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Қ. И. Сәтпаев атындағы Қазақ ұлттық техникалық зерттеу университеті

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Kazakh national research technical university
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ГЕОЛОГИЯ ЖӘНЕ ТЕХНИКАЛЫҚ ҒЫЛЫМДАР СЕРИЯСЫ

♦

СЕРИЯ ГЕОЛОГИИ И ТЕХНИЧЕСКИХ НАУК

♦

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SIMULATION OF AUTOCATALYTIC SYSTEMS WITH CHEMICAL OSCILLATIONS WITH ALLOWING FOR REACTION STAGES REVERSIBILITY

Abstract. The paper deals with the simulation of two auto-catalytic system are remarkable for chemical oscillations in the case of some reactions stages reversibility. The methods for engineering calculations of kinetic characteristics and the residence time applying to the dynamical processes of mass transfer in cascades of chemical reactors for various model environments have been studied. The preferred model schemes for describing various systems with vibrational chemical reactions have been established. For bromate systems and systems with oxidation-reduction reactions containing metal ions, it is proposed to use a Belousov-Zhabotinsky model with irreversibility kinetic stages. For systems with organic reducing agents the suitable model is a model of the type of the Belousov–Zhabotinsky system, taking into account the reversibility of the reaction stages. For systems with enzymatic reactions and biochemical systems it is a model with autocatalysis or "Brusselator".

Grounded on the criteria equations for the known flow pattern in each of the cascade reactors, the following calculation sequence is recommended. First, the required average velocity of the phase flows in the section of the tubular reactor is calculated. Then, according to this average velocity, the diameter of the apparatus is calculated for a given consumption of the processed substance. Then, at a given degree of the conversion in the reactor and at a certain average flow rate, the required residence time in the reactor is calculated according to the offered method. If the length of the reactor is specified for design reasons, the calculated residence time can be used to determine the required number of reactors in the cascade. If the length of the reactor is not specified, it can be selected by iterative calculation according to the described procedure to ensure a given degree of the conversion.

Key words: modelling of chemical reactors, recycle, two-stage reactor, Belousov-Zhabotinsky reaction, “brusselator” system.

1. Introduction. Description of concentration wave fronts and oscillatory processes in reaction-diffusion systems is an extremely topical scientific problem, which also has great practical significance. Many known methods for modeling chemical reactors completely ignore the possibility of forming wave fronts in physicochemical systems. At the same time, the intensity of the processes of heat and mass transfer in the presence of moving frontal sections changes significantly and cannot be correctly calculated without accounting these phenomena.

In this paper, we consider some fairly simple models that make it possible to describe the general characteristic features of propagation of wave fronts in reaction-diffusion systems.

2. Analysis of model systems.

2.1. Belousov-Zhabotinsky reaction. The Belousov-Zhabotinsky (BZ) reaction is the process of oxidation of malonic or bromomalonic acid by bromate ions in an acidic medium, catalyzed by ions of

transition metals (cerium, iron). Under different conditions in a flow-type reactor with mixing, one can observe long-term self-oscillations, several stationary states and periodic regimes, chaotic behavior, and for distributed systems the propagating concentration waves of oxidation and restoration can be occurred. It is well-known that this reaction serves as a model for studying nonlinear phenomena in chemical kinetics. Field, Köresh and Noyes developed a detailed scheme for the mechanism of this reaction, consisting of eleven basic reactions between twelve components. Later, Field and Noyes proposed a simplified scheme, consisting of five main stages.

In our analysis, we will use the reduced scheme of Field-Noyes. Let us denote the components of the reaction as follows: $A=BrO_3^-$, $B=BrMA$, $P=HOBr$, $X=HbrO_2$, $Y=Br^-$, $Z=Ce^{4+}$. The appropriate scheme of the reaction reads

$$A + Y \to X + P, \tag{1}$$

$$X + Y \to 2P, \tag{2}$$

$$A + X \to 2X + 2Z, \tag{3}$$

$$2X \to A + P, \tag{4}$$

$$B + Z \to hY. \tag{5}$$

As a rule, the known papers in mathematical models of the kinetics of individual stages do not take into account the reversibility of the reactions. However, under real conditions, many reaction stages of the Belousov-Zhabotinsky type can be reversible [1, 2]. This circumstance substantially complicates the kinetic equations and their analysis.

Further we consider the case of reversibility of the fourth stage of the reaction, i.e. decomposition of the product *X*.

Then the scheme of chemical equations is supplemented by the equation:

$$A + P \xrightarrow{k_6} 2X. \tag{6}$$

Let us assume that the components A and B are in a large excess and that their concentrations do not change noticeable on time. For the convenience of numerical experiments and the data interpretation, all reaction rate constants k_i, transfer coefficients D_{ij} and time t were dimensioned with respect to the relaxation time of the first stage $\tau_p = 1/k_1$, i.e. the dimensionless characteristics were determined according to the scheme:

$$t \to tk_1,\ k_i \to k_i/k_1,\ D_{ij} \to D_{ij}\ k_1. \tag{7}$$

Then the time variation of the concentrations of the remaining components in a closed system can be described using the equations:

$$\begin{aligned}
\frac{dX}{dt} &= k_1 AY - k_2 XY + k_3 AX - 2k_4 X^2 + 2k_6 AP,\\
\frac{dY}{dt} &= -k_1 AY - k_2 XY + hk_5 BZ,\\
\frac{dZ}{dt} &= 2k_3 AX - k_5 BZ,\\
\frac{dP}{dt} &= k_1 AY + 2k_2 XY + k_4 X^2 - 2k_6 AP
\end{aligned} \tag{8}$$

