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HYDROTREATING OF VARIOUS PETROL FRACTIONS OVER MODIFIED ALUMOCOBALTMOLYDBDENIC CATALYSTS

Abstract. The paper presents the results of a study of hydrotreating of various gasoline fractions (straight-run gasoline, catalytic cracking gasoline, coking gasoline) on alumo-cobalt-molybdenum catalysts modified with additives of zeolites ZSM-5 and HY, phosphorus and rare earth elements: $CoO-MoO_3-Ce_2O_3-P_2O_5-Al_2O_3-ZSM(KGO-18)$; $CoO-MoO_3-La_2O_3-P_2O_5-Al_2O_3-ZSM(KGO-20)$ µ $CoO-MoO_3-Ce_2O_3-P_2O_5-Al_2O_3-ZSM-HY$ (KGO-16).

The results obtained during the hydroprocessing of various types of gasoline on the catalyst KGO-18 show that the highest amount of isoalkanes is observed during the processing of catalytic cracking gasoline. In the catalyst obtained on the catalyst KGO-18 at 320-380°C 40,3-48,1% of isoalkanes were found, whereas when processing straight-run and gasoline coking, the content of isoalkanes is 34,0-39,7% and 33,9-37%. The octane number after hydrofining of straight-run gasoline and catalytic cracking gasoline is almost the same and is 90,7 and 90,5 (RM), respectively. In the case of coking gasoline, the octane number of fuel produced is lower than when processing other types of gasoline – 83,1 (MM). When studying the process of hydroprocessing of various gasoline fractions over the catalyst KGO-18, it was found that the highest degree of hydrodesulfurization is observed when processing coking gasoline – 87,3%, straight-run gasoline – 81,3% and catalytic cracking gasoline – 75,4%. Modified zeolite-containing alumo-cobalt-molybdenum catalysts for hydroprocessing gasoline fractions in one stage conduct hydrotreating, hydroisomerization, hydrogenation and allow to obtain low-sulfur high-octane gasoline.

Key words: zeolite, straight run gasoline, catalytic cracking gasoline, coking gasoline, catalyst, hydrotreatment.

Introduction

Different gasoline fractions of oil contain a significant amount of linear alkanes, which are characterized by low detonation resistance, which does not allow them to be used directly as motor gasoline. In the world production of motor gasoline, there is a constant tendency to increase the content of isoparaffins with high octane numbers and better ecological characteristics in comparison with aromatic hydrocarbons, limitations on the content of aromatic hydrocarbons, especially benzene ($\leq 1\%$), olefins and sulfur are provided.

To obtain high-quality motor fuels from sulphurous, paraffinic oils, the use of catalytic hydroprocessing processes is necessary: hydrotreatment, hydroisomerization, and hydrogenation. In this regard, the process of hydroisomerization is one of the methods for improving the performance characteristics of light gasoline fractions.

Toughening the requirements for the quality of motor fuels leads to the need for significant changes in available technologies for processing low-grade gasoline fractions using new high-performance catalysts. It is very promising to create one-stage technologies that allow one-stage hydrotreating, hydroisomerization and hydrogenation processes. Existing industrial catalysts for processing petroleum fractions do not meet the increased requirements for the quality of motor fuels. In this regard, the creation of new catalysts for directed single-stage hydroprocessing of oil and its fractions in high-quality fuel will remain an urgent task now and in the near future [1-20]. In this paper, we present the results of a study of the hydroprocessing of various gasoline fractions (straight-run

gasoline, catalytic cracking gasoline, coking gasoline) on alumo-cobalt-molybdenum catalysts modified with additives of zeolites ZSM-5 and HY, phosphorus and rare earth elements.

Experimental part

Zeolite-containing alumina catalysts were prepared. impregnation of a mixture of aluminum hydroxide and zeolites with aqueous solutions of cobalt, molybdenum and modifying additives. After molding, the catalyst was dried at 150°C and calcined at 550°C for 5 hours. Catalysts of the following compositions were prepared: CoO-MoO₃-Ce₂O₃-P₂O₅-Al₂O₃-ZSM (KGO-18); CoO-MoO₃-La₂O₃-P₂O₅-Al₂O₃-ZSM (KGO-20) and CoO-MoO₃-Ce₂O₃-P₂O₅-Al₂O₃-ZSM-HY (KGO-16).

