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SOLUTION OF A MODEL FOR TWO–DIMENSIONAL INTRADIFFUSION

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A system of partial differential equations corresponding to two-dimensional intradiffusion (Wicke) model for a weakly nonlinear isotherm of sorption is solved by reducing it to the Burgers' equation. Some limiting cases interesing from the physical point of view are considered in details.
Key words: diffusion, dynamics of sorption, packed-bed.

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I. INTRODUCTION

The microscopic physical description of the sorption/desorption phenomenon is a rather complicated problem. It is especially the case, when this phenomenon is connected with diffusion into a sorption medium. In this case one considers a flow of inactive liquid or gas (a mobile phase) through a bed homogeneously packed by small solid spheres covered by sorbing film (a sorption medium) with a packing characteristic porosity $\varepsilon = \frac{\text{void volume}}{\text{total volume}}$. The aim of this note is to study dynamics of sorption of an active marker introduced into a mobile phase. The process of permeability of this marker through a sorption medium is based essentially on a physical picture of multiply repeated sorption and desorption acts.

The mathematical description of dynamics of sorption has to take into account the following basic peculiarities of this complicated phenomenon: balance of an active marker in process of displacement into a sorption medium, kinetic and static properties of this marker, hydrodynamics of this process, dependence between thermodynamic condition parameters of a sorption medium, balance of heat and heat transfer in the process of dynamics of sorption in a mobile phase, etc. Because of complexity of this physical picture the theory develops via formulation of different simplifications and approximations. Usually they are the following (see, e. g., [1–4] and references therein):

- dynamics of sorption is isothermal;
- a mobile phase is incompressible and a concentration of an active marker is so small that variation of a density of sorbing marker can be neglected;
- a flow of a mobile phase is either one dimensional, or one considers an axial-symmetric problem;
- one can neglect the non-homogeneity of a sorption medium.

The main problem of the theory of dynamics of sorption now can be formulated as follows: to find the time (or space) distribution of concentration of sorbing marker for each length x (or for each moment of time t), if the initial concentration of the marker into mobile phase and character of interaction between marker and a sorption medium are known.

Usually a concentration of the sorbing marker into the mobile phase and a concentration of the sorbed marker into the sorption medium are averaged over the cross-section, thus they are functions of one variable x ([2–4]). But more precise consideration should take into account a variation of concentration of sorbed marker into a sorption medium along the radius of packing particles. Wicke [5] proposed a two-dimension intradiffusion model (the mathematical formulation is due to Tunitskij *et al.* [6]) where concentration of the sorbed marker results from diffusion of the sorbing marker into a layer of a sorption medium (see Fig. 1).

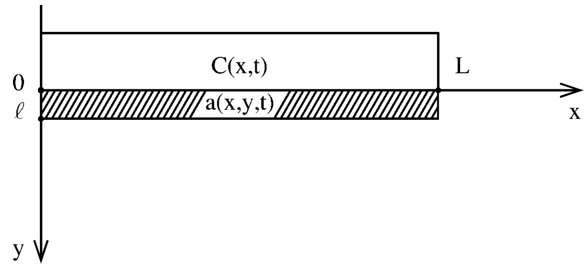

Fig. 1. Diffusion of a sorbing marker $c(x, t)$ into a layer of a sorption medium with thickness l.

In the present paper it is shown that the two-dimension intradiffusion Wicke model can be reduced to a partial differential equation for the concentration of the sorbing marker into the mobile phase. Moreover, this equation is solved explicitly for a weakly nonlinear isotherm of sorption.

II. THE MATHEMATICAL SET UP

The system of the partial differential equations corresponding to Wicke model has the form [6] (see Fig. 1):

$$\frac{\partial a}{\partial t} = \mathcal{D}_f \frac{\partial^2 a}{\partial y^2} , \tag{1}$$

$$\frac{s\mathcal{D}_f}{\varepsilon}\frac{\partial a}{\partial y}\bigg|_{y=0} = \frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x}\,, \tag{2}$$

$$\frac{\partial a}{\partial y}\bigg|_{y=l} = 0\,, \tag{3}$$

$$c(x,t)\bigg|_{t=0} = f(x) = \begin{cases} c_0, & 0 \le x \le x_0\,, \\ 0, & x > x_0\,; \end{cases} \tag{4}$$

$$a(x,y,t)\bigg|_{t=0} = \begin{cases} F(c(x,t))\Big|_{t=0} = \psi(x), & y = 0\,; \\ 0, & y > 0\,. \end{cases} \tag{5}$$