The above balance equations do not take into account the input and output streams. Thus, it is assumed that only the *Y* (Br^-) component is fed into the flow-type reactor with mixing, and the reaction components X *X*($HbrO_2$) and *Z*(Ce^{4+}) are intermediate products arising during the reaction. The analytical solution of the obtained system (8) is impossible. Therefore, we carried out a numerical study using the Runge-Kutta method. For the numerical experiment, the following sets of control parameters were selected:

The first series of experiments:

$$k_1 = 1,\ k_2 = 1,\ k_3 = 1,\ k_4 = 1,\ k_5 = 1,\ k_6 = 1/2,$$

The second series of experiments:

$$k_1 = 1,\ k_2 = 1,\ k_3 = 1,\ k_4 = 1,\ k_5 = 1/2,\ k_6 = 1/2.$$

Some results of the experiments are depicted in figures 1, 2.

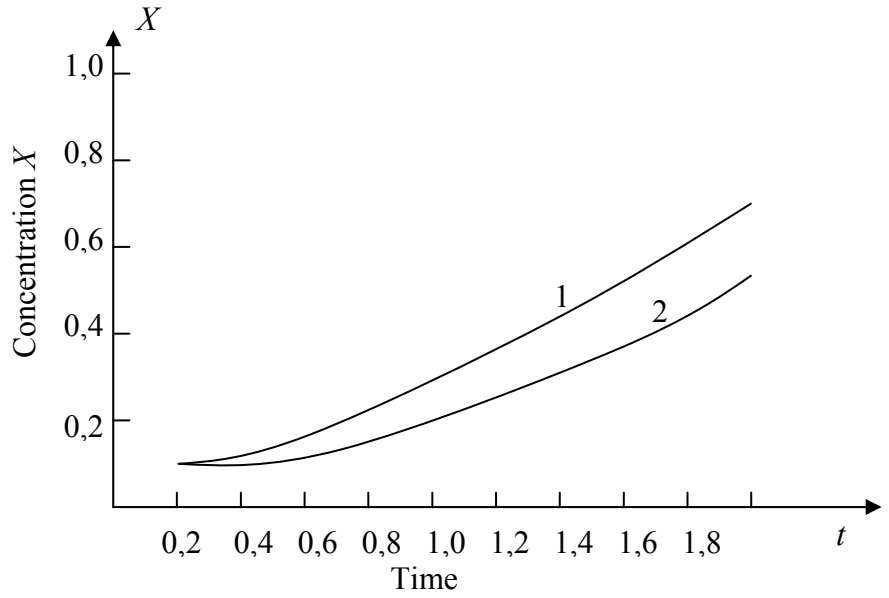


Figure 1 – The change in the concentration of intermediate reagent *X* in the reactor of flow type with mixing at *A* = 1; *B* = 1,4: 1 – reversible reaction; 2 – inreversible reaction

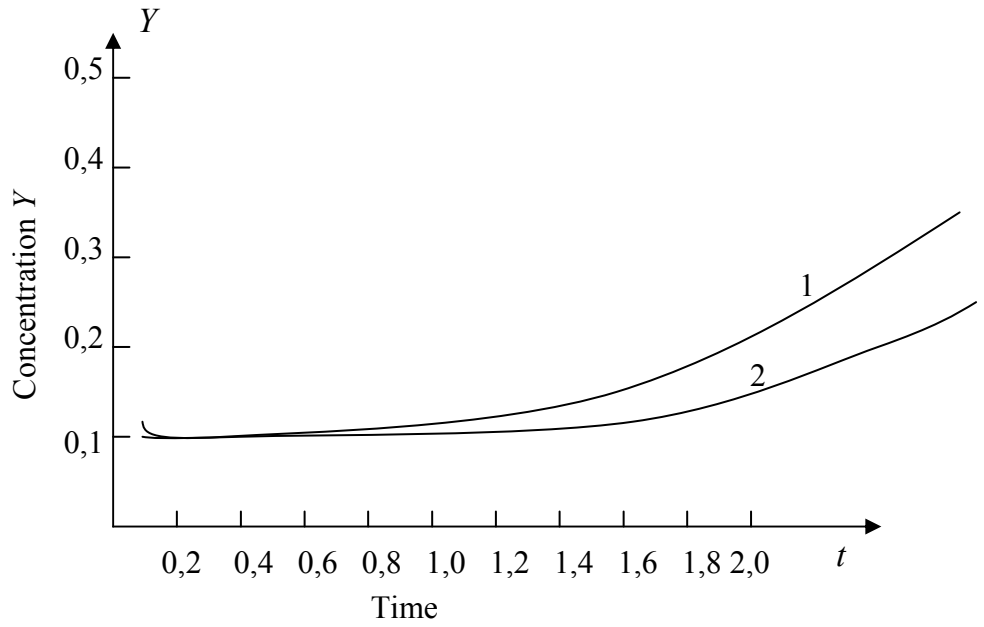


Figure 2 – The change in the concentration of intermediate reagent *Y* in the reactor of flow type with mixing at *A* = 1; *B* = 1,4:1 – reversible reaction; 2 – inreversible reaction

Numerical investigation has shown that taking into account the reversibility of the reaction stages can be important in the simulation of the considered process. In particular, it can be seen that the reversibility of the fourth stage alters the yield of the product *Z* by 25%.

