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2D GAS TRANSFER DYNAMICS  
IN AN ADSORBATE-ADSORBENT OPEN SYSTEM  
OF THE INTERACTING ADATOMS

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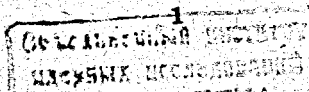
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## 1. Introduction

The experimental results of the gas sorption on a solid surface indicate the enough various behaviour of the physical adsorption heats with the growth of the surface coverage rate [1—4]. The decreasing of the adsorption heats with the surface coverage is often connected with the surface inhomogeneities. But the increasing of the heats is due to the interactions (attractions) among the adsorbing molecules or atoms [4—7]. A great number of the experimental works on the adsorption of gases and vapors leads to the conclusion that the adatom interactions in an adsorptional system must be taken into account. The latter is important in studying the adsorptional phenomena and their influence on the gas or liquid transfer processes. In [8—14] we proposed a model for the description of the gas transfer dynamics in an adsorbate—adsorbent open system. In our model, unlike the well-known sorption dynamics models, the interactions between adatoms and activated complexes (AC) are explicitly taken into account. Within the proposed model the gas isotherms at different values of the adatom interaction parameter  $\beta\epsilon$  ( $\beta = (k_B T)^{-1}$ ) were numerically calculated. The calculations were performed for three approximations of the particle correlation effects, namely, for the molecular field approximation (MFA), quasichemical approximation (QCA) and polynomial fission (PF) [8,9]. The analysis of the numerical results, obtained in [8,9], shows essential nonlinear dependence of the surface coverage rate  $\theta$  on the gas concentration  $\nu$  with the growth of  $\beta\epsilon$ . In [8—10], on the basis of the numerical solution the system of eqs.(1)–(3), for 1D physical adsorption, a "hidden mechanism" of the concentrational wave formation is found that is owed to the parameter  $\beta\epsilon$ . It is very important that the jumpy behaviour of the concentrations  $\theta$  and  $\nu$  in both gas and adsorbate phases exists during the period of  $t \leq \tau_0$ . For the  $t > \tau_0$  we have a uniform distribution of the concentrations in the adsorptional system. That is in the system under consideration "characteristic", "chemical" time  $\tau_{ch} = \tau_0$  of the interface mass exchange arises. The detailed analysis of the obtained results indicate existing a threshold for saturation values of the surface coverage rate  $\theta_s$ , depending on the parameter  $\beta\epsilon$ .

The paper is constructed as follows. Section 2 is devoted to the results of calculations of the basic characteristics of the 1D nonequilibrium adsorption dynamics. In section 3 we give an estimation of the "chemical" time  $\tau_{ch}$  for the gas nondissociative (physical) adsorption and desorption. Section



4 generalizes the obtained results for the 2D sorption dynamics assuming stochasticity of the transfer processes in the adsorbing system. The stochastic process in the adsorbate-adsorbent open system is simulated by introducing the spatial random adatom-atom, adatom-AC interactions. The paper concludes a brief summary.

## 2. The threshold for the surface coverage saturation in the adsorbing system of the interacting particles

The equations of the model of the interacting adatoms and activated complexes for the concentrations in the gas phase and in the sorbent look like [8-14]:

$$\frac{\partial \nu}{\partial \tau} + \frac{\partial \theta}{\partial \tau} + (\bar{v}\bar{\nabla})\nu = d\Delta\nu, \quad (1)$$

$$\bar{v}\bar{\nabla} = v_1 \frac{\partial}{\partial \xi_1} + \dots + v_D \frac{\partial}{\partial \xi_D}, \quad \Delta = \frac{\partial^2}{\partial \xi_1^2} + \dots + \frac{\partial^2}{\partial \xi_D^2},$$

$$\frac{\partial \theta}{\partial \tau} = v_{ads} - v_{des} = \left\{ \frac{\nu}{ap_0} - 1 \right\} \Phi(T, \varepsilon, \varepsilon_1, z, \theta), \quad (2)$$

$$\theta = \phi(ap_0)_T = \sum_{m=0}^z C_z^m (1-\theta)^{z-m} \theta^m \frac{ap_0}{ap_0 + \exp(-m\beta\varepsilon)}. \quad (3)$$

Here  $C_z^m = \frac{z!}{m!(z-m)!}$  is a number of adatom distribution from  $z$  to  $m$ .

In the equations (1)-(3)  $\nu(\bar{\xi}, \tau)$  is the dimensionless concentration of the adsorbed matter in gas or liquid stream;  $\theta(\bar{\xi}, \tau)$  - the adsorbate concentration in the adsorbent pores ( $0 \leq \theta \leq 1$ );  $v$  - the average linear velocity of the gas stream, carrying through a tube, containing adsorbent;  $d$  - the coefficient of longitudinal diffusion;  $\tau_0$  has the time dimension and it is defined by the characteristics of the adsorption transition states (viz., activated complexes (AC)), the adsorbate and temperature of the system (we call  $\tau_0$  "chemical" time);  $\Phi(T, \varepsilon, \varepsilon_1, z, \theta)$  expresses the adsorption and desorption velocities dependency on the medium temperature and the effective adatom-adatom  $\varepsilon$ , adatom-AC  $\varepsilon_1$  interaction parameters;  $p_0$  - the equilibrium pressure, which corresponds to the surface coverage  $\theta$  and the function  $\Psi(\theta)$  is defined by the adsorption and desorption isotherms.

In our previous papers [8-14], on the basis of the numerical solution the system of eqs.(1)-(3), for the 1D physical adsorption, a "hidden mechanism" of the concentrational wave formation is found that is owed to the parameter  $\beta\varepsilon$ . It should be noted that the numerical results in [8-10] were obtained for several selected values of the adatom interaction parameter, namely, for  $\beta\varepsilon =$

0.0, 1.0, 1.5, 2.0, 2.5. Here we present results of the computer simulations for full ranges of the values of the adatom interaction parameter  $\beta\varepsilon \in [0.0 \div 2.5]$ . Let us denote through the  $\theta_s$  a saturation, maximum value of the surface coverage rate  $\theta(\xi, \tau)$ . To  $\theta_s$  lead us the calculations of the dynamical eqs.(1)-(3) in the limit  $\tau = \frac{t}{\tau_0} \rightarrow 1$ . The time, at which the system reaches the saturation, ending value of concentration in the sorbent pores (viz.,  $\theta_s$ ), we regard as  $\tau_s$ . Table I shows the dependence of  $\theta_s$  and  $\beta\varepsilon$ , which are calculated with the step  $\Delta(\beta\varepsilon) = 0.1$ . These results are drawn in Fig.1 and Fig.2.