Here $a = a(x,y,t)$ is the concentration of the sorbed marker into the sorption medium, and $a(x, y=0, t) = F(c(x,t))$ is the equation of the isotherm of sorption; $c = c(x,t)$ is the concentration of the sorbing marker into the mobile phase; y is the coordinate inside the film of the sorption medium of thickness l; $\mathcal{D}_f$ is the diffusion coefficient into the sorption medium; u is the linear flow velocity which is a constant in the cross-section and along the column length; s is the specific area of the surface boundary between a mobile phase and a sorption medium (per unit bend length and per unit bend cross-section); ε is the porosity.

Notice that the Wicke model describes in fact a *two-dimensional diffusion* (into directions y and x) corresponding to the sorbed a and the sorbing c markers related by some isotherm equation.

III. EVOLUTION EQUATION FOR THE MODEL (1)–(5)

Taking into account (5) we can represent the solution $a(x,y,t)$ in the form:

$$a(x,y,t) = \Theta(x,y,t) + F(c(x,t))\,. \tag{6}$$

Then by (1) the new function $\Theta = \Theta(x,y,t)$ satisfies the differential equation

$$\frac{\partial \Theta}{\partial t} = \mathcal{D}_f\frac{\partial^2\Theta}{\partial y^2} - \frac{\partial F}{\partial t}\,, \tag{7}$$

with homogeneous boundary conditions (cf. (3),(5)):

$$\frac{\partial \Theta}{\partial y}\bigg|_{y=l} = 0\,,$$

$$\Theta(x,y,t)\bigg|_{y=0} = 0\,. \tag{8}$$

By virtue of (5) the initial condition for $\Theta(x,y,t)$ gets the form:

$$\Theta(x,y,t)\bigg|_{t=0} = \begin{cases} 0 & y = 0\,, \\ -\psi(x) & y > 0\,. \end{cases} \tag{9}$$

The solution of the corresponding homogeneous system (7)–(9) we represent as

$$\Theta(x,y,t) = Y(y)T(x,t)\,. \tag{10}$$

Then the corresponding eigenvalues and normalized eigenfunctions for this problem are:

$$\chi_n = \left(\frac{\pi(2n+1)}{2l}\right)^2, \quad n = 0,1,2,\dots, \tag{11}$$

$$Y_n(y) = \left(\frac{2}{l}\right)^{1/2}\sin\frac{\pi(2n+1)}{2l}y\,. \tag{12}$$

Representing the solution of nonhomogeneous differential equation (7) in the form

$$\Theta(x,y,t) = \sum_{n=0}^{\infty} U_n(x,t)Y_n(y) \tag{13}$$

and expanding the nonhomogeneous term with the help of the complete set of the eigenfunctions (12), we get the equation for $U_n(x,t)$:

$$\frac{\partial U_n}{\partial t} = -\mathcal{D}_f\chi_n U_n - \left(\frac{2}{l}\right)^{1/2}\frac{2l}{\pi(2n+1)}\frac{\partial F}{\partial t}\,, \tag{14}$$

with the initial condition:

$$U_n(x,0) = -\left(\frac{2}{l}\right)^{1/2}\frac{2l}{\pi(2n+1)}\psi(x)\,, \qquad n = 0,1,\dots. \tag{15}$$

Solution of the equation (14) with (15) has the form:

$$U_n(x,t) = -\left(\frac{2}{l}\right)^{1/2}\frac{2l}{\pi(2n+1)}\psi(x)\exp(-\mathcal{D}_f\chi_n t) - \int_0^t \left(\frac{2}{l}\right)^{1/2}\frac{2l}{\pi(2n+1)}\frac{\partial F(c(x,\tau))}{\partial \tau}\exp(-\mathcal{D}_f\chi_n(t-\tau))d\tau\,. \tag{16}$$

