

Theoretical basis of chemical technology
Теоретические основы химической технологии

UDC 66.011

<https://doi.org/10.32362/2410-6593-2026-21-2-143-156>

EDN VSRQYI



RESEARCH ARTICLE

Steady state analysis of the flow continuous stirred tank reactor on instance exothermic dimerization reaction

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Abstract

Objectives. Due to the complexity of their behavior, chemical process flowsheets are characterized by steady state multiplicity, in other words, the presence of multiple steady state operating modes having the same set of parameters. The steady states differ from each other in terms of their reagent conversion, selectivity, product flow composition, and stability. Therefore, in order to be able to identify the steady state having optimal technological indicators, the objective of searching all steady states of a chemical process flowsheet is relevant. The aim of the study is to research all possible steady states for a continuous stirred tank reactor (CTSR) according to the exothermic dimerization reaction and investigate the influence of different operation parameters on the technological indicators of found states.

Methods. Mathematical simulations of material and energy balance equations for reactor were used. The quantity of steady states was estimated by the number of energy balance discrepancy function intersection with the Ox axis. The Newton method in Microsoft Excel was used to solve nonlinear material balance equations of the reactor. The initial value of productivity was in range from zero to feed rate value of reagent of 100 kmol/h.

Results. It is shown that up to three steady states may exist for the reactor in dependence on the reaction volume, composition, and temperature feed flow, as well as the heat carrier flow rate. The results of this study correspond with earlier obtained results, which were obtained for irreversible reactions in adiabatic conditions. These states differ in productivity, internal reactor temperature, and stability. Steady state stability analysis of small parameter deviations was carried out. The analysis demonstrated that real characteristic values are in all found steady states of the reactor. Therefore, no oscillations in stable steady states of reactor and asymptotical operating time dependencies are implemented.

Conclusions. The technique of steady state analysis of a continuous stirred tank reactor developed over the course of this study, which reveals all steady states of the reactor with heat exchange, can be used to perform steady state analysis of recycled chemical process flowsheets, including continuous stirred tank reactors and separation blocks.

Keywords

reactors, exothermic reactions, steady states, steady state stability, mathematical simulation

Submitted: 27.07.2025

Revised: 09.09.2025

Accepted: 17.02.2026

For citation

Korol'kova N.A., Nazanskii S.L., Solokhin M.A. Steady state analysis of the flow continuous stirred tank reactor on instance exothermic dimerization reaction. *Tonk. Khim. Tekhnol. = Fine Chem. Technol.* 2026;21(2):143–156. <https://doi.org/10.32362/2410-6593-2026-21-2-143-156>

НАУЧНАЯ СТАТЬЯ

Анализ стационарных состояний проточного реактора идеального смешения на примере экзотермической реакции димеризации

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Аннотация

Цели. Химико-технологические системы в силу сложности своего поведения во многих случаях характеризуются полистационарностью, то есть наличием множественных стационарных режимов работы при одном и том же наборе рабочих параметров. Данные стационарные состояния отличаются конверсией, селективностью, составами продуктовых потоков и устойчивостью. В связи с этим, актуальной задачей является выявление всех возможных стационарных состояний химико-технологических систем для того, чтобы в дальнейшем была возможность выбора состояния с наилучшими технологическими показателями. Цель работы — поиск всех возможных стационарных состояний проточного реактора идеального смешения на примере реакции димеризации и анализ влияния различных параметров на технологические показатели реактора для найденных состояний.

Методы. Работа выполнялась методом математического моделирования с использованием материальных балансов по веществам и энергетического баланса реактора. Количество стационарных состояний оценивалось по числу точек пересечения функции невязки энергетического баланса с осью Ox . Нелинейные алгебраические уравнения материального баланса реактора решались методом Ньютона в среде Microsoft Excel. Начальное приближение по производительности выбиралось в диапазоне от нуля до количества реагента 100 кмоль/ч в питании реактора.

Результаты. В ходе работы установлено, что в зависимости от объема реактора, температуры и состава входящего потока и расхода теплоносителя в проточном реакторе идеального смешения может реализовываться до трех стационарных состояний, которые отличаются производительностью реактора и температурой в нем. Результаты настоящей работы согласуются с литературными данными, полученными ранее для случаев необратимых реакций, протекающих в адиабатическом режиме. Кроме этого, проведен анализ устойчивости стационарных состояний при малых изменениях параметров, который показал, что характеристические корни в стационарных состояниях действительны, следовательно, колебания при работе реактора не реализуются, и в окрестности устойчивого состояния характер изменения параметров асимптотический.

Выводы. Разработана методика, которая позволяет выявить все возможные стационарные состояния проточного реактора идеального смешения с внешним теплообменом. Методику, примененную в данной работе, можно использовать для анализа стационарных состояний рециркуляционных химико-технологических систем, включающих реактор идеального смешения и блок разделения.

Ключевые слова

реакторы, экзотермические реакции, стационарные состояния, устойчивость стационарных состояний, математическое моделирование

Поступила: 27.07.2025

Доработана: 09.09.2025

Принята в печать: 17.02.2026

Для цитирования

Королькова Н.А., Назанский С.Л., Солохин М.А. Анализ стационарных состояний проточного реактора идеального смешения на примере экзотермической реакции димеризации. *Тонкие химические технологии*. 2026;21(2):143–156. <https://doi.org/10.32362/2410-6593-2026-21-2-143-156>

INTRODUCTION

Any chemicotechnological system, regardless of its scale and product range, consists of a sequence of technological processes linked by direct and reverse connections. These processes include receipt and preparation of raw materials, chemical conversions, separation of the reaction mixture to obtain the target product, and formation of recirculation flows for unreacted raw materials. Experience in operating chemical plants shows that the main contribution to the economic efficiency criterion is made by the costs of chemical conversion processes (reactor subsystem) and reaction mass separation (separation subsystem) [1]. In addition to general economic indicators, an assessment of the efficiency of individual stages of the process also includes efficiency criteria that more fully reflect the chemical and physicochemical phenomena occurring in individual units of the technological system. One of the most important quantitative criteria reflecting the efficiency of a chemical reactor is the specific productivity and selectivity of the process.

