TWO CONSECUTIVE HETEROGENEOUS REACTIONS IN TWO-PHASE SYSTEM

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We consider diffusion of species into a semi-infinite domain, where two consequent reactions take place. The exact self-similar solutions are obtained both for initial absence and presence of the first reaction's product in the media. The inequalities determining the mutual positions of the fronts are also obtained.

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1. INTRODUCTION

The consequent reactions systems were studied in kinetics and chemical engineering mainly in three following contexts:

1. Well mixed (batch) systems;

2. Tubular reactors with/without dispersion;

3. Gas-liquid reactions during the saturation of liquids from their surface.

For the cases 1 and 2 reactions may be homogeneous or heterogeneous.

In the single-phase systems reactions and mass transfer are not segregated spatially; in the multiphase systems where dispersed phases are present, the reactions are confined either to volume or surface of dispersed phase particles, e.g. to the surface of catalytic particles. It is convenient to call these reaction microheterogeneous.

On the other hand, case 3 represents essentially multiphase system. At least two phases - gas and liquid - are present, separated by an interface. If the liquid does not contain a dispersed third phase, this type of reaction may be conveniently called macroheterogeneous. If the liquid is multiphase itself there is a combination of macro- and micro-heterogeneity.

Homogeneous reactions for cases 1 and 2 were studied intensively [1, 2].Consecutive reactions for the case 1were studied in [3].

Nevertheless, systematic theoretical investigation of the consecutive reactions systems was performed for first-order reactions in well mixed systems only [4]. The microheterogeneous systems are studied insufficiently. Systematic theoretical study of a single microheterogeneous reaction in the systems of type 1 and 2 was performed in [5-7]; both the local and global (for a tubular reactor) diffusional and hydrodynamical mass transfer were taken into account. The theoretical investigation of consecutive microheterogeneous reactions systems was started in [8, 9]. In these papers the qualitative difference between dynamics of global and local concentrations, as well as different regimes of changes for the local concentrations were considered. Two consecutive reactions systems were considered: one first order and one second order binary reaction; two second order binary reactions.

Macroheterogeneous reactions of type 3 were studied most systematically, maybe due to excellent book of Danckwerts [10]. In this book Danckwerts first considers absorption and homogeneous reactions in a quiescent liquid. The results thus obtained are applied to study of absorption and reactions in agitated liquids exploiting so called "film theory" and "penetration theory". That is the results for absorption into quiescent finite width layer and semi-infinite domain, respectively, are used. Regretfully, considering the consecutive reactions Danckwerts started some terminological confusion, calling "consecutive" reaction systems which in kinetics and engineering are usually called "parallel-consecutive", or "multiple substitutional" reactions [1,2]. Maybe because of this confusion the systems of strictly consecutive higher order reactions appeared beyond the range of research. As we will show, the theoretical investigation of such systems is possible for quite general situations. In the present communication we consider two consecutive macroheterogeneous reactions in a quiescent liquid; the results will be used for study combined macro-, and microheterogeneous as well as more complicated systems of consecutive reactions.

We consider diffusion in semi-infinite domain; therefore the results may be used to study absorption into agitated liquid within the approach of penetration theory.

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Let us consider absorption of two species V_1 and V_2 accompanied by two consecutive infinitely fast and irreversible reactions

$$U + V_1 \rightarrow W_1 \,, \\ W_1 + V_2 \rightarrow W_2 \,. \tag{1}$$
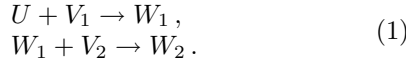
The reactions proceed in the liquid phase, which is filling the half-space $x > 0$. The reagents U and W_1 are initially dissolved in the liquid phase, $U(x,0) = U_\infty$, $W_1(x,0) = W_{1\infty}$.