2.2. "Brusselator" system with accounting two reaction stages reversibility. When modeling nonlinear physical and chemical systems, a system of nonlinear kinetic equations of the "brusselator" type is often used. This system simulates many real complex multi-stage reactions occurring in industrial reactors.

In accordance with this model, the starting reagents *A* and *B* are converted to reaction products *D* and *E* with the appearance of intermediates *X* and *Y*, and the kinetics of the reaction can be described by the following system of equations:

$$\begin{aligned} &A \xrightarrow{k_1} X, \\ &B + X \xrightarrow{k_2} Y + D, \\ &2X + Y \xrightarrow{k_3} 3X, \\ &X \xrightarrow{k_4} E. \end{aligned} \tag{9}$$

In the presented form, the system of kinetic equations is rather complicated for analytical investigation. The nonlinearity of the kinetic equations leads to a multiplicity of stationary states that differ from one another in the type of stability. In addition, under real conditions, the individual reaction stages can be reversible, which stipulates the formation of feedbacks and also complicates the theoretical analysis of the dynamics of the process.

At the same time, as it has already been shown, the question of reversibility is extremely important and requires special research.

In our work, a numerical study of the solutions of the system of kinetic equations describing the dynamics of the process (9) has been carried out, taking into account the reversibility of the two reaction stages, namely corresponding to the decomposition of the substance *A* entering the reactor and the product of the reaction between *X* and the other substance *B* entering the reactor inlet.

In this case the system of chemical equations is supplemented as follows:

$$\begin{aligned} &Y + D \xrightarrow{k_5} B + X; \\ &E \xrightarrow{k_6} X. \end{aligned} \tag{10}$$

With a constant concentration of the initial substances *A* and *B*, and the given set of reaction rate constants, we obtain the following mathematical model of the process kinetics in the reactor:

$$\begin{aligned} \frac{dX}{dt} &= k_1 A - k_2 BX + k_5 YD + k_3 X^2 Y - k_4 X + k_6 E, \\ \frac{dY}{dt} &= k_2 BX - k_5 YD - k_3 X^2 Y, \\ \frac{dD}{dt} &= k_2 BX - k_5 YD, \\ \frac{dE}{dt} &= k_4 X - k_6 E. \end{aligned} \tag{11}$$

Numerical investigation of the system (12) is carried out with the following values of reaction rate constants:

$$k_1 = 1,\ k_2 = 1,\ k_3 = 1,\ k_4 = 1,\ k_5 = 1/2,\ k_6 = 1/2\ .$$

Some results of the numerical experiment are presented in figures 3, 4.

An analysis of the results of a numerical study shows that the reversibility of the individual stages of the reaction can lead to a significant change in the parameters of the stationary state. This circumstance can radically change the type of stability of a stationary point. Moreover, the influence of the irreversibility factor increases with time and it is more significant not in the initial transition period, but in a period process stabilization corresponding to a completely irreversible system. As the rate constant of the reverse reaction increases, the influence of the reversibility factor increases too.

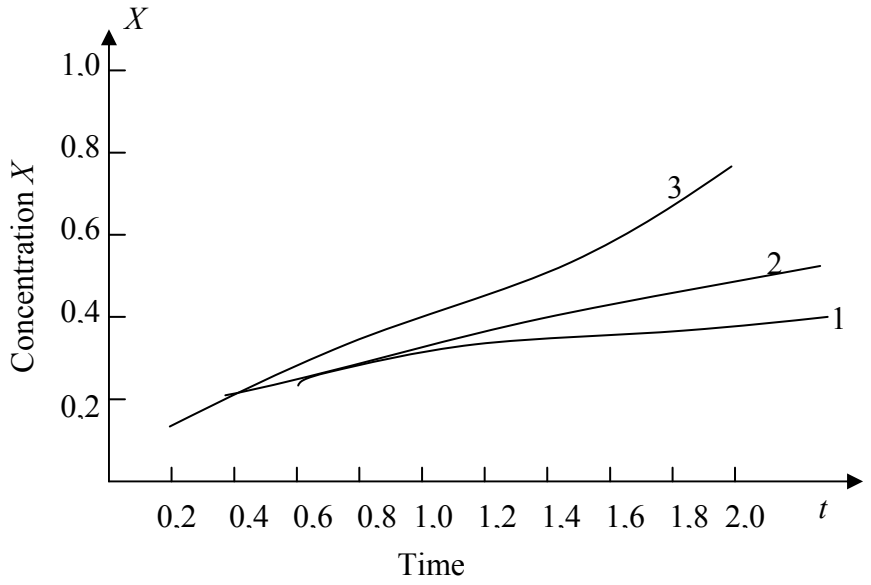


Figure 3 – The change in the concentration of intermediate reagent *X* in the reactor of flow type with mixing for "brusselator" at $A = 1$; $B = 1{,}4$:
1 – inreversible reaction; 2 – reversibility of the one reaction stage; 3 – reversibility of two reaction stages

