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SYSTEM KONTROLI PROCESU ALKILOWANIA BENZENU PROPYLENEM W FAZIE CIEKŁEJ

Streszczenie: W artykule ustalono efekt zmiany stanu katalizatora i opracowano model matematyczny. Ustalano, że stosowanie optymalnego systemu jest niepraktyczne ze względu na występowanie nieprzewidywalnych zaburzeń. Stwierdzono, że stosowanie chlorku glinu powoduje reakcje uboczne, co rezultacie zmienia wartość mnożnika przedwykładniczego i energii aktywacji. Rozważano również syntezę pewnego regulatora H∞.

Słowa kluczowe: alkilacja, chlorek glinu, reakcje uboczne, mnożnik wstępny, energia aktywacji, pewny regulator

SYSTEM OF CONTROL OF THE PROCESS OF ALKYL-BENZEL OF PROPYLENE IN THE LIQUID PHASE

Summary. The effect of changing the state of the catalyst is established. A mathematical model has been developed. The use of an optimal system is impractical due to the available unpredictable perturbations. It was found that in the presence of aluminum chloride side effects occur. As a result, the value of the preexponential multiplier and the activation energy change. The synthesis of a robust regulator H∞ is considered.

Keywords: alkylation, aluminum chloride, side reactions, preexponential multiplier, activation energy, robust regulator

Introduction

Modern alkylation technologies are represented by a wide class of various industrial processes, which, despite all the differences, have a fundamental mechanism

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in common. Among the problems common to all alkylation processes (obtaining components of gasoline, ethylbenzene, cumene, linear alkylbenzenes (LAB)), we can note the presence of concomitant adverse reactions that lead to reduced process selectivity and deterioration of product quality, as well as application to most existing plants obsolete liquid acid catalysts, which leads to faster wear of equipment due to corrosion, as well as a high risk of production and the need to strictly adhere to the rules of technological regulations. At the same time, the transition of existing alkylation plants to modern solid catalysts is often economically impractical due to the high cost of production reconstruction. Because of this, refineries are currently facing the problem of increasing the efficiency of alkylate production.

1. Structural synthesis of robust systems

Alkylation is carried out in different ways and is divided into separate types. This is due to the use of various catalysts and acids. Despite the large number of solid phase catalysts, there is no process that would use them. The current state of the study in solid acid alkylation is determined by the chaos of the synthesized and already tested catalysts. They are different in nature, crystallinity, acidity range. However, the operating time of the catalyst for which it is characterized by efficient operation will vary slightly, from a few minutes to several hours. The only exception is the work of the IBONH NAS of Ukraine, the duration of which is many times longer, due to the flow-circulation regime.

Aluminum chloride catalyst was used in this work. The use of this catalyst leads to better yields compared to acid catalysts. Aluminum chloride promotes the condensation of such compounds that do not react under the influence of other agents. The use of $AlCl_3$ in the cracking process is based primarily on the ability of this reagent to break down long-chain hydrocarbons into smaller molecules, thereby increasing the yield of gasoline from oil.

Since, in the presence of chloraluminium catalytic complex, the realkylation reaction proceeds simultaneously with the alkylation, in the reactor together with benzene recycles the fraction of polyalkylbenzene isolated from the reaction mass during distillation, and the amount of olefin introduced is reduced accordingly. As a result of all these reactions, a well-defined equilibrium composition of the reaction products is established, which depends only on the ratio of alkyl radicals and benzene nuclei in the reaction mixture.

To control the process of alkylation of benzene with propylene in the liquid phase, a mathematical model was developed:

1) Material balance for the accumulating capacity of the reaction mass in the alkylator at molar concentration values:

$$-F_a \cdot x_a + A \cdot v \cdot \rho \cdot \frac{M_a}{M_p} k \cdot \exp\left(-\frac{E}{R \cdot \Theta}\right) \cdot x_{p1} = v \cdot \rho \frac{dx_a}{dt}. \quad (1)$$