We presume that absorption into liquid does not change the concentrations of reagents in gaseous phase; that is, these concentrations are constants. At the gas/liquid surface equilibrium is achieved, that is the concentrations $V_1(0,t)$ and $V_2(0,t)$ of these reagents in liquid at the surface are constants as well

Let us consider the solution method for the first reaction[11-13]. Because reaction is infinitely fast and irreversible, the non-zero concentrations of reagents can not coexist. That is, the spatial domain with non-zero concentration of reagent V_1 and spatial domain with non-zero concentration of U are separated by a moving reaction front, see Fig.1, where both concentrations are equal to zero. Therefore the problem reduces to solution of two diffusion equations in two adjacent spatial domains and to matching solutions at the reaction front.

$$\frac{\partial V_1}{\partial t} = D_{V_1} \frac{\partial^2 V_1}{\partial x^2}, \quad x < X_1(t), \\ \frac{\partial U}{\partial t} = D_U \frac{\partial^2 U}{\partial x^2}, \quad x > X_1(t). \tag{2}$$

$$Q_1 \frac{dX_1(t)}{dt} = D_U \frac{\partial U}{\partial x}\bigg|_{x=X_1} = -D_{V_1} \frac{\partial V_1}{\partial x}\bigg|_{x=X_1}. \tag{3}$$

Where the latter equality is the stoichiometry condition, and

$$V_1|_{x=0} = V_{10}, \quad U|_{x\rightarrow\infty} = U_\infty, \\ V_1|_{x=X_1(t)} = U|_{x=X_1(t)} = 0. \tag{4}$$

Here D_{V_1}, D_U are diffusion coefficients of V_1 and U, respectively. And $X_1(t)$ - coordinate of reaction front. Introducing non-dimensional variables,

$$\lambda = \frac{x}{2\sqrt{D_U t}}, \tag{5}$$

$$\theta_1^2 = \frac{D_U}{D_{V_1}}, \quad \theta_2^2 = \frac{D_U}{D_{V_2}}, \quad \theta_3^2 = \frac{D_U}{D_W}, \tag{6}$$

and looking for self-similar solutions we get

$$\frac{d^2 U}{d\lambda^2} + 2\lambda \frac{dU}{d\lambda} = 0, \quad \lambda_1 < \lambda, \tag{7}$$

$$\frac{d^2 V_1}{d\lambda^2} + 2\lambda \theta_1^2 \frac{dV_1}{d\lambda} = 0, \quad \lambda_1 > \lambda, \tag{8}$$

$$\theta_1 \frac{\partial U}{\partial \lambda}\bigg|_{\lambda=\lambda_1} = -\frac{\partial V_1}{\partial \lambda}\bigg|_{\lambda=\lambda_1}, \tag{9}$$

$$U(\lambda_1) = 0, \quad U|_{\lambda\rightarrow\infty} = U_\infty, \tag{10}$$

$$V_1(0) = V_{10}, \quad V_1(\lambda_1) = 0. \tag{11}$$

Solutions of the above system are [11, 12, 13]:

$$V_1 = \begin{cases} V_{10}\left(1 - \frac{erf(\lambda\theta_1)}{erf(\lambda_1\theta_1)}\right), & \lambda < \lambda_1 \\ 0, & \lambda > \lambda_1 \end{cases} \tag{12}$$

$$U = \begin{cases} 0, & \lambda < \lambda_1 \\ U_\infty \frac{erf(\lambda)-erf(\lambda_1)}{1-erf(\lambda_1)}, & \lambda > \lambda_1 \end{cases} \tag{13}$$

where λ_1 is found from equation

$$\theta_1 U_\infty \frac{erf(\theta_1\lambda_1)}{erfc(\lambda_1)} = -V_{10}\exp\left(\lambda_1^2(1-\theta_1^2)\right). \tag{14}$$
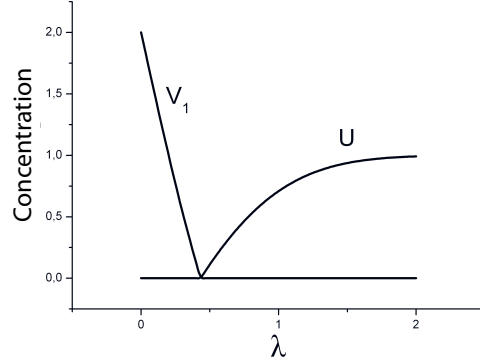

Fig.1. *First reaction's front. $\theta_1 = 1.3$, $V_{10} = 2$ and $U_\infty = 1$*

Due to reaction the inflow of reagent V_1 into the liquid phase is increased as compared to the case of pure diffusion. The enhancement factor which is defined as a ratio of the flow for the reactive case to the flow for purely diffusive case, taken at the gas/liquid interface [10], becomes for the present case

$$E_{V_1} = 1/erf(\theta_1\lambda_1). \tag{15}$$

Because the first reaction is irreversible, the second reaction does not influence the position of the front and correspondingly the position of the source of W_1.

