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## HYDROGENATION OF AROMATIC HYDROCARBONS ON MODIFIED METAL CATALYSTS SUPPORTED ON CARBON CARRIER

**Abstract.** The catalyst of the selection hydrogenation of benzene in engine fuels is developed. Methods of drawing low-percentage rhodium catalysts on the basis of natural and synthetic carriers are developed for hydrogenation of benzene in main products. It is established the optimum modes of carrying out hydrogenation of benzene on rhodium catalysts applied on zeolite and clay with the given structural characteristics. It is defined that 1 % the rhodium catalysts supported on zeolite increases selectivity on cyclohexane. Results showed that carrying out reaction of hydrogenation of benzene on the developed rhodium catalyst allows to receive pollution-free motor fuel with the low maintenance of aromatics.

**Keywords:** hydrogenation, aromatic hydrocarbons, benzene, catalysts, clay, zeolite.

**Introduction.** The rapid growth of road transport in the developed countries where car density has reached 10-20 km<sup>2</sup> resulted in severe contamination of the environment and especially emissions of harmful air basin exhaust gases [1]. A huge amount of pollutants produced during the combustion of motor gasoline, results in the fact that environmentally requirements are deduced in first place among all the requirements for gasoline. Ecological Problems of associated with environmental pollution at the expense exhaust emissions requires improving the quality of motor fuels. Aromatic hydrocarbons are subjected to catalytic hydrogenation to transfer them to naphthene for reduce the amount of them. The study of the process of catalytic hydrogenation of aromatic hydrocarbons and suitable catalysts improvement are an important task. The structure of aromatic hydrocarbons has a significant effect on carbon formation in combustion chambers and valves on engines, which impairs their performance indicators such as power, economic and environmental characteristics. Under the influence of high temperatures, aromatic hydrocarbons undergo oxidative conversion and are the main source of fouling [2]. Among the aromatic hydrocarbons the most low-boiling aromatic compound is benzene. Benzene damages many organs and vital systems, but, in general considering he is a poison blood [3]. Petrol fraction has to have several properties for using them as a component of engine fuel. So, for example, there is the extreme content of aromatic hydrocarbons and sulfur compounds above which the fraction cannot be used. This extreme content is caused by the fact that the considered fraction in termination products contains in particular quantity, and the compounding will not allow to receive the marketable products answering on aromatic and sulfur content are gray to requirements of standards [4].

Mainly, various hydrogenation processes are applied to decrease in content of sulfur and aromatic compounds in petrol fractions. Hydrotreating, hydrouppgrading, hydro-dearomatization are considered to them. The specified processes are carried out with use of sulfidic Ni-W and Co-Mo. Processes are carried out in flowing system at temperatures 350-450 °C at feed rate of raw materials 1-3 of the 1/hour and pressure of hydrogen, as a rule, to 50 atm. (sometimes even to 70-100 atm.) Reactions of catalytic

hydrogenation of benzene and dehydrogenation of cyclohexane can go only on the transitional metals having face-centered cubic structure or hexagonal structure and besides atomic radiuses of strictly particular sizes [5-6]. Palladium, platinum, iridium, rhodium, osmium are active catalysts of hydrogenation of benzene and a dehydrogenation to cyclohexane and meets the above requirements [7]. At the beginning of our century, Sabatye and Sanderan found that benzene is easily hydrogenated into cyclohexane in the presence of nickel catalysts. Hydrogenation carried out at  $T=120-250\text{ }^{\circ}\text{C}$  and  $P=2-6\text{ MPa}$ s and rate of volume flow on benzene  $0.5-2\text{ p}^{-1}$ . Conversion level reaches 95 %. On platinum catalysts hydrogenation of benzene proceeds under the same conditions, as on nickel, and the almost complete conversion at selectivity is reached, by the close to 100 % [8-9]. The advantages of platinum catalysts are necessary to carry them a little smaller sensitivity to sulfur compounds and a possibility of reactivation of the catalyst, than at nickel. The disadvantages are sensitivity to presence of moisture at raw materials that causes the necessity carefully to drain benzene [10].