2) Thermal balance of the reaction mass:

$$F_b \cdot c_b \cdot \Theta_b + F_p \cdot c_p \cdot \Theta_p - F_a \cdot c_a \cdot \Theta - K \cdot S_1 \left(\Theta - \frac{\Theta_v + \Theta_{v1}}{2} \right) -$$

$$- F_{b1} \cdot r + A \cdot S \cdot \rho \cdot \frac{M_a}{M_p} k \cdot \exp\left(- \frac{E}{R \cdot \Theta} \right) \cdot x_{p1} \cdot q = v \cdot \rho \cdot c_a \frac{d\Theta}{dt} \cdot \quad (2)$$

3) Heat balance by the temperature of the water supplied for cooling:

$$F_v \cdot c_v (\Theta_v - \Theta_{v1}) + KS_1 \left(\Theta - \frac{\Theta_{v1} + \Theta_v}{2} \right) = v_v \cdot \rho_v \cdot c_v \frac{d}{dt} \left(\frac{\Theta_v - \Theta_{v1}}{2} \right). \quad (3)$$

where F_b, F_{b1}, F_p, F_a, F_v, F_{v1} – the cost of benzene at the entrance to the device and evaporated; propylene; re-promotion products; water for cooling at the inlet and outlet, kg/s; k – preexponential multiplier; E – activation energy, kJ/mol; R – universal gas table, J/mol K; Θ_b, Θ_p, Θ, Θ_v, Θ_{v1} – temperatures of benzene and propylene; vapor-gas mixture; the reaction mass in the reactor; water at the inlet and outlet, K; x_b, x_p, x_a, x_{p1}, – concentrations of benzene and propylene at the inlet and alkylate at the outlet, propane-propylene fraction at the outlet, %; M_a, M_p – molar mass of alkylate and propylene, kg/mmol; q – specific heat of reaction, J/kg; c_v, c_a – specific heat of water and alkylate, J/(kg·K), r – specific heat of benzene vaporization, J/kg; ρ, ρ_v – density in the alkylator; water, kg/m^3; v, v_v – volume in the device; water, m^3; KS_1 – product of heat transfer coefficient per alkylator area, J / (s·K); S – heat transfer area,m^2; A – coefficient that takes into account the change in the state of the catalyst, range 0..1.

An optimal control system was developed for this model. Based on solving the Riccatti matrix differential equation and finding the optimal control. But the experience of using optimal systems according to the quadratic quality criterion has shown their sensitivity to the process parameters of a real object. Such systems turned out to be rude, which leads to their loss not only of optimality, but also of stability and quality. Therefore, it should be borne in mind that the system is affected by unpredictable disturbances.

The specific conductivity of the catalyst characterizes its activity, which is an important indicator for the process, because the technology of alkylation of benzene with propylene is built in such a way that the catalyst, which did not react, returns back to the process cycle.

As a result of processing a large array of experimental data, it was found that the maintenance of the activity of the returned catalyst complex is regulated by the consumption of fresh catalyst and as a result depends on the consumption of the returned catalyst, its activity and the activity of fresh catalyst complex in this way. As a result, there is a need for a parameter that takes into account changes in the state of the catalyst.

In this work, the actual values of the specific conductivity of the catalyst were normalized in the range from 0 to 1. Thus, the activity of fresh catalyst varies in the range from 0.27 to 1.00, and the activity of the rotary catalyst from 0.02 to 0.48.

It is established that in the presence of aluminum chloride $AlCl_3$ in the composition of the catalytic complex can be formed triple complexes not only with one, but also with two, three and so on hydrocarbon radicals. Complexes can enter into exchange reactions not only with benzene, but also with reaction products, for example, with dipropylbenzene, then there is a process of pe-realkylation and a significant number

of side reactions. As a result of these reactions, the value of the preexponential factor and the activation energy in the Arrhenius equation change. A mathematical model of the process with variable coefficients that take into account uncertainties is presented:

$$\frac{\partial x_a}{dt} = \left(-1{,}1 \cdot 10^{-26} \cdot e^{\frac{E}{8{,}31 \cdot \Theta}} \left(6{,}8 \cdot 10^{43} \cdot A \cdot k \cdot e^{-\frac{E}{8.31 \cdot \Theta}} + 5{,}43 \cdot A \cdot k^2 \times \right.\right.$$

$$\left.\left. \times e^{-\frac{2 \cdot E}{8.31{,}\Theta}} + 2{,}2 \cdot 10^{43} \right)^{1/2} - 7{,}1 \cdot 10^{-4} \cdot e^{\frac{E}{8{,}31 \cdot \Theta}} - 1{,}4 \cdot 10^{-4} \cdot A \cdot k^2 \right) \cdot x_a +$$