Synthesized modified zeolite-containing catalysts were used to study the hydroprocessing of various types of gasoline. The process was carried out in a high-pressure flow unit with a stationary catalyst bed at temperatures of 320-400°C, a pressure of 4,0 MPa and a space feed rate of 2h⁻¹. The hydrocarbon composition of the reaction products was analyzed on chromatographs "Chromatec-Crystal" and "Chrom-5". Analysis of sulfur content in feedstock and products was carried out in LLP "Oilsert International" (Almaty) and laboratory of physico-chemical methods of "D.V. SokolskyIFCE".

Results and discussion

On the catalyst KGO-18, the process of hydroprocessing various types of gasoline was studied. Hydrogen processing of straight-run gasoline on KGO-18 catalyst showed that in the temperature range 320-350°C, the resulting catalyst contains 39,7-38,2% of isoalkanes (Table 1). At higher temperatures, the yield of isoalkanes decreases to 34,4% at 400°C. The amount of aromatic hydrocarbons in the catalyst under these conditions increases from 15,2 to 24,8%, the yield of naphthenic hydrocarbons decreases from 26,1 to 24,0%.he concentration of olefinic hydrocarbons varies within the range of 4,6-7,6%. The yield of hydro-upgraded gasoline decreases from 70,7 to 60,5% with increasing temperature from 320 to 400°C. The octane number of hydrotreated gasoline is increased compared with the original from 78,9 to 90,7 (MM) and by the motor method from 60,9 to 71,7. The sulfur content of the catalyst decreased from 0,0080 (reference gasoline) to 0,0015% with an increase in temperature to 400°C.

Products,%	T°C						
	Source gasoline	320	350	380	400		
Paraffin C ₅ -C ₆	27,3	11,4	15,1	12,5	12,2		
Isoalkanes	36,8	39,7	38,2	34,0	34,4		
Oleffin	4,8	7,6	5,6	5,4	4,6		
Aromatichydrocarbons	9,2	15,2	18,7	20,9	24,8		
Naphthenichydrocarbons	21,9	26,1	22,3	27,2	24,0		
Yield of the liquid phase	i=i	70,7	68,3	65,0	60,5		
The octane number for research method	78,9	82,6	84,9	88,1	90,7		
Octane number by motor method	60,9	67,8	67,7	68,5	71,7		
Massfractionofsulfur,%	0,0080	0,0049	0,0026	0,0030	0,0015		
Note: $P = 4.0MPa$, $V = 2.0 h^{-1}$		•	•	•	•		

Table 1 - Effect of temperature on the process of hydroprocessing of straight-run gasoline on the catalyst KGO-18

The results of a study of the hydroprocessing of catalyticcracked gasoline using the catalyst KGO-18 showed that with an increase in temperature from 320 to 400°C, the yield of liquid catalyst varies from 80,0 to 68,5% (Table 2). When hydrolyzing catalyticcracked gasoline on the catalyst KGO-18 at 320-350°C, the amount of isoalkanes increases from 25,4 to 42,6-48,1% in comparison with the initial one. The content of aromatic and naphthenic hydrocarbons varies from 30.1 to 36,1%, from 7,0 to 10,4%, respectively. The amount of olefinic hydrocarbons under these conditions drops sharply from 31,2 to 4,3-3,3%. The octane number of the With an increase in temperature from 350 to 400oC, the content of isoalkanes decreases to 39,9% (400°C). The yield of naphthenic and olefinic hydrocarbons under these conditions also decreases to 6,7 and 2,9%, respectively, the amount of aromatic hydrocarbons increases to 43,1%.

The octane number of the refined gasoline obtained at 400° C is 90,5 (RM) and 83,4 (MM). It should be noted that in gasoline hydro-enriched on catalyst KGO-18 at 320° C, the sulfur content is 0,0110% (in the initial 0,0134%), and at 400° C - 0,0033%, which indicates a sufficiently high hydrodesulfurizing activity of this catalyst, produced gasoline practically does not change.