Selective hydrogenation of benzene to cyclohexane (SHBC) has attracted many attentions during the past decades [11-12], due to its simplicity, high yields and costs compared with other cyclohexane production methods. Hydrogenation of benzene in cyclohexane is the main method of receiving cyclohexane. Cyclohexane is used for production of caprolactam in the industry of synfils, for receiving hexane diacid and cyclohexanol. Clear benzene, free of sulfur compounds, hydrogenate in a liquid phase over the nickel or platinum catalyst at 4 MPas,  $200\text{ }^{\circ}\text{C}$  and rate of volume flow of supply of raw materials  $1-3\text{ p}^{-1}$ . The benzene containing sulfur and nitrogen compounds is hydrogenated over the nickel tungsten sulphide catalyst at 24 MPas,  $300\text{ }^{\circ}\text{C}$  and rate of volume flow  $0.5\text{ p}^{-1}$ . At hydrogenation of benzene in cyclohexane light-end and other by-products are practically not formed [13-14]. Nickel catalysts on carriers are most often used: alumina, chromium oxide, etc. The nickel-chrome catalyst, as well as other nickel contacts, easily gets poisoned with sulfur compounds. Poisoning is bound to strong, irreversible adsorption of the last, i.e. to blocking of the fissile surface, and at elevated temperatures and to formation of chemical combinations [15].

Catalysis using nanoscale materials has been one of the most active research areas in recent years because of its relevance to chemical and energy related applications. Recently, several excellent review articles have shown that nanocatalysts with high dispersion and narrow size distributions stabilized by appropriate supports or capping materials can work under milder conditions with higher activity and selectivity as compared to conventional heterogeneous catalysts [16-17]. It is known that transition metal nanoparticles are effective catalysts, and the catalytic activity depends on their shape, size, and surface structure of the solid supports [18-20]. Modifying of natural zeolites becomes particularly important in connection with a possibility of creation of catalysts on their basis for process of thermocatalytic transformation of low-quality hydrocarbon raw materials in alicyclic rings. It is established that the greatest positive effect gives modifying of zeolites platinum, palladium and rhodium.

In the real work optimum conditions of a dearomatization on the basis of benzene hydrogenation reaction, for receiving cyclohexane are picked up. Rhodium catalysts operate in more weak conditions and have more selectivity than the traditional systems based on other metals. Kazakhstan possesses huge reserves of natural zeolite and clay which can be used as catalysts of hydrogenation of aromatic hydrocarbons for receiving an ecological straight product – cyclohexane. The aims of the work were synthesis of new rhodium nanosized catalysts on the carbon carrier with the increased efficiency and having high selectivity at hydrogenation of aromatic hydrocarbons for receiving pollution-free engine fuel.

**Experimental.** Red clay, Tonkeris clay, zeolite are used, as a basis for body height and formation of nanocarbon composites. The choice of Tonkeris clay is caused by the fact that as a part of this clay oxides  $\text{Fe}_3\text{O}_4$ ,  $\text{TiO}_2$ ,  $\text{MgO}$ ,  $\text{Cr}_2\text{O}_3$  which have to promote education in the course of a carbonization of fibrous carbon and nanostructures were found. This catalytic carbon causes increase in a specific surface area and porosity that leads to formation of a transport time of system [21].

Rhodium catalysts on the basis of zeolite are prepared by an impregnation method with the subsequent calcination. The impregnated zeolite is exposed to process of a carbonization during 3-5 clocks. The received catalyst containing 0.5-2 % of rhodium is used for hydrogenation.

Carbon materials were prepared by a carbonization and activation at a temperature  $400\text{ }^{\circ}\text{C}$  for the carrier. Argon current is passed for increase in a specific surface area to exemplars during a carbonization. Carbonization was carried out at temperatures of  $650-750\text{ }^{\circ}\text{C}$  at an interval of  $25^{\circ}\text{C}$ . Propane is used as a source of carbon.

The research of activity of the synthesized catalysts was conducted in laboratory flowing installation of high pressure in the range of temperatures of 50-300 °C, pressure of 0.1-3.0 MPa, rate of volume flow 1.0-4.0 hour<sup>-1</sup>. Feed rate of hydrogen made 30-60 ml/min.