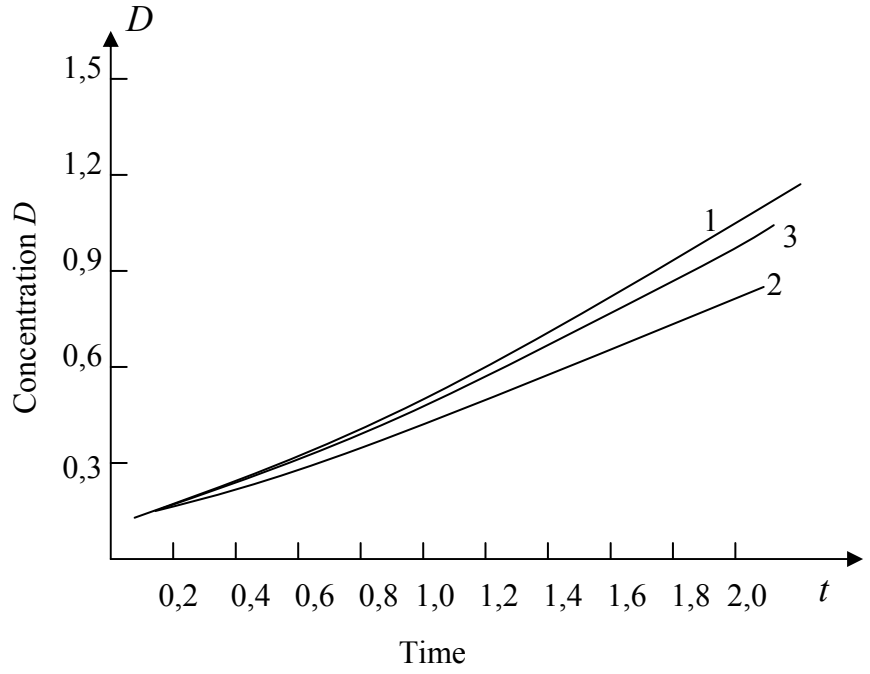


Figure 4 – The change in the concentration of intermediate reagent *D* in the reactor of flow type with mixing for "brusselator" at $A = 1$; $B = 1{,}4$:
1 – inreversible reaction; 2 – reversibility of the one reaction stage; 3 – reversibility of two reaction stages

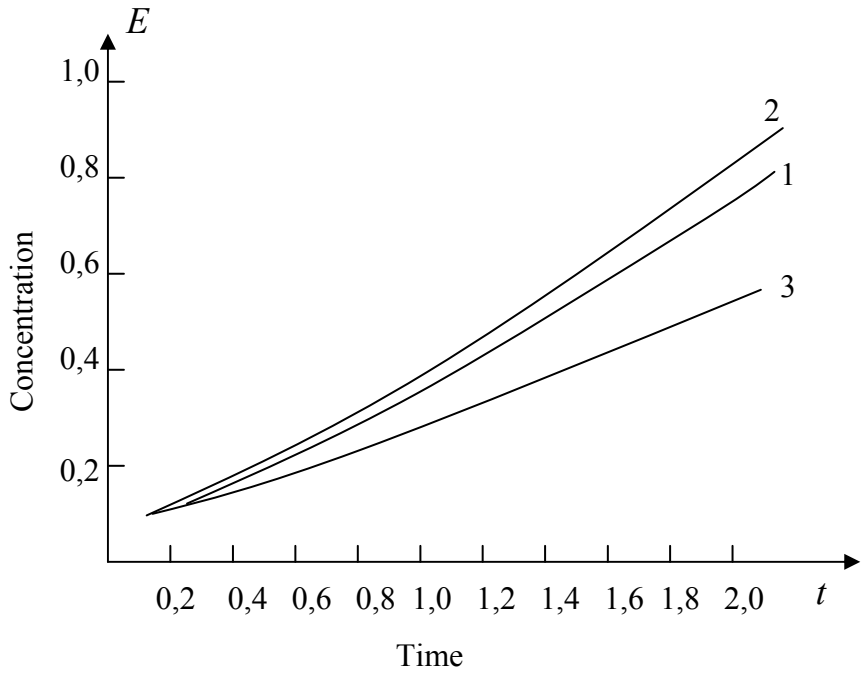


Figure 5 – The change in the concentration of intermediate reagent *E*
in the reactor of flow type with mixing for "brusselator" at $A = 1$; $B = 1{,}4$:
1 – inreversible reaction; 2 – reversibility of the one reaction stage; 3 – reversibility of two reaction stages

Further studies should be aimed at assessing the influence of the flow structure in the reactor and of the reversibility of various stages of the "brusselator type "reactions on the kinetics of the process.

3. Methods of calculation and discussion of the results.

3.1. Simulation of a two-cascade reactor with a partially reversible reaction in the "brusselator" system. Simulation of chemical reactors for the implementation of complex physicochemical processes requires taking into account the specific features of the kinetics of chemical transformations and the conditions for the heat and mass transfer. at the same time, the correct choice of the model system allows often for greater generality of the conclusions and the possibility to create methods for calculating a wide class of systems and reactors.

In this section, a mathematical model of two cascade reactors with a brusselator type reaction is constructed, taking into account the partial decomposition of the final product, that is, the reversibility of the last stage of the reaction:

$$E \xrightarrow{k_5} X$$

The numerical investigation of the developed mathematical model is carried out. The concentrations of the input substances *A* and *B* are assumed to be constant (that is, the assumption of their excess is assumed), and the reaction rate constants are assumed to be equal to the following values: $k_1 = 1$, $k_2 = 1$, $k_3 = 1$, $k_4 = 1$, $k_5 = 1/2$.