Products,%	T,°C						
	Source gasoline	320	350	380	400		
Paraffin C ₅ -C ₆	6,3	6,9	8,2	7,3	7,4		
Isoalkanes	25,4	42,6	48,1	40,3	39,9		
Oleffin	31,2	4,3	3,3	3,4	2,9		
Aromatichydrocarbons	30,1	36,1	30,0	40,3	43,1		
Naphthenichydrocarbons	7,0	10,1	10,4	8,7	6,7		
Yield of the liquid phase	-	80,0	75,0	71,0	68,5		
The octane number for research	88,7	87,9	88,7	90,4	90,5		
method			W				
Octane number by motor method	80,1	80,9	83,2	82,5	83,4		
Massfractionofsulfur,%	0,0134	0,0110	0,0077	0,0053	0,0033		
Note: $P = 4.0MPa$, $V = 2.0 h^{-1}$				•			

Table 2 - Effect of temperature on the process of hydroprocessing of catalytic cracking gasoline on catalyst KGO-18

When hydrolyzing coking gasoline on the catalyst KGO-18 with an increase in temperature from 320 to 400°C, the content of isoalkanes increases from 23,6% (initial), reaching a maximum value of 39,7% at 380°C, slightly decreasing to 37,1% at 400°C. Under these conditions, the yield of aromatic hydrocarbons increases to 18,6% (400°C). The amount of olefins in the resulting product is significantly reduced compared to the initial (24,2%) and varies within the range of 6,5-9,0%. The octane number of the refined gasoline obtained at 400°C is 82,6 (RM) and 66,8 (MM). The yield of liquid catalyst is 75,5-82,6%. The sulfur content of the catalyst after hydroprocessing decreased from 0,7127 in the initial gasoline to 0,0906% (Table 3).

Products,%	T,°C					
	Source gasoline	320	350	380	400	
Paraffin C ₅ -C ₆	27,2	26,2	20,9	24,7	19,1	
Isoalkanes	23,6	35,6	38,7	33,9	37,1	
Oleffin	24,2	9,0	8,0	8,1	6,5	
Aromatichydrocarbons	8,9	13,2	10,6	14,9	18,6	
Naphthenichydrocarbons	16,2	16,0	21,8	18,4	18,7	
Yield of the liquid phase	-,	82,6	78,2	76,1	75,5	
The octane number for research method	84,0	80,9	80,6	81,4	82,6	
Octane number by motor method	64,7	65,2	64,4	65,0	66,8	
Massfractionofsulfur,%	0,7127	0,0950	0,0930	0,0908	0,0906	

Table 3 - Effect of temperature on the process of hydroprocessing of coking gasoline on the catalyst KGO-18

Analysis of the results obtained during the hydroprocessing of various types of gasoline on the catalyst KGO-18 shows that the highest amount of isoalkanes is observed during the processing of catalytic cracking gasoline. In the catalyst obtained at 320-380°C 40,3-48,1% of isoalkanes were found, whereas when processing straight-run and gasoline coking, their quantity is 34,0-39,7% and 33,9-38,7%, respectively. The octane number of gasoline after hydrofining of straight-run gasoline and catalytic cracking gasoline is almost the same and equal to 90,7 and 90,5 (RM), respectively. In the case of coking gasoline, the octane number of fuel produced is lower than when processing other types of gasoline – 83,1 (MM).

The results obtained by studying the process of hydroprocessing of various gasoline fractions on the catalyst KGO-18 show that the highest degree of hydrodesulfurization is observed during the processing

of coking gasoline and is 87,3%, whereas for straight-run gasoline and catalytic cracking gasoline this figure is lower – 81,3% and 75,4% respectively.

The process of hydroprocessing straight-run gasoline was also investigated on the catalysts KGO-20 and KGO-16. Comparison of the composition of the initial gasoline and the resulting products of hydroprocessing of straight-run gasoline on the catalyst KGO-20 at 320°C shows that the content of isoalkanes increases from 36,8 to 39,3%, aromatic hydrocarbons from 9,2 to 14,9%, olefins from 4,8 to 6.8%, and naphthenes, from 21,9 to 24,5%. At the same time, there is a decrease in the number of paraffins from 27,3 to 14,5%. The octane number by the research method after hydrotreating the straight-run gasoline on the catalyst KGO-20 increased: according to the research method - from 78,9 to 81,4, on the motor - from 60,9 to 66,3. With an increase in temperature from 320 to 400°C, an insignificant decrease in the yield of isoalkanes to 36,2% is observed. The yield of aromatic hydrocarbons is increased to 20,1%. The content of olefins varies between 4,0-7,4%. Under these conditions, the octane number according to the research method after hydrotreating the straight-run gasoline reaches a maximum value of 89,3 for the motor one – 73,4 (Table 4).