Hydrocarbon part was analyzed on a chromatograph «Chromatec Cristall 5000». The detector is flame and ionization. Gas carrier is nitrogen, temperature from 90 °C to 180 °C, a stainless steel column (long 3m, diameter 3mm).

Electronic and microscopic studying of exemplars was carried out on the JEM-100CX device at the accelerating voltage of 100 kV. The device allows study objects at high-res (3.0E).

**Results and discussion.** The rhodium and cobalt carbon catalysts supported on various carriers were prepared and tested. Also low-percentage rhodium carbon catalysts on the carbonized apricot stones were synthesized. Different concentration of rhodium supported on a series of carriers: Al<sub>2</sub>O<sub>3</sub>, zeolite, clay. Cobalt contained carbon catalysts were prepared for comparison of catalytic activities.

At researches of clay breeds from various regions of Kazakhstan for production of catalysts, as it is established what the kaolinit of clays is inexpedient to use additives for carriers of carbon catalysts. Carbonization of clays in the range of temperatures 700-750 °C well develops a specific surface area. Chemical composition of clays is presented in table 1.

Studying chemical composition of clays and a research of process of a carbonization it is possible to tell all clays can be used as carriers of rhodium carbon catalysts. As a part of clays as appears from table 8, there are ion-exchange cations: Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup> which decrease almost twice at processing. Also the increased content of iron in TC is observed.

Table 1 – Chemical composition of clays

Clay	Mass %										
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>3</sub> O <sub>4</sub>	TiO <sub>2</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	Cr <sub>2</sub> O <sub>3</sub>	H <sub>2</sub> O	others
TC	52.4	15.6	8.2	1.7	1.3	3.22	1.2	4.4	0.08	8.3	3.6
RC	65.3	21.7	0.8	1.7	0.21	0.83	0.7	0.3	-	4.9	5.8
WC	56.8	15.6	8.9	0.9	0.7	2.9	0.5	3.2	0.1	7.5	2.9

Physical and chemical characteristics of the synthesized catalysts are shown in table 2. Rhodium salts applied on zeolites for creation of bifunctional catalysts of hydrogenation. The metal centers allow to maintain low equilibrium concentration of olefins in a reaction mixture and prevent coking up of a surface of the catalyst.

Table 2 – Main physical and chemical characteristics of the synthesized rhodium carbon catalysts on different carriers

Indicators	Zeolite	Zeolite	Clay	Zeolite+Clay	Al <sub>2</sub> O <sub>3</sub>
Content of Rh, mass. %	-	0.5	0.5	0.5	0.5
Content of C, mass. %	-	5.0	15.0	12.0	9.0
Surface area, m <sup>2</sup> /g	112.0	122.0	106.0	156.0	96.0
Carbonization, Hour	1.0	3.0	1.0	3.0	2.0

Results of hydrogenation of benzene on cobalt and rhodium catalysts at a temperature of 450 °C and 25 atm are shown in table 3. The composition of hydrogenates of benzene depends not only on a catalytic activity and also from conditions of supply of hydrogen and benzene. Follows from the received experimental results that with increase in supply of benzene in an original stock, a cyclohexane exit gradually decreases because of shortage of hydrogen. On cobalt catalysts conversion of benzene contains 51 % at temperature of 430 °C.

Table 3 – Conversion of benzene on different catalysts (CAS)

T, °C	Conversion of benzene %		
	0.5 % Rh	5 % Co	7 % Co
350	47	24	27
380	49	29	32
400	59	33	35
430	64	41	42
450	65	42	45
480	65	45	48

From all tested exemplars, the rhodium catalyst on zeolite differed in the highest activity. It is considered that for efficient operation of bifunctional catalysts the metal centers have to be as it is possible closer to the acid centers therefore most often the metal component is applied immediately on the surface of zeolite.

Further synthesized rhodium catalysts applied on clay. Their catalytic activity in benzene hydrogenation reaction was checked. It is by practical consideration defined that at a research of activity of 0.5-2 % of the rhodium put catalyst formation of methylcyclopentane, methylcyclopentane, cyclohexane is observed.