$$+ \left(-\frac{105 \cdot A \cdot k}{\Theta^2} \right) \cdot \Theta; \tag{4}$$

$$\frac{\partial \Theta}{dt} = \left(3{,}6 \cdot 10^{19} \cdot A \cdot k \cdot e^{-\frac{E}{8{,}1 \cdot \Theta}} + 2{,}4 \cdot 10^{20} \cdot A \cdot k^2 \cdot e^{-\frac{2 \cdot E}{8{,}1 \cdot \Theta}} - \right.$$

$$\left. -3{,}3 \cdot 10^{16} \cdot \Theta \cdot A \cdot k \cdot e^{-\frac{E}{8{,}31 \cdot \Theta}} + 4{,}7 \cdot 10^{17} \right)^{1/2} \left(-1{,}2 \cdot 10^{-14} e^{\frac{E}{8{,}31 \cdot \Theta}} - \right.$$

$$\left. -1{,}9 \cdot 10^{-4} \cdot A \cdot k - 8{,}6 \cdot 10^{-6} \cdot e^{\frac{E}{8{,}31 \cdot \Theta}} \right) \cdot x_a + \left(\frac{109{,}4 \cdot A \cdot k - 510{,}3}{\Theta^2} \right) \times$$

$$\times \Theta + 2{,}8 \cdot 10^{-3} \cdot \Theta_v + \left(0{,}081 - 1{,}995 \cdot 10^{-4} \cdot \Theta \right) \cdot F_p; \tag{5}$$

$$\frac{\partial \Theta_v}{dt} = -5{,}82 \cdot 10^{-3} \cdot \Theta + \left(-6{,}81 \cdot 10^{-5} \cdot \Theta + 4{,}75 \cdot 10^{-3} \right) \cdot \Theta_v. \tag{6}$$

The change in the properties of the catalyst and the presence of side reactions affect the rate of the alkylation reaction of benzene.

Analysis of the effect of catalyst uncertainties on the change in alkylate concentration showed that the uncertain parameters are the change in catalyst activity, the value of the redexpension factor and the activation energy in the Arrhenius equation, which significantly affect the quality of the process.

2. Minimization of H∞-norm

For clarity, we present a mathematical model of the process in the state space:

$$\begin{cases} \dfrac{d}{dt}\mathbf{x} = \mathbf{A}\cdot\mathbf{x} + \mathbf{B_1}\cdot\mathbf{w} + \mathbf{B_2}\cdot F_p, \\ \mathbf{z} = \mathbf{C_1}\cdot\mathbf{x} + \mathbf{D}_{12}\cdot F_p, \\ \Delta x_a = \mathbf{C_2}\cdot\mathbf{x} + \mathbf{D}_{21}\cdot\mathbf{w}, \end{cases} \tag{7}$$

where **A**, **B**, **C**, **D**, – object state space matrices;
z and **w** uncertainty outputs and inputs, respectively;

In this model, the adjustable value is the concentration of alkylate at the outlet of the apparatus – x_a, and the controlled effect is the consumption of propane-propylene fraction – F_p.

We present a mathematical model of the alkylation plant (7) in the operator form, which is the most convenient for the synthesis of robust control:

$$\Delta x_a = F_U\left(\mathbf{G}, \mathbf{\Delta}\right) F_p \tag{8}$$

where F_U – operator of the upper fractional-linear transformation (FLT);
G – process model;
Δ – uncertainty matrix.

Let's rewrite the model of alkylation installation in operator form using the following FLT:

$$\mathbf{z} = F_L\left(\mathbf{P}, K\right)\mathbf{w} \tag{9}$$

where F_L – lower operator FLT;
P – generalized mathematical model;
K – regulator.

