Development of the mathematical model of catalytic oxidation-reduction reaction kinetics for wastewater treatment technology using vortex layer devices

P E Lukashev, M I Sidorov, M E Stavrovskiy, S A Lazarev, A V Ragutkin, E I Korolev and N Sh Gazanova

MIREA - Russian Technological University, 78 Vernadsky Avenue, Moscow 119454, Russia
dyuzya@gmail.com

Abstract. The article considers the mechanisms of intensifying chemical reactions occurring in vortex layer devices. The competition of reagents used as oxidizing agents in water treatment and oxidizing agents appearing during the operation of vortex layer devices is considered. The article offers a description of wastewater treatment process kinetics both for the oxidizing process and the process caused by electrochemical (corrosive) synthesis of highly active reducing agents. The mathematical model is constructed and its analysis is given.

1. Introduction

The goal of the paper [1] was to evaluate the possibility of studying the peculiarities of kinetics in vortex layer devices (VLD) [2, 3] using the mathematical modelling of the “typical” technological process of wastewater treatment. For this purpose, a simple model was considered on the example of catalytic reduction of \( \text{Cr(VI)} \) compounds with salts of \( \text{Fe}^{2+} \). This model demonstrated that the independent additional process of producing highly active oxidizing agents \( (\text{OH}, \text{H}_2\text{O}_2 \text{ etc.}) \) during the cavitation sonolysis of the treated water can explain the high speed of the purification process in the VLD. As it was noted in [1], the designed simple model has wide possibilities for modification.

In order to set the next similar task of studying the wastewater treatment technological process using mathematical modelling, we need to note the assumption that was not specified in [1]. This assumption consists in the fact that the description of the process actually postulated that the amount of reducing agent (reagent dose) is excessive and always sufficient for the process of oxidizer (contamination) recovery. Therefore, its concentration was not mentioned in the model. However, the reagent dose is an important parameter for the technologist, since an insufficient dose would prevent achieving the required degree of purification, while an excessive amount would turn the reagent into a contaminant. This secondary contamination can pose a problem for the technologist. Calculations based on stoichiometry of simple “oxidizer — reducing agent” reaction can only estimate reagent dose. At the same time, the peculiarities of the kinetics of heterogeneous catalytic redox reactions can be unexpected.
2. Mathematical model

First of all, it should be noted that heterogeneous catalysis is associated with the concentration of reagents on the catalyst surface (naturally, this is not its only specifics). Therefore, this fact should be reflected in the mathematical model. Similarly, to the previous study, we will use the simplest models wherever possible. This approach will let us use the results of technological experiments to estimate the model parameters responsible for certain stages of the process and thus, to check the performance of the model by comparing it with the qualitative dynamics of the real object.

Let the following equilibriums of adsorption (desorption) be realized on the surface of the catalyst adsorbent

\[
A_N + \hat{A}_v \rightleftharpoons k_1 A_F; \quad \tag{1.a}
\]

\[
A_N + B_v \rightleftharpoons k_2 B_F, \quad \tag{1.b}
\]

where \(A_N\), \(A_F\), \(B_F\) — surface concentration of active adsorbent centers that are free of adsorbates and occupied by adsorbates: oxidizer and reducing agent, respectively; \(k_1\), \(k_2\), \(k_{-1}\), \(k_{-2}\) — velocity constants of adsorption and desorption of oxidizer and reducing agent.

From the scheme of reactions (1) follows a system of differential equations of kinetics

\[
\frac{dA_F}{dt} = k_1 A_N A_v - k_{-1} A_F; \quad \tag{2.a}
\]

\[
\frac{dB_F}{dt} = k_2 A_N B_v - k_{-2} B_F. \quad \tag{2.b}
\]

Within the quasi-stationary concentration method approach, let us initially assume that the surface area of the catalyst adsorbent is a constant value. This assumption allows us to accept that

\[
A_N + A_F + B_F = N_F, \quad \tag{3}
\]

i.e. the surface concentration of free and adsorbate occupied active centers of adsorbent-catalyst is a constant value \((N_F = const)\). In equilibrium (or for quasi-stationary approximation) \(\frac{dA_F}{dt} = \frac{dB_F}{dt} = 0\). From this follows

\[
K_{A,ads} = \frac{A_F}{(N_F - A_F - B_F)A_v}; \quad K_{B,ads} = \frac{B_F}{(N_F - A_F - B_F)B_v}. \quad \tag{4}
\]

