefficient. However, to gain a better understanding of the mechanism underlying the process of mass transfer, it is advisable in the first approximation to consider separate effect of different factors, as well as to neglect the radiation-induced nonuniformities of thermodynamic parameters.

Let us estimate the magnitude of radiation-induced flux of molecules passing through cylindrical capillaries that model the pores of a membrane in two cases: (1) where transfer in an adsorbed layer predominates and (2) where transfer in a gas phase alone is essential [63]. Eq. (19) aids in determining the case in hand. The densities of the fluxes of molecules incident on the end surfaces of a capillary at its both ends are assumed to be identical and it is considered that radiation acts on gas molecules near one of the surfaces of the porous membrane. Next, for simplicity the sticking coefficient of unexcited molecules is considered equal to unity. In the first case, where transfer in the adsorbed layer prevails, for the value of j_c the following estimating expression can be written (we assume that the molecules adsorbed on the surface are unexcited and that the values of P, T, τ and D_s are the same for both ends of the capillary) [63]:

$$j_\mathrm{c}=\frac{2\pi r}{L}\frac{\gamma P\tau D_\mathrm{s}}{(2\pi mkT)^{1/2}}(1-\alpha_2). \tag{28}$$

In the second case, when the surface diffusion can be neglected, the flux of molecules passing through the capillary in free-molecular gas flow can be written in the form

$$j_\mathrm{c}=\pi r^2\frac{P}{(2\pi mkT)^{1/2}}(W_1-W_2), \tag{29}$$

where W_1 and W_2 are the passage probabilities for different directions [90].

If the sticking coefficient of excited molecules differs from unity, according to the above-made assumptions in the diffuse-specular scheme of the scattering of molecules by a surface the fraction of specularly reflecting molecules that enter the capillary from the side of the zone exposed to resonance radiation will differ from zero. This leads to the fact that the probability of the passage of a molecule through the capillary in the direction from its mouth, which faces the zone exposed to radiation, W_1 will be higher than W_2 (which under the above-stipulated conditions is defined only by diffuse distribution of the molecules escaping from the capillary wall; when $L \gg r$ this quantity is equal to $\frac{8r}{3L}$ [90]).

Thus, the direction of the radiation-induced mass flux in these two cases can be opposite. In the first case, where transfer over the surface prevails, the flux of molecules will be directed into the zone exposed to radiation, since mass transfer is attributable to the drop in the concentration of adsorbed molecules; this concentration on the end surface of the capillary facing to the zone exposed to resonance radiation will be smaller. At the same time, as shown above in the second case, the direction of the resulting flux of molecules is opposite.

The difference between the average values of the sticking coefficients for the opposite sides of the porous membrane and between the values of W_1 and W_2 may also be due to the so-called velocity-selective excitation of gas molecules, when on detuning of the radiation frequency from the resonance frequency of gas molecules, only the molecules flying in a certain direction will pass into an excited state due to the Doppler effect [65,66]. Let us assume that in the direction of the coordinate X perpendicular to the side surfaces of a transparent porous plate the radiation flux is incident which is characterized by the frequency v. If the resonance frequency of gas molecules v_0 is somewhat higher than v, then only the molecules moving in the direction opposite to the direction of radiation flux, i.e., the molecules flying opposite to the X-axis, may pass into an excited state due to the Doppler effect [43]. Thus, a larger number of excited molecules will be incident on the plate surface at $X = L$ than on the surface at $X = 0$ (excited molecules will be incident on this surface only after intermolecular collisions in which they can be deactivated). Next, for simplicity we consider that excited molecules are not incident on the surface at $X = 0$ at all. The fraction of excited molecules can be estimated from Eq. (26). In the case considered, the cross-section for resonance absorption differs from zero, when the projection of the velocity of the particles meets the X-axis in a certain interval of values, which depends on the frequency of radiation. The problems of the resonance radiation-induced drift of molecules in capillaries in a free-molecular regime of gas flow on velocity-selective excitation of molecules were considered in [65,66].