On the other hand (9) has the form

$$\mathbf{z}(s) = \mathbf{T}_{\mathbf{z}\to\mathbf{w}}(s)\cdot\mathbf{w}(s), \tag{10}$$

where the matrix of transfer functions of the whole generalized system is defined by the expression:

$$\mathbf{T}_{\mathbf{z}\to\mathbf{w}}(s) = \left[\mathbf{I} + K(s)\cdot\mathbf{G}(s)\right]^{-1}\begin{bmatrix} W_e(s) \\ K(s)\cdot W_S(s) \\ K(s)\cdot\mathbf{G}(s)\cdot W_T(s) \end{bmatrix}, \tag{11}$$

where I – single matrix;
$W_e(s)$, $W_S(s)$, $W_T(s)$ – weight transfer functions by error e, controlling influence S and the original coordinate y, respectively.

Rewrite (5) in the form:

$$\begin{bmatrix} \mathrm{z}_e(s) \\ \mathrm{z}_{F_p}(s) \\ \mathrm{z}_{\Delta x_a}(s) \end{bmatrix} = \begin{bmatrix} \mathrm{W}_e(s)\cdot\mathbf{S}(s) \\ \mathrm{W}_S(s)\cdot\mathbf{R}(s) \\ \mathrm{W}_T(s)\cdot\mathbf{T}(s) \end{bmatrix}\cdot\mathbf{w}(s). \tag{12}$$

Equations (12) include the following matrices of transfer functions:
- from the input of process uncertainties $\mathbf{w}(t)$ to tracking error $z_e(t)$ is a function of sensitivity:

$$\mathbf{S}(s)=\left[\mathbf{I}+\mathbf{G}(s)\cdot K(s)\right]^{-1}; \tag{13}$$

- from the input of process uncertainties $\mathbf{w}(t)$ to control the flow of propane-propylene fraction $z_{Fp}(t)$ is a function of control sensitivity:

$$\mathbf{R}(s)=K(s)\left[\mathbf{I}+\mathbf{G}(s)\cdot K(s)\right]^{-1}; \tag{14}$$

- from the input of process uncertainties $\mathbf{w}(t)$ to change the concentration of alkylate at the outlet of the alkylator $z_{\Delta xa}(t)$ is a function of additional sensitivity:

$$\mathbf{T}(s)=K(s)\cdot\mathbf{G}(s)\cdot\left[\mathbf{I}+\mathbf{G}(s)\cdot K(s)\right]^{-1}. \tag{15}$$

Therefore, the matrix transfer function (15) establishes a relationship between the change in the concentration of alkylate at the outlet of the alkylator and its desired value. The matrix transfer function (13) describes the change in the concentration of alkylate at the exit of the alkylator as a function of the input of process uncertainties. From the definition of the transfer functions of sensitivity and additional sensitivity it is known that:

$$\mathbf{S}(s)+\mathbf{T}(s)=\mathbf{I}. \tag{16}$$

Sensitivity functions together with weight functions are widely used in H_∞-theory. For the alkylation process, it is important to take into account the process uncertainties that appear over the life of the catalyst. Taking into account (16), you can write the following quality criteria in the form of inequalities:
- to track errors:

$$\left\|\mathrm{W}_e(s)\cdot\mathbf{S}(s)\right\|_\infty < 1; \tag{17}$$

- to change the concentration of alkylate at the outlet of the alkylator:

$$\left\|\mathrm{W}_T(s)\cdot\mathbf{T}(s)\right\|_\infty < 1. \tag{18}$$

- to reduce control costs, the transfer function of the control effect is used $\mathbf{R}(s)$ with the appropriate weighting factor $W_S(s)$ to limit the amplitudes of the output signal:

$$\left\|\mathrm{W}(s)\cdot\mathbf{R}(s)\right\| < 1. \tag{19}$$

In this case, to minimize the sensitivity of the control system, it is necessary to maintain a small value of the control signal so that the monitoring system is not saturated, does not amplify the sensor noise and does not mix the disturbance signal with the control signal.

Weight transfer functions are used to provide the necessary quality characteristics of transients for system errors and its regulatory effects. To compensate for disturbances, it is necessary to have a small error **e** in the low frequency range, and to ensure the stability and suppression of high-frequency interference, it is desirable to have a small output value **y** in the high frequency range. For this error **e** in the low frequency range you need to "weigh" with more weight than at high frequencies, ie the amplitude of the frequency response $W_S(s)$ should decrease with increasing frequency ($W_S(s)$ – low pass filter). On the contrary, the amplitude of the frequency response $W_T(s)$ should increase ($W_T(s)$ – high frequency filter). It should be noted that there is no unambiguous algorithm for selecting weight functions, in fact it all comes down to trial and error. Weight functions were found by expressions:

$$W_e(s) = \frac{s/M + \omega_0}{s + \omega_0 \cdot A}, \tag{20}$$

$$W_S(s) = const, \tag{21}$$

$$W_T(s) = \frac{s + \omega_0/M}{A \cdot s + \omega_0}, \tag{22}$$

where A – the desired permissible error in the steady state; ω_0 – desired bandwidth; M – peak sensitivity.