The structure of the streams is represented as a cascade of two fully agitated reactors with mutual mass transfer (figure 8).

a b

Figure 6 – Cascade of two fully agitated reactors with mutual mass transfer:
a) – cascade with recycles; b) – stream in one direction

The corresponding systems of kinetic equations, provided that the resulting inflow and outflow of intermediate products are zero, and the concentrations of incoming components are kept constant, have the form:

$$\frac{dX_1}{dt} = k_1A_1 - k_2BX_1 - k_4X_1 + k_3X_1{}^2Y_1 + k_5E_1 + D_{11}(X_2 - X_1),$$
$$\frac{dY_1}{dt} = k_2BX_1 - k_3X_1{}^2Y_1 + D_{21}(Y_2 - Y_1), \frac{dE_1}{dt} = k_4X_1 - k_5E_1 + D_{31}(E_2 - E_1),$$
$$\frac{dX_2}{dt} = k_1A_2 - k_2BX_2 - k_4X_2 + k_3X_2{}^2Y_2 + k_5E_2 + D_{12}(X_1 - X_2), \quad (12)$$
$$\frac{dY_2}{dt} = k_2BX_2 - k_3X_2{}^2Y_2 + D_{22}(Y_1 - Y_2), \frac{dE_2}{dt} = k_4X_2 - k_5E_2 + D_{32}(E_1 - E_2),$$

Two reactors with mutual mass transfer can also be considered in the case of a stream flowing in only one direction from the reactor 1 to the reactor 2.

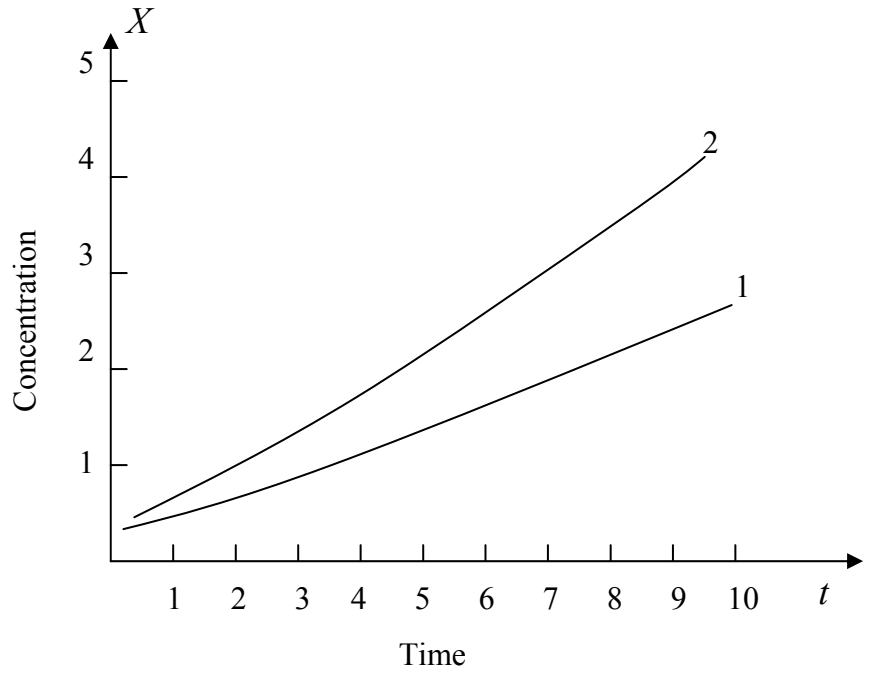
Then the system of equations (12) takes the form:

$$\begin{aligned}
\frac{dX_1}{dt} &= k_1A_1 - k_2BX_1 - k_4X_1 + k_3X_1{}^2Y_1 + k_5E_1,\\
\frac{dY_1}{dt} &= k_2BX_1 - k_3X_1{}^2Y_1,\\
\frac{dE_1}{dt} &= k_4X_1 - k_5E_1,\\
\frac{dX_2}{dt} &= k_1A_2 - k_2BX_2 - k_4X_2 + k_3X_2{}^2Y_2 + k_5E_2 + D_{12}(X_1 - X_2),\\
\frac{dY_2}{dt} &= k_2BX_2 - k_3X_2{}^2Y_2 + D_{22}(Y_1 - Y_2),\\
\frac{dE_2}{dt} &= k_4X_2 - k_5E_2 + D_{32}(E_1 - E_2),
\end{aligned} \quad (13)$$

The analytical solution of the obtained systems (12) and (13) is impossible. Therefore, we conducted a numerical study. The reaction rate constants in the numerical experiment are assumed to be equal to the following values: *A1 = 1; A2 = 1.2; D11 = 0.1; D21 = 0.1; D31 = 0.1; D12 = 0.05; D22 = 0.05; D32 = 0.05.* The initial conditions can be taken for *t = 0 X1 = X2 = Y1 = Y2 = E1 = E2 = 0*. Some results of the numerical experiment are shown in figures 9–12.

The analysis of the data of a numerical experiment showed that the concentration of the intermediate active complex Y arising during the reaction reaches a maximum value after the start of the process, and then decreases, trending to a stable value.

a)