In [49], the problem of the influence of heating of a porous body by laser radiation on the permeability of gas molecules was investigated. We will briefly consider the influence of thermal effects on the radiation-induced transfer processes in porous bodies [61]. The heating of a porous membrane by incident radiation may influence the mass transfer through a porous membrane in two ways. First, a flux of electromagnetic radiation incident on one of the surfaces of the membrane may create there a temperature gradient, which in turn will lead to the appearance of the gradient of concentration of adsorbed molecules and realization of the resulting mass flux in an initially equilibrium system. On the other hand, the heating of the membrane (as a whole) will lead to a decrease in the concentration of the molecules adsorbed in the membrane and, correspondingly, to a decrease in the resulting flux of molecules through the membrane. Thus, in principle, there may exist a situation where with an increase in the intensity of radiation the mass flux induced by it will pass through a maximum. We note that mass transfer through a porous body due to surface diffusion causes also energy transfer. In [91], the problem of the appearance of a temperature drop in the porous plate was considered; it is associated with the difference in the densities of the fluxes of molecules desorbed from the opposite sides of the plate. This difference in the flux densities of desorbed molecules is in turn attributable to the surface flux of molecules.

6. Description of gas flow in channels with account for surface diffusion on the basis of an integro-differential equation

Above we considered the general laws governing the influence of surface diffusion on free-molecular gas flow in long channels with the use of a simple model, in which the volumetric and surface fluxes are taken into account additively, with the condition of local equilibrium between the adsorbed and gas phases in each section of the channel being assumed. The use of the approach based on the integro-differential equation for the number density of adsorbed molecules allows us to remove limitation on the ratio of the channel length to its radius, as well as the assumption of local equilibrium. This approach was used in [53,54,61,92–94]. With regard to Eqs. (3) and (11) and the assumption of low values of θ to determine the concentration of the molecules n_a adsorbed on the surface of the cylindrical capillary, we may write equations [61]

$$\frac{1}{L^2}\frac{\mathrm{d}}{\mathrm{d}x}\left(D_\mathrm{s}\frac{\mathrm{d}n_\mathrm{a}}{\mathrm{d}x} - LbFn_\mathrm{a}\right) = \frac{n_\mathrm{a}}{\tau} - \alpha\left[\int_0^1\left(\frac{n_\mathrm{a}}{\tau} + I\right)K_1(|x-\xi|)\,\mathrm{d}\xi + I_0K(x) + I_1K(1-x)\right], \tag{30}$$

$$I = (1-\alpha)\left[\int_0^1\left(\frac{n_\mathrm{a}}{\tau} + I\right)K_1(|x-\xi|)\,\mathrm{d}\xi + I_0K(x) + I_1K(1-x)\right], \tag{31}$$

where I_0 and I_1 are the densities of the fluxes of molecules entering the capillary at $x = 0$ and $x = 1$, the functions K_1 and K characterize the probabilities of the transition of molecules from one surface element to the other [90,94]. The first term on the right-hand side of Eq. (30) defines the density of the flux of the molecules that desorb from the inner surface of the capillary, the integral term in square brackets characterizes molecules flying out from one element of the inner surface of the capillary and arriving at its other element. The terms in square brackets outside the integral describe the molecules entering the capillary through open ends and arriving at the element of the side surface in the vicinity of the point x. The quantity I describes the density of the flux of elastically reflected molecules. The flux of molecules exiting the capillary at $x = 1$ is defined as

$$N(1) = -\frac{2\pi r}{L}\left(D_\mathrm{s}\frac{\mathrm{d}n_\mathrm{a}}{\mathrm{d}x} - LbFn_\mathrm{a}\right)\bigg|_{x=1} + 2\pi rL\int_0^1\left(\frac{n_\mathrm{a}}{\tau} + I\right)K(1-x)\,\mathrm{d}x + \pi r^2 I_0K_2(1), \tag{32}$$

where the value of $K_2(x)$ is determined from the expression

$$\frac{\mathrm{d}K_2}{\mathrm{d}x} = -2lK, \quad l = \frac{L}{r}. \tag{33}$$

The boundary conditions for Eq. (30), as well as an approximate method of its solution, are given in [61].

In the case of an opaque porous body, its exposure to resonance radiation may lead to an asymmetry in the values of τ or α on the opposite sides of the body (near the opposite ends of capillaries). The first case (at $F = 0$) is considered on the basis of the solution of an integro-differential equation in [53] and the second case – in [54]. Note that the surface diffusion induced by resonance radiation in porous bodies placed in a gas phase may cause an additional force acting on the body due to the difference in the densities of the fluxes of the molecules desorbed from the opposite sides of the body [56].

7. Conclusions

A review of the papers concerning the influence of surface diffusion of adsorbed molecules on the transfer phenomena in heterogeneous (in particular, in nanoscale) systems, including also the case of their exposure to resonance radiation, is given. It is shown that surface diffusion may serve as a means by which the radiation-induced transfer processes in heterogeneous systems can be realized.

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