$\beta\varepsilon$	0.0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8
$\theta_s$	0.12	0.13	0.14	0.15	0.16	0.17	0.19	0.22	0.26
$\tau_s$	1	1	1	1	1	1	1	1	1

$\beta\varepsilon$	0.9	1.0	1.02	1.05	1.08	1.1	1.15	1.18	1.2
$\theta_s$	0.32	0.48	0.50	0.58	0.68	0.81	0.91	0.93	0.94
$\tau_s$	1	1	1	1	1	1	1	1	1

$\beta\varepsilon$	1.25	1.3	1.4	1.5	0.6	1.7	1.8	1.9	2.0
$\theta_s$	0.95	0.96	0.98	0.99	0.99	0.99	0.99	0.99	0.99
$\tau_s$	0.9	<0.8	0.6	<0.6	0.4	0.2	<0.2	<0.2	<0.2

From these results it is easy to see that there exists a threshold  $\eta_{cr} \equiv (\beta\varepsilon)_{cr} > 1.0$  of the values of  $\beta\varepsilon$ , depending on which the saturation surface coverage reaches  $\theta \rightarrow 1$ . It should be noted that the time  $\tau_s$  for reaching such a saturation of the adsorbate concentration decreases essentially with the growth of  $\beta\varepsilon$ . That is the interactions of atoms or molecules in the nonequilibrium adsorbing system lead to the fast adsorption of a matter (gas or liquid) on the surface and to greater values of the coverage rates  $\theta_s$ . It is very important to regard the application of the obtained results to interpretate some experimental data of the gas adsorptions on solid surface, for example, on crystalline zeolites [5-7]. As it is known, the adsorption of many gases (for example,  $O_2$ ,  $N_2$ ,  $Ar$ ,  $Xe$  and so on) on the crystalline zeolites is characterized by the fast velocities. Here the pressure, temperature, particles size and other degidration conditions influence on the adsorption velocity. Among these factors the interactions of the adsorbates play a considerable role and the numerical results in Table I coincides with the fast adsorption of gases. It is well known that the crystalline zeolites have a strict periodical structure and the adsorption and desorption processes on them goes physically (i.e. with adsorption heats essentially less than in the case

of chemisorption). To describe such a regularly periodical structure, we use a quantum-statistical Ising type Hamiltonian for an adsorbate-adsorbent open system, on the basis of which the above nonlinear system of equations (1)–(3) is derived. The Hamiltonian for the inhomogeneous periodical surface, which delivered a definite number of adsorption centers, looks like [8–10]:

$$H = -\mu \sum_f n_f - \sum_{\langle fg \rangle} n_f n_g, \quad (4)$$

$\mu$  is a chemical potential of the system;  $\varepsilon$  (eV) – the effective adatom interaction parameter ( $\varepsilon > 0$  – corresponds to physical adsorption,  $\varepsilon < 0$  – to chemisorption) and the summation is carried out over all adsorption active sites ( $f$ ) on the surface, and all nearest to the ( $f$ ) neighbours ( $g = 1, 2, \dots, z$ ) ("active element").

The parameter  $\varepsilon$ , as it is shown in papers [12–14], can be calculated from the experimental, calorimetric measurements of the adsorption heats  $\varepsilon = \frac{(\Delta Q)_{exp}}{N_A z n}$ , where  $n = 1, 2$  – for nondissociative (one-central, physical) and dissociative adsorptions from the gas phases, respectively. The coordination number  $z$  expresses also the dimension of the "active element", which adsorbed (or desorbed) particles from a carrying gas stream. In the term of the "active element" we imply, for example, polymer  $z = 2$  ( $D = 1$ ), surface  $z = 3, 4, 6$  ( $D = 2$ ) and crystall (in particularly, zeolite)  $z = 6, 8, 12$  ( $D = 3$ ). The physical adsorption heats lie in the interval  $\Delta Q \approx 1.0 \div 10 \frac{kcal}{mol}$  and for an active surface ( $z = 4$ ) we have  $\varepsilon \in [0.01 \div 0.1]$  eV. At the normal temperatures  $T \approx 300$  K,  $\beta \approx 40$  (eV) $^{-1}$  we get that  $\beta\varepsilon \in [0.4 \div 4.0]$ . Thus, the values of  $\beta\varepsilon$ , under which we numerically simulate the nonequilibrium sorption dynamics, correspond to the area of physical adsorption and desorptions.

### 3. The estimation of the "chemical" time

It is very important that the jumpy behaviour of the concentrations  $\theta$  and  $\nu$  in both gas and adsorbate phases exists during the period of  $t \leq \tau_0$ . For the  $t > \tau_0$  we have a uniform distribution of the concentrations in the adsorptional system. That is in the system under consideration the "characteristical", "chemical" time of the interface mass exchange arises. The chemical time  $\tau_0$  is characterized by the temperature of the system, the adatoms and the transition state partition functions  $J_a$  and  $J^*$ , respectively, and it looks like [8–10]:

$$\tau_0 = \frac{J_a h \beta}{J^*} e^{\beta\varepsilon_k}. \quad (5)$$

In eq.(5)  $h$  is the Planck constant and  $\varepsilon_k$  – the adsorption activation energy.

Let us estimate, even though approximately, the order of the chemical time  $\tau_0$ .

As it is known [2,4], the relation  $\frac{J^*}{J_a}$  expresses the internal freedom degrees of the AC and adsorbing molecules and there may exist, for example, the following cases: a) If the AC and adsorbing molecules have the same degrees of freedom, then  $J^* = J_a$ , i.e.  $\frac{J^*}{J_a} = 1$ ; b) Inasmuch as the AC are binded to the surface less strongly then the adsorbing particles, they may have more freedom to move, and, consequently,  $J^* \gg J_a$ . In the limit cases, when the AC have both directional and rotational degrees of freedom, we have  $\frac{J^*}{J_a} = 10^3 \div 10^4$ . Thus, for the approximate estimation of  $\tau_0$ , we choose, without severity, that  $J^* \approx J_a$ , i.e.  $\frac{J^*}{J_a} \approx 1$ .