In the case of kinetically reversible reactions, the overall reaction rate decreases as the chemical transformation proceeds due to an increase in the rate of reverse processes; as a result, a state of equilibrium is eventually achieved. Such a chemical equilibrium state comprises the main thermodynamic limitation that prevents the complete conversion of reactants into products in a single pass through a chemical reactor [2, 3]. In practice, many reactions require a long contact time and large reactor volume to achieve chemical equilibrium due to kinetic difficulties (low reaction rate). Under industrial conditions, the process should be carried out in such a way as to prevent the achievement of an equilibrium state of the chemical reaction, which would result in a lower yield of products. These kinetic difficulties and thermodynamic limitations of the process necessitate the use of chemicotechnological methods to overcome these difficulties. One such chemicotechnological method involves the organization of a recirculation flow of unreacted raw materials into the reactor. This reduces the residence time of the reagents in the reaction zone and consequent conversion in the reactor, which leads to an increase in the concentration of the reagents and corresponding chemical reaction rate, resulting in improved productivity [4]. However, in order to organize the recirculation flow, the chemical reactor must be supplemented with another separation unit, in which unreacted raw materials and reaction products will be

separated. Using such an independent recirculation system, which is inherently more complex than a basic chemical reactor, it is possible to achieve a conversion higher than chemical equilibrium due to the absence of thermodynamic limitations on the maximum conversion value. In the case of reversible chemical reactions, recirculation can be used to achieve a degree of raw material conversion that exceeds the maximum permissible value for any type of reactor up to a theoretical maximum of 100% conversion. Thus, despite a low degree of reagent conversion in a single pass, the use of recirculation significantly increases a reactor's productivity. Similarly, with the proper organization of recirculation flows, it is possible to reduce the rate of side reactions to increase the overall selectivity of the process [5].

The presence of recirculating flows in the system causes feedback, which can lead to the emergence of multiple steady states under the same operating parameters of the system¹ [6]. In certain cases, steady state continua can be realized in recirculating reaction-rectification processes, which however disappear when transitioning from infinite separation capacity to a finite number of plates [7, 8]. In this regard, it is relevant to study the evolution of steady states of a given chemicotechnological scheme subject to changing design and operating parameters. Furthermore, in the presence of multiple steady states, the task of analyzing the stability of these states arises. The solution of this task in turn determines the strategy for starting the chemicotechnological process and the ability to maintain the operating parameters set for the desired steady state.

In view of the above, the present study set out to analyze the steady states of the reactor, taking into account heat exchange and changes in design and operating parameters, as well as the composition of the feed stream.

MATHEMATICAL MODEL OF THE REACTOR

In the first stage of the computational part of the study, research was conducted of the steady states of a continuous stirred tank reactor (CSTR) in which a liquid-phase reversible exothermic dimerization reaction $2A \leftrightarrow B$ takes place. The reaction was considered hypothetical, with the thermophysical properties of the components (heat capacity, density) used for the reaction under study corresponding to acetone (A) and diacetone alcohol (B).

¹ Blagov S.A. *Razrabotka metoda analiza statsionarnykh sostoyanii retsirkulyatsionnykh reaktsionno-rektifikatsionnykh processov (Development of a method for analyzing stationary states of recirculation reaction-distillation processes)*. Cand. Sci. Thesis. (Eng.). Moscow: MITHT; 1999, 195 p.

Figure 1 shows a flow diagram of the CSTR for this process.

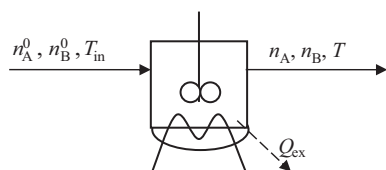


Fig. 1. Flow diagram of CSTR (all designations are given at the end of the article)

The physicochemical properties of the reaction mixture components, such as heat capacity, were taken from reference literature [9]. The thermal effect of the reaction under consideration was calculated taking into account the change in the heat capacity of the reaction mixture [10]:

$$\Delta H_T = \Delta H_{298.15}^0 + \Delta C_p (T - 298.15), \quad (1)$$

where ΔH_T is the heat effect of the reaction, kJ/kmol; $\Delta H_{298.15}^0$ is the standard heat effect of the reaction at $T = 298.15$ K, kJ/kmol; ΔC_p is the change in the heat capacity of the reaction during the conversion of acetone to diacetone alcohol, kJ/kmol·K; T is the absolute temperature, K.

The standard heat effect of the reaction at $T = 298.15$ K was calculated based on the values of standard heat effects of substance formation available in the NIST TDE² Aspen Plus database and is equal to -95000 kJ/kmol.

The change in heat capacity during a chemical reaction is calculated using the formula:

$$\Delta C_p = C_{pB} - C_{pA}, \quad (2)$$

where C_{pA} and C_{pB} are heat capacities of components A and B, respectively, in the liquid state, kJ/(kmol·K).

The dependence of heat capacity on temperature is expressed by a power series:

$$C_p = a + bT + cT^2 + dT^3, \quad (3)$$

The coefficients a , b , c , d of Eq. (3) are presented in Table 1.