Substituting the expression (16) into (13) and taking into account (3) and (6) we get:

$$\left.\frac{\partial a}{\partial y}\right|_{y=0} = \left(-\frac{2}{l}\right)\psi(x)\sum_{n=0}^{\infty}\exp(-\mathcal{D}_f\chi_n t) - \left(\frac{2}{l}\right)\sum_{n=0}^{\infty}\int_0^t \frac{\partial F(c(\tau,x))}{\partial\tau}\exp(-\mathcal{D}_f\chi_n(t-\tau))d\tau\ , \tag{17}$$

where (due to (4),(5)):

$$\psi(x) = \begin{cases} F(f(x)) = F(c_0), & 0 \le x \le x_0\ , \\ 0, & x > x_0\ . \end{cases}$$

Let us consider the limit $x \gg x_0$. Then by the above conditions the first term in the right-hand side is negligible. Let $\mathcal{D}_f t \gg l^2$ (where l is the film thickness of the sorption medium) and let $x \gg x_0$. Then for the next term in (17) we get:

$$-\int_0^t \frac{\partial F(c(\tau,x))}{\partial\tau}\Delta(t-\tau)d\tau$$
$$= -\int_0^t \frac{\partial F(c(t-\xi,x))}{\partial t}\Delta(\xi)d\xi\ , \tag{18}$$

where $\xi = t-\tau,\ \Delta(\xi) = \dfrac{2}{l}\sum_{n=0}^{\infty}\exp(-\mathcal{D}_f\chi_n\xi)$. Using the fast decay of the function of $\Delta(\xi)$ for $\xi > 0$, the expression (18) can be represented in the form of expansion:

$$-\int_0^t \frac{dF}{dc}\frac{\partial c(x,t)}{\partial t}\Delta(\xi)d\xi$$
$$+\int_0^t \frac{dF}{dc}\frac{\partial^2 c(x,t)}{\partial t^2}\xi\Delta(\xi)d\xi + \mathcal{O}\left(\frac{l^5}{\mathcal{D}_f^3}\right)\ . \tag{19}$$

Consequently the first and the second terms in (19) for $\mathcal{D}_f t \gg l^2$ are given by:

$$-\frac{dF}{dc}\frac{\partial c(x,t)}{\partial t}\int_0^t \frac{2}{l}\sum_{n=0}^{\infty}\exp(-\mathcal{D}_f\chi_n\xi)d\xi$$
$$\simeq -\frac{dF}{dc}\frac{\partial c(x,t)}{\partial t}\frac{2}{l}\sum_{n=0}^{\infty}(\mathcal{D}_f\chi_n)^{-1} = -\frac{l}{\mathcal{D}_f}\frac{dF}{dc}\frac{\partial c(x,t)}{\partial t}\ ; \tag{20}$$

and

$$\frac{dF}{dc}\frac{\partial^2 c(x,t)}{\partial t^2}\int_0^t \xi\frac{2}{l}\sum_{n=0}^{\infty}\exp(-\mathcal{D}_f\chi_n\xi)d\xi \simeq$$
$$\frac{dF}{dc}\frac{\partial^2 c(x,t)}{\partial t^2}\frac{2}{l}\sum_{n=0}^{\infty}(\mathcal{D}_f\chi_n)^{-2} = \frac{l^3}{3\mathcal{D}_f^2}\frac{dF}{dc}\frac{\partial^2 c(x,t)}{\partial t^2}\ . \tag{21}$$

Then the derivation (17) takes the form:

$$\left.\frac{\partial a}{\partial y}\right|_{y=0} = -\frac{l}{\mathcal{D}_f}\frac{dF}{dc}\frac{\partial c}{\partial t} + \frac{l^3}{3\mathcal{D}_f^2}\frac{dF}{dc}\frac{\partial^2 c}{\partial t^2}\ . \tag{22}$$

Hence, for $\mathcal{D}_f t \gg l^2$ and $x \gg x_0$ the boundary condition (2) for the model (1)–(5) reduces to the equation:

$$\frac{u\varepsilon}{s\mathcal{D}_f}\frac{\partial c}{\partial x} + \left(\frac{\varepsilon}{s\mathcal{D}_f} + \frac{l}{\mathcal{D}_f}\frac{dF}{dc}\right)\frac{\partial c}{\partial t} = \frac{l^3}{3\mathcal{D}_f^2}\frac{dF}{dc}\frac{\partial^2 c}{\partial t^2}\ , \tag{23}$$

which is nothing but the evolution equation for the concentration of sorbing marker.