Studies have shown that the residual sulfur content after hydroprocessing straight-run gasoline on the catalyst KGO-20 has decreased from 0,0080 in the initial gasoline to 0,0028% (Table 4).

Products.%	T,°C					
110440.5,70	Source gasoline	320	350	380	400	
Paraffin C ₅ -C ₆	27,3	14,5	7,1	4,3	16,7	
Isoalkanes	36,8	39,3	40,4	33,4	36,2	
Oleffin	4,8	6,8	7,0	7,4	4,0	
Aromatichydrocarbons	9,2	14,9	17,8	23,1	20,1	
Naphthenichydrocarbons	21,9	24,5	27,7	31,8	23,0	
Yield of the liquid phase	-	77,0	49,0	44,5	55,0	
The octane number for research method	78,9	81,4	84,9	88,3	89,3	
Octane number by motor method	60,9	66,3	69,4	69,2	73,4	
Mass fraction of sulfur,%j	0,0080	0,0072	0,0062	0,0056	0,0028	

Table 4 - Effect of temperature on the process of hydroprocessing of straight-run gasoline on the catalyst KGO-20.

Table 5 - Effect of temperature on the process of hydroprocessing straight-run gasoline on the catalyst KGO-16

Products,%	T°C					
	Source gasoline	320	350	380	400	
Paraffin C ₅ -C ₆	27,5	17,0	12,7	12,8	14,2	
Isoalkanes	39,8	41,3	40,0	36,9	39,8	
Oleffin	5,8	5,6	6,3	5,6	6,2	
Aromatichydrocarbons	8,7	12,9	15,7	22,8	19,7	
Naphthenichydrocarbons	18,2	23,2	25,3	21,9	20,1	
Yield of the liquid phase	-	77,0	67,0	65,0	60,0	
The octane number for research method	79,2	84,0	86,4	85,2	87,3	
Octane number by motor method	61,1	68,7	71,0	70,8	72,7	
Massfractionofsulfur,%	0,0080	0,0036	0,0027	0,0023	0,0016	
Note: $P = 4.0MPa$, $V = 2.0 h^{-1}$						

At P = 4.0 MPa, V = 2.0 h-1, the process of hydroprocessing a straight-run gasoline fraction with a sulfur content of 0,0080% on a catalyst KGO-16 was studied (Table 5). The yield of the liquid phase decreases from 83.5 to 70.0% with increasing temperature from 320 to 400° C. When hydrolyzing gasoline on KGO-16 catalyst in the temperature range 320-400°C, the maximum isoalkane content in the resulting catalyst is observed at 320° C and is equal to 41.3%. It should be noted that the amount of isoalkanes in the catalyst obtained after hydroprocessing depends little on the process temperature and is equal to 36.9-41,3%, which is higher than in the original gasoline fraction. The amount of aromatic hydrocarbons rises

from 8.7 to 19.7% with an increase in the process temperature to 400°C. The amount of olefins slightly increases compared with the original from 5,8 to 6.2%. The amount of naphthenic hydrocarbons in the resulting catalyst varies between 20,1-25,3%. The octane number of enriched gasoline increases in comparison with the initial from 19,2 to 87,3 (IM) and from 61,1 to 72,7 (MM). The content of sulfur in the catalyst with an increase in temperature to 400°C decreased compared with the initial from 0,0080 to 0,0016%.

Comparison of the results obtained in the study of the process of hydroprocessing the straight-run gasoline fraction shows that the catalysts KGO-16 and KGO-18 possess the highest hydrodesulfurizing activity. The degree of hydrodesulfurization on the catalyst KGO-18 is 81,3%, for KGO-16 80,0%, for KGO-2065%.

When hydrotreating straight-run gasoline (octane number 79,2) on the catalysts KGO-16, KGO-18 and KGO-20, an increase in the octane number compared with the original: 87.3; 90,7 and 89,3 respectively. This is mainly due to the increase in the content of isoalkanes in hydro-upgraded gasoline.