Table 1. Coefficients of the temperature dependence (3) of heat capacity for components A and B

Component	a	b	c	d
A	135.6	-0.177	$2.837 \cdot 10^{-4}$	$6.890 \cdot 10^{-7}$
B	59.7	0.542	0.00	0.00

For the dimerization process, it was assumed that the chemical reaction rate obeys the law of active masses and, thus, the reaction rate W has the form [12]:

$$W = k^+ \cdot C_A^2 - k^- \cdot C_B, \quad (4)$$

where k^+ ($\text{m}^3/(\text{kmol} \cdot \text{h})$) and k^- (h^{-1}) are rate constants for the forward and reverse reactions, respectively; C_A and C_B (kmol/m^3) are concentrations of components A and B, respectively.

The dependence of reaction rate on temperature obeys the Arrhenius law, according to which the following expression is valid for the reaction rate constant [11]:

$$k = A e^{-\frac{E_a}{RT}}, \quad (5)$$

where A is the pre-exponential factor; E_a is activation energy, J/mol; R is the universal gas constant, J/(mol·K); T is absolute temperature, K.

At the preliminary calculation stage, velocity constant values were selected to ensure a sufficiently high velocity in the range of 300–400 K. The parameters of the temperature dependence of the constants (pre-exponential factors A and activation energies E) are given in Table 2.

Table 2. Arrhenius law parameters

A^+	E^+	A^-	E^-
$5.00 \cdot 10^9$	60000	$2.67 \cdot 10^{10}$	63200

The mathematical model of the CSTR shown in Fig. 1 consists of material balance equations:

$$n_A^0 - n_A - 2P = 0, \quad (6)$$

$$n_B^0 - n_B + P = 0, \quad (7)$$

where are feed rates of raw material into the reactor, kmol/h; n_A and n_B are product flow rates, kmol/h; P is reactor performance, kmol/h.

Reactor productivity depends on the volume V of the CSTR and the chemical reaction rate W . Thus, using formula (4) for W , we obtain the kinetic equation for productivity [12]:

$$P = W \cdot V = (k^+ \cdot C_A^2 - k^- \cdot C_B) \cdot V, \quad (8)$$

where V is a reactor volume, m^3 ; k^+ and k^- are constants of the forward and reverse reaction rates, respectively.

In order to reduce the problem to a single unknown parameter, we will relate the molar concentrations in the reactor to the molar fractions and molar flows using the molar volume of the mixture.

² NIST Standard Reference Data (SRD); © 2022 by the U.S. Secretary of Commerce on behalf of the United States of America.

Let us assume that the molar volume of the mixture obeys Amaga's law [9]:

$$v = v_A x_A + v_B x_B, \quad (9)$$

where v_A and v_B are the molar volumes of components A and B, respectively, in the mixture, m^3/kmol ; x_A and x_B are mole fractions of components A and B, respectively.

The molar volume of a component depends on temperature and is calculated using the formula:

$$v_i = \frac{1}{a} \cdot b \cdot \left[1 + \left(1 - \frac{T}{c} \right)^d \right], \quad i = A, B. \quad (10)$$

The coefficients a , b , c , d in Eq. (10) are given in Table 3.

Table 3. Coefficients for the mole volume calculation by (10)

Component	a	b	c	d
A	1.2298	0.2576	508.2	0.29903
B	0.6727	0.2603	606	0.2511

Molar concentrations in the reactor are related to molar fractions by the ratio:

$$C_A = \frac{x_A}{v}, \quad (11)$$

$$C_B = \frac{x_B}{v}. \quad (12)$$

Molar fractions are related to molar flows in the reactor by the following ratios:

$$x_A = \frac{n_A}{N}, \quad (13)$$

$$x_B = \frac{n_B}{N}, \quad (14)$$

$$N = n_A + n_B. \quad (15)$$

From Eqs. (6) and (7) of the component material balance of the reactor, we express the product molar flows through the known input flows and the unknown reactor productivity P :

$$n_A = n_A^0 - 2P, \quad (16)$$

$$n_B = n_B^0 + P. \quad (17)$$

Thus, using (9), (11)–(17), we obtain relations reflecting the connection between molar concentrations and molar fractions and molar flows in the reactor through one unknown quantity P :

$$C_A = \frac{x_A}{v_A x_A + v_B x_B} = \frac{\frac{n_A}{N}}{v_A \cdot \frac{n_A}{N} + v_B \cdot \frac{n_B}{N}} = \frac{n_A}{v_A n_A + v_B n_B} = \frac{n_A^0 - 2P}{v_A (n_A^0 - 2P) + v_B (n_B^0 + P)}, \quad (18)$$

$$C_B = \frac{x_B}{v_A x_A + v_B x_B} = \frac{\frac{n_B}{N}}{v_A \cdot \frac{n_A}{N} + v_B \cdot \frac{n_B}{N}} = \frac{n_B}{v_A n_A + v_B n_B} = \frac{n_B^0 + P}{v_A (n_A^0 - 2P) + v_B (n_B^0 + P)}. \quad (19)$$

To find the reactor performance, we will transform Eq. (8) using the obtained expressions for the chemical reaction rate (4) and molar concentrations (18) and (19).