Following [1], these expressions can be transformed using the scaled surface concentrations (unit of measurement: \(\frac{kg}{m^2}\)) instead of surface concentrations (unit of measurement: \(\frac{kg}{m^3}\)), i.e. instead of (2)–(4) we can write

\[
\frac{d\hat{A}_F}{dt} = k_1 \hat{A}_N \hat{A}_v - k_{-1} \hat{A}_F; \quad \tag{5.a}
\]

\[
\frac{d\hat{B}_F}{dt} = k_2 \hat{A}_N \hat{B}_v - k_{-2} \hat{B}_F. \quad \tag{5.b}
\]

(kinetic constants in (2) and (5) have different dimensions);

\[
\hat{A}_N + \hat{A}_F + \hat{B}_F = \hat{N}_F; \quad \tag{6}
\]
\[
\hat{K}_{A,\text{ads}} = \frac{\hat{A}_F}{(\hat{N}_F' - \hat{A}_F - \hat{B}_F)A_Y} ; \quad \hat{K}_{B,\text{ads}} = \frac{\hat{B}_F}{(\hat{N}_F' - \hat{A}_F - \hat{B}_F)B_Y} \quad \tag{7}
\]

Let us introduce dimensionless variables \( x = \frac{A_F}{A_Y,0} \); \( y = \frac{B_Y}{A_Y,0} \); \( u = \frac{B_Y}{A_Y,0} \); \( v = \frac{B_Y}{A_Y,0} \); \( N_F' = \frac{\bar{N}_F}{A_Y,0} \). After this the equations (7) with dimensionless variables will look as following

\[
K_{A,\text{ads}}^* = \frac{y}{(N_F' - y - v)x} ; \quad K_{B,\text{ads}}^* = \frac{v}{(N_F' - y - v)u}, \quad \tag{8}
\]

where \( K_{A,\text{ads}}^* = \hat{K}_{A,\text{ads}}A_Y,0 \) and \( K_{B,\text{ads}}^* = \hat{K}_{B,\text{ads}}A_Y,0 \) — dimensionless variables of adsorption equilibriums. From (8) it is possible to derive equations for dimensionless reduced surface concentrations through volumetric ones

\[
y = \frac{K_{B,\text{ads}}^* N_F' u - (K_{B,\text{ads}}^* u + 1) v}{K_{B,\text{ads}}^* u} ; \quad y = \frac{K_{A,\text{ads}}^* x(N_F' - v)}{K_{A,\text{ads}}^* x + 1}, \quad \tag{9}
\]

\[
v = \frac{K_{A,\text{ads}}^* N_F' x - (K_{A,\text{ads}}^* x + 1) y}{K_{A,\text{ads}}^* x} ; \quad v = \frac{K_{B,\text{ads}}^* u(N_F' - y)}{K_{B,\text{ads}}^* u + 1}. \quad \tag{10}
\]

Equations (9) and (10) allow to express the given surface concentrations \( y \) and \( v \) via volumetric \( x \) and \( u \)

\[
y = N_F' \frac{K_{A,\text{ads}}^* x}{1 + K_{A,\text{ads}}^* x + K_{B,\text{ads}}^* u} ; \quad v = N_F' \frac{K_{B,\text{ads}}^* u}{1 + K_{A,\text{ads}}^* x + K_{B,\text{ads}}^* u}. \quad \tag{11}
\]

Removal of the contaminant (oxidizer) \( A \) and consumption of the reagent (reducing agent) \( B \) is accompanied by the process of catalyst adsorbent production through hydrolysis of the oxidized form of the reducing agent \((Fe^{3+})\) and to some extent is possible due to hydrolysis of the reduced form of the oxidant \((Cr^{3+})\). During the hydrolysis of \( Cr^{3+} \) colloidal solutions [5]. By analogy with [1] let’s write down the equation of kinetics for this process

\[
\frac{dG}{dt} = k\hat{A}_F \hat{B}_F G, \quad \tag{12}
\]

where \( \mu = k\hat{A}_F \hat{B}_F \) — specific production rate of catalytic adsorbent. In dimensionless variables (12) it looks as follows

\[
\frac{dG}{dt} = k\hat{A}_{Y,0} \hat{B}_{Y} \mu G, \quad \tag{13}
\]