IV. DISCUSSION: LIMITING CASES

We showed that the system of partial differential equations (1)–(5) for the sorbing marker $c(x,t)$ and sorbed marker $a(x,y,t)$ can be reduced (in the long-time limit $\mathcal{D}_f t \gg l^2$ and $x \gg x_0$) to unique partial differential equation corresponding the evolution of sorbing marker (23). It is the concentration which experimenter measures at the end $x = L \gg x_0$ of packed bed. This equation can be solved explicitly for the case of a weakly nonlinear isotherm of sorption known in the gas-liquid dynamics of sorption [1]. To this end we use approximation for the isotherm of sorption (cf. Eq. (5)):

$$a(x,y=0,t) = F(c(x,t)) = k_1 c(x,t) + k_2 c^2(x,t)\ , \tag{24}$$

up to quadratic term, where $\mid k_2 \mid c(x,t) \ll k_1$.
Then the equation (23) gets the form:

$$v\frac{\partial c}{\partial x} + (1+\lambda c)\frac{\partial c}{\partial t} = \tau\frac{\partial^2 c}{\partial t^2}\ , \tag{25}$$

where

$$v = \frac{u\varepsilon}{\varepsilon+\mu k_1},\ \lambda = \frac{2k_2\mu}{\varepsilon+\mu k_1},\ \tau = \frac{\mu k_1 l^2}{3\mathcal{D}_f(\varepsilon+\mu k_1)},\ \mu = sl.$$

Using the change of variables $\xi = t - x/v$, $\eta = x/v$ and $c^* = \lambda c(x,t)$ we get the Burgers' equation for $c^* = \lambda c(x,t)$:

$$\frac{\partial c^*}{\partial\eta} + c^*\frac{\partial c^*}{\partial\xi} = \tau\frac{\partial^2 c^*}{\partial\xi^2}\ . \tag{26}$$

Its solution is well-investigated (see, e.g. [7]).

Therefore, we reduced the two-dimensional intradiffusion Wicke model (1–5) to the Burgers' equation (26).

Below we consider the Cauchy problem for the equation (25) with initial condition with respect to variable x:

$$c(x=0,t) = \begin{cases} f(t) \geq 0, & \text{if } t \geq 0, \\ f(t) = 0, & \text{if } t < 0 , \end{cases} \qquad \int_{-\infty}^{+\infty} f(t)dt = M < +\infty \, . \tag{27}$$

Then the solution of the equation (25) has the form:

$$c(x,t) = \frac{v}{\lambda x} \cdot \frac{\int_{-\infty}^{+\infty} d\omega (t - \frac{x}{v} - \omega) \exp\left[-\frac{(t-\frac{x}{v}-\omega)^2}{4\tau x/v}\right] \cdot \exp\left[-\frac{1}{2\tau}\int_0^\omega dy \lambda f(y)\right]}{\int_{-\infty}^{+\infty} d\omega \exp\left[-\frac{(t-\frac{x}{v}-\omega)^2}{4\tau x/v}\right] \exp\left[-\frac{1}{2\tau}\int_0^\omega dy \lambda f(y)\right]} \, . \tag{28}$$

For the simplest case

$$f(t) = c(x=0,t) = \begin{cases} c_0, & 0 \leq t \leq t_0 \, , \\ 0, & t < 0; t > t_0 \, , \end{cases}$$

one gets from (28):

$$c(x,t) = \frac{c_0}{\lambda} \cdot \frac{[\phi(a+d) - \phi(b+d)]\exp c}{[\phi(a+d) - \phi(b+d)]\exp c + [2 + \phi(b)]\exp h - \phi(a)} \, , \tag{29}$$

where $\phi(z) = \dfrac{2}{\sqrt{\pi}} \displaystyle\int_0^z e^{-\xi^2} d\xi$ and

$$a = \frac{vt - x}{\sqrt{4\tau x}}, \qquad c = \frac{-\lambda c_0 (2x(vt - x) - \lambda c_0 x^2 + v t_0 x)}{4\tau x} \, , \tag{30}$$

$$b = \frac{vt - x - vt_0}{\sqrt{4\tau x}}, \qquad d = -\frac{\lambda c_0 x}{\sqrt{4\tau x}}, \qquad h = -\frac{\lambda c_0 v t_0}{2\tau} \, . \tag{31}$$

