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Analysis of the Efficiency of Catalysts for Isomerisation of Light Gasoline Fractions by the Mathematical Modelling Method

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Abstract

An increase in the octane number of straight-run fractions of normal hydrocarbons C₅–C₆ up to 92 points by research octane number (RON) through their catalytic isomerisation appears to be an effective solution of the critical applied task of changing the structure of the domestic gasoline pool. Isomerisates have a high octane number and do not contain hazardous sulphur compounds, aromatic hydrocarbons and particularly, benzene. Herewith, selection of the optimum technology for isomerisation of light gasoline fractions considering a peculiarity of a specific refinery is an important scientific and technical issue. Additionally, a number of complex multiple-factor tasks, such as ensuring of a specified quality and quantity of raw materials, maintaining the optimum activity of the used catalysts, and also providing the optimum operation modes for each complex block considering energy and resource efficiency in manufacturing might arise. To describe nonstationary catalytic processes of catalytic processing of light alkanes the mathematical modelling method is used. This paper assesses the operating efficiency of catalysts for isomerisation of light gasoline fractions based on Pt/SO₄²⁻/ZrO₂ and Pt/Cl⁻/Al₂O₃ using the mathematical modelling method for nonstationary catalytic processing of hydrocarbon raw materials. Catalysts based on sulphated zirconium oxide retain high activities and stabilities during the entire operating cycle, while catalytic systems based on chlorinated alumina are deactivated faster. Isomerisation process scheme with the recycle of unreacted C₅ and C₆ hydrocarbons involves significant capital and operating costs, however, its use will allow increasing the RON of isomerisates in 10–12 points.

Key words: isomerisation, catalyst, mathematical modelling, activity

INTRODUCTION

Straight-run gasoline fractions obtained by primary distillation of oil and gas condensate contain substantial amounts of linear paraffin hydrocarbons with low octane numbers and cannot be used as motor fuel components. The modern environmental requirements are pretty tough on the content of aromatic hydrocarbons, particularly, benzene in the composition of motor fuels. The introduction of catalytic isomerization process in a hydrogen-containing gas medium into a

scheme of a refinery enterprise is one of the most promising methods of increasing the octane number of straight-run gasoline fractions by research octane number (RON) [1–3]. The works on the development and improvement of highly efficient catalytic systems of isomerization of light alkanes are of great scientific and practical interest [4, 5]. However, the introduction of these processes into the structure of the operational refinery is complicated by a number of technological challenges, such as ensuring a specified quality and quantity of raw materials, maintaining the optimum activ-

ity of the used catalysts, and also providing optimum operation modes of each unit of the complex considering energy and resource efficiency of production. One can solve similar complex chemical-technological problems most efficiently using the mathematical modelling method considering the physicochemical component of industrial processes.

This paper uses the mathematical modelling method to describe non-stationary catalytic processes of isomerization of hydrocarbon raw materials. According to the methodology of [6–9], mathematical model building on the physicochemical base of catalytic reactor process consists in the sequential execution of the following stages: thermodynamic analysis of the process, assessment of kinetic parameters of thermodynamically possible reactions, construction of models of the reactor and chemical-technological system considering recycles.

EXPERIMENTAL

Study object

A plant for catalytic isomerization of light gasoline fractions using a sulphated catalyst with various organizations of recirculating flows, and also isomerization process setup with the recycle of low-branched hexanes using a platinum-containing catalyst are the study objects. Table 1 describes the technological parameters of the isomerization process of light gasoline fractions.

Isomerization catalysts based on sulphated metal oxides combine high activities and resistance to

catalytic poisons, have the ability to regenerate [1–3]. Isomerization process scheme of light gasoline fractions “for the passage” is simplest, characterized by the minimum capital and operating costs and ensures production of isocomponent with RON of 80–83 points. Introduction to a scheme “for the passage” of recycles for non-reacted pentanes and low-branched hexanes significantly complicates its operation conditions, however, allows achieving the rise in RON in 10–12 points. Catalysts based on chlorinated aluminium oxide lose halogen during continuous operation, with the result that their activity is decreased. Herewith, alkaline washing of organic chlorine in special scrubbers is required. Chlorinated catalysts are not regenerated, and their durability is 3–5 years.