The values of the adsorption activation energy  $\varepsilon_k$ , as it is well known, change in wide intervals, depending on the type of the adsorption processes. But for the two existing types of adsorption (viz., physical adsorption and chemisorption) we have that  $\varepsilon_k \sim q = \frac{\Delta Q}{N_A}$ , where  $q$  is the adsorption heat. To illustrate the heat capacities of these two types of adsorption, we bring Fig.3. From Fig.3 we see that chemisorption is characterized by the large heats (curve 1). For the nondissociative, physical adsorption (curve 1) we have, consequently, small values of the adsorption heats. Using the values of the physically adsorption heats, from the previous section, at the normal temperatures we get  $\beta\varepsilon_k \in [1.5 \div 15]$ . Therefore, the values of the characteristical adsorption time  $\tau_0$  in our model lie in the interval:

$$\tau_0 \in [10^{-12} \div 10^{-7}](sec).$$

It is important to note that this interval fully includes the whole experimentally realized temporary area of physical adsorption processes [15]. Indeed, the adsorption time in the dynamical processes is defined as time, during which an adsorbing atom or molecule is located on an adsorbent surface [16]. The average lifetime of a molecule on the surface can be estimated by the Frenkel equation

$$\tau_0 = \bar{\tau} e^{\frac{Q}{RT}}, \quad (6)$$

where  $\bar{\tau}$  is the time of molecular oscillations ( $\bar{\tau} \in [10^{-13} \div 10^{-12}](sec)$ ), and  $Q$  is the interaction energy of a molecule with the surface (viz., the adsorption heat). The estimation of  $\tau_0$  in the areas of physical adsorption, on the basis of eq.(6), leads us to the above temporary interval for  $\tau_0$ . I.e. we come to a good agree of the two independently calculated results for  $\tau_0$ . This indicates that the proposed approach and the model of the interacting adatoms may be usefull to describe the nonequilibrium adsorptional processes.

#### 4. Stochastic gas transfer dynamics on an active surface

In this section we simulate the two-dimensional gas transfer dynamics on the active surface, assuming the stochasticity of the adsorptional processes in the system. The stochastic process in the adsorbate-adsorbent open system is simulated by introducing the spatial random adatom-adatom, adatom-AC interactions (further we will use the designations  $\eta \equiv \beta\varepsilon$  and  $\eta_1 \equiv \beta\delta\varepsilon = \beta(\varepsilon_1 - \varepsilon)$ ):

$$\eta \rightarrow \eta(\vec{\xi}) = \eta_0 + \tilde{\eta}(\vec{\xi}), \quad \eta_1 \rightarrow \eta_1(\vec{\xi}) = \eta_{10} + \tilde{\eta}_1(\vec{\xi}), \quad (7)$$

where  $\eta_0 = \langle \eta \rangle$ ,  $\eta_{10} = \langle \eta_1 \rangle$  and  $\langle \tilde{\eta} \rangle = 0$ ,  $\langle \tilde{\eta}_1 \rangle = 0$ . In expression (7)  $\tilde{\eta}$  and  $\tilde{\eta}_1$  are the random variables, and their values are given by the uniform pseudorandom number generator. Fig.4 shows the examples of the introduced random adatom interactions in the system.

The calculations for the difference values of the parameters  $\eta_0 = \langle \eta(\vec{\xi}) \rangle$  and  $\eta_{10} = \langle \eta_1(\vec{\xi}) \rangle$  are fulfilled. Averaging the expressions  $\nu(\xi_1, \xi_2, \tau)$  and  $\theta(\xi_1, \xi_2, \tau)$  over all realizations of the random variable  $\eta$  were carried out usually

$$\langle A \rangle = \sum_{i=1}^N A(\eta_i) P_A(\eta_i).$$

Here  $A \equiv \{ \nu(\xi_1, \xi_2, \tau); \theta(\xi_1, \xi_2, \tau) \}$ , and the equal probability at each realization is assumed as  $P_A(\eta_1) = P_A(\eta_2) = \dots = P_A(\eta_N) = \frac{1}{N}$ . The evolutions of concentrations  $\langle \theta(\vec{\xi}, \tau) \rangle$  (left) and  $\langle \nu(\vec{\xi}, \tau) \rangle$  (right) are shown in Figs.5, 6, 7, 8, 9 and 10. The analysis of the obtained results shows that the dynamics of the system depends essentially on the quantity of  $\eta_0 = \langle \eta(\vec{\xi}) \rangle$ , as in the case  $\eta = \beta\varepsilon = const$ . That is:

1) when  $\eta_0 < \eta_{cr}$  (see section 2), we observe stable oscillations (for large times  $\tau \gg 1$  ( $t \gg \tau_0$ )) of the adsorbate concentration  $\langle \theta(\vec{\xi}, \tau) \rangle \sim f(\eta(\vec{\xi}))$ ;

2) when  $\eta_0 \geq \eta_{cr}$ , the oscillations of  $\langle \theta(\vec{\xi}, \tau) \rangle \sim f(\eta(\vec{\xi}))$  exist during the period  $\tau \leq \tau_{ch}$ ; for the  $\tau > \tau_{ch}$  we have a uniform, equilibrium distribution of concentration  $\langle \theta(\vec{\xi}, \tau) \rangle$  in the sorbent pores.

But in both cases the wave-like evolution of  $\langle \nu(\vec{\xi}, \tau) \rangle$ , similar to a "hidden mechanism" of the concentrational wave formation in 1D physical adsorption [8-14], is observed in the interval  $\tau \in [0 \div 1]$  ( $t \in [0 \div \tau_{ch}]$ ). For the  $\tau > \tau_{ch}$  we come to an equilibrium distribution of concentration  $\langle \nu(\vec{\xi}, \tau) \rangle$  in the moving phases.