Figure 1 shows the graphical dependence of benzene conversion on temperature. It can be seen from the figure that among all the modified catalysts, the catalyst based on clay containing 0.5 % rhodium exhibits the least catalytic activity.

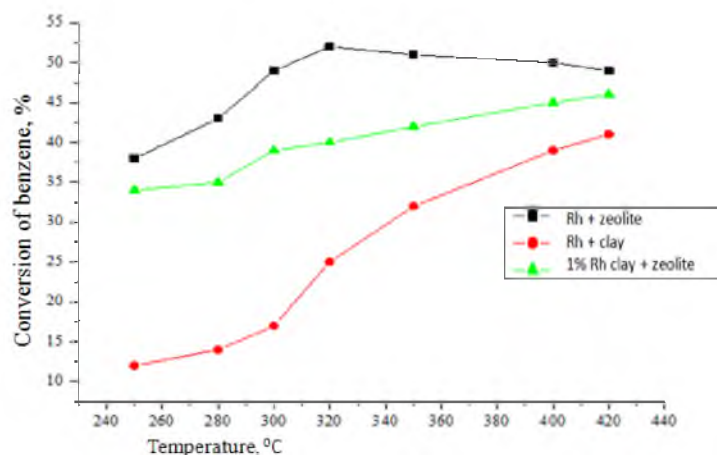


Figure 1 – Dependence of benzene conversion on temperature on different catalyst

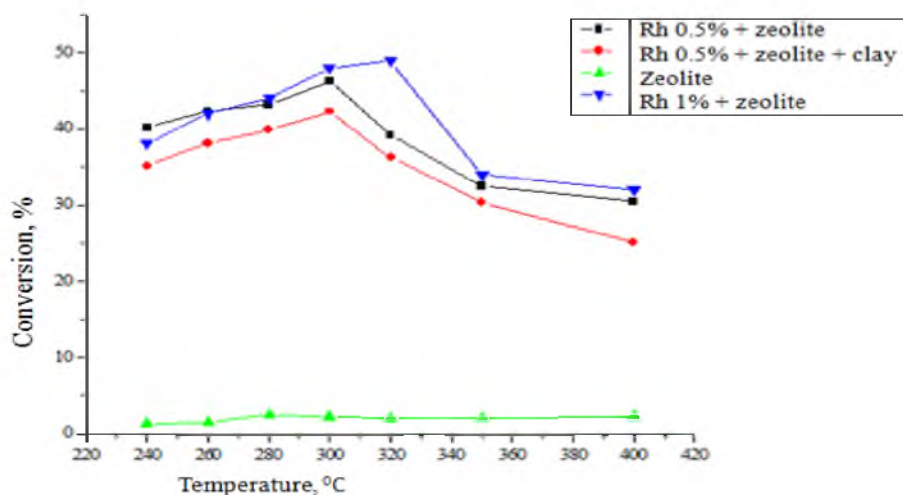


Figure 2 – Dependence of the yield of cyclohexane on various catalysts

Dependence of yield of cyclohexane on amount rhodium in catalysts are shown in Figure 2. The catalytic activity increases in the benzene hydrogenation reactions, when the zeolite is modified with 1 % rhodium. According to data figure 2, Rh 1% catalyst supported on zeolite is the most active catalysts. As noted above, the zeolite modified with 0.5 rhodium allows one to obtain dimethylbutane (2,2-DMB), methylcyclopentane and cyclohexane in the mixture of isomers. A comparative physicochemical analysis of the carbonized modified rhodium zeolite showed that catalytic hydrogenation of benzene catalyzed in the reactions for obtaining valuable by-products.

The obtained results showed that in the study of the catalytic activity of rhodium catalysts, active and selective catalyst was 1% rhodium supported on zeolite, the maximum amount of the desired products was observed in the reaction mixture.