(dimensions \( k : \frac{1}{c \left( \frac{m^3}{kg} \right)^2} \)); the introduction of dimensionless variables \( z = \frac{G}{A_Y,0} \) and \( \tau = \frac{t_0}{kA_Y,0} \) makes it possible to write equation (13) as

\[
\frac{dz}{d\tau} = \mu Gz. \quad \tag{14}
\]
Besides, the specific reaction rate $\mu$ can be defined via dimensionless volumetric concentrations $x$ and $u$ by substituting for it with dimensionless $\mu^* = \frac{\mu}{k}$,

$$
\mu = \frac{kN_F^2K'_{A,ads}K'_{B,ads}xu}{(1 + K'_{A,ads}x + K'_{B,ads}u)^2};
\mu^* = \frac{N_F^2K'_{A,ads}K'_{B,ads}xu}{(1 + K'_{A,ads}x + K'_{B,ads}u)^2}.
$$  (15)

The equation (15) can be written in a generalized form

$$
\mu^* = \frac{\alpha x}{\beta + \gamma x + \delta x^2},
$$  (16.a)

where parameters in (16) are defined for a given amount of dimensionless reagent dose $u$ with the equations:

$$
\alpha = N_F^2K'_{A,ads}K'_{B,ads}u; \beta = 1 + 2K'_{B,ads}u + K'_{B,ads}u^2;
$$  (16.b)

$$
\gamma = 2K'_{A,ads}(1 + K'_{A,ads}u); \delta = K'_{B,ads}.
$$

besides, (15) can be presented as

$$
\mu^* = \frac{x}{a + bx + cx^2},
$$  (16.a)

where $a = \beta/\alpha$, $b = \gamma/\alpha$, $c = \delta/\alpha$ (in this case for constants can be substituted for with three ones, which can be more convenient for evaluations and calculations).

In formulas (16) and (17) parameters $\alpha$, $\beta$, $\gamma$, $\delta$ and $a$, $b$, $c$ are the functions of volumetric concentration of oxidizing agent and the thermodynamic affinity of the oxidizing and reducing agents to the adsorptive catalyst surface in the form of equilibrium constants of two-component Langmuir adsorption ($K'_{A,ads}$, $K'_{B,ads}$). The dependence type (17) is illustrated in figure 1 (figure 1a demonstrates the coefficient variation for quadratic terms, figure 1b — for linear terms). These dependencies reflect the fact that the oxidizing and reducing agents compete for the spare active sites of the adsorptive catalyst. If one of the agents prevails, this leads to the reduction of overall oxidation-reduction process speed that results in hydrolysis and the production of adsorptive catalyst. The maximum value of specific speed $\mu^*$ is provided by a certain optimal ratio of reagent surface concentrations, which are dependent on their volumetric concentrations.

Formula (17) allows to determine the volumetric concentration of the contaminant component, which corresponds to the maximum specific speed of the treatment process. Analyzing the function

$$
\mu^*(x) = \frac{x}{g(x)}
$$

to the maximum leads to a simple ration: $x^*_\mu = \sqrt[4]{\frac{\alpha}{c}}$ (which is derived from equation $(\mu^*(x))^\prime_x = 0$, i.e. $a - cx^2 = 0$).

Thus, for example, for the curve $\alpha(x)$ (figure 1a), which represents $\mu^*(x)$ at $a = 0.05; b = 0.5; c = 2.0$ the dimensionless volumetric concentration that corresponds to the maximum $\mu^*(x)$, equals $x^*_\mu = 0.158$. 
Figure 1. Dependence (17) — specified by $\alpha(x)$, $\beta(x)$, $\gamma(x)$:

(a) — $a = 0.05; b = 0.5; c_{\alpha} = 2.0; c_{\beta} = 5.0; c_{\gamma} = 10.0$;
(b) — $a = 0.05; b_{\alpha} = 0.5; b_{\beta} = 0.75; b_{\gamma} = 1.0; c = 2.0$.

