



Effect of electromagnetic fields on transfer processes in heterogeneous systems

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Abstract

Some effects are analyzed which are related to the influence of electromagnetic radiation on transfer processes in heterogeneous systems. The influence exerted by the excitation of internal degrees of freedom of molecules in a field of the resonance laser radiation is considered, as well as the effect of volumetric heating of a porous catalyst particle in the field of microwave radiation. It is shown that on exposure to microwave radiation heterogeneous reaction in a particle of the porous catalyst can occur more uniformly over the particle volume. An expression is found for temperature distribution in a spherical particle of the porous catalyst resulting in a uniform course of chemical reaction over the particle volume. © 2001 Elsevier Science Ltd. All rights reserved.

1. Introduction

An electromagnetic radiation can affect transfer processes in heterogeneous systems ‘gas-condensed phase’ due to both the change in the regularities of interaction of gas molecules with the surface of a condensed phase and system heating by incident radiation. The influence of electromagnetic radiation on a substance is manifested in different ways for different ranges of wavelengths. In the wavelength region corresponding to laser radiation one can observe not only ordinary heating by incident radiation, but also excitation of the internal degrees of freedom of molecules, resulting in a change in the interaction of gas molecules with the condensed phase [1–3]. On the other hand, in the wavelength region of microwave range the factor related to fast volumetric selective heating of the condensed phase becomes controlling [4]. In a number of cases this leads to the appearance of the so-called inverse temperature gradient (when the temperature in the centre of

the system is higher than on its periphery). The paper deals with the problems of heat and mass transfer related to the above-mentioned characteristics of the electromagnetic radiation effect on the heterogeneous systems consisting of gas and condensed phase. A scheme illustrating some possible effects due to the influence of electromagnetic fields on the given systems is presented in Fig. 1.

2. Influence of excitation of gas molecules in a field of resonance radiation on transfer processes in heterogeneous systems

At first let us consider the effects related to excitation of internal degrees of freedom of gas molecules. Such excitation in a field of resonance radiation can lead both to a change in the probability of gas molecule sticking to the surface and to varying regularities of molecule desorption from the surface. In the case of internal problems of gas flow in channels and capillary-porous bodies this can result in radiation-induced mass transfer in the initially equilibrium system [5–11]. For aerosol systems the indicated effects can lead to the appearance of new components of the photophoretic force acting on the

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Nomenclature			
b	adsorbed molecule mobility	q	power of microwave radiation absorbed in unit volume of a porous particle
c_p	specific heat of porous particle	R	radius of an aerosol particle
D_e	effective diffusion coefficient of the molecules of reactant in the porous particle	R_c	radius of a capillary
D_s	coefficient of surface diffusion	R_p	radius of a porous particle
E_r	activation energy of heterogeneous chemical reaction	r	radial coordinate reckoned from the particle centre, $\bar{r} = r/R_p$
E_{rms}	rms electric field strength	T	surface temperature
F	force acting on an adsorbed molecule along the capillary	T_g	gas temperature
F_p	force acting on an aerosol particle	T_p	temperature of a porous particle
H_r	heat of chemical reaction per molecule	v_B	mean thermal velocity of molecules of component B
I_0	density of molecular fluxes entering the capillary at $x = 0$ and $x = 1$	v_R	rate of condensation growth of a particle
K, K_1, K_2	functions characterizing probabilities of the transitions of molecules from one surface element to another	x	dimensionless coordinate directed along the capillary axis.
k	Boltzmann constant	<i>Greek symbols</i>	
k_r	rate constant of chemical reaction in gas phase	α and α'	sticking coefficients of nonexcited and excited molecules, respectively
k_{rp}	rate constant of heterogeneous chemical reaction in a porous catalytic particle	γ	fraction of excited molecules
k_{rp0}	pre-exponential factor in expression for constant of reaction rate in porous particle	ε_e''	effective loss factor
L	length of a capillary	ε_0	permittivity of a free space
n_a	number density of adsorbed molecules	λ_p	effective thermal conductivity of a porous particle
n_{rp}	number density of reactant molecules in a porous particle	ρ_p	mass density of a porous particle
P_s	recoil pressure of molecules escaping from the particle surface	τ_a	adsorption time
		ω	angular frequency
		<i>Subscripts</i>	
		a	adsorbed molecules
		c	capillary
		p	porous particle
		r	reacting substance

aerosol particle [12,13]. The mentioned effect of radiation is most pronounced in the free-molecular regime of gas flow, when the effect of surface processes is found to be the strongest.