2. CONSECUTIVE REACTIONS

The self-similar solutions for the concentration of reagents, participating in the second reaction, will now depend on mutual positions of first and second fronts of reactions.

Let us consider the case when the first reaction's product is initially absent in the liquid $W_{1\infty} = 0$. We presume $\lambda_2 < \lambda_1$, which corresponds to inequality

$$V_{20} < \theta_2 U_\infty \exp\left(-\lambda_1^2\left(1-\theta_2^2\right)\right)\frac{erf(\theta_2\lambda_1)}{erfc(\lambda_1)}. \tag{16}$$

For this case equations look:

$$\frac{d^2 W_1}{d\lambda^2} + 2\lambda\theta_3^2\frac{dW_1}{d\lambda} = 0, \quad \lambda_2 < \lambda < \lambda_1, \tag{17}$$

$$\frac{d^2 W_1}{d\lambda^2} + 2\lambda\theta_3^2\frac{dW_1}{d\lambda} = 0, \quad \lambda_1 < \lambda, \tag{18}$$

$$\frac{d^2 V_2}{d\lambda^2} + 2\lambda\theta_2^2\frac{dV_2}{d\lambda} = 0, \quad 0 < \lambda < \lambda_2, \tag{19}$$

$$\theta_2 \frac{\partial W_1}{\partial \lambda}\bigg|_{\lambda=\lambda_2} = -\theta_3 \frac{\partial V_2}{\partial \lambda}\bigg|_{\lambda=\lambda_2}, \qquad (20)$$

$$\frac{\partial W_1}{\partial \lambda}\bigg|_{\lambda=\lambda_1-0} - \frac{\partial W_1}{\partial \lambda}\bigg|_{\lambda=\lambda_1+0} = \theta_3 \frac{\partial U}{\partial \lambda}\bigg|_{\lambda=\lambda_1}, \quad (21)$$

$$W_1(\lambda_2) = 0, \quad W_1|_{\lambda \to \infty} = 0, \qquad (22)$$

$$V_2(0) = V_{20}, \quad V_2(\lambda_2) = 0. \qquad (23)$$

And the solution is:

$$V_2 = \begin{cases} V_{20}\left(1 - \frac{erf(\theta_2\lambda)}{erf(\theta_2\lambda_2)}\right), & \lambda < \lambda_2 \\ 0, & \lambda > \lambda_2 \end{cases} \qquad (24)$$

$$W_1 = \begin{cases} 0, & \lambda < \lambda_2 \\ C_{22}\left(erf(\theta_3\lambda) - erf(\theta_3\lambda_2)\right), & \lambda_2 < \lambda < \lambda_1 \\ C_{21}erfc(\theta_3\lambda), & \lambda > \lambda_1 \end{cases}$$
$$(25)$$

Where

$$C_{21} = \frac{\theta_3 U_\infty}{e^{\left(\lambda_1^2\left(1-\theta_3^2\right)\right)}} \frac{erf(\theta_3\lambda_1) - erf(\theta_3\lambda_2)}{erfc(\theta_3\lambda_2)\,erfc(\lambda_1)}, \quad (26)$$

$$C_{22} = \frac{\theta_3 U_\infty}{\exp\left(\lambda_1^2\left(1-\theta_3^2\right)\right)} \frac{erfc(\theta_3\lambda_1)}{erfc(\theta_3\lambda_2)\,erfc(\lambda_1)}, \quad (27)$$

And λ_2 is determined by equation:

$$e^{\lambda_2^2\left(1-\theta_2^2\right)} = \frac{\theta_2 U_\infty}{V_{20}} \frac{erfc(\theta_3\lambda_1)\,erf(\theta_2\lambda_2)}{erfc(\theta_3\lambda_2)\,erfc(\lambda_1)}. \quad (28)$$