As a result of the transformations, we will obtain a nonlinear equation expressing the material balance of the CSTR through one unknown quantity P :

$$P - V \left[\frac{k^+ \cdot (n_A^0 - 2P)^2}{\left(v_A (n_A^0 - 2P) + v_B (n_B^0 + P) \right)^2} - \frac{k^- \cdot (n_B^0 + P)}{v_A (n_A^0 - 2P) + v_B (n_B^0 + P)} \right] = 0. \quad (20)$$

The mathematical model of CSTR also includes the reactor heat balance equation (ΔQ is the heat balance discrepancy, kJ/h):

$$\Delta Q = \left(n_A^0 \cdot C_{P_A}^{\text{in}} + n_B^0 \cdot C_{P_B}^{\text{in}} \right) T_{\text{in}} - \left(n_A \cdot C_{P_A} + n_B \cdot C_{P_B} \right) T - \Delta H_T \cdot P + Q_{\text{ex}} = 0 \quad (21)$$

where C_{P_A} and C_{P_B} are heat capacity of components A and B at inlet temperature, $\text{kJ}/(\text{kmol} \cdot \text{K})$; T_{in} is the temperature at the reactor inlet, K ; Q_{ex} is the amount of heat supplied by external heat exchange, kJ/h .

To simplify the study, let us assume that the CSTR has a cylindrical shape with a flat bottom enclosed in a jacket. The flow structure in the reactor jacket is assumed to correspond to ideal mixing. The amount of heat supplied by external heat exchange is calculated based on the temperature difference between the reactor and the jacket [13].

The equation for calculating the amount of heat supplied by external heat exchange is as follows:

$$Q_{\text{ex}} = K_t \cdot F(T - T^{\text{in}}), \quad (22)$$

where K_t is the heat transfer coefficient, $\text{kJ}/(\text{h} \cdot \text{m}^2 \cdot \text{K})$; F is the heat exchange surface area, m^2 ; T is the reactor

temperature, K; T'' is the refrigerant temperature in the jacket, K.

To solve the problem of analyzing steady states under conditions of external heat exchange, it is necessary to specify the design characteristics, comprising the diameter and height of the reactor, the heat exchange surface, as well as the amount of refrigerant supplied to the jacket, its inlet temperature, and its thermophysical properties.

The heat balance equation for the refrigerant is expressed as follows:

$$G_{\text{ref}} \cdot C_{P_{\text{ref}}} \cdot T' - G_{\text{ref}} \cdot C_{P_{\text{ref}}} \cdot T'' + K_t \cdot F(T - T'') = 0, \quad (23)$$

where G_{ref} is the refrigerant consumption, kmol/h; $C_{P_{\text{ref}}}$ is the heat capacity of refrigerant, kJ/(kmol·K); T' is the refrigerant inlet temperature, K.

When calculating the material balance of the reactor and the thermal balance mismatch, the input parameters were the feed flow rate, its composition and temperature, as well as the refrigerant flow rate and its inlet temperature. The influence of the feed flow input parameters on the steady-state conditions when varying the temperature and composition of the feedstock, as well as the influence of heat exchange, were studied. The following parameters were set: reactor volume, refrigerant quantity, and refrigerant temperature.

Thus, we obtained a problem in which the known parameters are: reactor volume V ; feed rate of the raw material n_A^0 and n_B^0 ; inlet flow temperature T_{in} ; standard heat effect of reaction $\Delta H_{298.15}^0$; refrigerant consumption G_{ref} ; refrigerant inlet temperature T' . The unknown parameters are: the temperature in the reactor and the composition of the product stream.

To solve this problem, the current temperature value in the CSTR was set and Eq. (20) was solved to find the reactor performance value. Then, using Eqs. (16) and (17), the material balance of the reactor was calculated at a given temperature. To solve Eq. (20), the Newton method was used, which is implemented in Microsoft Excel in the Solver module. Next, based on the information obtained, the reactor heat balance discrepancy (21) was calculated. By graphically plotting the dependence $\Delta Q = f(T)$ according to the number of points of intersection of this dependence with the Ox axis, the number of steady states of the reactor was determined. For this purpose, the thermal balance discrepancy was calculated in the temperature range of 200–500 K. When selecting the temperature range, the possibility of lowering the temperature in the reactor by removing heat with an external heat carrier, as well as heating the reaction mass due to the thermal effect, was taken into account.

Analysis of steady states of a flow adiabatic CSTR at different inlet temperatures of reagents

Let us consider the influence of the inlet flow temperature on the steady-state parameters. To do this, we will specify the design parameters of the reactor and the composition of the reagent flow, and its temperature will act as a variable.

Initial data for the calculation were as follows: reactor volume $V = 1.4 \text{ m}^3$; feed rate of raw material $n_A^0 = 100 \text{ kmol/h}$; $n_B^0 = 0$; no external heat exchange, $Q_{\text{ex}} = 0$.

Next, we obtain the dependencies of the thermal balance discrepancies of the adiabatic reactor on temperature at various values of the inlet flow temperature. The calculation results are shown in Fig. 2.

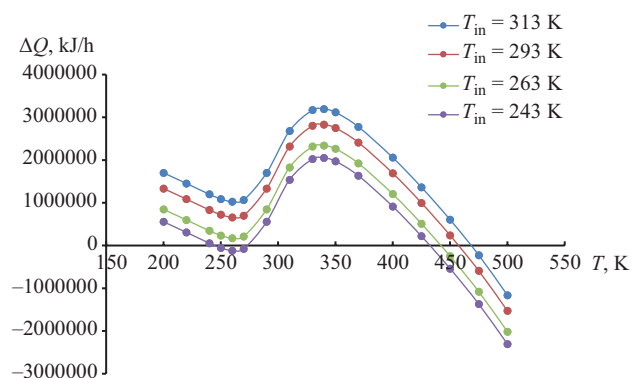


Fig. 2. Reactor temperature dependencies of the energetic balance discrepancy for adiabatic reactor with different feed temperature

Based on the data obtained from the thermal balance discrepancies from the temperature, the parameters of the reactor's steady states were determined, the results of which are shown in Fig. 3.