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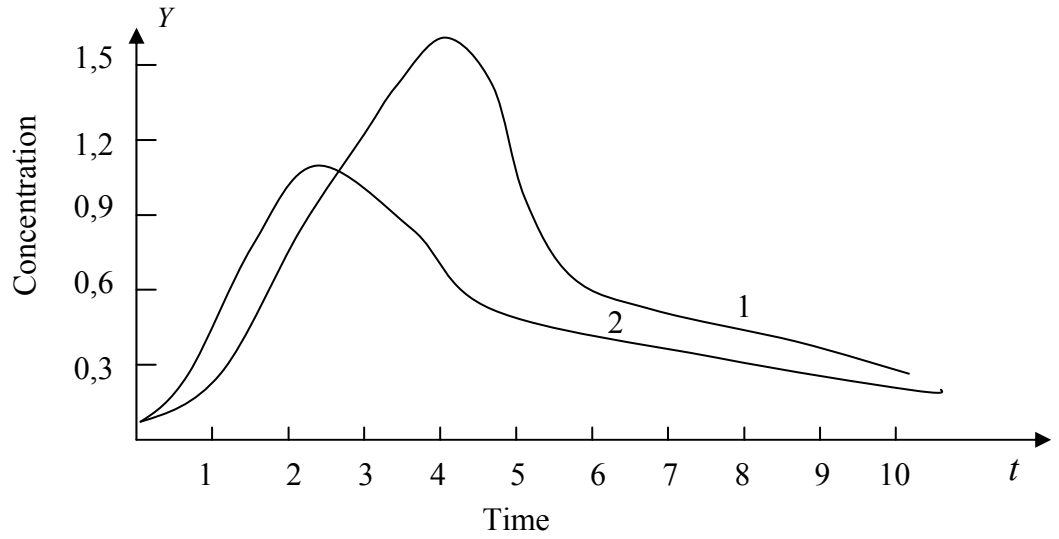

Figure 7 – The change in the concentration of intermediate reagents *X*- a) and *Y*- b)
in the cascade with recycle ("brusselator"):
1 – the first reactor of the cascade; 2 – the second reactor of the cascade

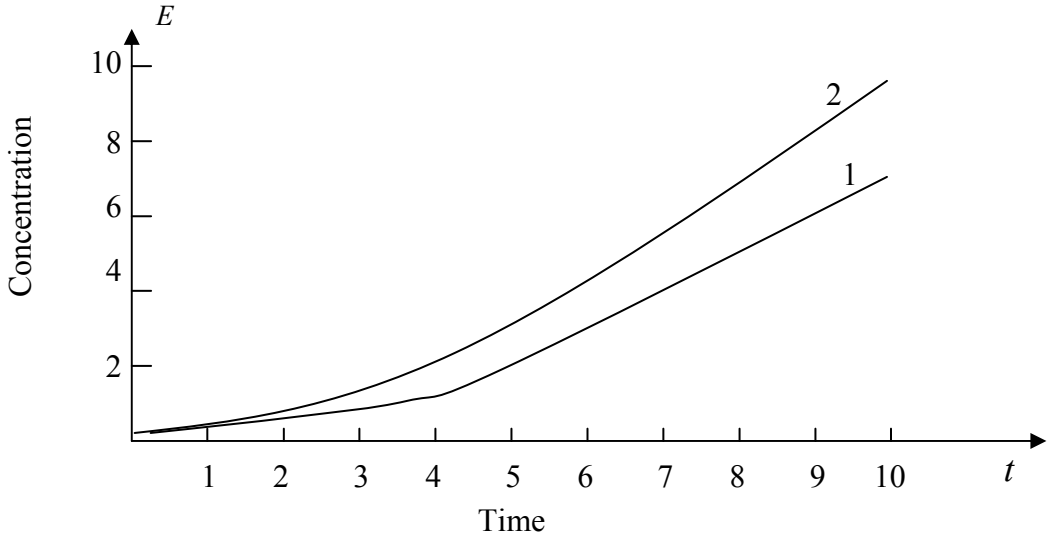

Figure 8 – The change in the concentration of the end-product *E* in the cascade reactors with recycle ("brusselator"): 1 – the first reactor of the cascade; 2 – the second reactor of the cascade

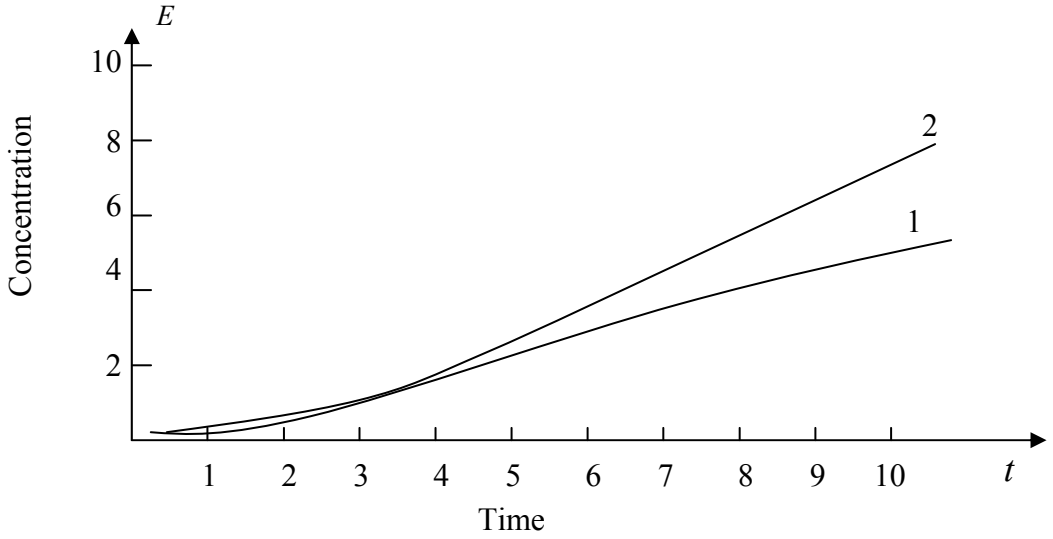

Figure 9 – The change in the concentration of the end-product E in the cascade reactors without recycle ("brusselator")

The dependencies of the concentration of the intermediate complex X and the final product E on time are monotonic and reach stable values after a long time. The partial decomposition of the final product in the case when the rate constant of the decomposition reaction is comparable with the constants of the other stages, can remove the oscillatory character of the concentrations of intermediate complexes characteristic to the "brusselator". A significant effect of recycling on the nature of the process was not found in the investigated range of parameters.