While increasing the concentration (partial pressure) of V_2 in gas phase and, correspondingly, the concentration V_{20}, the second reaction's front is approaching λ_1 and will merge with the first reaction's front for

$$V_{20}^* = \theta_2 U_\infty \exp\left(-\lambda_1^2\left(1-\theta_2^2\right)\right) \frac{erf(\theta_2\lambda_1)}{erfc(\lambda_1)}. \quad (29)$$
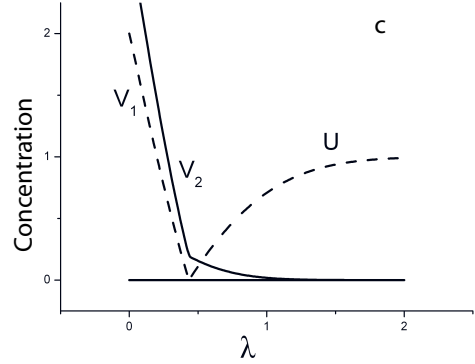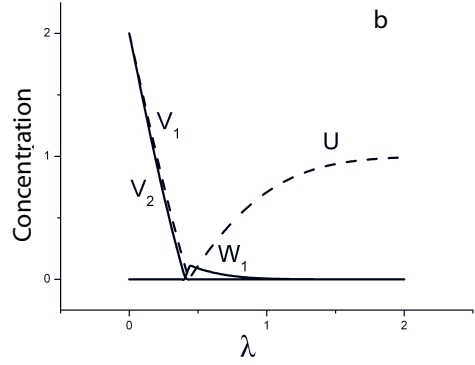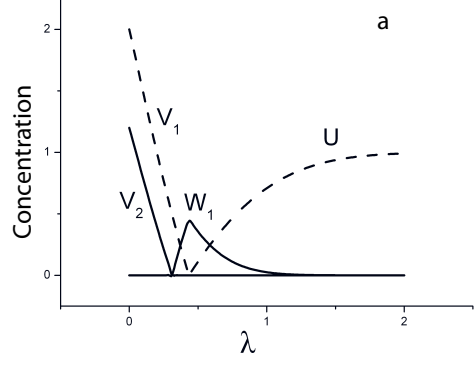

Fig.2. *Second reaction's front.* $\theta_1 = 1.3$, $\theta_2 = 1.7$, $\theta_3 = 1.5$, $V_{10} = 2$, $U_\infty = 1$, $\theta_2 = 1.7$, $\theta_3 = 1.5$, $V_{10} = 2$, $U_\infty = 1$, $V_{20} = 1.2$ (a), $V_{20} = 2$ (b), $V_{20} = 2.8$ (c).

By merging of two reactions fronts concentration W_1 becomes zero in the whole domain and remains zero when V_{20} is further increased, see Fig.2. That is even by continuing increase of V_{20} the front of the second reaction can not advance faster then the front of the first(such self-similar solution does not exist).

That is if

$$V_{20} > \theta_2 U_\infty \exp\left(-\lambda_1^2\left(1-\theta_2^2\right)\right) \frac{erf(\theta_2\lambda_1)}{erfc(\lambda_1)}, \quad (30)$$

equations take the form

$$\frac{d^2V_2}{d\lambda^2} + 2\lambda\theta_2^2 \frac{dV_2}{d\lambda} = 0, \quad 0 < \lambda < \lambda_1, \quad (31)$$

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$$\frac{d^2 V_2}{d\lambda^2} + 2\lambda\theta_2^2 \frac{dV_2}{d\lambda} = 0, \quad \lambda_1 < \lambda, \qquad (32)$$

$$\left.\frac{\partial V_2}{\partial \lambda}\right|_{\lambda=\lambda_1+0} - \left.\frac{\partial V_2}{\partial \lambda}\right|_{\lambda=\lambda_1-0} = \theta_2 \left.\frac{\partial U}{\partial \lambda}\right|_{\lambda=\lambda_1}, \qquad (33)$$

$$V_2(0) = V_{20}, \quad V_2(\lambda_1) = 0. \qquad (34)$$

And solution becomes respectively:

$$W_1 = 0. \qquad (35)$$

$$V_2 = \begin{cases} V_{20} - C_{11} erf(\theta_2 \lambda), & 0 < \lambda < \lambda_1 \\ C_{12} erfc(\theta_2 \lambda), & \lambda_1 < \lambda. \end{cases} \qquad (36)$$