Research method

This work uses a valid formalization of the scheme of transformations of hydrocarbon raw materials during catalytic isomerization based on the reactivity of individual components. The mathematical model building consisted in the sequential execution of the following stages:

- assessment of thermodynamic parameters of possible reactions during isomerization;
- a formalization of the scheme for transformations of hydrocarbon raw materials during the isomerization process based on the thermodynamic analysis;
- assessment of kinetic parameters of the isomerization process by solving the inverse kinetic problem based on the accepted formalized scheme of transformations;

TABLE 1

Technological parameters of the isomerization process

Parameters	Catalytic system Pt/SO ₄ ²⁻ /ZrO ₂		Catalytic system Pt/Cl ⁻ /Al ₂ O ₃
	Scheme “for the passage”	Scheme with recycles by unreacted pentanes and hexanes	Scheme with recycles by low-branched hexanes
Temperature, °C	120–160		120–180
Pressure, MPa	2.5–2.8		3.0–4.0
Volume rate, h ⁻¹	2.5–2.5		1.5
Molar ratio H ₂ /CH	(1.5–2.5) : 1		(0.3–0.5) : 1
Compressor	Required	Required	Absent
Feed of chlorine and alkaline treatment	–	–	Required
Drying of raw materials	–	–	Required
Service cycle	2–3 years, with pre hydrotreatment (9 years)*	2–3 years, with pre hydrotreatment	Capable of regeneration
Octane number (RON), points	80–82	91–92	88

Note. The dash indicates absent

* According to the data of Kinef OOO, Gazpromneft Omsk Refinery OAO, and Rosneft-Komsomolsky refinery OAO

- the transition to a reactor model based on the calculation of the Peclet and Reynolds criteria;
- account for the unsteadiness of the isomerization process of hydrocarbon raw materials due to side coking processes in the catalyst surface.

When building the mathematical model experimental data of industrial run of the isomerization unit for isomerization of light alkanes were used. The variability interval of technological parameters: a temperature of the inlet of the isomerization reactor is 120–140 °C, the pressure at the inlet of the isomerization reactor is 2.5–3.0 MPa, the consumption of raw materials is 60–90 m³/h, the multiplicity of circulation of a hydrogen-containing gas is 300–500 m³ (NC.)/m³.

The composition of raw materials processed in the isomerization setup varies within the following limits, mass %: isopentane 11.56–18.01, *n*-pentane 25.89–40.36, 2,2 dimethylbutane 0.22–0.40, cyclopentane 2.28–3.50, 2,3 dimethylbutane 2.11–2.75, 2-methylpentane 13.90–17.36, 3-methylpentane 7.18–10.41, *n*-hexane 13.41–24.73, methylcyclopentane 2.23–5.45, benzene 0.99–3.0, cyclohexane 0.05–1.36.

The Gibbs energy and first-order rate constants (for the Pt/SO₄²⁻/ZrO₂ system) proceeding during isomerization are given in Table 2.

Assessment of the thermodynamic mode of isomerization reactor operation was carried out by calculations of the Peclet and Reynolds criteria (Re). According to the calculations of Re = 6.26; the diffusion and thermal Peclet criteria exceed 100, reflecting the predominance of the convective streams of heat and mass in the catalyst layer over diffusion ones and allows accepting a model of a plug-flow reactor. A full mathematical description of the catalytic isomerization process of hydrocarbons is based on the component material and heat balances, and in the general form has the following view:

$$\begin{cases} G \frac{\partial C_i}{\partial z} + G \frac{\partial C_i}{\partial V} = \sum_{j=1}^m a_j r_{ij} \\ G \frac{\partial T}{\partial z} + G \frac{\partial T}{\partial V} = \frac{1}{\rho C_p^m} \sum_{j=1}^m Q_j a_j r_{ij} \end{cases}$$