Consider a question related to radiation-induced mass transfer in a cylindrical capillary with free-molecular gas flow in it. Assuming the fulfillment of the Henry law, we can write the equation for the density n_a of the molecules adsorbed on the internal surface of a capillary [14]:

$$\frac{1}{L^2} \frac{d}{dx} \left(D_s \frac{dn_a}{dx} - LbFn_a \right) = \frac{n_a}{\tau_a} - \alpha \left[\int_0^1 \left(\frac{n_a}{\tau_a} + I \right) K_1(|x - \xi|) d\xi + I_0K(x) + I_0K(1-x) \right], \quad (1)$$

$$I = (1 - \alpha) \left[\int_0^1 \left(\frac{n_a}{\tau_a} + I \right) K_1(|x - \xi|) d\xi + I_0K(x) + I_0K(1-x) \right], \quad (2)$$

where L is the length of the capillary; $x = X/L$ the dimensionless coordinate directed along the capillary axis; D_s the surface diffusion coefficient; F the force acting on the adsorbed molecules in the direction of the x coordinate and being due to the influence of external fields; b the mobility of the adsorbed molecules; τ_a the adsorption time; α the sticking coefficient of molecules; I_0 the density of the molecular flux entering into the capillary at $x = 0$ and $x = 1$; the functions K and K_1 characterize the probabilities of the transition of molecules from one surface element to another. The first and second terms on the left-hand side of Eq. (1) are related to transfer of adsorbed molecules along the surface by means of surface diffusion, and also under the influence of the external force acting on adsorbed molecules (e.g., in the case of the effect of external electric field on the adsorbed molecules, that possess an electric charge or a dipole moment). The first term on the right-hand side of Eq. (1) defines the density of the flux of molecules desorbing from the internal surface of the capillary; the integral term in the square brackets characterizes the density of

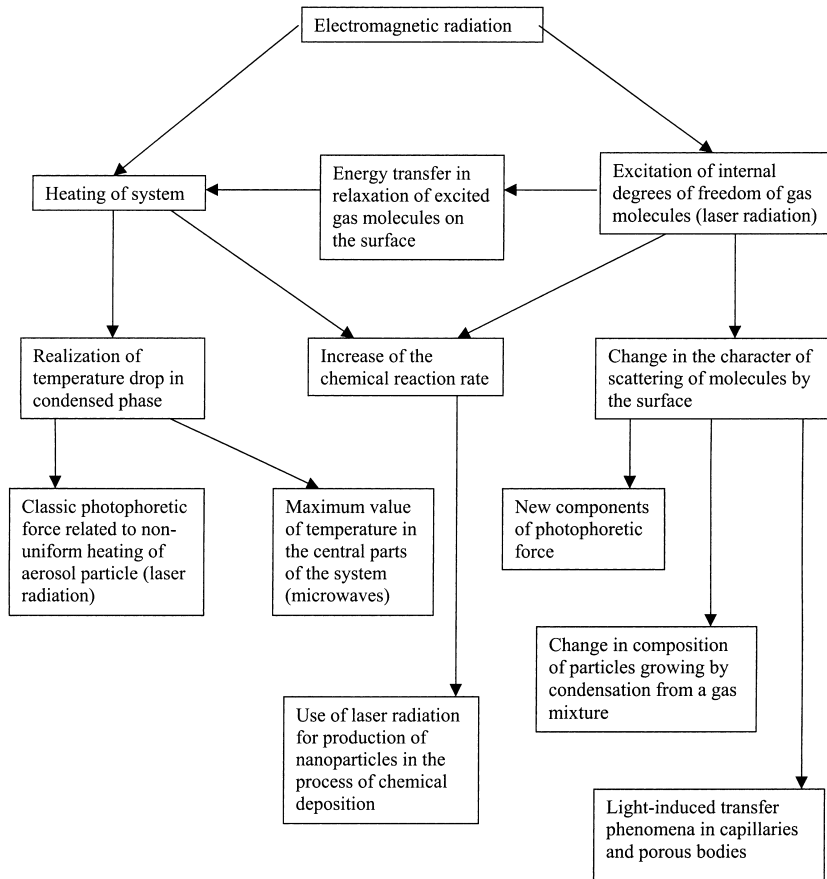


Fig. 1. Influence of electromagnetic radiation on heterogeneous systems.