If we consider the case $t_0 \to 0$ ($c_0 t_0 = M = \text{const}$) we get

$$c(x,t) = \frac{1}{\lambda}\sqrt{\frac{\tau}{x/v}} \frac{(e^R - 1)\exp\left(-\frac{(t-\frac{x}{v})^2}{4\tau x/v}\right)}{\sqrt{\pi} + \frac{\sqrt{\pi}}{2}(e^R - 1)\left\{1 - \phi\left(\frac{t-\frac{x}{v}}{\sqrt{\frac{4\tau x}{v}}}\right)\right\}} \, , \tag{32}$$

where $R = \dfrac{\lambda c_0 t_0}{2\tau}$ is the parameter of the non-linearity. It is positive for concave isotherm of sorption (the case of $k_2 > 0$ in (24)) and negative for convex isotherm of sorption (the case of $k_2 < 0$).

To investigate the form of the time distribution of the sorbing marker (32) for $t \gg l^2/\mathcal{D}_f$ and for a fixed length $x = L \gg x_0$ one should to consider the following two limiting cases.

Case of small parameter of non-linearity $|R| \ll 1$

In this case we would expect the diffusion in time to dominate over the non-linearity. For $|R| \ll 1$ the denominator in (32) is $\sqrt{\pi} + \mathcal{O}(|R|)$, uniformly in x,t,τ. Hence $c(x,t)$ may be approximated by

$$c(x,t) = \frac{1}{\lambda}\sqrt{\frac{\tau}{\pi x/v}} R \exp\left(-\frac{(t - x/v)^2}{4\tau x/v}\right)$$
$$= \frac{c_0 t_0}{\sqrt{4\pi\tau x/v}} \exp\left(-\frac{(t - x/v)^2}{4\tau x/v}\right) \, . \tag{33}$$

This is the source solution of the "heat" equation, close to the Gaussian shape. So the expectation is verified.

Case of large parameter of non-linearity $|R| \gg 1$

To discuss the behaviour of the solution (32) for large $|R|$ (i. e., when the non-linearity dominates over the diffusion) it is convenient to introduce the similarity variable $T = \frac{t-x/v}{\sqrt{2|\lambda|c_0t_0x/v}}$. Then we analyze the behaviour of $c(T,x)$, when $|R| \gg 1$ for different ranges of T.

1). Let us consider the case when the isotherm of sorption is concave, $R > 0$.

Writing (32) as

$$c(T,x) = \sqrt{\frac{2c_0t_0v}{\lambda x}} \cdot g(T,R), \qquad g(T,R) = \frac{e^R-1}{2\sqrt{R}} \cdot \frac{e^{-T^2R}}{\sqrt{\pi} + (e^R-1)\int\limits_{T\sqrt{R}}^{\infty} e^{-\xi^2}d\xi}\,, \tag{34}$$

we discuss the behaviour of g as $R \to \infty$ (i. e. $\tau \to 0$) for different ranges of T. In all cases, $e^R - 1 \sim e^R$ and we may use

$$g(T,R) = \frac{1}{2\sqrt{R}} \cdot \frac{e^{R\left(1-T^2\right)}}{\sqrt{\pi} + e^R\int\limits_{T\sqrt{R}}^{\infty} e^{-\xi^2}d\xi}\,, \tag{35}$$

If $T < 0$, the integral in (35) tends to $\sqrt{\pi}$; therefore, $c \to 0$ at least like $1/\sqrt{R}$. If $T > 0$, the integral in (35) becomes small and we use for the asymptotic expansion

$$\int_\eta^\infty e^{-\xi^2}d\xi \sim \frac{e^{-\eta^2}}{2\eta} \qquad \text{as} \qquad \eta \to \infty\,. \tag{36}$$