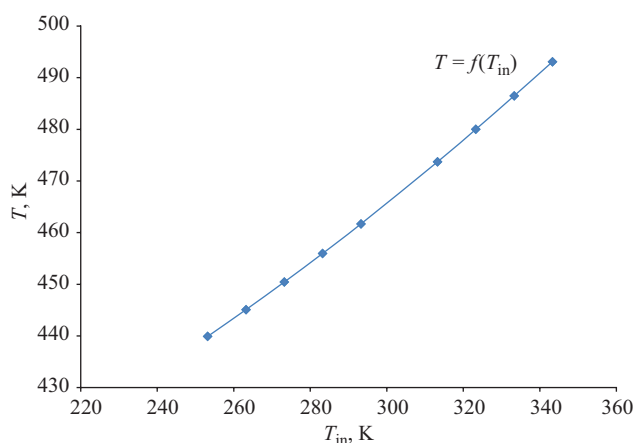


Fig. 3. Feed flow temperature dependence of the internal reactor temperature

The results shown in Fig. 2 indicate that when the inlet flow temperature decreases below a certain value (below 248 K), three steady states become possible, differing in reactor temperature and productivity. For example, at $T_{in} = 243$ K, three steady states are realized: $T_1 = 244.70$ K, $P_1 = 0.229$ kmol/h; $T_2 = 274.79$ K, $P_2 = 4.545$ kmol/h, and $T_3 = 432.46$ K, $P_3 = 37.263$ kmol/h. The calculations showed that the temperature in the reactor increases with an increase in the inlet flow temperature, while the productivity decreases. The decrease in productivity is due to the fact that as the temperature in the reactor increases, the reverse reaction rate constant begins to prevail, which leads to a significant increase in its rate.

Analysis of steady states of a flow adiabatic CSTR at different values of its volume

Let us consider the effect of the CSTR volume on the steady-state parameters. This can be achieved by setting the input values for the reagent flow (its composition and temperature), while the reactor volume will vary.

Initial data for the calculation were as follows: inlet flow temperature $T_{in} = 263.15$ K, feed rate of raw material $n_A^0 = 100$ kmol/h; $n_B^0 = 0$; $Q_{ex} = 0$.

Next, we will obtain the dependencies of the thermal balance discrepancies of an adiabatic reactor on temperature at different values of its volume, m^3 . The calculation results are shown in Fig. 4.

From Fig. 4, it can be concluded that, with a decrease in the reactor volume, three steady states can be achieved. At high volume values, one steady state is achieved with a high temperature in the reactor (approximately 450 K). At low volume values, two more steady states appear with lower temperatures and, accordingly, lower reactor productivity.

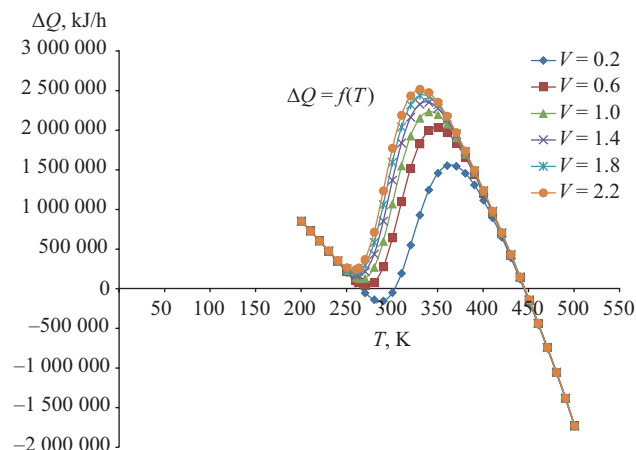


Fig. 4. Reactor temperature dependencies of the energetic balance discrepancy for adiabatic reactor with different reaction volumes

Analysis of steady states in a flow adiabatic CSTR at various inlet flow compositions

Let us consider the influence of the inlet stream composition on the steady-state parameters. This can be achieved by setting the design parameters of the reactor and the inlet stream temperature, while its composition will vary.

Initial data for calculation: reactor volume $V = 1.4$ m^3 ; inlet flow temperature $T_{in} = 263.15$ K; $Q_{ex} = 0$. The calculation results are shown in Fig. 5.

The data obtained (Fig. 5) show that, as the content of component A in the input stream decreases, the heat balance discrepancy curve decreases relative to the Oy axis and straightens. The temperature and productivity of the reactor decrease as the molar fraction of component A in the initial mixture decreases. Furthermore, when the

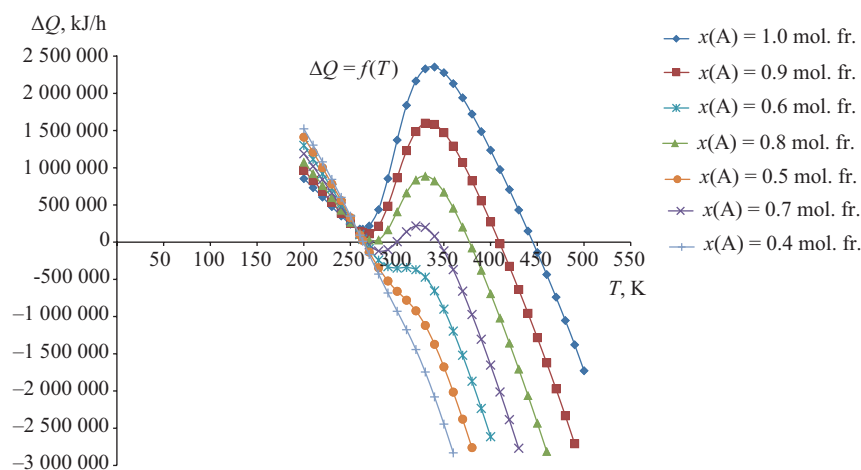


Fig. 5. Reactor temperature dependencies of the energetic balance discrepancy for adiabatic reactor with different composition of feed flow. $x(A)$ is the mole fraction A in the feed flow

ratio of reagents changes, there is a range of values (0.65–0.80 molar fractions of component A) in which three steady states are realized.