In Figs.5 and 6 the results of the numerical solution of eqs.(1)-(3) for five moments of time in the interval  $\tau = \frac{t}{\tau_0} \in [0, 1]$ , with the step  $\Delta\tau = 0.2$ ,

for  $\eta_0 = 1.0$  ( $\eta_0 < \eta_{cr}$ ) and  $\eta_0 = 2.0$  ( $\eta_0 > \eta_{cr}$ ) are shown, respectively. From Figs.5 and 6 it is easy to see the irregular evolutions of concentrations  $\langle \theta(\vec{\xi}, \tau) \rangle$  (left) and  $\langle \nu(\vec{\xi}, \tau) \rangle$  (right). It should be noted that such a regime of the wave formation in the case of  $\eta_0 = 2.0$  exists for the period of time  $\tau \rightarrow 1$ , i.e. for  $t \rightarrow \tau_0$ . In the limits  $t > \tau_0$ , as in the case of  $\eta = const$ , we have a uniform, equilibrium distribution of concentrations in the system (see Fig.7 for  $\eta_0 = 2.0$  and  $\tau = 1.0, 2.0, 3.0, 4.0$  and 5.0). As regards to  $\eta_0 < \eta_{cr}$  in the moments  $t > \tau_0$ , we will have stable oscillations of the adsorbate phase, which do not influence the evolutions of gas concentration  $\nu(\vec{\xi})$ . In Fig.8 we see the example of such regime for  $\eta_0 = 1.0$  and  $\tau = 1.0, 2.0, 3.0, 4.0$  and 5.0. The above results, regarding to the jumpy wave-like concentrational formations in the intervals  $t \leq \tau_0$  and  $t > \tau_0$ , we can observe more clearly in one dimension. In Figs.9, 10 and 11 the evolutions of the concentrations  $\langle \theta(\xi, \tau) \rangle$  and  $\langle \nu(\xi, \tau) \rangle$  in 1D stochastic adsorptional system are shown (Fig.9 corresponds to  $\eta = 1.0$  ( $\eta < \eta_{cr}$ ); Figs.10 and 11—to  $\eta = 2.0$  ( $\eta > \eta_{cr}$ )).

#### 5. Conclusion

We investigate the gas transfer dynamics under the influence of the "active element". In the term of the "active element" we imply quasi-one-dimensional system (for example, polymer) ( $D = 1$ ), surface ( $D = 2$ ) and crystal (in particularly, zeolite), ( $D = 3$ ). The active element may adsorb (or desorb) particles from a carrying gas stream. For the description of the gas transfer dynamics in the adsorbate-adsorbent open system we proposed a model in which, unlike the well-known sorption dynamics models, the interactions between adatoms and activated complexes (AC) are explicitly taken into account. From the results obtained here it is found that there exists a threshold of the values of the adatoms interaction parameters, depending on which the surface coverage reaches the saturation values  $\theta \rightarrow 1$  very fast. The time  $\tau_s$  for reaching such a saturation of the adsorbate concentration decreases essentially with the growth of  $\beta\varepsilon$ . I.e. the interactions of atoms or molecules in the nonequilibrium adsorbing system lead to the fast adsorption of a matter (gas or liquid) on surface and to larger values of the coverage rates  $\theta_s$ . As an example, we regard the obtained results to the adsorption of gases on the crystalline zeolites and the adsorbate interactions factor may play an essential role on the adsorption velocity here. It is very important that the jumpy behaviour of the concentrations  $\theta$  and  $\nu$  in both gas and adsorbate phases exists during the period of  $t \leq \tau_0$ . For the  $t > \tau_0$  we have a uniform distribution of the concentrations in the adsorptional system. That is in the system under consideration "characteristical", "chemical" time

$\tau_{ch} = \tau_0$  of the interface mass exchange arises. The estimation of "chemical" time in the areas of physical adsorption was carried out. It is shown that the temporary interval for the adsorption processes fully includes the whole experimentally realized temporary area of physical adsorption processes. The 2D gas transfer dynamics on the active surface, assuming the stochasticity of the adsorptional processes in the system was numerically studied. The stochastic process in the adsorbate-adsorbent open system is simulated by the introducing of the spatial random adatom-adatom, adatom-AC interactions. On the basis of the comparison and analysis of the results for the 2D stochastic processes obtained here and for the 1D ones (see [12-14]) we come to the conclusion that

- 1) the dynamics of concentrations in the adsorbing system, under the influence of the spatial random adatom-adatom interactions, depends on the relations of  $\eta_0 < \eta_{cr}$  and  $\eta_0 > \eta_{cr}$  ( $\eta_0 = \langle \eta(\xi) \rangle = \langle \beta \epsilon(\xi) \rangle$ );
- 2) in the case of  $\eta_0 < \eta_{cr}$  we observe oscillations of concentration  $\theta$  in the adsorbate for  $t > \tau_0$ , and they do not influence the evolution of concentration  $\nu$  in the gas stream;
- 3) for  $\eta_0 > \eta_{cr}$  the wave-like oscillations of concentrations  $\theta$  and  $\nu$  are limited by the temporary interval  $t \leq \tau_{ch}$ ;
- 4) the nonlinear wave-like oscillations of the gas concentration  $\nu$  exist to the moments  $t \rightarrow \tau_0$ , i.e., as in the case of  $\beta \epsilon = const$ , the dynamics of the system depends on the "chemical" time.

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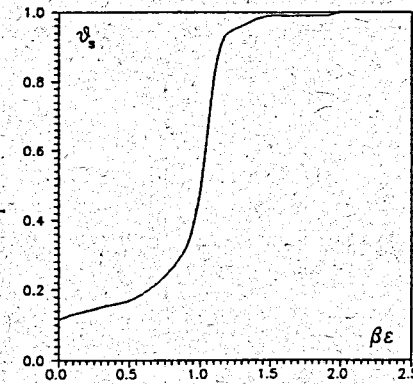


Fig.1. The surface coverage saturation values  $\theta_s$  versus the adatoms interaction parameter  $\eta = \beta \epsilon$ .

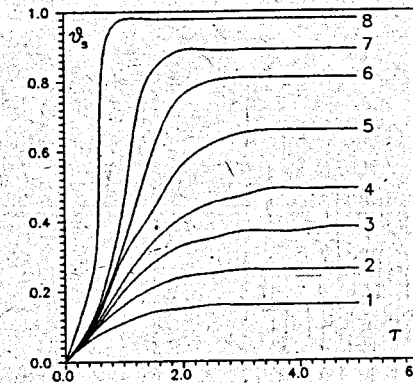


Fig.2. The evolutions of surface coverage saturation values  $\theta_s$  versus the time  $\tau$  for the values of  $\beta \epsilon = 0.0(1), 0.1(2), 0.3(3), 0.5(4), 0.6(5), 0.8(6), 0.9(7), 1.0(8)$ .

