**Авторлар туралы мәліметтер**

**Гульсым Кабаевна Буркеева** – PhD доктор, қауымд. профессоры, академик Е.А. Бөкетов атындағы Қарағанды ұлттық зерттеу университеті, Қарағанды, Қазақстан Республикасы; e-mail: guls_b@mail.ru. ORCID: https://orcid.org/0000-0003-1993-7648.

**Анна Константиновна Ковалева*** – PhD доктор, жетекші ғылыми қызметкер, академик Е.А. Бөкетов атындағы Қарағанды ұлттық зерттеу университеті, Қазақстан Республикасы; e-mail: cherry-girl1899@mail.ru. ORCID: https://orcid.org/0000-0001-9758-648X.

**Нұркен Мейрамұлы Жұмабек –** 2-ші курс магистранты, академик Е.А. Бөкетов атындағы Қарағанды ұлттық зерттеу университеті, Қазақстан Республикасы; e-mail: zhumabek2003@list.ru. ORCID: https://orcid.org/0009-0004-5318-0110.

**Жансая Мирханкызы Ибраева –** Аға оқытушы, Академик Е. А. Бөкетов атындағы Қарағанды ұлттық зерттеу университеті, Қарағанды, Қазақстан Республикасы; e-mail: zhansaya.ibraieva@mail.ru. ORCID: https://orcid.org/0000-0002-1419-1384.

**Information about authors**

**Gulsym Kabayevna Burkeyeva** – PhD, Associate Professor of Chemical Sciences, Karaganda National Research University named after academician Ye.A. Buketov, Republic of Kazakhstan; e-mail: guls_b@mail.ru. ORCID: https://orcid.org/0000-0003-1993-7648.

**Anna Konstantinovna Kovaleva*** – PhD, Lead researcher, Karaganda National Research University named after academician Ye.A. Buketov, Republic of Kazakhstan; e-mail: cherry-girl1899@mail.ru. ORCID: https://orcid.org/0000-0001-9758-648X.

**Nurken Meiramuly Zhumabek** – 2nd year master student, Karaganda National Research University named after academician Ye.A. Buketov, Republic of Kazakhstan; e-mail: zhumabek2003@list.ru. ORCID: https://orcid.org/0009-0004-5318-0110.

**Zhansaya Mirkhankyzy Ibrayeva** – Senior Lecturer, Karaganda National Research University named after academician Ye.A. Buketov, Karaganda, Republic of Kazakhstan; e-mail: zhansaya.ibraieva@mail.ru. ORCID: https://orcid.org/0000-0002-1419-1384.

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**D.B. Karimova[1,2*], A.A. Vosmerikov[2], S.R. Konuspaev[3], L.N. Vosmerikova[2], A.V. Vosmerikov[2]**
[1]Al-Farabi Kazakh National University,
050040, Al-Farabi Avenue, 71, Almaty, Kazakhstan
[2]Institute of Petroleum Chemistry, SB RAS,
634055, Academic Avenue 4, Tomsk, Russia
[3]A.B. Bekhturov Institute of Chemical Sciences,
050010, Sh.Ualikhanov str.,106, Almaty, Kazakhstan
*e-mail: dana.tleugabylova@mail.ru

# INVESTIGATION OF CONVERSION OF N-BUTANE TO LIGHT OLEFINS ON CATALYSTS Pd/γ-$Al_2O_3$ AND Pd/$SiO_2$

***Abstract:*** *The results of the conversion of n-butane on supported palladium catalysts in a stationary mode in the temperature range of 400-600 °C, at atmospheric pressure and a volumetric feed rate of n-butane of 250 $h^{-1}$ are presented. The catalysts were obtained by the method of incipient wetness impregnation using γ-$Al_2O_3$ and $SiO_2$ (silica gel) as supports, the palladium loading was 1 and 3%. It was found that, along with the dehydrogenation of n-butane, cracking processes occur in parallel, leading to the formation of both olefins (ethylene, propylene) and alkanes (methane, ethane, propane), isomerization of n-butane to isobutane, and other reactions. Hydrogen contained in contact gases is equivalent to the amount of dehydrogenation and degradation products. In trace amounts, non-condensed liquid products such as pentane and aromatics are present in the gas phase. The catalysts were studied by SEM-EDS to determine the morphology of catalysts*