at $Z = 0$, $C_i = C_{i0}$, at $V = 0$, $C_i = C_{i0}$, at $Z = 0$, $T = T_{int}$, at $V = 0$, $T = T_{int}$,

where G is a load of raw materials, m³/s; C_i is the concentration of i -th component, mol/m³; Z is the volume of processed raw materials, m³; $i = 1, \dots, N$; $j = 1, \dots, M$; N is the number of substances involved in the reaction; M is the number of reactions; r_{ij} is the rate of occurrence of i -th hydro-

TABLE 2

Gibbs energy and rate constants of reactions proceeding during isomerization (T = 130 °C, P = 3 MPa)

Reactions	Rate constant of reaction, s ⁻¹		ΔG , kJ/mol
	direct	inverse	
Isomerization			
$n\text{-C}_5\text{H}_{12} \leftrightarrow i\text{-C}_5\text{H}_{12}$	0.717	0.025	-6.46
$n\text{-C}_6\text{H}_{14} \leftrightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3$	0.279	0.210	-4.75
$n\text{-C}_6\text{H}_{14} \leftrightarrow \text{CH}_3\text{-CH}_2\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_3$	0.590	0.484	-1.85
$\text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3 \leftrightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}(\text{CH}_3)\text{-CH}_3$	0.029	0.039	-0.97
$\text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3 \leftrightarrow \text{CH}_3\text{-CH}_2\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_3$	0.323	0.526	-2.9
$\text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}(\text{CH}_3)\text{-CH}_3 \leftrightarrow \text{CH}_3\text{-C}(\text{CH}_3)_2\text{-CH}_2\text{-CH}_3$	0.058	0.127	-5.53
$n\text{-C}_7\text{H}_{16} \leftrightarrow i\text{-C}_7\text{H}_{16}$	0.718	0.215	-3.38
$\text{C}_5\text{H}_9\text{CH}_3 \leftrightarrow \text{C}_6\text{H}_{12}$	0.002	0.0004	-3.92
Benzene saturation			
$\text{C}_6\text{H}_6 + 3\text{H}_2 \rightarrow \text{C}_6\text{H}_{12}$	0.021	-	-215
$\text{C}_6\text{H}_6 + 3\text{H}_2 \rightarrow \text{C}_5\text{H}_9\text{CH}_3$	0.021	-	-211
Naphthene ring expansion			
$\text{C}_6\text{H}_{12} + \text{H}_2 \rightarrow n\text{-C}_6\text{H}_{14}$	0.001	-	-71.2
$\text{C}_5\text{H}_9\text{CH}_3 + \text{H}_2 \rightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3$	0.001	-	-79.9
$\text{C}_5\text{H}_9\text{CH}_3 + \text{H}_2 \rightarrow \text{CH}_3\text{-CH}_2\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_3$	0.001	-	-76.9
$\text{C}_5\text{H}_9\text{CH}_3 + \text{H}_2 \rightarrow \text{CH}_3\text{-C}(\text{CH}_3)_2\text{-CH}_2\text{-CH}_3$	0.002	-	-84.4
$\text{C}_5\text{H}_9\text{CH}_3 + \text{H}_2 \rightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}(\text{CH}_3)$	0.002	-	-78.9
Hydrocracking			
$n\text{-C}_6\text{H}_{14} + \text{H}_2 \rightarrow \text{Gas}$	0.0002	-	-198
$n\text{-C}_7\text{H}_{16} + \text{H}_2 \rightarrow \text{Gas}$	0.021	-	-165
$i\text{-C}_7\text{H}_{16} + \text{H}_2 \rightarrow \text{Gas}$	0.042	-	-162

carbon of the j -th reaction, $\text{mol}/(\text{m}^3 \cdot \text{s})$, in accordance with the scheme of thermodynamically possible reactions [8, 9]; V is the isomerization reactor volume, m^3 ; T is the reactor temperature, $^\circ\text{C}$; ρ is density, mol/m^3 ; Q_j is the heat effect of the j -th reaction, J/mol ; C_p is the specific heat of the gas mixture, $\text{J}/(\text{mol} \cdot ^\circ\text{C})$; a is the catalyst activity.