When hydrolyzing catalyticcracked gasoline on KGO-16 catalyst with an increase in temperature to 320-350°C, the content of isoalkanes increases from 25,4 to 47,5% (Table 6). With a further increase in temperature, there is a decrease in the amount of isoalkanes formed to 40,3% (400°C). Under these conditions, the amount of aromatic hydrocarbons in the produced gasoline rises from 30,1 to 35,0%. The content of naphthenic hydrocarbons in the catalyst is small and varies between 6,1 and 8,3%. The content of olefins in the catalyst decreases from 31,2 to 8,0%. The yield of the liquid phase with an increase in temperature in the range 320-400°C varies in the range 95.0-100%. The octane number of catalyticcracked gasoline enriched at 400°C is 86,1 (RM) and 79,6 (MM). When hydrotreating gasoline catalytic cracking, the sulfur content in the final product is reduced from 0,0134% (reference gasoline) to 0,0029%.

8,8 47,5 12,1	350 9,2 47,3 7,6	380 10,6 43,0 5,0	400 8,1 40,3 8,0
47,5 12,1	47,3 7,6	43,0 5,0	40,3
12,1	7,6	5,0	
			8,0
25,5	29,4	32,6	35,0
6,1	6,6	8,1	8,3
100	97,5	95,5	95
86,2	85,2	85,4	86,1
81,0	79,0	78,0	79,6
0.0048	0,0047	0,0035	0,0029
	86,2	86,2 85,2 81,0 79,0	86,2 85,2 85,4 81,0 79,0 78,0

Table 6 - Effect of temperature on the process of hydroprocessing of catalytic cracking gasoline on the catalyst KGO-16

In the study of the process of hydroprocessing of catalytic cracking gasoline, it was shown that the catalysts of KGO have a sufficiently high hydrodesulfurizing activity. The degree of hydrodesulfurization on the catalyst KGO-18 is 75,4%, at KGO-16 – 78,4%. In the hydrotreating of catalytic cracking gasoline (octane number 88,7 (RM), a slight decrease in the octane number to 86,1 (RM) is observed on the catalyst KGO-16, while at the KGO-18 the octane number of the upgraded gasoline increases to 90,5 (RM)

We have previously shown that the activity of hydroprocessing catalysts of various petroleum fractions is related to the surface structure, phase composition and the state of the modifying additives [8]. An electron microscopic study was made of the structure and state of the active centers of the KGO catalysts promoted by Ce, Co, Mo, etc. Studies have shown that on the surface of these catalysts, there are several types of surface structures that differ significantly in both size and chemical state of the components.

According to electron microscopy, the thermodesorption of ammonia and X-ray diffraction catalysts are highly dispersed, the metal components of the active phase are predominantly in the oxidized state, forming cluster-associates on the surface whose dispersion and structure and state are determined by the nature of the catalyst components [8].

Conclusions. When hydroprocessing various types of gasoline on the catalyst KGO-18, the highest content of isoalkanes is observed during the processing of catalytic cracking gasoline. 40,3-48,1% of isoalkanes were found in the catalyst obtained on the catalyst KGO-18 at 320-380°C, whereas in the process of straight-run and gasoline coking, the content of isoalkanes is 34,0-39,7% and 33,9 – 38,7%. The octane number of gasoline after hydrofining of straight-run gasoline and catalytic cracking gasoline is almost the same and is 90,7 and 90,5 (RM), respectively. Modified zeolite-containing alumo-cobalt-molybdenum catalysts for hydroprocessing gasoline fractions in one stage conduct hydrotreating, hydroisomerization, hydrogenation and allow to obtain low-sulfur high-octane gasoline.

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БЕНЗИННІҢ ӘРТҮРЛІ ФРАКЦИЯЛАРЫН МОДИФИЦИРЛЕНГЕН АЛЮМОКОБАЛЬТМОЛИБДЕН КАТАЛИЗАТОРАЛАРЫНДА ГИДРОЖАҚСАРТУ

Аннотация. Жұмыста ZSM-5 және HY цеолит қоспалары, фосфор мен сирек кездесетін элементтермен модифицирленген алюмокобальтмолибден катализаторларында: CoO-MoO₃-Ce $_2$ O₃-P $_2$ O₅-Al $_2$ O₃-ZSM (KГО-18); CoO-MoO₃-La $_2$ O₃-P $_2$ O₅-Al $_2$ O₃-ZSM(KГО-20) және CoO-MoO₃-Ce $_2$ O₃-P $_2$ O₅-Al $_2$ O₃-ZSM-HY(КГО-16) бензиннің әртүрлі фракцияларын (тура айдалған бензин, каталитикалық крекинг бензині және кокстеу бензині) гидроөндеу зерттеулерінің нәтижелері берілген.