Analysis of steady states in a flow isothermal CSTR at different amounts of heat removal

The results of the analysis of the steady states of the adiabatic reactor demonstrate the possibility of three points of intersection of the $\Delta Q(T)$ curve with the Ox axis and consequent likelihood of three steady states of the reactor. The achievement of multiple steady states in the presence of heat removal Q_{ex} can be assumed, since the presence of an additional negative term in (21) will lead to the appearance of new points of intersection of the mismatch curve with the Ox axis. In this regard, calculations of a mathematical model of a reactor with heat removal were performed.

Let us consider the effect of the amount of heat supplied to the refrigerant jacket on the steady-state parameters. This can be achieved by setting the design parameters of the reactor, the temperature and composition of the inlet flow, while the refrigerant supply rate will vary.

Initial data for solving the problem were as follows: inlet temperature of the reactants $T_{in} = 263.15$ K; reactor

volume $V = 1.4$ m³; reactor diameter $d = 1.2$ m; reactor height $h = 1.24$ m; feed rate of the raw material $n_A^0 = 100$ kmol/h; $n_B^0 = 0$.

The heat transfer coefficient K_t generally depends on the hydrodynamic conditions in the reactor and jacket and the composition of the reaction mass. To simplify calculations, we will assume K_t to be constant in accordance with the recommended parameters [13]: $K_t = 5400$ kJ/(h·m²·K); heat capacity of the refrigerant $C_{P_{ref}} = 83.4$ kJ/(kmol·K). Refrigerant inlet temperature $T' = 250.15$ K.

We can calculate the heat exchange surface area F knowing the diameter d and height h of the reactor:

$$F = \pi dh + \frac{\pi d^2}{4} = 3.14 \cdot 1.2 \cdot 1.24 + \frac{3.14 \cdot 1.2^2}{4} = 5.80 \text{ m}^2.$$

To calculate the outlet temperature of the refrigerant and the amount of heat removed, we will use Eqs. (22) and (23). Using the reactor model, we will obtain the dependencies of the heat balance discrepancies of the isothermal reactor on the temperature at different refrigerant flow rates. The calculation results are shown in Fig. 6.

Based on the obtained dependencies of heat balance discrepancies on temperature, the parameters of steady states of an isothermal reactor were determined as given in Table 4.

Table 4. Steady states of the isothermal reactor with different flow rates of heat carrier

G_{ref} , kmol/h	T , K	P , kmol/h	Q_{ex} , kJ/h	T'' , K
0	445.07	37.06	0.00	–
200	267.60	2.48	$1.90 \cdot 10^5$	261.53
	271.22	3.40	$2.29 \cdot 10^5$	263.90
	388.78	38.01	$1.51 \cdot 10^6$	340.59
400	260.75	1.30	$1.71 \cdot 10^5$	255.28
	282.60	7.98	$5.24 \cdot 10^5$	265.86
	369.94	37.95	$1.93 \cdot 10^5$	308.14

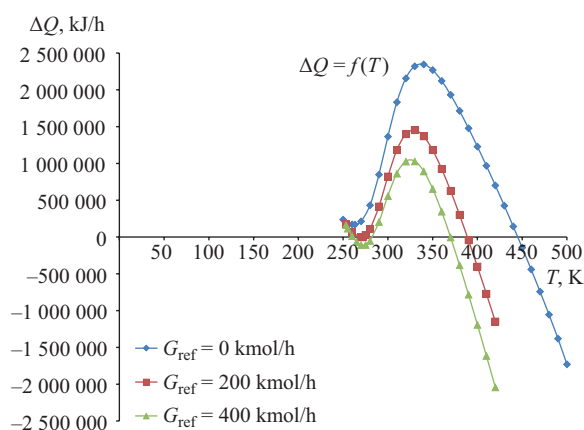


Fig. 6. Reactor temperature dependencies of the energetic balance discrepancy for isothermal reactor with different flow rates of heat carrier

The results show that, in the case of heat exchange with an increased quantity of refrigerant, three steady states of the reactor occur, differing in temperature and performance. The presence of several steady states is realized at refrigerant flow rates above 200 kmol/h.

Thus, numerical modeling of the reactor made it possible to identify its possible steady states. If we assume that the reactor under consideration is an element of a recirculation system, then the reactor feed flow is formed by mixing some external flow with the recirculation flow. Thus, it can be assumed that the recirculation system, including the reactor and the separation unit, can also implement several steady states with different reactor capacities and, consequently, conversion throughout the system.

Stability of steady states in a dimerization reactor under small parameter changes

For a CSTR, whose mathematical model is a system of ordinary differential equations, the concentrations of reactants and the temperature in the reactor play the role of variables. The behavior of the object is described by a system of first-order differential equations [14]:

$$\begin{cases} \frac{dC_A}{dt} = \frac{n_A^0 - qC_A}{V} - 2k^+C_A^2 + 2k^-C_B \equiv f_1, \\ \frac{dC_B}{dt} = \frac{n_B^0 - qC_B}{V} + k^+C_A^2 - k^-C_B \equiv f_2, \\ \frac{dT}{dt} = \frac{(n_A^0C_{PA} + n_B^0C_{PB})T_{in} - qT - \Delta H_T(k^+C_A^2 - k^-C_B)}{V(C_A C_{PA} + C_B C_{PB})} \equiv f_3, \end{cases} \quad (24)$$

where q is the volume flow rate of the mixture, m^3/h , which is calculated using the expression:

$$q = (v_A x_A + v_B x_B)L, \quad (25)$$

where L is the flow rate at the reactor outlet, $kmol/h$.