*and the elemental composition of individual sections of their surface, and the specific surface area of the catalysts was also determined.*

***Key words:*** *catalyst, palladium, alumina, silica, dehydrogenation, alkanes, n-butane, olefins.*

## Introduction

Light alkanes are the main components of natural gas and crude oil. Despite the long-standing interest in hydrocarbon dehydrogenation reactions, the development of new active, selective, and energy-efficient catalysts remains an urgent problem. Catalytic dehydrogenation of light alkanes can effectively produce olefins and hydrogen. Light olefins, such as ethylene, propylene, and butenes, are bulk commodities and important building blocks for the production of value-added polymers [1, 2]. The demand for these olefins has been growing rapidly in recent years [3]. Today, this problem is especially due to the necessity of processing associated petroleum gases, which are still flared in large quantities in oil-producing regions. The processing of hydrocarbon gases can be carried out via two main routes: oxidative dehydrogenation, producing light olefins and water, and non-oxidative dehydrogenation, yielding light olefins and hydrogen. The last route is preferable, since both products are highly demanded by the petrochemical industry. In reviews [4, 5], existing dehydrogenation catalysts were reviewed; however, very limited information is available in the literature on the use of palladium catalysts in the non-oxidative dehydrogenation of light alkanes.

In a study [6], the addition of Ga to $Pd/Al_2O_3$ significantly affects palladium dispersion, catalytic activity, and selectivity in the n-butane dehydrogenation reaction at 500 °C. Ga changes the surface properties of Pd, inhibiting hydrogenolysis and carbon deposit coking, leading to higher selectivity for n-butenes compared to monometallic Pd catalysts.

Miller and co-workers conducted ethane and propane dehydrogenation on Pd intermetallic alloy catalysts: Pd-Zn [7], Pd-In [8], Pd-Fe [9]. All catalysts exhibited higher dehydrogenation activity, olefin selectivity, and stability compared to monometallic Pd catalysts. It was proposed that the high olefin selectivity arises from geometrically isolated Pd active sites located near inactive metallic promoters (geometric effect). At the same time, inactive metal changes the electronic structure of the Pd 4d orbital (electronic effect) [7-9]. In another study [10], well-dispersed Pd nanoparticles were prepared and encapsulated within IPC-2 and IPC-4 zeolites using the ADOR approach based on a 3D–2D–3D structural transformation. Pd nanoparticles inside zeolite channels prevent their agglomeration and sintering during the reaction of butane dehydrogenation, whereas Pd NPs in catalysts synthesized by the impregnation method tend to grow in size under reaction conditions [11]. A series of Pt-, Pd-, and Ru-based metals promoted by a second non-noble metal (Zn, Cu, Sn, or Fe) were synthesized and confined inside silicalite-1 crystals using a ligand-protected method. The second metal effectively stabilized and dispersed the noble metal atoms within the rigid S-1 zeolite framework via Si–O–M bonds, in this way enabling a reduction in the loading of expensive noble metals. The resulting bimetallic catalysts exhibited excellent performance in the non-oxidative propane dehydrogenation reaction.

Studies directly comparing $Pd/\gamma\text{-}Al_2O_3$ and $Pd/SiO_2$ catalysts in the non-oxidative dehydrogenation of n-butane under the same reaction conditions are still limited. In particular, there is insufficient information on how palladium dispersion, its real content on the support, and the distribution of metal particles affect the activity and selectivity of the reaction. For this reason, further investigation of the physicochemical properties of these catalysts and their behavior during the reaction is necessary.

The aim of this work is to study how the type of support and palladium loading influence the catalytic performance in non-oxidative n-butane dehydrogenation, with special attention to the relationship between metal dispersion, metal-support interaction, and the formation of $C_2$-$C_4$ olefins.