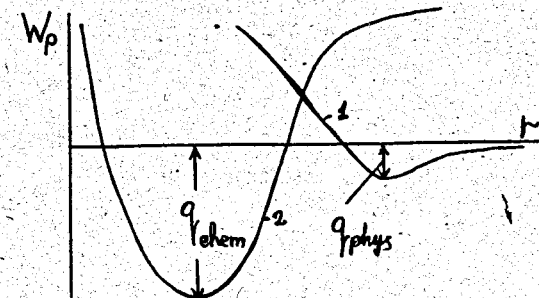


Fig.3. The potential energy in the physical adsorption (curve 1) and chemisorption (curve 2).

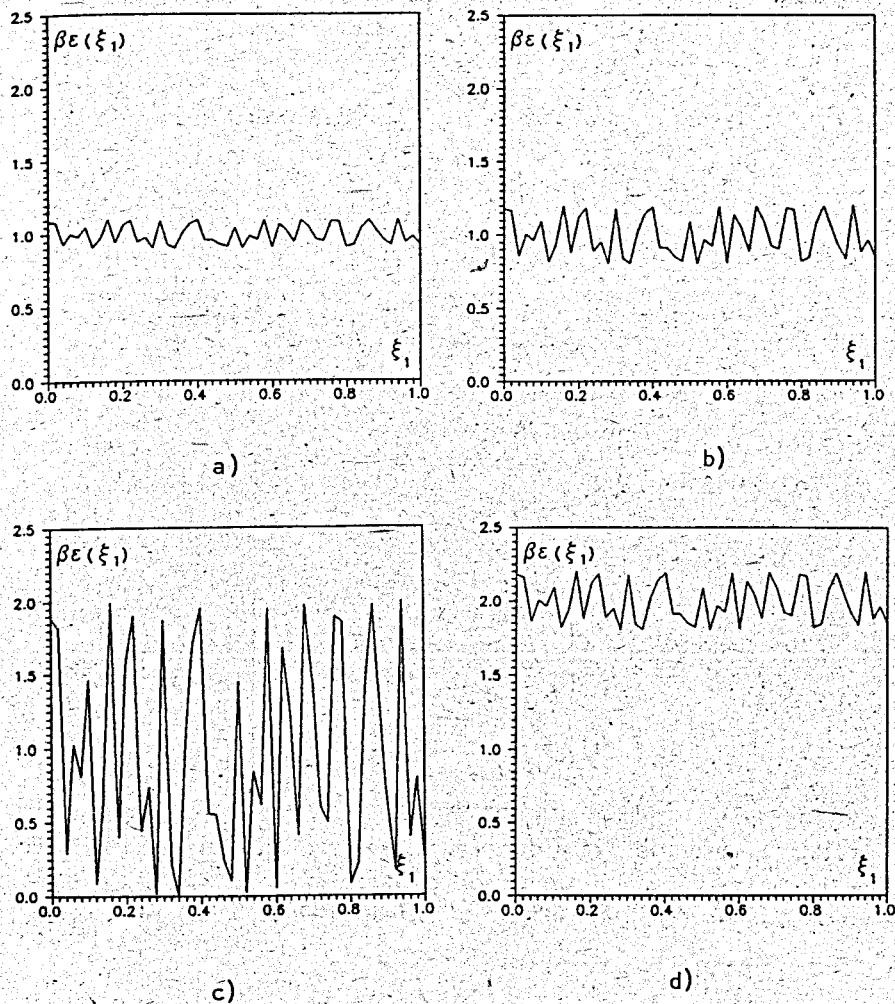


Fig.4. The examples of the introduced spatial random adatom interactions in the adsorbing system.

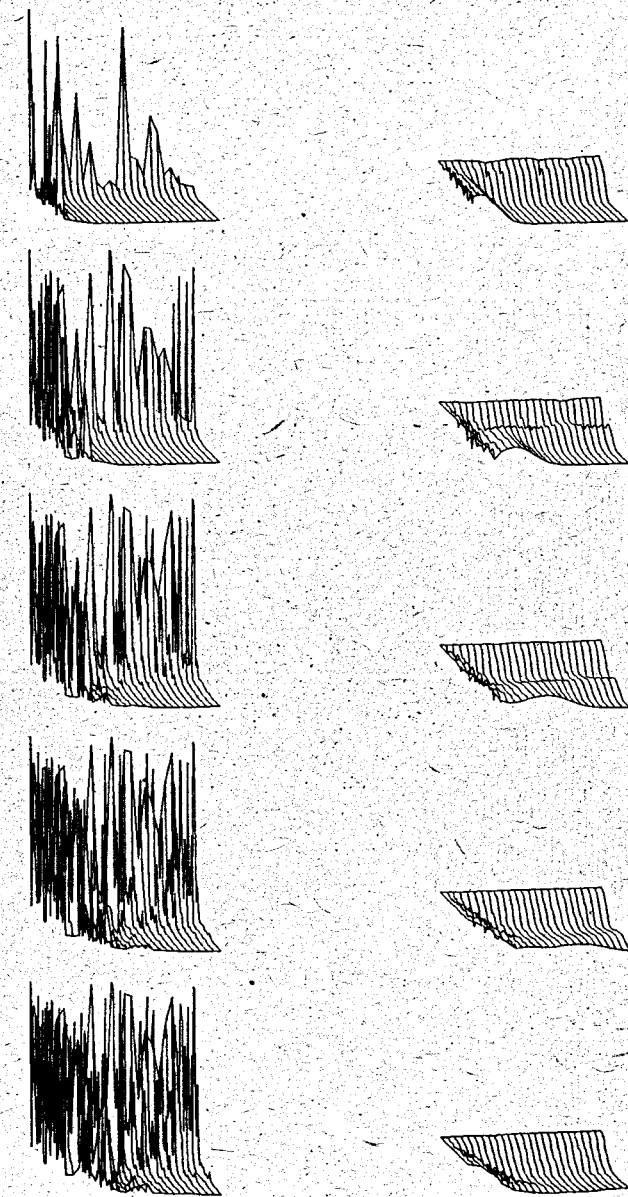


Fig.5. The evolutions of concentrations  $\langle \theta(\vec{\xi}, \tau) \rangle$  (left) and  $\langle \nu(\vec{\xi}, \tau) \rangle$  (right) in 2D adsorbing system at the moments  $\tau = 0.2, 0.4, 0.6, 0.8, 1.0$  ( $t \leq \tau_0$ ) for  $\eta_0 = 1.0$  ( $\eta_0 < \eta_{cr}$ ).