To calculate RON of the isomerization product the assumption has been taken that the contribution of each component is proportional to its content in a mixture, that is the octane number of the mixture is additive by volume:

$$\text{ON}_{\text{add}} = \sum_{i=1}^n (\text{ON}_i C_i)$$

where ON_{add} is octane number without considering mixing nonadditivity; ON_i is octane number of the i -th component of the mixture; C_i is volume fraction of the i -th component.

A software implementation of the model has been performed in Delphi 7 integrated medium. The composition of processed raw materials and technological parameters of the process are the initial data for calculations.

When calculating technological schemes of the isomerization process with a recycle modelling of the reactor unit was carried out using the computer modelling system Izomer developed in the Tomsk Polytechnic University at the Department of Solid State Chemistry and Chemical Kinetics; modelling of fractionating columns is made through the Aspen HYSYS software. The hydrocarbon compositions of material flows, technological parameters of the rectification process, and also constructive characteristics of column equipment (numbers and types of plates, feed plate, etc.) were input parameters.

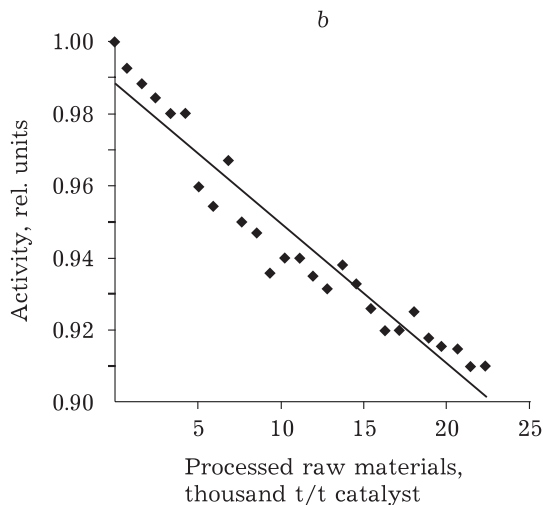
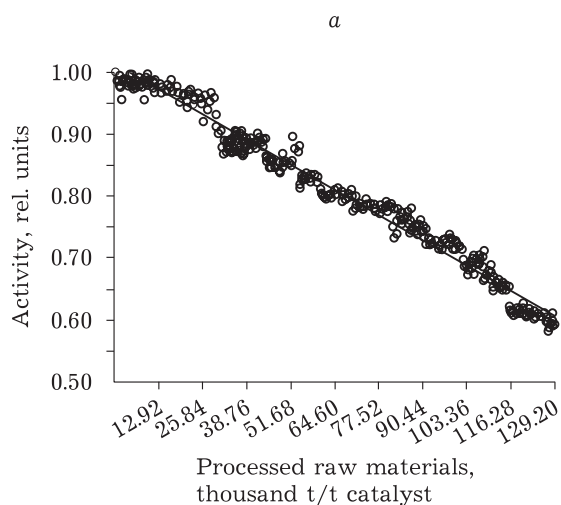


Fig. 1. Dynamics of decreasing $\text{Pt}/\text{SO}_4^{2-}$ catalyst activity: a – scheme “for the passage”; b – scheme with recycles by unreacted pentanes and hexanes.

Model adequacy was verified by comparison of experimental data from the industrial isomerization plant L-35-11/300 with calculated values [8, 9]. It is known that catalyst activity is decreased resulting from occurring processes, such as coking, poisoning, and ageing [6–9].