the flux of molecules escaping from one element of the internal surface of the capillary and arriving at its another element. The terms outside the integral in the square brackets describe molecules entering into the capillary through the open ends and arriving at the element of the side surface in the vicinity of the x point. The quantity I characterizes the density of the flux of elastically reflected molecules. The flux of molecules leaving the capillary at $x = 1$ is determined as

$$N(1) = -\frac{2\pi R_c}{L} \left(D_s \frac{dn_a}{dx} - LbFn_a \right) \Big|_{x=1} + 2\pi R_c L \times \int_0^1 \left(\frac{n_a}{\tau_a} + I \right) K(1-x) dx + \pi R_c^2 I_0 K_2(1). \quad (3)$$

The value of $K_2(x)$ is obtained from the expression

$$\frac{dK_2}{dx} = -2lK(x), \quad (4)$$

where $l = L/R_c$. The boundary conditions for Eq. (1) and the approximate method of its solution are considered in [14,15].

In the case of an opaque porous body asymmetry may appear in the values of τ_a and α on the opposite sides of the porous body (at the opposite ends of capillaries) under the influence of resonance radiation due to radiation-induced desorption of molecules on one side of the body or to excitation of molecules in the gas phase, respectively. In turn, it leads to a difference between the values of n_a near both sides of the porous body (both ends of the capillary) and to an appearance of the resultant mass flux in the initially equilibrium system. The cases related to the asymmetry in τ_a and α were considered in [5,6], respectively. In the case of a porous body which is transparent relative to incident radiation, a situation can occur when the resultant mass flux appears in the previously equilibrium system due to velocity-selective excitation of molecules. Here, with detuning of the radiation frequency from the resonance frequency of gas molecules only the molecules that move towards one of the capillary ends can transit into excited state owing to the Doppler effect. The initiation of the resultant mass flux in a capillary due to the mentioned effect for the Maxwell scheme of scattering of molecules

by the surface and the free-molecular regime of gas flow was studied theoretically in [9,10] and experimentally in [11]. The effect of the change in the scattering indicatrix of the molecules being desorbed (of its difference from the cosine law) on capillary permeability under the action of resonance radiation was considered in [16]. The dependence of the radiation-induced mass flux on the relationship between the volumetric and surface fluxes, which is determined by the capillary radius, was discussed in [8]. The initiation of mass transfer under the action of the external force F (the second term on the left-hand side of Eq. (1)) was investigated in [17].

As it was noted above, new components of the force, acting on the aerosol particle, can appear in the field of resonance radiation. Further we will consider briefly the photophoretic force related to a change, in the field of the resonance radiation, of the recoil pressure of gas molecules emerging from the aerosol particle sides, opposite to the incident radiation. The mentioned force can take place, in particular, on velocity-selective excitation of gas molecules. It differs from zero also in the case of uniform heating of the aerosol particle (note that the classical photophoretic force is realized only when the particle is heated nonuniformly). The value of this force for the particle of radius R can be estimated as

$$F_p = 2\pi R^2 \int_0^\pi P_s \sin \theta \cos \theta d\theta, \quad (5)$$

where P_s is the recoil pressure of molecules emerging from the particle surface. The influence of resonance radiation on the asymmetry appearing in the values of P_s , which results in new components of the photophoretic force was considered in [18–20]. The effects appearing in simultaneous manifestation of resonance and thermal effects (i.e., of velocity-selective excitation of gas molecules and of uniform heating of the particle by radiation) were analyzed in [18]. The change in the scattering indicatrix of molecules emerging from the aerosol particle surface in the radiation field was discussed in [19], and relaxation of excited molecules on the aerosol particle was considered in [20].