Consequently the solution of the equation (25) for $R \gg 1$ is

$$c(T,x)|_{R\gg1} = \begin{cases} O(R^{-1/2}) & , \; T < 0\,, \\ \sqrt{\frac{2c_0t_0v}{\lambda x}} \cdot \frac{T}{1+2T\sqrt{\pi R}\exp(R(T^2-1))} & , \; T \geq 0\,. \end{cases} \tag{37}$$

If $0 < T < 1$, we have from (35)

$$g \sim T\,, \quad 0 < T < 1\,, \quad R \to \infty\,, \tag{38}$$

whereas if $T > 1$, $g \to 0$ as $R \to \infty$. Thus $g \to 0$ except in $0 < T < 1$, and in that range $g \sim T$. In the original variables the result reads:

$$c(x,t)|_{R\to\infty} = \begin{cases} \frac{v}{\lambda x}(t - x/v) & , \; 0 < \frac{t-x/v}{\sqrt{(2\lambda c_0t_0x)/v}} < 1\,, \\ 0 & , \; \text{for the other } \; x, t\,. \end{cases} \tag{39}$$

This is the appropriate solution of (25) with a *shock* (i. e., with a jump) at the point $t = x/v + \sqrt{(2\lambda c_0t_0x)/v}$. The concentration c jumps from zero to $\sqrt{(2c_0t_0v)/(\lambda x)}$. The shock is located at $T = 1$ and for large but finite R (37) shows a rapid transition from exponentially small values in $T > 1$ to $(1/\lambda)\sqrt{(4R\tau v)/x}$ in $T < 1$. The transition layer is of thickness $\mathcal{O}(\mathcal{R}^{-1})$ around $T = 1$ (see Fig. 2)

2). In the case when the isotherm of sorption is a convex the solution of (25) is (see Fig. 3):

$$c(T,x)|_{|R|\gg1} = \begin{cases} O(|R|^{-1/2}) & , \; T > 0\,, \\ \sqrt{\frac{2c_0t_0v}{|\lambda|x}} \cdot \frac{|T|}{1+2|T|\sqrt{\pi|R|}\exp(|R|(T^2-1))} & , \; T \leq 0\,. \end{cases} \tag{40}$$

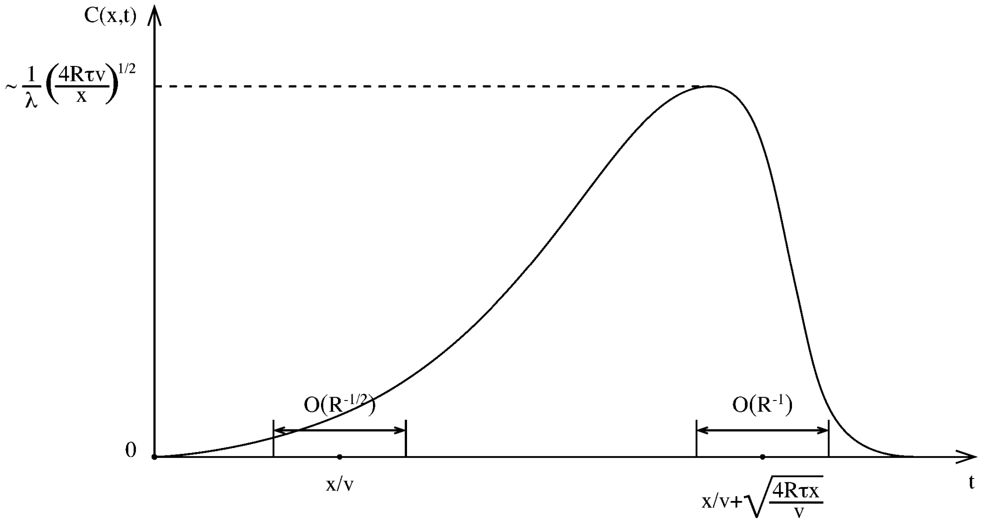

Fig. 2. Shock-wave type solution of Burgers' equation for concave isotherm of sorption.