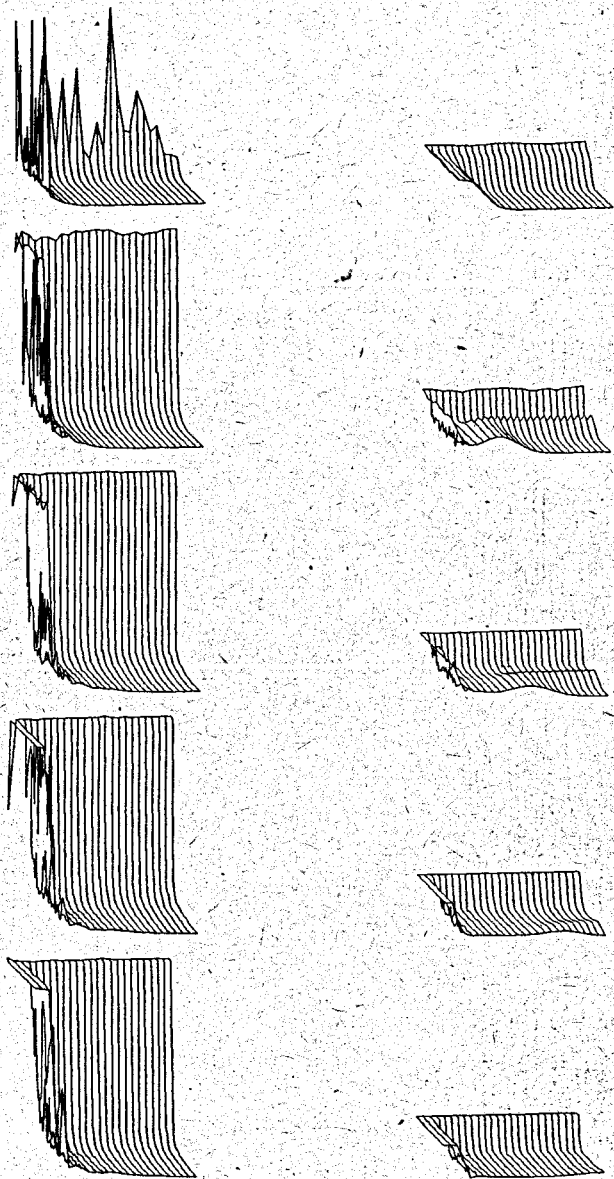


Fig.6. The evolutions of concentrations  $\langle \theta(\vec{\xi}, \tau) \rangle$  (left) and  $\langle \nu(\vec{\xi}, \tau) \rangle$  (right) in 2D adsorbing system at the moments  $\tau = 0.2, 0.4, 0.6, 0.8, 1.0$  ( $t \leq \tau_0$ ) for  $\eta_0 = 2.0$  ( $\eta_0 > \eta_{cr}$ ).

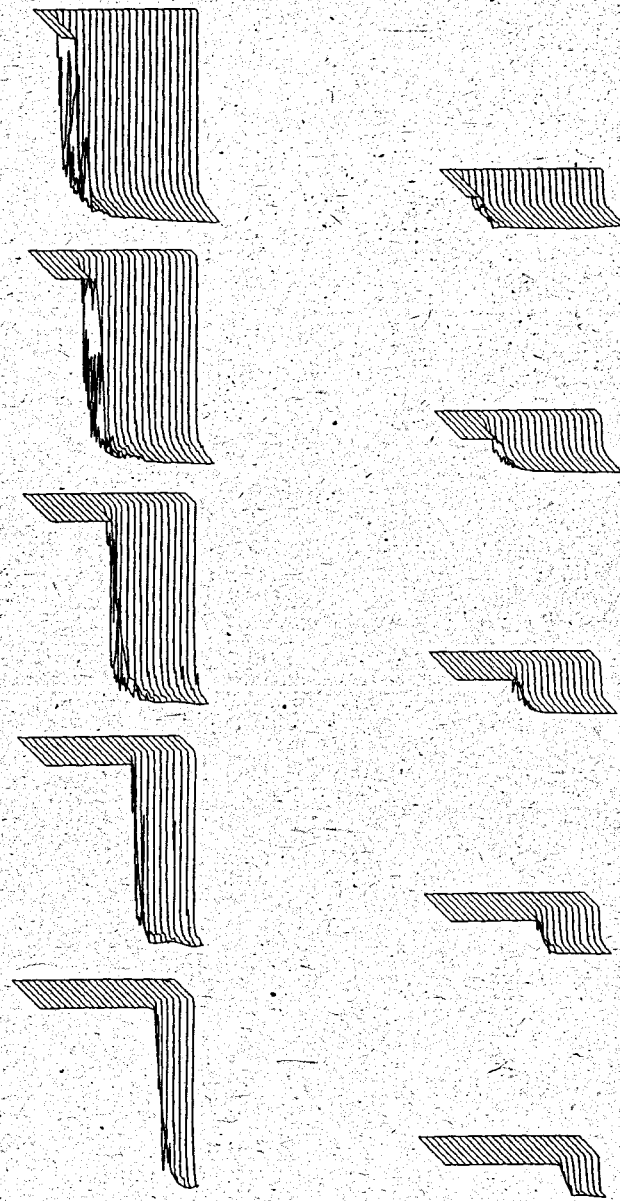


Fig.7. The evolutions of concentrations  $\langle \theta(\vec{\xi}, \tau) \rangle$  (left) and  $\langle \nu(\vec{\xi}, \tau) \rangle$  (right) in 2D adsorbing system at the moments  $\tau = 1.0, 2.0, 3.0, 4.0, 5.0$  ( $t > \tau_0$ ) for  $\eta_0 = 2.0$  ( $\eta_0 > \eta_{cr}$ ).