## Materials and methods

The catalysts with a palladium content of 1 % and 3 % were synthesized on $\gamma\text{-}Al_2O_3$ and $SiO_2$ supports by the impregnation method, where a salt with the active metal is dissolved in water, with the solution volume equal to the pore volume of the support. The work began with the preparation of the supports: fractions with a size of 1.5-1.8 mm were selected by sieving and calcined in a muffle furnace at 400°C for 3 hours. Then, the pore volume of the prepared supports was determined. The support was impregnated with the resulting solution and left overnight. The process of n-butane conversion is carried out in a flow-reactor with a fixed bed catalyst. The volume of the catalyst was

5.2 ml. The experiment was carried out in the temperature range from 400 to 600 °C at atmospheric pressure, with a feed rate of 250 $h^{-1}$. Gas supply from the cylinder was carried out from top to bottom with flow control by means of a flow regulator. Uniform heating of the reaction zone was ensured by an electric furnace. At each temperature point, the process was maintained continuously for 30 minutes, and the composition of the reaction products was analyzed at fixed time intervals for all investigated catalysts. Under the conditions of the experiment, liquid products were absent. Gaseous products entered the analytical system and were analyzed by gas liquid chromatography (GLC) on a chromatograph “Chromatek Kristall 5000.2” with a flame ionization detector.

Scanning electron microscopy (SEM) was used to assess the uniformity of the distribution of the metal on the surface of the carrier and for the local determination of the elemental composition. Photomicrographs were obtained using an electron microscope JSM 6610LV. The accelerating voltage was 20 kV, with a resolution from 1 to 500 μm. This method is based on the registration of characteristic X-ray radiation generated by irradiation of the surface with an electron beam, which allows for the precise determination of the elemental composition at selected points of the sample.

The BET (Brunauer-Emmet-Teller) method was used to determine the specific surface area of the catalysts.

**Results**

The results of conversion of n-butane over Pd-containing catalysts supported on γ-$Al_2O_3$ are presented in Tables 1 and 2. As can be seen, from the data in Table 1, for the 1% Pd/γ-$Al_2O_3$ sample, an increase of both the conversion of n-butane and the yield of olefin hydrocarbons is observed with increasing temperature reaching 45 and 31%, respectively, at 600 ° C. The selectivity towards olefin formation in the studied temperature range varies from 68.9% to 76.3%.

Sample 3% Pd/γ-$Al_2O_3$ exhibits lower catalytic activity compared to sample 1% Pd/γ-$Al_2O_3$. Thus, the yield of olefins at 600 °C is 23.9%, which is 7.1% lower than for the 1% Pd/γ-$Al_2O_3$ sample.

Table 1 – Influence of temperature on the main indicators of the process of conversion of n-butane on Pd/γ-$Al_2O_3$ catalysts

| Catalyst | T, °C | X, % | Y, % | S, % | Selectivity, % | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | Hydrogen | Alkanes $C_1$-$C_2$ | Ethylene | Propylene | Butenes | Alkanes $C_3$-$C_5$ | Arenes |
| 1% Pd/γ-$Al_2O_3$ | 400 | 4 | 3.1 | 76,3 | 1,4 | 3,7 | 0,5 | 5,7 | 70,0 | 18,6 | – |
| | 450 | 3 | 2,5 | 73,0 | 1,6 | 3,6 | 0,9 | 5,1 | 67,0 | 21,7 | – |
| | 500 | 5 | 3,7 | 71,8 | 3,0 | 8,2 | 2,2 | 10,0 | 59,6 | 14,6 | 2,4 |
| | 550 | 16 | 12,3 | 78,4 | 3,2 | 12,1 | 5,5 | 13,0 | 59,9 | 5,0 | 1,3 |
| | 600 | 45 | 31,0 | 68,9 | 3,4 | 21,4 | 9,5 | 17,0 | 42,4 | 4,3 | 1,9 |
| 3% Pd/γ-$Al_2O_3$ | 400 | 7 | 4,7 | 68,0 | 1,8 | 13,3 | 0,2 | 6,4 | 61,4 | 16,9 | – |
| | 450 | 4 | 3,0 | 69,0 | 2,7 | 10,5 | 0,7 | 5,8 | 62,5 | 17,9 | – |
| | 500 | 6 | 4,2 | 73,3 | 2,8 | 7,0 | 1,2 | 5,5 | 66,6 | 13,3 | 3,6 |
| | 550 | 14 | 10,5 | 76,9 | 3,2 | 12,4 | 5,5 | 12,1 | 59,3 | 5,6 | 2,0 |
| | 600 | 36 | 23,9 | 67,0 | 3,9 | 23,4 | 9,9 | 19,5 | 37,6 | 3,8 | 1,9 |