RESULTS AND DISCUSSION

The dynamics of decreasing the activity of the $\text{Pt}/\text{SO}_4^{2-}/\text{ZrO}_2$ catalytic systems in realisation of technological scheme “for the passage” and with the recycle by unreacted pentanes and hexanes, and also $\text{Pt}/\text{Cl}^-/\text{Al}_2\text{O}_3$ in a scheme with the recycle by low-branched hexanes was studied using a mathematical model of the isomerization process of light gasoline fractions. Research findings are given in Fig. 1 and 2.

A catalyst for the industrial isomerization process (see Fig. 1, a) is operated for 11 years. The volume of processed raw materials is 129.2 thou-

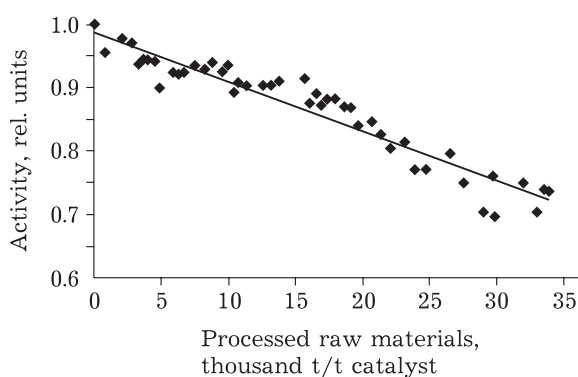


Fig. 2. Dynamics of decreasing the activity of the $\text{Pt}/\text{Cl}^-/\text{Al}_2\text{O}_3$ catalyst, isomerization process scheme with the recycle of low-branched hexanes.

TABLE 3

Comparison of kinetic parameters of the main reactions in catalytic isomerization process

Reactions	Rate constant, rel. units	
	Pt/SO ₄ ²⁻ /ZrO ₂	Pt/Cl ⁻ /Al ₂ O ₃
$n\text{-C}_5\text{H}_{12} \leftrightarrow i\text{-C}_5\text{H}_{12}$	1	0.31
$n\text{-C}_6\text{H}_{14} \leftrightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3$	1	0.47
$\text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3 \leftrightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}(\text{CH}_3)\text{-CH}_3$	1	0.16
$\text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_2\text{-CH}_3 \leftrightarrow \text{CH}_3\text{-CH}_2\text{-CH}(\text{CH}_3)\text{-CH}_2\text{-CH}_3$	1	1.72
$\text{CH}_3\text{-C}(\text{CH}_3)_2\text{-CH}_2\text{-CH}_3 \leftrightarrow \text{CH}_3\text{-CH}(\text{CH}_3)\text{-CH}(\text{CH}_3)\text{-CH}_3$	1	0.85

sand tons of raw materials per 1 ton of catalyst. The activity of this catalyst is 60 % of that of a fresh catalyst, which indicates its low deactivation and an operation mode of the isomerization plant close to optimum proceeding from the volume of processed raw materials. The volume of processed raw materials is 22.31 thousand tons per 1 ton of catalyst for the Pt/SO₄²⁻/ZrO₂ catalytic system of a plant with two recycles by unreacted pentanes and hexanes (see Fig. 1, b). Herewith, catalyst activity is maintained at a high level and amounts to 90 % of the activity of the fresh catalyst.

The activity of the Pt/Cl⁻/Al₂O₃ catalyst is 75 % of the activity of the fresh catalyst with the volume of processed raw materials of 33.88 million tons per 1 ton of catalyst. The activity of the chlorinated catalyst is lower and non-homogeneous compared to the Pt/SO₄²⁻/ZrO₂ systems with a comparable volume of processed raw materials,

which is related to uneven feeding of organochlorine compound.

Kinetic parameters for the target reactions of the isomerization process of light gasoline fractions for chlorinated and sulphated catalysts are given in Table 3.

One can see that the rate constants for the main reactions of the isomerization process for the sulphated catalyst are taken for one, and those for the chlorinated catalyst are presented as a percentage of one. Thus, the target reactions of the isomerization process proceed faster over the sulphated catalyst.

The compositions of the isomerization product for different technologies of the isomerization process of light gasoline fractions are given in Table 4.