The results of the study show the importance of taking into account the reversibility of individual reaction stages in the analysis of the kinetics in cascades of chemical reactors.

3.2. Stages of modeling and calculation of cascades of through-flowing reactors. To obtain an approximate estimation of the necessary residence time of the reactants in the cascade of reactors operating in the dynamic regimes of the traveling concentration waves, we consider separately the cases of a model medium of the "Brusselator" type and a medium with three stationary states (to this case, a reaction-diffusion medium with the Belousov-Zhabotinsky system can be attributed).

For a model medium of the "Brusselator type", we use the expressions from [8] to characterize the concentration profile, since the numerical experiment confirms the qualitative agreement with such an approximation.

In estimating the minimum residence time, we take the waves front velocity to be equal to the critical propagation velocity of the traveling wave.

Then we can rewrite the mentioned expression in the form:

$$C \approx 3\left(C_{01} - \frac{R_2}{R_1}\right)\mathrm{sch}^2\left(\sqrt{\frac{(C_{01}R_1 - R_2)}{4D}}\left(L - v^*\tau\right)\right), \tag{14}$$

where L – a reactor length; τ –residence time.

We introduce the degree of conversion in the reactor as the main initial criterion for the calculation

$$\eta = \frac{C_0 - C}{C_0}\ . \tag{15}$$

Then (14) can be rewrite as follows:

$$\eta = 1 - \frac{3}{C_0}\left(C_{01} - \frac{R_2}{R_1}\right)\mathrm{sch}^2\left(\sqrt{\frac{(C_{01}R_1 - R_2)}{4D}}\left(L - v^*\tau\right)\right). \tag{16}$$

Let us denote $S = \sqrt{\frac{(C_{01}R_1 - R_2)}{4D}}\left(L - v^*\tau\right)$ and rewrite (16) in the form:

$$\eta = 1 - \frac{3}{C_0}\left(C_{01} - \frac{R_2}{R_1}\right)\mathrm{sch}^2(S) = 1 - \frac{3}{C_0}\left[\frac{2}{\exp(S) + \exp(-S)}\right]^2. \tag{17}$$

We now use the expansion of exponents in a Taylor series, restricting ourselves to a minimum estimation of the residence time by two terms of the expansion:

$$\exp(S) \approx 1 + S + \frac{S^2}{2};\ \ \exp(-S) \approx 1 - S + \frac{S^2}{2}\ .$$

After rearrangements the following expression linking the residence time in the reactor, its length and the given conversion can be obtained:

$$\tau \approx \left[L - \sqrt{\frac{D_{ef}}{C_0 k_1}\left(\frac{3}{\eta} - \frac{1}{g}\right)}\right] \Big/ v^*\ . \tag{18}$$

Here

$$g = \frac{R_2}{R_1 C_0}\ . \tag{19}$$

For a medium with three stationary states (or a reaction-diffusion medium with the Belousov-Zhabotinsky system), we use expression from [8] to estimate the necessary residence time of reagents in a dynamic mode, since numerical experiments and published data confirm the validity of such an approximation.

$$C = \frac{C_{01} + C_{02}}{2} + \frac{C_{01} - C_{02}}{2}\,\mathrm{th}\left(-\frac{1}{2}\left[\sqrt{\frac{k}{2D_{ef}}}(C_{01} - C_{02})\left(L - v^*t\right) + C(0)\right]\right). \tag{20}$$

Let us use the notation

$$S = -\frac{1}{2}\left[\sqrt{\frac{k}{2D_{ef}}}\left(C_{01} - C_{02}\right)\left(L - v^* t\right) + C(0)\right]. \quad (21)$$

Thus from the expression for hyperbolic tangent

$$\mathrm{th}(S) = \frac{\exp(S) - \exp(-S)}{\exp(S) + \exp(-S)}. \quad (22)$$

and Taylor expansions we can offer the following calculation scheme for optimal residence time:

A) Calculation of the subsidiary parameter **M**:

$$\mathbf{M} = C_{01} + C_{02} - 2C_0\left(1 - \eta\right). \quad (23)$$

B) Calculation of the subsidiary parameter **N** from the equation:

$$\mathrm{M} \cdot \mathrm{N}^2 + \mathrm{N}\left(C_{01} - C_{02}\right) + 2\mathrm{M} = 0. \quad (24)$$

C) Obtaining the estimation of the residence time:

$$\tau \approx \frac{C_0 + 2\mathrm{N}}{\alpha\left(C_{01} - C_{02}\right)v^*} + \frac{L}{v^*}. \quad (25)$$

For the cascade of reactors, the estimations of the necessary residence time in an individual diffusion cell for different systems with a lot of stationary states and self-oscillating dynamic regimes should be used in the general system of design equations, taking into account the known structure of the flows.

Conclusions. Thus, the following general scheme of simulation and calculation is proposed:

1. Choice of the model kinetic scheme.

For bromate systems and systems with oxidation-reduction reactions containing metal ions, the optimal model is the BZ type model.

For systems with organic reducing agents, the optimal model is the model of Belousov-Zhabotinsky type with allowing for the reversibility of the reaction stages.

For systems with enzymatic reactions and biochemical systems the optimal model is the model with autocatalysis or "Brusselator" type.