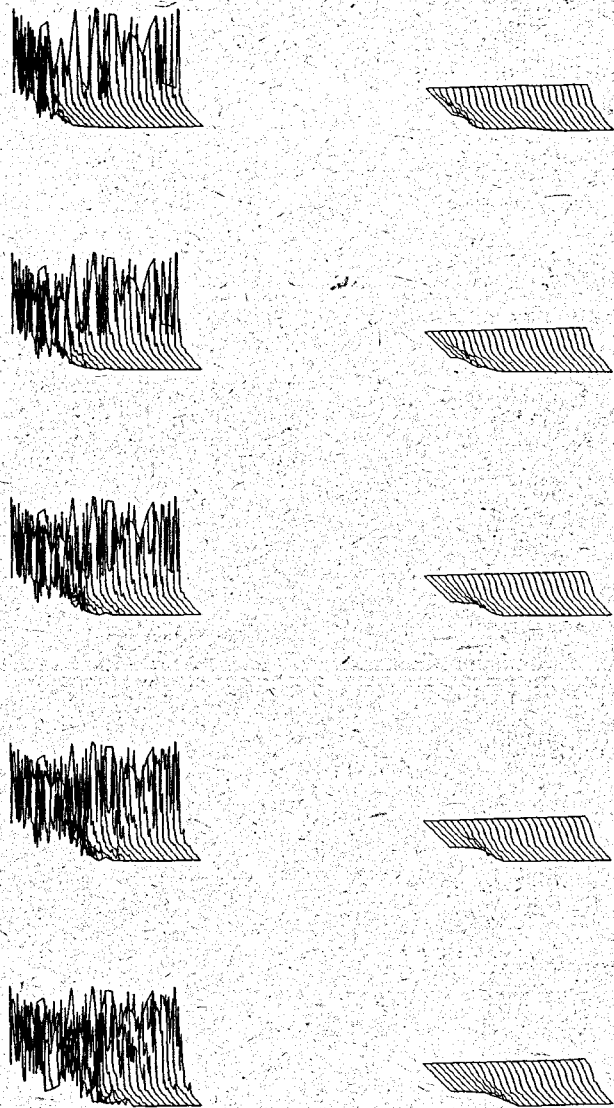


Fig.8. The evolutions of concentrations  $\langle \theta(\xi, \tau) \rangle$  (left) and  $\langle \nu(\xi, \tau) \rangle$  (right) in 2D adsorbing system at the moments  $\tau = 1.0, 2.0, 3.0, 4.0, 5.0$  ( $t > \tau_0$ ) for  $\eta_0 = 1.0$  ( $\eta_0 < \eta_{cr}$ ).

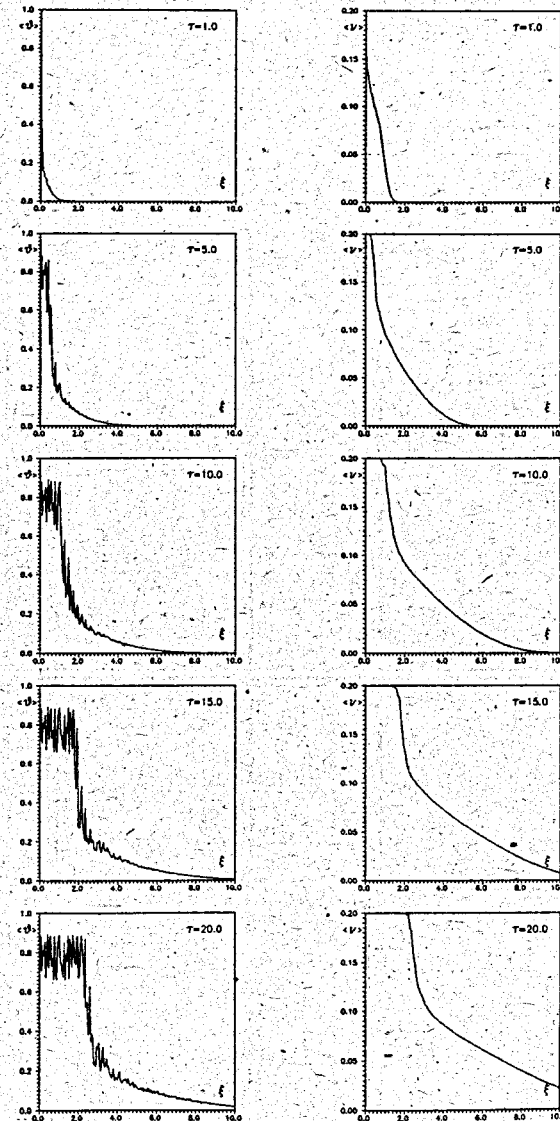


Fig.9. The evolutions of concentrations  $\langle \theta(\xi, \tau) \rangle$  (left) and  $\langle \nu(\xi, \tau) \rangle$  (right) in 1D adsorbing system at the moments  $\tau = 1.0, 5.0, 10.0, 15.0, 20.0$  ( $t > \tau_0$ ) for  $\eta_0 = 1.0$  ( $\eta_0 < \eta_{cr}$ ).