The coefficients of system (24) are the values of partial derivatives from the right-hand sides of the equations corresponding to the equilibrium position coordinates, namely matrix \mathbf{B} :

$$\mathbf{B} = \begin{pmatrix} \frac{\partial f_1}{\partial C_A} & \frac{\partial f_1}{\partial C_B} & \frac{\partial f_1}{\partial T} \\ \frac{\partial f_2}{\partial C_A} & \frac{\partial f_2}{\partial C_B} & \frac{\partial f_2}{\partial T} \\ \frac{\partial f_3}{\partial C_A} & \frac{\partial f_3}{\partial C_B} & \frac{\partial f_3}{\partial T} \end{pmatrix} \quad (26)$$

The equation of system (24) with respect to characteristic roots λ has the form of a third-degree polynomial [15]:

$$\lambda^3 + \sigma\lambda^2 + \Delta\lambda + \theta = 0, \quad (27),$$

whose coefficients are calculated in accordance with the following expressions:

$$\sigma = -\left(\frac{\partial f_1}{\partial C_A} + \frac{\partial f_2}{\partial C_B} + \frac{\partial f_3}{\partial T}\right), \quad (28)$$

$$\Delta = \begin{vmatrix} \frac{\partial f_1}{\partial C_A} & \frac{\partial f_2}{\partial C_A} \\ \frac{\partial f_1}{\partial C_B} & \frac{\partial f_2}{\partial C_B} \end{vmatrix} + \begin{vmatrix} \frac{\partial f_1}{\partial C_A} & \frac{\partial f_3}{\partial C_A} \\ \frac{\partial f_1}{\partial T} & \frac{\partial f_3}{\partial T} \end{vmatrix} + \begin{vmatrix} \frac{\partial f_2}{\partial C_B} & \frac{\partial f_3}{\partial C_B} \\ \frac{\partial f_2}{\partial T} & \frac{\partial f_3}{\partial T} \end{vmatrix} \quad (29)$$

$$\theta = -\begin{vmatrix} \frac{\partial f_1}{\partial C_A} & \frac{\partial f_1}{\partial C_B} & \frac{\partial f_1}{\partial T} \\ \frac{\partial f_2}{\partial C_A} & \frac{\partial f_2}{\partial C_B} & \frac{\partial f_2}{\partial T} \\ \frac{\partial f_3}{\partial C_A} & \frac{\partial f_3}{\partial C_B} & \frac{\partial f_3}{\partial T} \end{vmatrix} \quad (30)$$

$$\Omega = -\sigma^2\Delta^2 + 4\sigma^3\theta + 4\Delta^3 - 18\sigma\Delta\theta + 27\theta^2. \quad (31)$$

To solve system (24), we express the partial derivatives of matrix \mathbf{B} in analytical form:

$$\frac{\partial f_1}{\partial C_A} = -\frac{q}{V} - 4k^+C_A, \quad (32)$$

$$\frac{\partial f_1}{\partial C_B} = 2k^-, \quad (33)$$

$$\frac{\partial f_1}{\partial T} = -2C_A^2 \frac{\partial k^+}{\partial T} + 2C_B \frac{\partial k^-}{\partial T}, \quad (34)$$

$$\frac{\partial f_2}{\partial C_A} = 2k^+C_A, \quad (35)$$

$$\frac{\partial f_2}{\partial C_B} = -\frac{q}{V} - k^-, \quad (36)$$

$$\frac{\partial f_2}{\partial T} = C_A^2 \frac{\partial k^+}{\partial T} - C_B \frac{\partial k^-}{\partial T}, \quad (37)$$

$$\frac{\partial k}{\partial T} = \frac{\partial \left[A \cdot e^{\frac{-E_a}{RT}} \right]}{\partial T} = A \cdot e^{\frac{-E_a}{RT}} \frac{E_a}{RT^2}. \quad (38)$$

We express the derivative functions f_3 as:

$$\begin{aligned} \frac{\partial f_3}{\partial C_A} &\approx \frac{\Delta f_3}{\Delta C_A} = \\ &= \frac{f_3(C_{As} + 0.001; C_{Bs}; T_S) - f_3(C_{As}; C_{Bs}; T_S)}{0.001}, \end{aligned} \quad (39)$$

$$\frac{\partial f_3}{\partial C_B} \approx \frac{\Delta f_3}{\Delta C_B} = \frac{f_3(C_{AS}; C_{BS} + 0.001; T_S) - f_3(C_{AS}; C_{BS}; T_S)}{0.001}, \quad (40)$$

$$\frac{\partial f_3}{\partial T} \approx \frac{\Delta f_3}{\Delta T} = \frac{f_3(C_{AS}; C_{BS}; T_S + 0.001) - f_3(C_{AS}; C_{BS}; T_S)}{0.001}. \quad (41)$$

Here, C_{AS} and C_{BS} are concentrations of components A and B at steady states, respectively, kmol/m³; T_S is the reactor temperature at steady states, K.

We will analyze the obtained steady states of the adiabatic reactor for stability; the parameters of the steady states are given in Table 5.

For these values, we will analyze the stability of steady states in the small. We will calculate the values of the derivatives using formulas (32)–(41) and find the values of the coefficients of the characteristic Eq. (27) using formulas (28)–(31). The results are presented in Table 6.