КГО-18 катализаторында бензиннің әртүрлі фракцияларын гидроөңдеуден алынған нәтижелері көрсеткендей,изоалкандардың ең жоғары үлесі каталитикалық крекинг бензинін өңдегенде байқалды. КГО-18 катализаторындағы 320-380°С-та алынған катализаттағы изоалкандар мөлшері 40,3-48,1%болса, ал тура айдалған бензин мен кокстеу бензиндерін өңдегенде алынған изоалкандар 34,0-39,7% және33,9-38,7%-ды құрады. Тура айдалған бензин мен каталитикалық крекинг бензинін гидрожақсартудан соң октан саны іс жүзінде бірдей, яғни 90,7 мен 90,5-ке (3 Θ) тең. Ал өңделген кокстеу бензинінің октан саны басқа өңделген бензин фракцияларынан қарағанда төмен, яғни 83,1-ге тең (3 Θ).КГО-18 катализаторында бензиннің әртүрлі

фракцияларынгидроөндеу процесінің зерттеулері байқатқандай,ең жоғары гидрокүкіртсіздендіру кокстеу бензинін өңдеуде жүзеге асты, яғни 87,3%-ға тең болды. Ал тура айдалған бензинде 81,3% болса, каталитикалық крекинг бензинінде 75,4%-ды құрады.

Түйін сөздер: цеолит, тура айдалған бензин, каталитикалық крекинг бензині, кокстеу бензині, катализатор, гидротазалау

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ГИДРООБЛАГОРАЖИВАНИЕ РАЗЛИЧНЫХ БЕНЗИНОВЫХ ФРАКЦИЙ НАМОДИФИЦИРОВАННЫХ АЛЮМОКОБАЛЬТМОЛИБДЕНОВЫХКАТАЛИЗАТОРАХ

Аннотация. В работе приведены результаты исследования гидропереработки различных бензиновых фракций (прямогонный бензин, бензин каталитического крекинга, бензин коксования) на алюмокобальтмолибденовых катализаторах, модифицированных добавками цеолитов ZSM-5 и HУ, фосфора и редкоземельных элементов:CoO-MoO₃- Ce_2O_3 - P_2O_5 - Al_2O_3 -ZSM (KГO-18);CoO-MoO₃- La_2O_3 - P_2O_5 - Al_2O_3 -ZSM(KГО-20) и CoO-MoO₃- Ce_2O_3 - P_2O_5 - Al_2O_3 -ZSM-HY (КГО-16).

Результаты, полученные при гидропереработке различных видов бензина на катализаторе КГО-18, показывают, что наиболее высокое количество изоалканов наблюдается при переработке бензина каталитического крекинга. В катализате, полученном на катализаторе КГО-18 при 320-380°C обнаружено 40,3-48,1% изоалканов, тогда как при переработке прямогонного и бензина коксования содержание изоалканов составляет 34,0-39,7% и 33,9 - 38,7%. Октановое число после гидрооблагораживания прямогонного бензина и бензина каталитического крекинга практически одинаково и составляет 90,7 и 90,5 (ИМ) соответственно. В случае бензина коксования октановое число получаемого топлива ниже, чем при переработке других видов бензинов - 83,1(ИМ). При исследовании процесса гидропереработкиразличных бензиновых фракции на катализаторе КГО-18 установлено, что наиболее высокая степень гидрообессеривания наблюдается при переработке бензина коксования - 87,3%, прямогонного бензина - 81,3% и бензина каталитического крекинга - 75,4%. Бензин фракцияларын гидроондеуде модифицирленген цеолиткұрамды алюмокобальтмолибден катализаторлары бір сатыда гидротазалау, гидроизомерлеу, гидрлеу реакцияларын жургізіп, аз күкіртті жоғары октанды бензин алуға мүмкіндік береді.

Ключевые слова: цеолит, прямогонный бензин, бензин каталитического крекинга, бензин коксования, катализатор, гидроочистка

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