3. Synthesis of H_∞-regulator

The H_∞ synthesis procedure involves sequentially solving two Riccati equations for the regulator and the observer. The synthesis of the H_∞ regulator takes place using an

iterative procedure, which consists in finding the smallest parameter $\gamma > 0$. At the beginning of the search is set $\gamma > 1$ two algebraic Riccati equations are solved.
For the regulator:

$$\mathbf{A}^T \cdot \mathbf{x} + \mathbf{x} \cdot \mathbf{A} + \mathbf{C}_1^T \cdot \mathbf{C}_1 - \mathbf{x} \cdot \left(\mathbf{B}_2 \cdot \mathbf{B}_2^T - \frac{1}{\gamma^2} \mathbf{B}_1 \cdot \mathbf{B}_1^T \right) \cdot \mathbf{x} = 0 , \tag{23}$$

and an observer:

$$\mathbf{A} \cdot \mathbf{y} + \mathbf{y} \cdot \mathbf{A}^T + \mathbf{B}_1 \cdot \mathbf{B}_1^T - \mathbf{y} \cdot \left(\mathbf{C}_2^T \cdot \mathbf{C}_2 - \frac{1}{\gamma^2} \cdot \mathbf{C}_1^T \cdot \mathbf{C}_1 \right) \cdot \mathbf{y} = 0 . \tag{24}$$

The assessment of the condition is as follows:

$$\frac{d}{dt} \hat{\mathbf{x}} = \left(\mathbf{A} - \frac{1}{\gamma^2} \cdot \mathbf{B}_1 \cdot \mathbf{B}_1^T \cdot \mathbf{x} \right) \cdot \hat{\mathbf{x}} + \mathbf{B}_2 \cdot F_p + \mathbf{Z} \cdot \mathbf{L} \cdot \left(\mathbf{C}_2 \cdot \hat{\mathbf{x}} - \mathbf{y} \right). \tag{25}$$

where substitutions

$$\mathbf{L} = \mathbf{y}^* \cdot \mathbf{C}_2^T ; \ \mathbf{Z} = \left(\mathbf{I} - \frac{1}{\gamma^2} \cdot \mathbf{y}^* \cdot \mathbf{x}^* \right)^{-1} . \tag{26}$$

where $\mathbf{Z}$ – vector used to assess quality;
$\mathbf{L}$ – matrix of observer gain.
'*' the solutions of the equations are marked (23) and (24).

The search continues until the minimum value of γ is found which can solve 2 Riccati equations, satisfy the quality criteria and conditions:

$$\begin{cases} \mathrm{Re}\left\{ \lambda_i \cdot \left[\mathbf{A} + \mathbf{y} \cdot \left(\frac{1}{\gamma^2} \cdot \mathbf{C}_1 \cdot \mathbf{C}_1^T - \mathbf{C}_2^T \cdot \mathbf{C}_2 \right) \right] \right\} < 0, \quad \forall i, \\ \mathrm{Re}\left\{ \lambda_j \cdot \left[\mathbf{A} + \left(\frac{1}{\gamma^2} \cdot \mathbf{B}_1 \cdot \mathbf{B}_1^T - \mathbf{B}_2 \cdot \mathbf{B}_2^T \right) \cdot \mathbf{x} \right] \right\} < 0, \quad \forall j, \\ \rho\left(\mathbf{x}^* \cdot \mathbf{y}^* \right) < \gamma^2, \end{cases} \tag{27}$$

where λ – own values of the system;