This illustration demonstrates that the “kinetic” behavior of the system in the case of competitive adsorption significantly differs from the behavior of a system in which the adsorbates do not compete for the spare active sites of the adsorptive catalyst [1]. Such “kinetic” behavior reflects significant peculiarities of the studied system and makes it interesting to perform a more thorough analysis of the kinetics differential equation system for competitive adsorption using numerical methods. In order to do this, we will supplement the system of adsorption equations (1) with a catalytic reaction between reagents (oxidizing and reducing agents) adsorbed on the surface of the catalyst adsorbent. In the result of this reaction the surface is filled with new spare active sites,

$$ A_r + B_r \rightarrow fA_N, \quad (18) $$

where $f$ is the stoichiometric coefficient (which is an adjustable parameter in terms of the technological process).

In this case the complete system of kinetic equations (in normalized mass concentrations) looks as following

$$ \frac{dA_r}{dt} = -k_1 \hat{A}_r A_r + k_{-1} \hat{A}_r; \quad (19.a) $$

$$ \frac{d\hat{A}_r}{dt} = k_1 \hat{A}_r A_r - k_{-1} \hat{A}_r - k_2 \hat{A}_r \hat{B}_r; \quad (19.b) $$

$$ \frac{dB_r}{dt} = -k_2 \hat{A}_r \hat{B}_r + k_2 \hat{B}_r; \quad (19.c) $$

$$ \frac{d\hat{B}_r}{dt} = k_2 \hat{A}_r B_r - k_{-2} \hat{B}_r - k_3 \hat{A}_r \hat{B}_r; \quad (19.d) $$

$$ \frac{d\hat{A}_N}{dt} = -k_1 \hat{A}_N A_r + k_{-1} \hat{A}_N + k_2 \hat{A}_N B_r + k_2 \hat{B}_r + f k_3 \hat{A}_r \hat{B}_r. \quad (19.e) $$

The introduction of dimensionless normalized concentration $z = \hat{A}_N/A_{N,0}$ (in addition to the previously introduced $x, y, u, v$) and time $\tau = t/t_0$ ($t_0 = 1/k_1 A_{r,0}$) allows to write the system (19) as

$$ \dot{x} = -\alpha x z + \beta y; \quad (20.a) $$
Due to the reaction of formation of new active sites (18), the volumetric concentration slows down (figure 3). Because of this, the surface concentration of the second component increases faster, but it also peaks at later times (also because of the competition with the first component; figure 3.d).

The comparison of Figures 2 and 3 makes it possible to trace the influence of competition on the studied process. The only difference between the situations illustrated in figure 3 and in figure 2 is that the adsorption equilibrium constant of the second component is 5 times higher than the equilibrium constant of the first component \((\alpha/\beta < \gamma/\delta)\). This leads to higher thermodynamic affinity of the second component with the adsorbent catalyst surface, and its consequent competitive advantage. This results in the fact that the process speed both for the first and for the second component decreases (depletion happens at significantly longer durations — figure 3.a, c).

Thus, it can be noted that the kinetics of the studied process in the conditions of competitive adsorption is more complicated than the kinetics of the process when the reagents do not compete for the active surface of the adsorptive catalyst ([1]; figure 2–5).

The preliminary system analysis (20) shows that it has no stationary solution, because if the right parts of equations e.g. (20.a) and (20.b) are equal to zero and then to each other, this leads to the result \((\varepsilon yv = 0)\), which is not assumed by the conditions of the problem. This means the problem has no stationary solution. Therefore, the stationary state of the studied system does not exist in the batch reactor regime, which corresponds with the batch reactor dynamics [5–7].

The results of numerical calculations (MathCad) with varying parameters of the equation system (20) are shown in figure 2–4. Figure 2 demonstrates the situation when the adsorption equilibrium constants of both reagents equal \((\alpha/\beta = \gamma/\delta = 10)\), i.e. they are on par in terms of competing for the surface (for the spare active sites). In the beginning of the process the volumetric concentration of both reagents drastically decrease (figure 2a, c) due to the adsorption on the surface that is free of them (this decrease is not significantly high in absolute figures; figure 2e, f). A similar change is demonstrated by the curves of surface concentrations increase in figure 2b, d (not shown in larger scale). After this stage the decrease of their volumetric concentration slows down (figure 2e, f), but then the speed of this process increases (figure 2a, c) until the moment when it starts to slow down due to the depletion of reagents in the volume of the reactor. The process completes simultaneously for both reagents.