Where

$$C_{11} = V_{20} + \frac{\theta_2 U_\infty}{\exp(\lambda_1^2(1-\theta_2^2))} \frac{erfc(\theta_2 \lambda_1)}{erfc(\lambda_1)}, \qquad (37)$$

$$C_{12} = V_{20} - \frac{\theta_2 U_\infty}{\exp(\lambda_1^2(1-\theta_2^2))} \frac{erf(\theta_2 \lambda_1)}{erfc(\lambda_1)}. \qquad (38)$$

For $V_{20} < V^*$ the enhancement factor is

$$E_{V_2} = 1/erf(\theta_2 \lambda_2). \qquad (39)$$

And for $V_{20} \geq V^*$ it becomes

$$E_{V_2} = \left(1 + \frac{\theta_2 U_\infty}{V_{20}} \frac{erfc(\theta_2 \lambda_1)}{erfc(\lambda_1) e^{-\lambda_1^2(\theta_3^2-1)}}\right). \qquad (40)$$

On the other hand, if $W_{1\infty}$ is non-zero it is possible to move the second reaction's front into $\lambda > \lambda_1$ domain and, correspondingly, to control its position by varying $W_{1\infty}$

If the second reaction's front is advancing faster then the front of first reaction, see Fig.3

$$\lambda_1 < \lambda_2, \qquad (41)$$

which corresponds to inequality

$$\frac{V_{20}}{\theta_2} \frac{e^{-\lambda_1^2\theta_2^2}}{erf(\theta_2\lambda_1)} > \frac{W_{1\infty}}{\theta_3} \frac{e^{-\lambda_1^2\theta_3^2}}{erfc(\theta_3\lambda_1)} + U_\infty \frac{e^{-\lambda_1^2}}{erfc(\lambda_1)}, \qquad (42)$$

then equations become

$$\frac{d^2 W_1}{d\lambda^2} + 2\lambda\theta_3^2 \frac{dW_1}{d\lambda} = 0, \quad \lambda_2 < \lambda, \qquad (43)$$

$$\frac{d^2 V_2}{d\lambda^2} + 2\lambda\theta_2^2 \frac{dV_2}{d\lambda} = 0, \quad 0 < \lambda < \lambda_1, \qquad (44)$$

$$\frac{d^2 V_2}{d\lambda^2} + 2\lambda\theta_2^2 \frac{dV_2}{d\lambda} = 0, \quad \lambda_1 < \lambda < \lambda_2, \qquad (45)$$

$$\theta_2 \left.\frac{\partial W_1}{\partial \lambda}\right|_{\lambda=\lambda_2} = -\theta_3 \left.\frac{\partial V_2}{\partial \lambda}\right|_{\lambda=\lambda_2}, \qquad (46)$$

$$\left.\frac{\partial V_2}{\partial \lambda}\right|_{\lambda=\lambda_1+0} - \left.\frac{\partial V_2}{\partial \lambda}\right|_{\lambda=\lambda_1-0} = \theta_2 \left.\frac{\partial U}{\partial \lambda}\right|_{\lambda=\lambda_1}, \qquad (47)$$

$$W_1(\lambda_2) = 0, \quad W_1|_{\lambda\to\infty} = W_{1\infty}, \qquad (48)$$

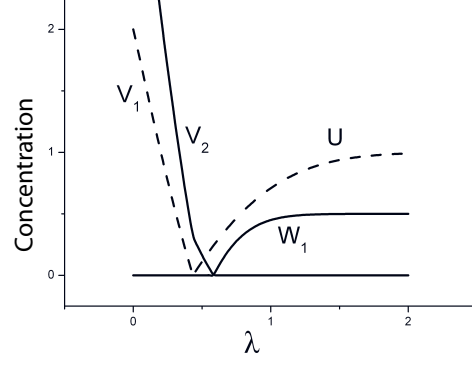$$V_2(0) = V_{20}, \quad V_2(\lambda_2) = 0. \qquad (49)$$

Fig.3. *Second reaction's front is advancing faster then the front of first reaction, $\theta_1 = 1.3$, $\theta_2 = 1.7$, $\theta_3 = 1.5$, $V_{10} = 2$, $U_\infty = 1$, $W_{1\infty} = 0.5$, $V_{20} = 4$.*