The laser radiation which is resonant relative to gas molecules can also affect the process of chemical decomposition of some substance in a gas phase. The molecules of one of the reaction products appearing as a result of the reaction mentioned can condense forming aerosol particles [21]. Assuming the model of a reactor of ideal mixing and first-order reaction for the process of decomposition of a gas-precursor, we can write, similarly to [22], the following expression for the rate v_R of condensation growth of particles (further the parameters relating to the precursor and to the molecules of the condensing component are denoted by indexes A and B , respectively, and the parameters that characterize the excited molecules are primed):

$$\begin{aligned} v_R &= \frac{dR}{dt} \\ &= \frac{n_{A0}d}{\{1 + 1/[\tau k_r(1 - \gamma_A) + \tau k'_r \gamma_A]\}[1 + \tau/\tau_B]} \\ &\quad - d_1 \exp\left\{\frac{2\sigma V_m}{kTR}\right\}, \end{aligned} \quad (6)$$

where

$$\begin{aligned} d &= [\alpha_B - \gamma_B(\alpha_B - \alpha'_B)] \frac{v_B}{4n_s} \\ &\quad \times \left\{1 + [\alpha_B - \gamma_B(\alpha_B - \alpha'_B)] \frac{v_B R}{4D_B}\right\}^{-1}, \\ d_1 &= \frac{\alpha_{Be} P_{Be}}{n_s (2\pi m_B kT)^{1/2}} \left\{1 + [\alpha_B - \gamma_B(\alpha_B - \alpha'_B)] \frac{v_B R}{4D_B}\right\}^{-1}. \end{aligned}$$

Here t is the time; n_s the number density of molecules in the condensed phase; n_{A0} is the number density of the precursor molecules at the reactor inlet; τ the residence time of the gas in the reactor; k_r and k'_r are constants of the reaction rates for the nonexcited and excited molecules of the precursor; γ_i a part of the excited molecules of component i determined from the kinetic equations for transitions of the molecules from one state to another; σ the surface tension; V_m is the volume per molecule in the condensed phase; m_B the mass of the molecule of the component B ; α and α'_B are the condensation coefficients of nonexcited and excited molecules of the component B , D_B the diffusion coefficient of the component B in the gas phase; v_B the mean thermal velocity of the molecules of the component B ; P_{Be} is the saturated vapour pressure of the component B for a flat surface; α_{Be} is the evaporation coefficient of the molecules of the component B (it is supposed in Eq. (6) that the ordinary process of molecule evaporation–condensation occurs on the particle surface). The quantities k_r and k'_r can be presented as:

$$k_r = k_{r0} \exp\{-E_r/kT\}, \quad (7)$$

$$k'_r = k_{r0} \exp\{-(E_r - \Delta E_r)/kT\}, \quad (8)$$

where E_r is the activation energy of the precursor decomposition reaction in the absence of radiation; ΔE_r characterizes a change in the activation energy of this reaction in the radiation field; k_{r0} is the pre-exponential factor assumed constant.

It follows from Eq. (6) that one can control the growth of aerosol particles by means of excitation of the precursor molecules ($\gamma_A \neq 0$), as well as of the molecules of the condensing component ($\gamma_B \neq 0$) in a field of resonance (e.g., laser) radiation. In particular, it is seen from Eq. (6) that for $k'_r > k_r$ the rate of the growth of a particle increases with increasing γ_A . Note that the resonance laser radiation can also change composition of

the aerosol particle in its growth from a gas mixture [23,24].

3. Influence of microwave radiation on heterogeneous chemical reactions in porous particles

It is known that influence of the microwave radiation on the systems, where chemical reactions take place, can increase the reaction rate. For example, a possibility of using microwave radiation as a heating source is studied in [25,26] for ultrapyrolysis of chlorodifluoromethane in a fluidized-bed reactor. Advantages of the mentioned method of heating as compared with traditional ones are discussed. The reactor with porous catalyst particles in a gas-carrier is considered in [27]. Heating of microparticles-inclusions (active centres) in a particle of a porous catalyst in the field of microwave radiation is investigated. Note that temperature distribution in porous particles themselves and, correspondingly, the influence of this distribution on the course of heterogeneous chemical reactions in particles are not studied in [27]. Further these problems are considered in relation to a spherical particle of a porous catalyst.

We can write the following expression for the number of molecules N_p that undergo reaction in the porous particle:

$$N_p = 4\pi \int_0^{R_p} s_p k_{rp} n_{rp} r^2 dr, \quad (9)$$

where r is the radial coordinate reckoned from the particle centre; R_p the radius of the porous particle; s_p the specific surface per unit volume of the porous particle.