At first the surface concentration of both reagents increases (figure 2b, d), but as they get depleted in the reactor volume it starts to decrease; however, at almost zero concentration in the volume, the surface concentration does not reach zero. Due to the reaction of formation of new active sites (18), their surface concentration grows along the S-curve (figure 2g) almost throughout the whole process except for the initial period (figure 2h) when the concentration reduction is conditioned by the adsorption.

Thus, it can be noted that the kinetics of the studied process in the conditions of competitive adsorption is more complicated than the kinetics of the process when the reagents do not compete for the active surface of the adsorptive catalyst (11; figure 2–5).

\[
\dot{y} = \alpha xz - \beta y - \varepsilon yv; \quad (20.b)
\]
\[
\dot{u} = -\gamma uz + \delta v; \quad (20.c)
\]
\[
\dot{v} = \gamma uz - \delta v - \varepsilon yv; \quad (20.d)
\]
\[
\dot{z} = -\alpha xz + \beta y - \gamma uz + \delta v + f e yv, \quad (20.e)
\]

where \(\alpha = k_1/k_2; \beta = k_1/k_3; \gamma = k_2/k_3; \delta = k_2/k_4; A_k; \varepsilon \approx 1; f \) — adjustable (stoichiometric) parameter; \(\dot{x} = dx/d\tau; \dot{y} = dy/d\tau; \dot{u} = du/d\tau; \dot{v} = dv/d\tau; \dot{z} = dz/d\tau\). Here \(\alpha\) and \(\beta\) are the relative adsorption and desorption rates (i.e. related to the rate of oxidation-reduction reaction).
Figure 2. Variation of dimensionless concentrations over time; here $x \rightarrow Z_{n,1}$; $y \rightarrow Z_{n,2}$; $u \rightarrow Z_{n,3}$; $v \rightarrow Z_{n,4}$; $z \rightarrow Z_{n,5}$; figure (e), (f) and (h) — same as figure (a), (c) and (g), but at the initial period; here $\alpha = 1; \beta = 0.1; \gamma = 1; \delta = 0.1; \varepsilon = 1; f = 3$; initial conditions: $x_{i} = 1; y_{i} = 0.01; u_{i} = 1; v_{i} = 0.01; z_{i} = 0.1$. 
Figure 3. Variation of dimensionless concentrations over time; here $x \rightarrow Z_{n,1}$, $y \rightarrow Z_{n,2}$, $u \rightarrow Z_{n,3}$, $v \rightarrow Z_{n,4}$, $z \rightarrow Z_{n,5}$; figure (e), (f) and (h) — same as figure (a), (c) and (g), but at the initial period; here $\alpha = 1; \beta = 0.1; \gamma = 5; \delta = 0.1; \varepsilon = 1; f = 3$; initial conditions: $x = 1; y = 0.01; u = 1; v = 0.01; z = 0.1$. 
Figure 4. Variation of dimensionless concentrations over time; here \( x \rightarrow Z_{n,1}; \ y \rightarrow Z_{n,2}; \ u \rightarrow Z_{n,3}; \ v \rightarrow Z_{n,4}; \ z \rightarrow Z_{n,5}; \) figure (e), (f) and (h) — same as (a), (c) and (g), but at the initial period; here \( \alpha = 1; \beta = 0.1; \gamma = 1; \delta = 0.1; \varepsilon = 1; \ f = 3; \ g = 0.05 \) initial conditions: \( x_0 = 1; \ y_0 = 0.01; \ u_0 = 1; \ v_0 = 0.01; \ z_0 = 0.1. \)
Previously, the mathematical model of the kinetics of heterogeneous catalysis of oxidation-reduction reactions showed \[1\] that an additional independent process that produces highly active oxidizing agents \( (\cdot \cdot \cdot \cdot \cdot H_\cdot \cdot H \cdot \cdot \cdot \cdot O_\cdot \cdot \cdot O \cdot \cdot \cdot O \cdot \cdot \cdot ) \) and includes cavitation sonolysis reactions [6, 7], which supposedly can take place in VLD [2], leads to a sharp increase in the rate of the main process.