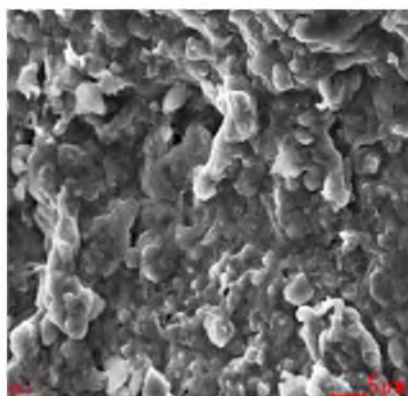
Catalysts with a different ratio of components of the fissile phase were prepared and investigated in the course of benzene hydrogenation. The catalyst synthesized rhodium 0.5 % worked within 20 hours in an interval of temperatures 400-420 °C under pressure hydrogen of 18 atm. before activity loss.

Catalysts with a different ratio of components of the fissile phase were prepared and investigated in the course of benzene hydrogenation. The catalyst synthesized rhodium 0.5 % worked within 20 clocks in an interval of temperatures 400-420 °C under pressure hydrogen of 18 atm. before activity loss. The deactivated catalyst was investigated by physical and chemical methods of the analysis. Comparisons of specific surface areas and activity of prepared, the deactivated and regenerated exemplars shows, that at thermal regenerations in the atmosphere of hydrogen catalytic activity is restored (table 4).

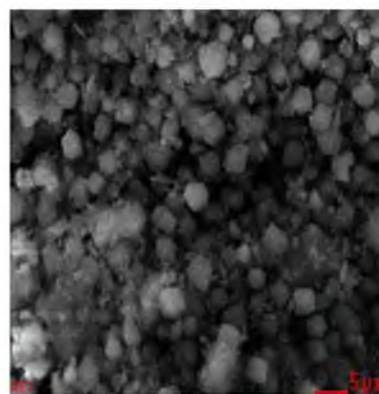
After regeneration in argon current at temperatures of 650 °C on rhodium catalysts activity for 50 % allowed to restore catalytic carbon. And regeneration of rhodium catalysts in the atmosphere of hydrogen restored catalytic activity for 80 %.

Table 4 – Characteristics of synthesized Rh catalysts

Catalysts	Surface area. m <sup>2</sup> /g
0.5 % Rh on carbon	535
0.5 % Rh on deactivated carbon	180
0.5 % Rh on deactivated carbon (in argon flow)	322
0.5 % Rh on regenerated carbon (in hydrogen flow)	480



a) – 1% of Rh/zeolite



b) – 0.5% of Rh/zeolite

Figure 3 – SEM pictures of catalysts

The synthesized rhodium catalysts were investigated by method by an electronic and microscopic method. It is shown in Figure 3, that exemplars of the rhodium catalyst put zeolite are in an amorphous crystalline state, and amorphous is understood as an amorphous state, i.e. in this state there can be also crystal ultra-dispersible particles with sizes less than 500 nanometers. Rhodium the applied zeolite with clay raises dispersion of a surface. Apparently from the figure 3 (b), rhodium catalysts nanoparticles of 50 nanometers in size, and were formed less.

**Conclusion.** The catalyst of the selection hydrogenation of benzene in engine fuels is developed. Methods of drawing low-percentage rhodium catalysts on the basis of natural and synthetic carriers are developed for hydrogenation of benzene in main products.

It is established the optimum modes of carrying out hydrogenation of benzene on rhodium catalysts applied on zeolite and clay with the given structural characteristics. It is defined that 1% the rhodium catalysts supported on zeolite increases selectivity on cyclohexane.

Influence of the rhodium catalyst on effectiveness in reactions of hydrogenation of benzene in cyclohexane and main products is defined. Conversion of 1 % the rhodium catalyst makes 75-80 %, with ultimate yield of isomers of 65 % (methyl cyclohexane, methylcyclopentane).

Results showed that carrying out reaction of hydrogenation of benzene on the developed rhodium catalyst allows to receive pollution-free motor fuel with the low maintenance of aromatics.

As a result it is possible to improve significantly ecological characteristics of engine fuel for achievement of parameters, conditioned Euro-4.