Note: T – The reaction temperature; X – conversion; Y – yield of $C_2$-$C_4$ olefins; S – selectivity for the formation of $C_2$-$C_4$ olefins

Data on the composition of the products of n-butane conversion over 1% Pd/γ-$Al_2O_3$ and 3% Pd/γ-$Al_2O_3$ catalysts are given in Table 2. The main products are hydrogen, methane, ethane and olefins. Normal pentane and isopentanes are presented in small quantities and at a temperature of 500 °C and higher, the formation of aromatic hydrocarbons in small amounts (benzene, toluene) are observed. The obtained reaction products contain ethylene, propylene and butene isomers. The concentration of methane, ethane and olefins in the reaction products grows with increasing temperature of process, and the total content of $C_3$-$C_5$ alkanes decreases, mainly due to the conversion of n-butane. The concentration of aromatic hydrocarbons varies slightly with increasing temperature. The largest amount of olefin hydrocarbons is formed over the 1% Pd/γ-$Al_2O_3$ sample. At 600 °C, butenes (19%) prevail, while the yields of propylene are 7.7%, and ethylene is 4.3%.

Based on the experimental findings, the conversion of n butane is not limited to dehydrogenation. The reaction system also involves parallel cracking processes, leading to the formation of lower alkanes ($CH_4$, $C_2H_6$, $C_3H_8$), the isomerization of n butane isobutane, and a series

of consecutive reactions involving intermediate compounds, namely aromatization and dehydrocyclization.

Table 2 – The composition of the products of the conversion of n-butane on catalysts Pd/γ-$Al_2O_3$

| Catalyst | T,°C | X,% | Yield of products,% | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | Hydrogen | Methane | Ethane | Ethylene | Propylene | Butenes | Alkanes $C_3$-$C_5$ | Arenes |
| 1% Pd/γ-$Al_2O_3$ | 400 | 4 | 0,06 | 0,1 | 0,04 | 0,02 | 0,2 | 2,9 | 96,7 | – |
| | 450 | 3 | 0,06 | 0,1 | 0,03 | 0,03 | 0,2 | 2,3 | 97,4 | – |
| | 500 | 5 | 0,15 | 0,3 | 0,12 | 0,11 | 0,5 | 3,0 | 95,6 | 0,1 |
| | 550 | 16 | 0,50 | 1,2 | 0,69 | 0,87 | 2,0 | 9,4 | 85,1 | 0,2 |
| | 600 | 45 | 1,52 | 5,6 | 3,97 | 4,28 | 7,7 | 19,0 | 57,0 | 0,9 |
| 3% Pd/γ-$Al_2O_3$ | 400 | 7 | 0,1 | 0,7 | 0,2 | 0,01 | 0,4 | 4,3 | 94,2 | – |
| | 450 | 4 | 0,1 | 0,4 | - | 0,03 | 0,3 | 2,7 | 96,4 | – |
| | 500 | 6 | 0,2 | 0,3 | 0,1 | 0,07 | 0,3 | 3,8 | 95,1 | 0,2 |
| | 550 | 14 | 0,4 | 1,1 | 0,6 | 0,7 | 1,6 | 8,1 | 87,2 | 0,3 |
| | 600 | 36 | 1,4 | 5,2 | 3,1 | 3,5 | 7,0 | 13,4 | 65,7 | 0,7 |
| Note: T – Temperature of reaction; X – Conversion | | | | | | | | | | |

The results of n-butane conversion on $SiO_2$ samples containing 1 and 3% palladium are presented in Tables 3 and 4. According to Table 3, samples 1% Pd/$SiO_2$ and 3% Pd/$SiO_2$ are characterized by almost the same total catalytic (estimated by the degree of conversion of n-butane) and dehydrogenating activity. The conversion of n-butane at 600 °C for both catalysts is 23%, while the selectivity to olefin hydrocarbons on the 1% Pd/$SiO_2$ sample is slightly higher than for the 3% Pd/$SiO_2$ sample.