Assessment of the effectiveness of various technological schemes for the isomerization process of the light gasoline fraction has been per-

TABLE 4

Calculated compositions of the isomerized product, mass %

Components	Isomerization process technology		
	Pt/SO ₄ ²⁻ /ZrO ₂ System		Pt/Cl ⁻ /Al ₂ O ₃ System
	"For the passage"	With recycles by $n\text{-C}_5$ and C_6	With the recycle of unreacted C_6
<i>n</i> -Butane	0.16	0.42	0.44
<i>iso</i> -Butane	0.13	0.00	0.06
<i>n</i> -Pentane	14.14	0.00	11.56
<i>iso</i> -Pentane	32.97	2.32	30.96
<i>n</i> -Hexane	5.94	6.86	0.54
2-Methylpentane	14.94	0.00	5.24
3-Methylpentane	7.60	9.86	0.76
2,2-Dimethylbutane	11.59	61.69	36.40
2,3-Dimethylbutane	4.36	14.89	5.24
<i>n</i> -Heptane	0.04	0.13	0.00
Total <i>iso</i> -C ₇	3.24	3.74	0.00
Total C ₃	1.77	0.00	0.00
Cyclopentane	0.83	0.00	9.18
Methylcyclopentane	1.05	0.00	0.40
RON of isomerized product	80.20	91.50	83.50

TABLE 5

Initial compositions of raw materials for the isomerization process in various sampling dates, 2015

Components	Content, %							
	Sampling dates							
	04.05	28.05	21.06	15.07	08.08	01.09	25.09	19.10
<i>i</i> -C ₄	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
<i>n</i> -C ₄	0.02	0.02	0.01	0.1	0.00	0.00	0.00	0.00
<i>i</i> -C ₅	11.82	11.79	10.19	11.18	11.36	10.59	8.29	9.09
<i>n</i> -C ₅	33.29	32.31	31.95	31.65	29.15	32.86	33.13	34.57
2,2-Dimethylbutane	0.34	0.29	0.34	0.31	0.39	0.52	0.46	0.39
Cyclopentane	3.57	3.79	3.50	3.67	3.67	3.38	3.54	3.92
2,3-Dimethylbutane	1.91	1.92	2.01	1.91	1.64	1.88	1.86	1.85
2-Methylpentane	14.77	15.10	15.00	14.91	14.42	14.63	14.81	14.95
3-Methylpentane	8.62	8.75	8.86	8.72	8.35	8.65	8.70	8.72
<i>n</i> -C ₆	16.70	17.52	18.18	18.02	18.95	17.91	17.91	17.60
Methylcyclohexane	5.89	6.25	6.60	6.64	7.41	6.05	6.37	6.16
2,4-Dimethylpentane	0.13	0.06	0.15	0.12	0.23	0.11	0.24	0.09
3,3-Dimethylpentane	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
Cyclohexane	1.32	0.72	1.58	1.37	2.39	1.24	2.34	1.01
Benzene	1.58	1.46	1.56	1.45	1.83	2.15	1.97	1.62
C ₇₊	0.04	0.02	0.07	0.04	0.21	0.03	0.37	0.03
Naphthenes	10.79	10.76	11.70	11.68	13.52	10.68	12.34	11.10

formed using a computer modelling system. Calculations were carried out with the industrial composition of raw materials that were IBP 62 °C fraction (Table 5) with various dates of sampling.

As follows from the data of Fig. 1, the use of catalysts based on sulphated zirconium oxide in the industrial process of isomerization of light gasoline fractions realised in the technological scheme with the recycle of unreacted pentanes and low-branched hexanes allows producing the finished isomerization product with high RON. It is 10–12 points higher compared to that of the isomerizate obtained by the traditional scheme of the isomerization process “for the passage” and 8–9 points higher compared to RON in case of a catalyst based on chlorinated aluminium oxide in the scheme with the recycle by unreacted hexanes (Fig. 3).

The use of the scheme with two recycles and deisopentanisation allows increasing the degree of conversion of C₆ paraffins into branched dimethylbutanes, which increases RON of the resulting product by 10–12 points.