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### **АРОМАТТЫ КӨМІРСУТЕКТІ КӨМІРТЕК ТАСЫМАЛДАҒЫШТАҒЫ МОДИФИЦИРЛЕНҒЕН МЕТАЛЛ КАТАЛИЗАТОРЫНДА ҒИДРЛЕУ**

**Аннотация.** Соңғы жылдары көп ел мотор отының сапасына қатысты қатаң талап қоюда. Мотор отынына қойылған талаптар автомобильді бензин құрамындағы күкірт мөлшері, буланғыштығы мен бензиннің көміртекті қоспасы: ароматты және олефинді көмірсутектер мөлшерін азайтуға негізделеді. Көмірқышқыл газы, күкірт диоксиді, күйе сынды зиянды заттар олардың жануы барысында қозғалтқышқа көп мөлшерде түседі. Сонымен қатар, көптеген ароматты көмірсутектер аса улы зат болып саналады.

Осыған байланысты мотор отыны құрамындағы ароматты көмірсутектің, әсіресе, бензолдың мөлшерін азайту мәселесі өзекті болып есептеледі. Бензол және оның шала жану өнімі бензопирен-канцерогенді зат, адам және жануар өміріне қауіп тудырады. Ароматты көмірсутекті каталитикалық гидрлеуді зерттеу және қазіргі таңда нарықтағы катализаторлардың сапасын арттыру да маңызды. Бензолды каталитикалық гидрлеу реакциясы тек қана атомдық радиусы нақты бір өлшемдерге ие, түйіршіктелген кубтік немесе гексагоналды құрылымға ие ауыспалы металдарда жүреді. Палладий, платина, иридий, родий, осмий бензолды гидрлеу үдерісінің активті катализаторы және жоғарыдағы аталған талаптарға толықтай сай келеді.

Циклогександы бензолды гидрлеу реакциясы арқылы алуға негізделген деароматтау үрдісінің тиімді шарттары осы жұмыста таңдалынды. Өзге металдарға негізделген дәстүрлі жүйелерден гөрі, родий катализаторлары жоғары селективті және жеңіл жағдайда қызмет атқарады. Экологиялық таза өнім циклогександы алуға арналған ароматты көмірсутектерді гидрлеу үрдісі катализаторларының тасымалдағышы ретінде қолданыла алатын саз бен цеолиттің үлкен қоры Қазақстанда бар. Экологиялық таза мотор жанармайын алу үшін ароматты көмірсутектерді гидрлеуге арналған жоғары селективті көміртекті тасымалдағышта өнімі жоғары отандық родийлі катализаторды алудың жаңа әдістері жұмыстың мақсаты болып саналады.

Карбондау үдерісі кезінде талшықталған көміртегі мен наноқұрылымның пайда болуына себеп болатын  $Fe_3O_4$ ,  $TiO_2$ ,  $MgO$   $Cr_2O_3$ , оксидтері болғандықтан Төңкер глинасы таңдалды. Цеолит негізіндегі родий катализаторларын отырғызу әдісі арқылы алады. Синтезделген катализаторлардың белсенділігі жоғары қысымдағы зертханалық қондырғыда, 50-300° С температура және 0,1-3,0 МПа қысым диапазонында зерттелді, көлемдік ағым жылдамдығы 1,0-4,0 сағ<sup>-1</sup>.

Мотор отын құрамындағы бензолді селективті гидрлеуге қолданылатын катализатор дайындалды. Бензолды мақсатты өнімге гидрлеу үшін табиғи және синтетикалық тасымалдағыштар негізінде төменгі мөлшердегі родий отырғызу әдісі қолданылды. Құрылымдық сипаттамалары белгілі цеолит пен глинаға отырғызылған родий катализаторларында бензолды гидрлеудің тиімді режимі анықталды. Цеолитке отырғызылған 1% родий катализаторы циклогексан бойынша селективтілікті арттыратыны анықталды. Бензолды циклогексанға дейін гидрлеу реакциясының эффективтілігіне родий катализаторларының әсері анықталды. 1% родий катализатор қатысында, үдеріс нәтижесінде 65% (метилциклогексан, метилциклопентан) изомерлер шығымы болған жағдайда конверсия 75-80% көрсетті.

Нәтижелерге сәйкес, жасалынған родий катализаторында бензолды гидрлеуді жүргізу құрамында ароматты көмірсутектің мөлшері төмен экологиялық таза мотор отының алуға мүмкіндік беретіндігін көрсетті. Нәтижесінде Еуро-4 стандарттарына сай параметрлерге жететін дәрежеде мотор отындарының экологиялық сипаттамаларын арттыруға болады.