2. Modelling the flows structure and defining the number of cascades.

3. Analysis of the multiplicity of stationary states for each cascade.

4. Investigation of the stability of stationary states.

5. Analysis of conditions for the formation of wave regimes of mass transfer and estimation of the parameters of wave fronts.

6. Calculation of the conversions of reagents.

The calculation sequence is the same.

On the base of the criterial equations for the known flow structure in each of the cascade reactors, the required average flow rate of the phases in the section of the tubular reactor is calculated.

Then, at this average speed, the diameter of the apparatus can be determined for a given flow rate of the processed substance.

Then, at a given degree of conversion in the reactor and a certain average flow rate, the required residence time in the reactor is calculated according to the procedure described above.

If the length of the reactor is specified for design reasons or it is also specified, the calculated residence time is used to determine the required number of reactors in the cascade.

If the length of the reactor is not specified, it can be selected by iterative calculation according to the described procedure to ensure a given degree of conversion.

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РЕАКЦИЯНЫҢ ҚАЙТЫМЫН ЕСЕПКЕ АЛА ОТЫРЫП ХИМИЯЛЫҚ ОСЦИЛЛЯЦИЯМЕН АВТОКАТАЛИТИКАЛЫҚ ЖҮЙЕЛЕРДІ МОДЕЛЬДЕУ

Аннотация. Мақалада химиялық осцилляциямен екі автокаталитикалық жүйені модельдеу қарастырылады: Белоусов–Жаботинский реакциясы және «брюсселятор» жүйесі, қайтымды реакция жағдайында. Түрлі моделдік орта үшін химиялық реакторлар какскадында және келу уақытын анықтау әдістемесі жалпы тасымалдаудың динамикалық процестері үшін кинетикалық сипаттамалардың инженерлік есептеу сұлбасы ұсынылған. Тербелмелі химиялық реакциямен түрлі жүйелерді сипаттау үшін қолайлы модельдің сұлбасы орнатылған. Ионды металы бар броматты жүйе және тотығу – қалпына келтіру реакциясы үшін БЖ түріндегі модельді қолдану ұсынылады. Органикалық қалпына келтіру жүйесі үшін – Белоусов–Жаботинский жүйесі түріндегі модель реакциялардың қайтымды сатыларын есепке алу. Ферментативті реакциялар мен биохимиялық жүйелері үшін – автокатализ немесе «Брюсселятор» мен модель.

Әр каскад реакторынан белгілі ағындар құрылымында критериальды теңдеуден шығатыны келесі есептеу тізбегі. Бастапқыда құбырлы реактордың қимасындағы фазалар ағындарының қажетті орташа жылдамдығы есептеледі. Ары қарай осы орташа жылдамдықпен өңделетін субстанциялардың берілген шығыны бойынша аппарат диаметрі анықталады. Сонан соң берілген айналдыру дәрежесінде реакторды және жоғарыда аталған әдіс бойынша ағындардың анықталған орташа жылдамдығымен реактордағы қажетті келу уақыты есептеледі. Егер реактор ұзындығы конструктивті тұрғыдан берілсе, онда есептелген келу уақыты каскадтағы реакторлардың қажетті санын анықтау үшін қолданылады. Егер реактор ұзындығы берілмесе, онда ол берілген айналдыру дәрежесін қамтамасыз ету үшін сипатталған әдіс бойынша итерациялық есептеу жолымен таңдала алады.

Түйін сөздер: химиялық реакторларды модельдеу, екі сатылы реактор, Белоусов–Жаботинский реакциясы, «Брюсселятор» жүйесі.

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МОДЕЛИРОВАНИЕ АВТОКАТАЛИТИЧЕСКИХ СИСТЕМ С ХИМИЧЕСКИМИ ОСЦИЛЛЯЦИЯМИ С УЧЕТОМ ОБРАТИМОСТИ РЕАКЦИЙ

Аннотация. В статье рассматривается моделирование двух автокаталитических систем с химическими осцилляциями: реакция Белоусова-Жаботинского и система «брюсселятор» в случае обратимости реакций. Предложена схема инженерного расчета кинетических характеристик для динамических процессов массопереноса в каскадах химических реакторов и методика определения времени пребывания для различных модельных сред. Установлены предпочтительные модельные схемы для описания различных систем с колебательными химическими реакциями. Для броматных систем и систем с окислительно-восстановительными реакциями, содержащих ионы металлов, предлагается использовать модель типа БЖ. Для систем с органическими восстановителями – модель типа системы Белоусова–Жаботинского с учетом обратимости стадий реакций. Для систем с ферментативными реакциями и биохимических систем – модель с автокатализом или «Брюсселятор».

Исходя из критериальных уравнений при известной структуре потоков в каждом из реакторов каскада рекомендована следующая последовательность расчета. Вначале рассчитывается необходимая средняя скорость потоков фаз в сечении трубчатого реактора. Далее по этой средней скорости при заданном расходе

обрабатываемой субстанции определяется диаметр аппарата. Затем при заданной степени превращения в реакторе и определенной средней скорости потоков по описанной выше методике рассчитывается необходимое время пребывания в реакторе. Если длина реактора задается из конструктивных соображений, то рассчитанное время пребывания используется для определения необходимого числа реакторов в каскаде. Если же длина реактора не задана, то она может подбираться путем итерационного расчета по описанной методике для обеспечения заданной степени превращения.

Ключевые слова: моделирование химических реакторов, двухкаскадный реактор, реакция Белоусова–Жаботинского, система «брюсселятор».

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