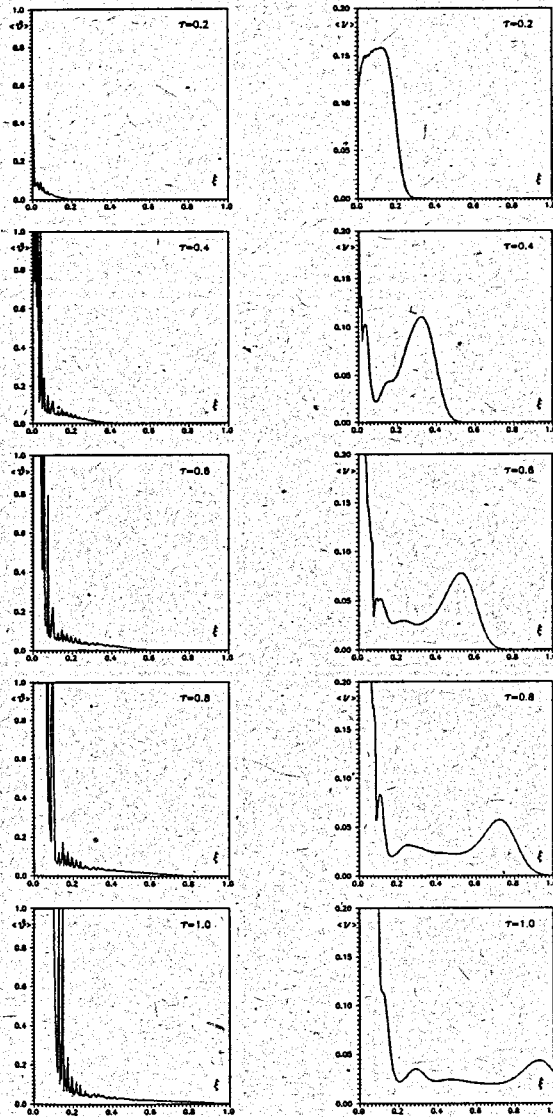


Fig.10. The evolutions of concentrations  $\langle \theta(\xi, \tau) \rangle$  (left) and  $\langle \nu(\xi, \tau) \rangle$  (right) in 1D adsorbing system at the moments  $\tau = 0.2, 0.4, 0.6, 0.8, 1.0$  ( $t \leq \tau_0$ ) for  $\eta_0 = 2.0$  ( $\eta_0 > \eta_{cr}$ ).

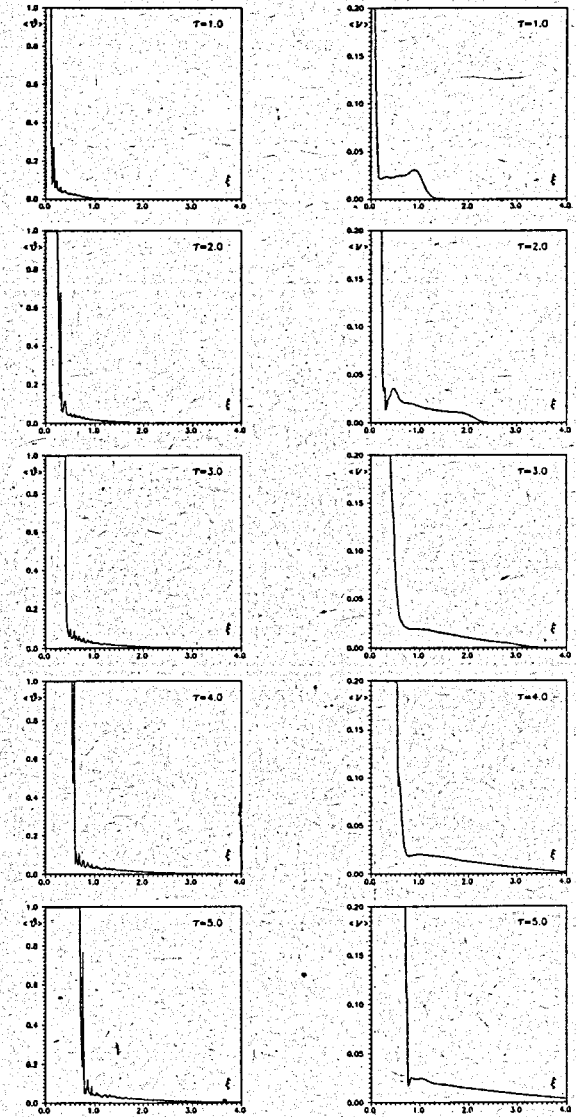


Fig.11. The evolutions of concentrations  $\langle \theta(\xi, \tau) \rangle$  (left) and  $\langle \nu(\xi, \tau) \rangle$  (right) in 1D adsorbing system at the moments  $\tau = 1.0, 2.0, 3.0, 4.0, 5.0$  ( $t > \tau_0$ ) for  $\eta_0 = 2.0$  ( $\eta_0 > \eta_{cr}$ ).

## References

1. McBain J.W. The sorption of gases and vapours by solids. London, George Routledge and Sons, Ltd., 1932.
2. Glasstone S., Laidler J.K., Eyring H. The theory of rate processes. New York and London, 1941.
3. Brunauer S. The adsorption of gases and vapors. I. Physical Adsorption. Princeton, USA, 1945.
4. Trapnell B.M.W. Chemisorption. London, Butterworths Scientific Publications, 1955.
5. Breck D.W. Zeolite molecular sieves. A Wiley Interscience Publication, John Wiley and Sons, New York, London, Sydney, Toronto, 1974.
6. Fomkin A.A., Serpinskiy V.V. Izvestiya AN SSSR, Ser. Chem., 1975, No.6, p.1244.
7. Ustinov E.A. Russian Journal of Physical Chemistry, 1995, v.69, No.4, p.673-676.
8. Fedyanin V.K., Puzynin I.V., Kholmurodov Kh.T. JINR Rapid Comm. 1994. N 1-64. P.26.
9. Puzynin I.V, Fedyanin V.K., Kholmurodov Kh.T. Preprint JINR, P17-94-135, Dubna, 1995; Journal of physical chemistry. 1995. V.69. N 3. P.492.
10. Puzynin I.V, Fedyanin V.K., Kholmurodov Kh.T. Journal of physical chemistry. 1995. V.69. N 8. P.1480.
11. Puzynin I.V, Fedyanin V.K., Kholmurodov Kh.T. Preprint JINR, P17-95-405, Dubna, 1995; Journal of physical chemistry (to be published).
12. Puzynin I.V, Fedyanin V.K., Kholmurodov Kh.T. Preprint JINR, P17-95-406, Dubna, 1995; Journal of physical chemistry (to be published).
13. Kholmurodov Kh.T., Fedyanin V.K., Puzynin I.V. Proceedings of the International Conference PLDS-2, Dubna, 1995; Journal "Physics of Low-Dimensional Structures" (to be published).
14. Kholmurodov Kh.T. Preprint JINR, P17-95-437, Dubna, 1995.
15. Adamson A.W. Physical chemistry of surfaces. A Wiley Interscience Publication, John Wiley and Sons, New York, London, Sydney, Toronto, 1979.
16. Coltharp M.T., Hackerman N. J. Phys. Chem., 72, 1171, 1968.

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