From the Routh–Hurwitz conditions, it follows that the equilibrium position under investigation is stable if the following inequalities are satisfied [15]:

$$\sigma > 0; \Delta > 0; \theta > 0; \sigma\Delta - \theta > 0. \quad (42)$$

From this, it can be concluded that systems characterized by a single steady state are stable. For example, steady states 1, 2 and 3, which when occurring in isolation (as shown by qualitative analysis), are characterized by stable reactor operation. However, when several steady states are realized, not all of them correspond to a stable reactor state. In a reactor with a volume of 0.2 m³, three steady states are realized, two

Table 5. Steady states of the reactor

No.	V , m ³	T_{in} , K	x_A , mol. fr.	T , K	P , kmol/h
1	1.4	253.15	1.0	439.89	37.18
2	1.4	263.15	1.0	445.07	37.06
3	1.4	273.15	1.0	492.98	35.72
4	0.2	263.15	1.0	265.22	0.30
5				302.73	6.31
6				444.22	36.88
7	1.4	263.15	0.7	266.85	0.75
8				299.77	7.90
9				344.27	18.99

Table 6. Coefficients of the characteristic Eq. (27)

No.	σ	Δ	θ	$\sigma\Delta - \theta$	Ω
1	6020	$2.11 \cdot 10^6$	$1.19 \cdot 10^7$	$1.27 \cdot 10^{10}$	$-1.16 \cdot 10^{20}$
2	7270	$3.07 \cdot 10^6$	$1.75 \cdot 10^7$	$2.23 \cdot 10^{10}$	$-3.64 \cdot 10^{20}$
3	8800	$4.47 \cdot 10^6$	$2.58 \cdot 10^7$	$3.93 \cdot 10^{10}$	$-1.14 \cdot 10^{21}$
4	119	4670	59900	498000	-224000
5	15.3	-4510	-136000	67300	$-4.13 \cdot 10^{10}$
6	6990	$1.76 \cdot 10^6$	$5.99 \cdot 10^7$	$1.23 \cdot 10^{10}$	$-6.14 \cdot 10^{19}$
7	20.3	135	293	2440	-22.9
8	4.78	-157	-925	172	$-5.98 \cdot 10^6$
9	52.8	-1340	-10400	-60300	$-3.11 \cdot 10^{10}$

of which (Nos. 4 and 6) are stable, and one (No. 5) is a saddle [15]. When a mixture of components A and B in a ratio of 0.7/0.3 is fed into the reactor, it is also possible to implement three steady states, where one state is a stable node (No. 7) and the other two are saddles (Nos. 8 and 9).

DISCUSSION OF RESULTS AND CONCLUSIONS

By calculating the thermal balance discrepancies, possible steady states of a CSTR were identified. The influence of the inlet temperature and composition of the reagent flow, reactor volume, heat exchange, and inlet flow composition on the parameters of steady states was considered along with a determination of their type.

As the inlet flow temperature increases, the steady-state temperature also increases along with a decrease in their productivity. This is due to the fact that, as the temperature in the reactor increases, the reverse reaction rate begins to increase.

A decrease in the reactor volume leads to the realization of three steady states (when the reactor volume is less than 0.6 m³), which can lead to unstable operation or low productivity values.

As the product content in the feed stream increases due to the composition approaching chemical equilibrium, the reactor productivity decreases, and two more steady states appear in the range of 0.65–0.8 molar fractions of component A.

The analysis of the steady states demonstrates that the realized steady states are stable nodes and saddles. It follows that oscillatory modes for the reaction under consideration are not realized in a CSTR.

Symbols

A, B — reaction mixture components;
 C_A , C_B — concentrations of components A and B, respectively, kmol/m³;
 C_{A_S} , C_{B_S} — concentrations of components A and B at steady states, respectively, kmol/m³;

C_{P_A} , C_{P_B} — heat capacity of components A and B, respectively, kJ/(kmol·K);
 $C_{P_{ref}}$ — heat capacity of refrigerant, kJ/(kmol·K);
 E_a — activation energy, J/mol;
 F — surface area of heat exchange, m²;
 G_{ref} — refrigerant consumption, kmol/h;
 ΔH_T — change in enthalpy during the reaction, kJ/kmol;
 k — speed constant;
 K_t — heat transfer coefficient, kJ/(h·m²·K);
 L — reactor outlet flow, kmol/h;
 n_A^0 and n_B^0 — feed rate of raw material into the reactor, kmol/h;
 n_A and n_B — product flow rates, kmol/h;
 P — reactor performance, kmol/h;
 q — volume flow rate of the mixture, m³/h;
 Q_{ex} — heat removal from the reactor, kJ/h;
 ΔQ — energy imbalance, kJ/h;
 R — universal gas constant, 8.314 J/(mol·K);
 T_{in} — inlet flow temperature to the reactor, K;
 T' — refrigerant inlet temperature, K;
 T'' — refrigerant outlet temperature, K;
 T — reactor temperature, K;
 T_S — reactor temperature at steady states, K;
 V — reactor volume, m³;
 v — molar volume of mixture, m³/kmol;
 v_A and v_B — molar volumes of components A and B, respectively, in the mixture, m³/kmol;
 x_A and x_B — molar fractions of components A and B in the mixture, respectively;
 W — chemical reaction rate.

Authors' contributions

N.A. Korol'kova—search for data for reaction mixture physical and chemical properties, development of the reactor mathematical model, development of the steady state analysis for the reactor.

S.L. Nazanskii—statement the problem, development of the reactor mathematical model, development of the steady state stability analysis of reactor, conclusions.

M.A. Solokhin—search for data for reaction mixture physical and chemical properties, thermodynamical consistency of the reaction rate model, development of the steady state stability analysis of the reactor.

The authors declare no conflicts of interest.

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Translated from Russian into English by H. Moshkov

Edited for English language and spelling by Thomas A. Beavitt