ρ – spectral radius.
The suboptimal feedback is defined by the expression:

$$\mathbf{K} = \left[\begin{array}{c|c} \mathbf{A}_\infty & -\mathbf{Z}\cdot\mathbf{L} \\ \hline \mathbf{F} & \mathbf{0} \end{array}\right], \tag{28}$$

where substitutions

$$\mathbf{A}_\infty = \mathbf{A} + \frac{1}{\gamma^2}\cdot\mathbf{B}_1\cdot\mathbf{B}_1^T\cdot\mathbf{x}^* + \mathbf{B}_2\cdot\mathbf{F} + \mathbf{Z}\cdot\mathbf{L}\cdot\mathbf{C}_2, \quad \mathbf{F} = -\mathbf{B}_2\cdot\mathbf{x}^*. \tag{29}$$

where **F –** matrix of gain of the regulator.

4. Analysis of results

After 9-iterations, the H∞ regulator of the 5th order with the final value is obtained γ = 0,9863 (initial value γ = 1.5698). The transfer function of the regulator has the form:

$$K(s) = \frac{-43,59\cdot s^4 - 5,379\cdot s^3 - 0,1064\cdot s^2 - 0,0004508\cdot s - 3,23e-07}{s^5 + 0,6981\cdot s^4 + 0,14\cdot s^3 + 0,01212\cdot s^2 + 0,0001868\cdot s + 4,595e-08}. \tag{30}$$

The frequency characteristics of a closed system are presented in Fig. 1.

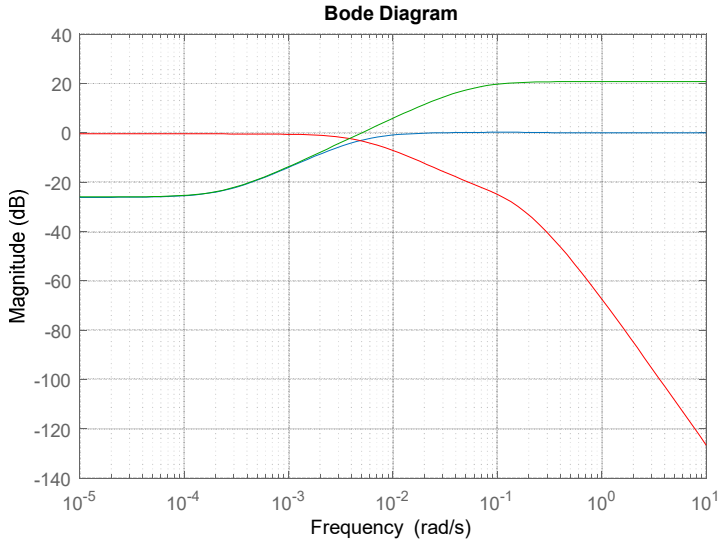

Figure 1. Amplitude-frequency characteristic

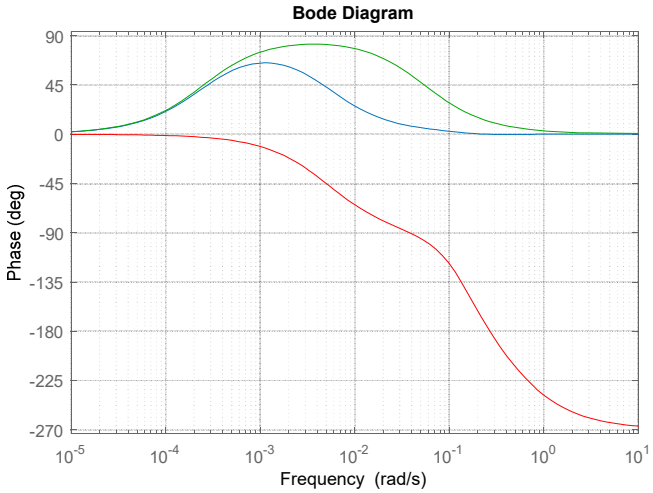

Figure 2. Phase-frequency characteristics

For one-dimensional systems H_∞, the norm is the maximum value of the ordinate of the amplitude characteristic of the closed system at the initial ordinate equal to one, ie the relative height of the resonant peak.

The smaller the margin of stability, the greater the susceptibility of the system to oscillations and the greater the resonant peak. The smaller the resonant peak, the higher the robustness of the system.