The solution is

$$W_1 = \begin{cases} 0, & \lambda < \lambda_2 \\ W_{1\infty} \frac{erf(\theta_3\lambda)-erf(\theta_3\lambda_2)}{1-erf(\theta_3\lambda_2)}, & \lambda > \lambda_2 \end{cases} \qquad (50)$$

$$V_2 = \begin{cases} V_{20} + C_{11} erf(\theta_2\lambda), & \lambda < \lambda_1 \\ C_{12}(erf(\theta_2\lambda) - erf(\theta_2\lambda_2)), & \lambda_1 < \lambda < \lambda_2 \\ 0, & \lambda > \lambda_2 \end{cases}, \qquad (51)$$

where λ_2 is determined by equation

$$\theta_2 W_{1\infty} e^{-\lambda_2^2(\theta_3^2-\theta_2^2)} = -\theta_3 C_{12} erfc(\theta_3\lambda_2). \qquad (52)$$

It is worth noticing that now second reaction proceeds not only at $\lambda = \lambda_2$, but at $\lambda = \lambda_1$, that is at the first reaction's front as well. This is manifested by discontinuous derivative of $V_2(\lambda)$ at this point.
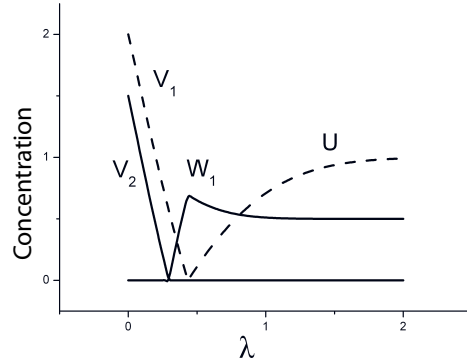

Fig.4. *First reaction's front is advancing faster then the front of second reaction, $\theta_1 = 1.3$, $\theta_2 = 1.7$, $\theta_3 = 1.5$, $V_{10} = 2$, $U_\infty = 1$, $W_{1\infty} = 0.5$, $V_{20} = 1.5$*

If the first reaction's front is advancing faster then the front of second reaction, see Fig.4,

$$\lambda_1 > \lambda_2, \qquad (53)$$

which corresponds to inequality

$$\frac{V_{20}}{\theta_2} \frac{\exp(-\lambda_1^2\theta_2)}{erf(\theta_2\lambda_1)} > \frac{W_{1\infty}}{\theta_3} \frac{\exp(-\lambda_1^2\theta_3^2)}{erfc(\theta_3\lambda_1)} + U_\infty \frac{\exp(-\lambda_1^2)}{erfc(\lambda_1)},$$

Then equations look

$$\frac{d^2 W_1}{d\lambda^2} + 2\lambda\theta_3^2 \frac{dW_1}{d\lambda} = 0, \quad \lambda_2 < \lambda < \lambda_1, \qquad (54)$$

$$\frac{d^2 W_1}{d\lambda^2} + 2\lambda\theta_3^2 \frac{dW_1}{d\lambda} = 0, \quad \lambda_1 < \lambda, \qquad (55)$$

$$\frac{d^2 V_2}{d\lambda^2} + 2\lambda\theta_2^2 \frac{dV_2}{d\lambda} = 0, \quad 0 < \lambda < \lambda_2, \qquad (56)$$

$$\theta_2 \frac{\partial W_1}{\partial \lambda}\bigg|_{\lambda=\lambda_2} = -\theta_3 \frac{\partial V_2}{\partial \lambda}\bigg|_{\lambda=\lambda_2}, \qquad (57)$$

$$\frac{\partial W_1}{\partial \lambda}\bigg|_{\lambda=\lambda_1-0} - \frac{\partial W_1}{\partial \lambda}\bigg|_{\lambda=\lambda_1+0} = \theta_3 \frac{\partial U}{\partial \lambda}\bigg|_{\lambda=\lambda_1}, \qquad (58)$$

$$W_1(\lambda_2) = 0, \quad W_1|_{\lambda\to\infty} = W_{1\infty}, \qquad (59)$$

$$V_2(0) = V_{20}, \quad V_2(\lambda_2) = 0. \qquad (60)$$

The solution is:

$$V_2 = \begin{cases} V_{20}\left(1 - \frac{erf(\theta_2\lambda)}{erf(\theta_2\lambda_2)}\right), & \lambda < \lambda_2 \\ 0, & \lambda > \lambda_2 \end{cases} \qquad (61)$$