The number density of the reacting molecules in the porous spherical particle n_{rp} in the case of the first-order reaction is determined from the equation [28]

$$\frac{\partial n_{rp}}{\partial t} = \frac{D_e}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial n_{rp}}{\partial r} \right) - s_p k_{rp0} \exp \left\{ -\frac{E_{rp}}{kT_p(r)} \right\} n_{rp}. \quad (10)$$

Here E_{rp} is the activation energy of a heterogeneous chemical reaction on the surface of the porous catalyst; D_e the effective diffusion coefficient of the reacting molecules in the particle; $T_p(r)$ the particle temperature.

The boundary conditions for Eq. (10) can be written as:

$$\left. \frac{\partial n_{rp}}{\partial r} \right|_{r=0} = 0, \quad (11)$$

$$n_{rp}|_{r=R_p} = n_0. \quad (12)$$

The equation for temperature distribution in the porous particle is given by

$$\rho_p c_p \frac{\partial T_p}{\partial t} = \frac{\lambda_p}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T_p}{\partial r} \right) + q + H_r s_p k_{rp0} \exp \left\{ -\frac{E_{rp}}{kT_p(r)} \right\} n_{rp}, \quad (13)$$

where λ_p is the effective thermal conductivity of the porous particle; ρ_p and c_p are the mass density and specific heat of the porous particle, respectively; H_r the heat of the chemical reaction per molecule. The last term in Eq. (13) is related to the heat source which is due to chemical reaction; q the power of the microwave radiation dissipated per unit volume of the porous particle, which is expressed as [4]

$$q = \omega \varepsilon_0 \varepsilon_e'' E_{rms}^2, \quad (14)$$

where ω is the angular frequency, ε_0 the permittivity of a free space, ε_e'' the effective loss factor, E_{rms} the rms electric field strength, the value of q is further assumed to be constant over the particle volume.

The boundary conditions for Eq. (13) under the assumption on heat transfer with the surrounding medium according to the Newton law have the form

$$\left. \frac{\partial T_p}{\partial r} \right|_{r=0} = 0, \quad (15)$$

$$-\lambda_p \left. \frac{\partial T_p}{\partial r} \right|_{r=R_p} = h_p (T_p - T_g), \quad (16)$$

where h_p is the heat transfer coefficient.

Below some effects related to heating of the spherical particle of the porous catalyst by microwave radiation will be discussed. With neglecting the last term on the right-hand side of Eq. (13) the stationary solution of this equation with the boundary conditions (15) and (16) is given by

$$T_p = T_g + \frac{Q_p}{4\pi R_p^2 h_p} + \frac{Q_p}{8\pi R_p \lambda_p} (1 - \bar{r}^2) = T_{ps} + \frac{Q_p}{8\pi R_p \lambda_p} (1 - \bar{r}^2), \quad (17)$$

where $\bar{r} = r/R_p$, T_g is the gas temperature; Q_p the total power of the microwave radiation absorbed by the porous particle ($Q_p = 4\pi R_p^3 q/3$).

It follows from Eq. (17) that the temperature of the porous particle increases from the periphery to the particle centre; the difference between the temperatures at $r = 0$ and $r = R_p$ is determined as

$$\Delta T_p = T_p(0) - T_p(R_p) = \frac{Q_p}{8\pi R_p \lambda_p}. \quad (18)$$

It is seen from expression (18) that ΔT_p increases with increasing value of Q_p and with decreasing λ_p .

The solution of the stationary heat conduction equation with energy supply through the particle surface

for the same boundary conditions and the same radiation power absorbed by the particle gives for the temperature of the particle the following value:

$$T_p = T_{ps} = T_g + \frac{Q_p}{4\pi R_p^2 h_p}. \quad (19)$$

As it follows from Eqs. (17) and (19), the particle surface temperature is the same for both considered variants of heating in the case of the same values of Q_p but the mean stationary temperature of the particle for volumetric absorption of radiation (for example, with microwave particle heating) is higher than the particle temperature for surface absorption (at the same character of heat exchange with the surrounding medium). This results in a larger number of reacted molecules in the porous particle in the first case than in the second.