CONCLUSION

Various types of catalytic systems for the isomerization process of light gasoline fractions

have been studied using the mathematical modelling method.

As established, catalysts based on sulphated zirconium oxide retain high activities and stabilities during the entire operating cycle, while catalytic systems based on Pt/Cl-/Al₂O₃ lose their activities faster.

Isomerization process scheme with the recycle of unreacted C₅ and C₆ hydrocarbons involves significant capital and operating costs, however, its use will allow increasing the RON of the isomerized product in 10–12 points. Isomerization process scheme with two recycles in the absence of high-octane nonaromatic components at an enterprise is economically feasible and recoups its operating costs in two-three years.

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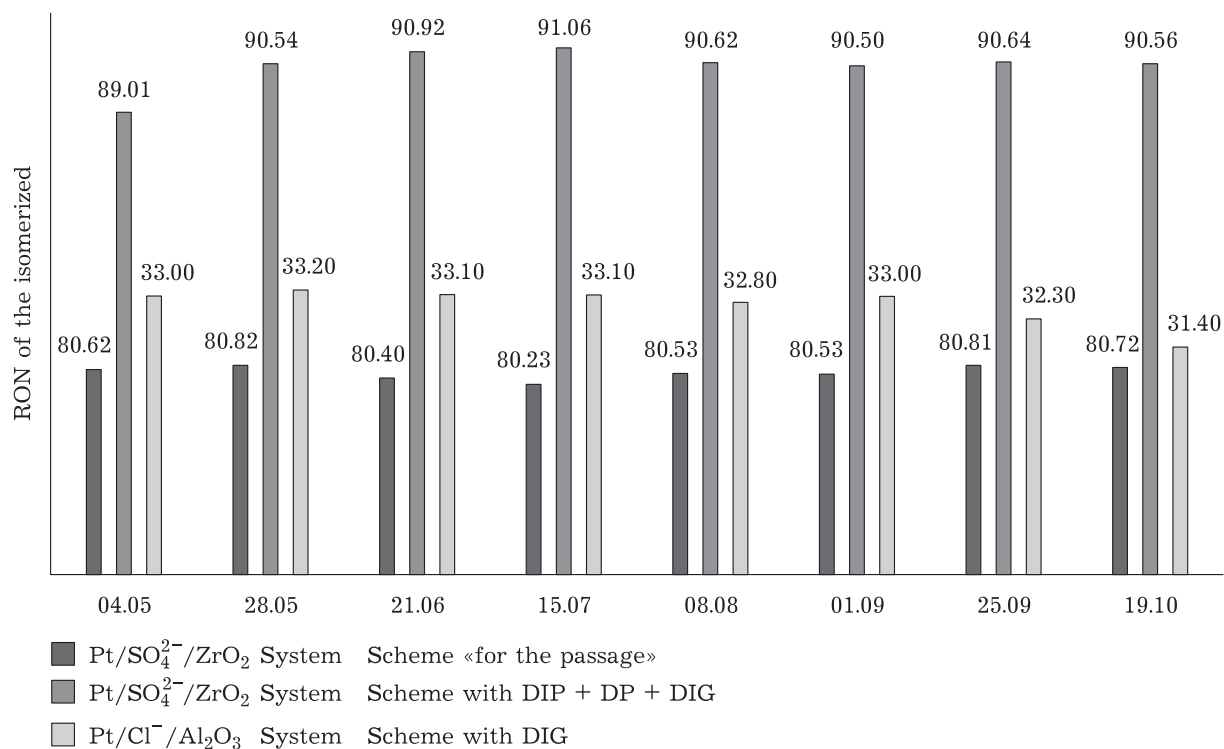


Fig. 3. RON of the isomerized product depending on catalyst type and technological scheme structure. The scheme with DIP + DP + DIG includes deisopentanzation, depentanzation, and deisohexanzation columns, the scheme with DIG – deisohexanzation column.

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