Transients through the channel "consumption of propane-propylene fraction - the concentration of alkylate at the outlet of the alkylator" are presented in Fig. 3:

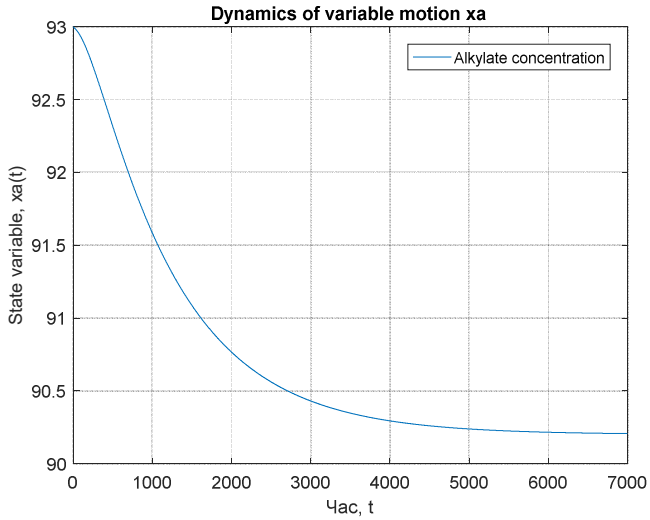

Figure 3. Transition characteristic on the channel "consumption of propane-propylene fraction - the concentration of alkylate at the outlet of the alkylator"

From fig. 3. it is seen that a closed system in the absence of uncertainties has an aperiodic nature of the transition process. The transition time is slightly more than 6000 s, which is considered normal for the alkylation process. For comparison, the graphs in Fig. 3, which take into account the effect of changes in the pre-exponential multiplier and the activation energy (at k = 1, k = 0.75, k = 0.5).

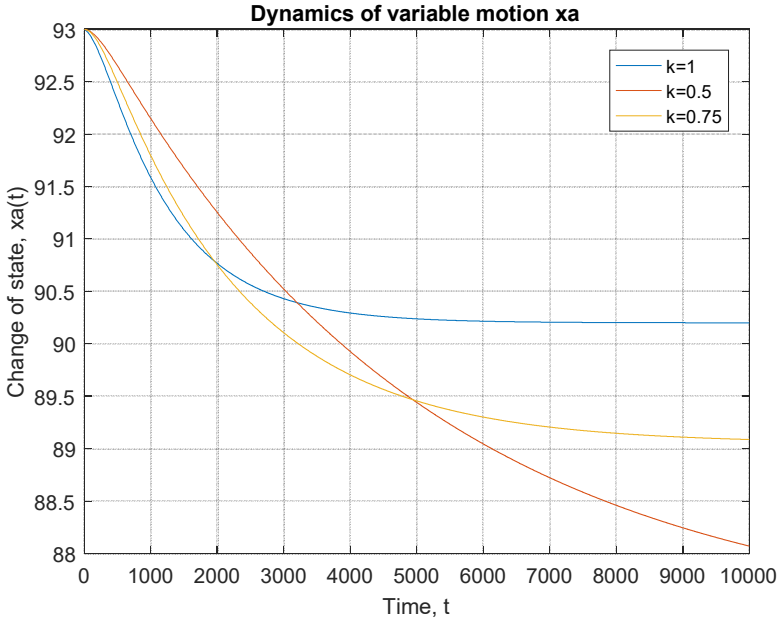

Figure 4. Comparison of transient characteristics at different values of the preexponential factor and activation energy

From the graph you can see that with a larger value of k, the concentration has a better value, and with a smaller value – worse. At k = 0.5, k=0.75 the concentration is less than 90%, and this is not a quality product. Because, the concentration of alkylate is in the range from 90% to 95% for the product to be high quality. It is the regulator that helps us maintain this at the right level.

Conclusion

When alkylating aromatic hydrocarbons (benzene, toluene, etc.), aluminum chloride is of the greatest industrial importance, having a number of significant advantages over other catalysts.

It is established that at a larger value of k the transient characteristic has better indicators. The analysis of the influence of the uncertainty of the parameters of the mathematical model of the process is carried out. The synthesis of a robust regulator is considered, which ensures the operation of the apparatus in the conditions of changing the properties of the catalyst and side reactions during operation. Maintaining a given concentration of alkylbenzene at the outlet is ensured by changing the flow rate of propylene.

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