$$W_1 = \begin{cases} 0, & \lambda < \lambda_2 \\ C_{22}(erf(\theta_3\lambda) - erf(\theta_3\lambda_2)), & \lambda_2 < \lambda < \lambda_1 \\ W_{1\infty} + C_{21}erfc(\theta_3\lambda), & \lambda > \lambda_1 \end{cases} \qquad (62)$$

$$\theta_3 V_{20}\exp\left(-\lambda_2^2\left(\theta_2^2 - \theta_3^2\right)\right) = \theta_2 C_{22} erf(\theta_2\lambda_2). \qquad (63)$$

That is, one can change the second reaction's front position and, correspondingly the distribution of the product by varying the concentration of $W_{1\infty}$.

The analytical expression for the enhancement factor also depends on mutual position of the fronts. If $\lambda_2 < \lambda_1$

$$E_{V_2} = 1/erf(\theta_2\lambda_2). \qquad (64)$$

On the other hand, if $\lambda_2 > \lambda_1$, then

$$E_{V_2} = \frac{\left(1 + \frac{\theta_2 U_\infty}{V_{20}} \frac{erf(\theta_2\lambda_2) - erf(\theta_2\lambda_1)}{erfc(\lambda_1)\exp\left(-\lambda_1^2\left(\theta_3^2-1\right)\right)}\right)}{erf(\theta_2\lambda_2)}. \qquad (65)$$

For $\lambda_2 \to \infty$ expression (65) transforms to (40) .

3. CONCLUSIONS

The enhancement factor for the second reaction depends not only on the surface concentration of V_{20}, but on the initial concentration $W_{1\infty}$ of the first reaction's product. While the V_{20}-dependence is explicit, the $W_{1\infty}$ influences the boundary inflow indirectly, via λ_2. For $W_{1\infty} = 0$ we get maximal values of λ_2 as function of V_{20}, see Fig.5; increasing $W_{1\infty}$ decreases λ_2, and (formally) $W_{1\infty} \to \infty$ yields $\lambda_2 \to 0$.

That is, the absorption of V_2 may be controlled by variation of initial value of W_1.

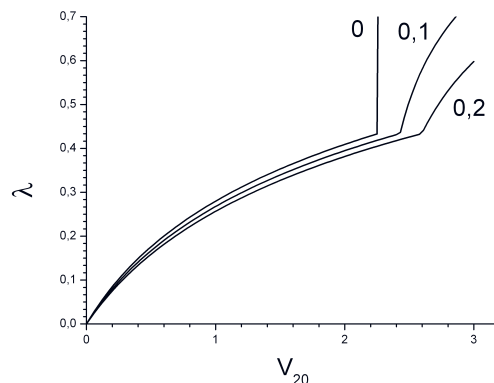**Fig.5.** λ_2 dependence on V_{20} for $W_{1\infty} = 0$, $W_{1\infty} = 0,1$ and $W_{1\infty} = 0,2$. The other parameters are the same $\theta_1 = 1.3$, $\theta_2 = 1.7$, $\theta_3 = 1.5$, $V_{10} = 2$, $U_\infty = 1$

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ДВЕ ПОСЛЕДОВАТЕЛЬНЫЕ ГЕТЕРОГЕННЫЕ РЕАКЦИИ В ДВУХФАЗНОЙ СИСТЕМЕ

Д.Ю. Копейченко, П.О. Мчедлов-Петросян

В работе рассмотрена диффузия веществ в полубесконечное пространство, в котором протекают две последовательные реакции. Получены аналитические автомодельные решения для случаев с изначальным наличием и отсутствием в среде продукта первой реакции. Получены неравенства, определяющие взаимное расположение фронтов реакций.

ДВІ ПОСЛІДОВНІ ГЕТЕРОГЕННІ РЕАКЦІЇ В ДВОФАЗОВІЙ СИСТЕМІ

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У роботі розглянуто дифузію речовин у напівбезконечний простір, у якому протікають дві послідовні реакції. Отримано аналітичні автомодельні рішення для випадків із початковою наявністю і відсутністю в середовищі продукту першої реакції. Отримано нерівності, що визначають взаємне розташування фронтів реакцій.