**Түйін сөздер:** гидрлеу, ароматты көмірсутектер, бензол, катализаторлар, саз, цеолит

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### ГИДРИРОВАНИЕ АРОМАТИЧЕСКИХ УГЛЕВОДОРОДОВ НА МОДИФИЦИРОВАННЫХ МЕТАЛЛ–КАТАЛИЗАТОРАХ НА УГЛЕРОДНОМ НОСИТЕЛЕ

**Аннотация.** В последние годы все более ужесточаются требования к качеству моторного топлива во всех странах. Строгие условия на автомобильный бензин ограничивают содержание серы, испаряемость и углеводородный состав бензина: ароматических и олефиновых углеводородов. Вредные вещества, такие как окись углерода, двуокись серы, сажа попадают в двигатели в больших количествах при их сгорании. Кроме того, многие ароматические углеводороды также являются высокотоксичными веществами.

В связи с этим, уменьшение количества ароматических углеводородов, особенно бензола, в моторных топливах является актуальной и неотложной задачей. Бензол и продукт неполного окисления бензопирен – канцерогенное вещество, накапливающееся в окружающей среде, оказывает негативное влияние на безопасность жизни людей и животных. Важной задачей является изучение процесса каталитического гидрирования ароматических углеводородов и улучшение соответствующих катализаторов. Реакции каталитического гидрирования бензола могут протекать только на переходных металлах, имеющих гранцентрированную кубическую или гексагональную структуру и, кроме того, атомные радиусы строго определенных размеров. Палладий, платина, иридий, родий, осмий являются активными катализаторами гидрирования бензола циклогексана и отвечают вышеуказанным требованиям.

В данной работе подобраны оптимальные условия деароматизации на основе реакции гидрирования бензола для получения циклогексана. Родиевые катализаторы работают в более мягких условиях и обладают большей селективностью, чем традиционные системы, основанные на других металлах. Казахстан обладает огромными запасами природного цеолита и глины, которые могут быть использованы в качестве носителя катализаторов гидрирования ароматических углеводородов для получения экологического чистого продукта – циклогексана. Целью работы являются новые методы получения родиевых наноразмерных катализаторов с повышенной продуктивностью на углеродном носителе, обладающем высокой селективностью при гидрировании ароматических углеводородов для получения экологически чистого моторного топлива.

Выбор тонкерской глины вызван тем, что в составе определены оксиды  $Fe_3O_4$ ,  $TiO_2$ ,  $MgO$   $Cr_2O_3$ , которые способствуют образованию волокнистого углерода и наноструктур в процессе карбонизации. Родиевые катализаторы на основе цеолита получают методом пропитки с последующим прокаливанием. Исследование активности синтезируемых катализаторов проводилось в лабораторной установке высокого давления в диапазоне температур 50-300° С, давления 0,1-3,0 МПа, скорости объемного потока 1,0-4,0 час<sup>-1</sup>.

Разработан катализатор селективного гидрирования бензола в моторных топливах. Разработаны способы нанесения низкопроцентных родиевых катализаторов на основе природных и синтетических носителей для гидрирования бензола в целевые продукты. Установлены оптимальные режимы проведения гидрирования бензола на родиевых катализаторах, нанесенных на цеолит и глину с заданными структурными характеристиками. Определено, что 1% родиевых катализаторов, нанесенных на цеолит, увеличивает селективность по циклогексану. Влияние родиевого катализатора на эффективность реакции гидрирования бензола в циклогексан и в основные продукты были определены. Конверсия 1% родиевого катализатора составляет 75-80% при конечном выходе изомеров 65% (метилциклогексан, метилциклопентан).

Результаты показали, что проведение реакции гидрирования бензола на разработанном родиевом катализаторе позволяет получать экологически чистое моторное топливо с низким содержанием ароматических углеводородов. В результате можно значительно улучшить экологические характеристики моторного топлива для достижения параметров, соответствующих стандартам Евро-4.

**Ключевые слова:** гидрирование, ароматические углеводороды, бензол, катализаторы, глина, цеолит.

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