Let us consider this situation when the chemical reaction is the controlling process. The diffusion is assumed to be very fast and the concentration of reactant is considered to be constant over the particle and to be equal to n_0 .

We assume also for simplicity that the following conditions take place:

$$\frac{Q_p}{8\pi R_p \lambda_p T_{ps}} \ll 1, \quad \frac{E_{rp} Q_p}{8\pi R_p \lambda_p k T_{ps}^2} \ll 1. \quad (20)$$

Under above mentioned assumptions an expression for N_p can be obtained from Eqs. (9) and (17)

$$N_p = \frac{4}{3} \pi R_p^3 n_0 s_p k_{rp} (T_{ps}) \left(1 + \frac{1}{20} \frac{E_{rp} Q_p}{k T_{ps}^2 \pi R_p \lambda_p} \right). \quad (21)$$

It is seen from Eq. (21) that in the case of volumetric absorption of radiation the quantity of molecules that react in the catalyst particle is higher than in the case of the surface absorption of radiation in the same power of absorbed energy.

Moreover, in accordance with what has been said above, in volumetric absorption the rate constant of the heterogeneous chemical reaction in the particle centre is larger than on its surface. On the other hand, in the general case, where diffusion process is essential, it is more difficult for the reacting molecule, due to diffusion resistance, to reach the particle centre than the point which is closer to the external surface of the particle. Because of simultaneous manifestation of these factors, the course of the chemical reaction over the particle volume with microwave radiation heating can be more uniform than for an isothermal particle, or a situation can be realized when the reaction rate at the particle centre is maximum. The latter case (especially with increase in the value of e'_c with temperature, which is often observed in practice [4]) can lead to the initiation of combustion process inside the porous particle or to its explosion. An increase in the reaction rate inside the porous particle can be also substantial, when the reac-

tion products block an access of the reacting molecules to an internal part of the porous catalytic particle. In particular, such a case can be realized in the process of pyrolytic chemical deposition of substance when depositing molecules form a solid layer on the surface, which decreases the permeability of the pores and creates a possibility, in principle, of complete blocking the transfer of the reacting molecules to the internal part of the porous particle. This decreases the effectiveness of the particle of the porous catalyst.

The condition of a uniform course of a heterogeneous chemical reaction over the volume of a spherical catalytic particle consists in the constancy of the quantity $J_p = k_{rp} n_{rp}$. Let us find this condition for the stationary case with constant values of the quantities q and s_p . The stationary solution of Eq. (10) with the boundary conditions (11) and (12) at $J_p = \text{const}$ has the form

$$n_{rp} = n_0 + \frac{J_p s_p}{6D_e} (r^2 - R_p^2). \quad (22)$$

Taking into account Eq. (22) and the expression for the constant of the reaction rate, we obtain the condition of uniform course of a heterogeneous chemical first-order reaction over the volume of a porous spherical particle

$$T_p = \frac{E_{rp}}{k \ln \frac{k_{rp} n_0}{J_p} \left[1 - \frac{J_p s_p R_p^2}{6D_e n_0} (1 - \bar{r}^2) \right]}. \quad (23)$$

It follows from Eq. (23) that the realization of the condition $J_p = \text{const}$ is possible only on condition that $(J_p s_p R_p^2 / 6D_e n_0) < 1$. The particle temperature must rise to the particle centre (as it is shown earlier, this condition is realized in the case of the particle heating by microwave radiation). This case is analogous to the one considered in [29] where the problem of uniform course of a heterogeneous chemical reaction was investigated for a single cylindrical capillary with the free-molecular regime of gas flow. It follows from the results of [29] that the temperature of the internal capillary surface must be higher in the middle part of the capillary for uniform course of a heterogeneous chemical reaction.

4. Conclusions

The paper analyzes the possibility to control transfer phenomena in heterogeneous systems gas-condensed phase by means of electromagnetic radiation. The processes considered are related both to excitation of internal degrees of freedom of molecules in the resonance laser radiation field and heating of the system in the microwave radiation field, which leads to the origin of the so-called inverse gradient of temperature (when a maximum value of temperature takes place in the system centre). An expression is obtained for temperature distribution in a spherical particle of a porous catalyst

when uniformity of course of a heterogeneous chemical reaction over the particle volume takes place.

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