In the mathematical model, this effect is reflected in the modified equation (20.e)

\[
\dot{c} = -axz + \beta y - \gamma uz + \delta v + f v y v + g v,
\]  

where component \( g v \) represents the following mechanism: an independent catalytic reaction of oxidizing \( Fe^{2+} \) to \( Fe^{3+} \), on the one hand, releases an active site for the second component and on the other hand, produces new active sites through hydrolysis, thus accelerating the total process. This mechanism is confirmed by the results of calculations (figure 4, figure 5). The greatest influence of this independent process is shown in the second case, when the reducing agent occupies the most part of adsorbent-ta-catalyst surface and therefore plays the role of inhibitor (similar to substrate inhibition in enzymatic kinetics [9–11]).

Thus, this variant of demonstrating the contribution of processes initiated in VLD, as well as the previous case [1], shows the possibility of taking into account a number of specific processes, in particular cavitation water sonolysis.

On the other hand, in addition to cavitation and sonolysis, VLD should activate the processes of electrochemical corrosion of the loading particles material (“needles”), especially if they are made of steel [2]. It is known [10] that corrosion processes in the electrolyte medium are strongly activated by friction due to constant removal of electrolysis products (e.g., passive films) as well as due to the elimination of diffusion restrictions on the supply of active solution components (oxidizing agents and reducing agents) to anode and cathode sections of galvanic corrosion pairs. Highly active oxidizing agents formed in sound-chemical reactions can also significantly accelerate this electrochemical process, during which anodic oxidation of metallic iron \( (Fe^0) \) may produce \( Fe^{2+} \), which performs the same function as the reagent introduced for cleaning.

In [2] it was also noted that the electromagnetic field that rotates the loaded ferromagnetic particles could also contribute to the activation of electrochemical corrosion by induced electrical charges. In this case, the additional process of corrosion production of active forms of reducing agent \( (Fe^0, Fe^{2+}) \) will have an additional increase in speed compared to the rate of tribochemical corrosion. Thus, additional processes conditioned by the cavitation synthesis of highly active oxidizing agents may run in VLD, but also processes conditioned by electrochemical (corrosion) synthesis of highly active reducing agents. This requires a separate qualitative study similar to the one presented above.

The contribution of these additional processes to the total process of water treatment can be reflected within the framework of kinetics models in the form of values of kinetic constants. Therefore, the study makes it possible to conclude that the mathematical models considered in [1] and in this work can be accepted as the basis for the development of the kinetics of wastewater treatment processes that include both sequential and parallel reactions. In this regard, a more in-depth parametric analysis of these models is required.

It should be also noted that chemical and technological systems (CTS) of sewage treatment are usually operated in a continuous mode. This requires mathematical models that correspond to these modes. In its simplest variant this task is solved by the model of continuous stirred-tank reactor (CSTR). The CSTR model may be the most appropriate for the actual process, since in VLD the mixing is very intensive and the retention time is short.

Mathematical models make it is much easier to study the stability of the technological process than the experiment, because the processes that include reactions with complex kinetics often demonstrate unstable behavior. For example, instability is typical for reactions with substrate inhibition (enzyme kinetics) [9–11], which, as noted above, have a description formalism similar to that of reactions involving stages of competitive adsorption. Unstable modes are extremely undesirable for CTS.
Figure 5. Variation of dimensionless concentrations over time; here $x \rightarrow Z_{n,1}$; $y \rightarrow Z_{n,2}$; $u \rightarrow Z_{n,3}$; $v \rightarrow Z_{n,4}$; $z \rightarrow Z_{n,5}$; figure (e), (f) and (h) — same as (a), (c) and (g), but at the initial period; here $\alpha = 1; \beta = 0.1; \gamma = 5; \delta = 0.1; \varepsilon = 1; f = 3; g = 0.05$ initial conditions: $x_i = 1; y_i = 0.01; u_i = 1; v_i = 0.01; z_i = 0.1$. 
All these tasks (within the framework of the general task of XTS mathematical model synthesis) can be studied with the help of the reviewed kinetic model that takes into account both basic (1), (18) and additional reactions, which may require further modifications.

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