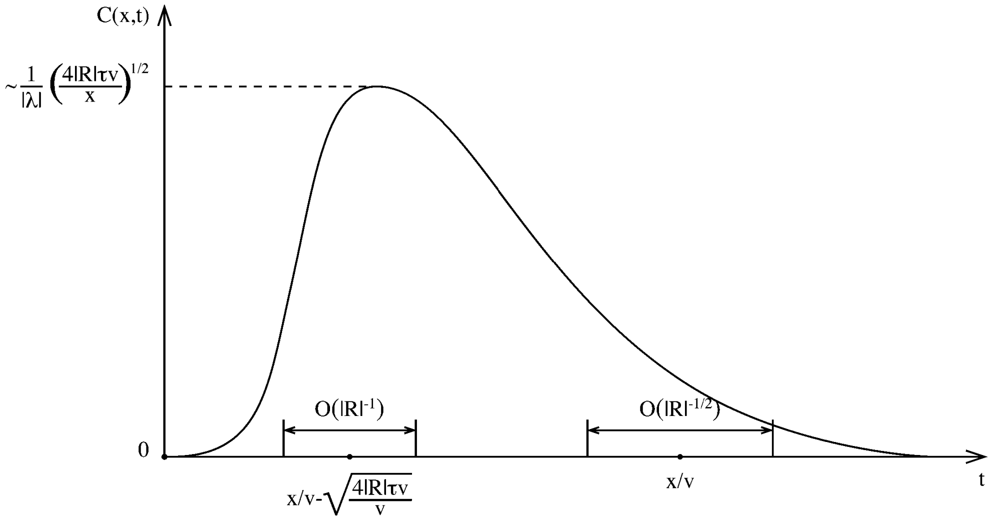

Fig. 3. Shock-wave type solution of Burgers' equation for convex isotherm of sorption.

If $|R| \to \infty$ (x is fixed) we get again a solution of the shock-wave type:

$$c(x,t)|_{|R|\to\infty} = \begin{cases} \dfrac{v}{|\lambda|x}|t - x/v| & , \ -1 < \frac{t-x/v}{\sqrt{(2|\lambda|c_0t_0x)/v}} < 0 \\ 0 & , \text{ for the other } \ x, t \ . \end{cases} \tag{41}$$

V. CONCLUSION

- A system of partial differential equations corresponding to two-dimensional intradiffusion (Wicke) model for a weakly nonlinear isotherm of sorption is solved by reducing it to the Burgers' equation. Therefore, in the present paper we solve a model for *two-dimensional diffusion*. Some limiting cases interested from the physical point of view are considered in details.

- The expression for the concentration (37)–(39) of the sorbing marker for each fixed x has a slanting left slope and a steep right slope in the vicinity of the point $t = x/v + \sqrt{(4R\tau x)/v}$ in the case of concave sorption isotherm (see Fig. 2).

- In this case of (40)–(41) the concentration of the sorbing marker for a fixed $x \gg x_0$ has a slanting right slope and the a steep left slope in the vicinity of the point $t = x/v - \sqrt{(4|R|\tau x)/v}$ in the case of convex sorption isoterm (see Fig. 3).
- The solutions of (37)–(39) and (40)–(41) have a clear physical interpretation.
  - From equation (25) we conclude that the sorbing concentration $c(x,t)$ moves with the velocity $v' \sim v/(1+\lambda c)$. The latter means that higher concentrations move slower than the small ones for $\lambda > 0$, i. e., $k_2 > 0$. Therefore, the small concentrations arrive to the packed-bed output earlier than the high concentrations (see Fig. 2).
  - For the case $\lambda < 0$ (i. e., $k_2 < 0$) higher concentrations arrive to the packed-bed output earlier than small concentrations (see Fig. 3).
- We neglected the non-homogeneity of a sorption medium. In most cases, however, the stationary phase film covering the support surface is non-uniform and fills mainly randomly narrow pores of the support. Latter means that the thickness l of sorption film (see Fig. 1) is stochastic. We return to this problem elsewhere.

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РОЗВ'ЯЗОК ДЛЯ МОДЕЛІ ДВОВИМІРНОЇ ІНТРАДИФУЗІЇ

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Систему диференціяльних рівнянь у частинних похідних, що відповідає двовимірній моделі інтрадифузії (моделі Віке) для слабо нелінійних ізотерм сорбції, розв'язано способом зведення її до рівняння Бурґерса. У деталях розглянуто деякі граничні випадки, цікаві з точки зору застосування у фізиці.