Table 3 – Influence of temperature on the main indicators of the process of conversion of n-butane on catalysts Pd/$SiO_2$

| Catalyst | T, °C | X, % | Y, % | S, % | Selectivity, % | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | Hydrogen | Alkanes $C_1$-$C_2$ | Ethylene | Propylene | Butenes | Alkanes $C_3$-$C_5$ | Arenes |
| 1% Pd/$SiO_2$ | 400 | 4 | 3,5 | 81,9 | 0,3 | 0,5 | 0,1 | 0,4 | 81,4 | 17,2 | – |
| | 450 | 4 | 3,6 | 82,1 | 0,3 | 0,6 | 0,2 | 0,5 | 81,4 | 17,1 | – |
| | 500 | 5 | 3,7 | 79,8 | 0,5 | 3,1 | 1,5 | 4,8 | 73,5 | 15,6 | 1,1 |
| | 550 | 7 | 5,5 | 75,0 | 0,5 | 13,4 | 8,1 | 21,8 | 45,1 | 10,0 | 1,1 |
| | 600 | 23 | 16,2 | 70,2 | 0,5 | 25,0 | 18,0 | 37,3 | 14,9 | 3,2 | 1,1 |
| 3% Pd/$SiO_2$ | 400 | 5 | 3,7 | 82,7 | 0,2 | 0,5 | - | 0,1 | 82,6 | 16,6 | – |
| | 450 | 4 | 3,6 | 82,2 | 0,4 | 0,7 | 0,1 | 0,4 | 81,7 | 16,8 | – |
| | 500 | 5 | 3,8 | 80,8 | 0,4 | 2,3 | 1,0 | 3,2 | 76,6 | 15,7 | 0,8 |
| | 550 | 7 | 4,7 | 71,4 | 2,2 | 13,1 | 6,4 | 19,8 | 45,2 | 11,6 | 1,7 |
| | 600 | 23 | 15,4 | 67,8 | 1,6 | 26,1 | 15,7 | 37,6 | 14,5 | 3,2 | 1,3 |
| Note: T – Temperature of reaction; X – conversion; Y – yield of $C_2$-$C_4$ olefins; S – selectivity for the formation of $C_2$-$C_4$ olefins | | | | | | | | | | | |

Analysis of the n-butane transformation process using 1% Pd/$SiO_2$ and 3% Pd/$SiO_2$ catalysts reveals a similar product profile for both systems. The principal reaction outputs are olefin hydrocarbons in the $C_2$-$C_4$ range and saturated hydrocarbons (alkanes) from $C_3$ to $C_5$. Secondary products, including hydrogen, methane/ethane ($C_1$-$C_2$ alkanes), and aromatic species, are present only in small quantities, as detailed in Table 4.

At the same time, it can be noted that significant differences are observed in the content of hydrogen, methane, ethane and olefins in gaseous products obtained on Pd-containing catalysts prepared on the carrier of γ-$Al_2O_3$ and $SiO_2$: over Pd/γ-$Al_2O_3$ samples more cracking products, especially at high temperature, while over Pd / $SiO_2$ catalysts show much higher selectivity to $C_4$ alkanes, meaning butenes are the dominant products (Tables 2 and 4).

Table 4 – The composition of the products of the conversion of n-butane on $Pd/SiO_2$

| Catalyst | T, °C | X, % | Yield of products, % | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | Hydrogen | Methane | Ethane | Ethylene | Propylene | Butenes | Alkanes $C_3$-$C_5$ | Arenes |
| 1% $Pd/SiO_2$ | 400 | 4 | 0,01 | 0,01 | 0,01 | - | 0,02 | 3,5 | 96,4 | – |
| | 450 | 4 | 0,01 | 0,02 | 0,01 | 0,01 | 0,02 | 3,5 | 96,4 | – |
| | 500 | 5 | 0,02 | 0,09 | 0,05 | 0,07 | 0,22 | 3,4 | 96,1 | 0,05 |
| | 550 | 7 | 0,04 | 0,62 | 0,36 | 0,59 | 1,59 | 3,3 | 93,4 | 0,08 |
| | 600 | 23 | 0,12 | 3,73 | 2,05 | 4,16 | 8,61 | 3,4 | 77,6 | 0,26 |
| 3% $Pd/SiO_2$ | 400 | 5 | – | – | – | – | – | 3,7 | 96,2 | – |
| | 450 | 4 | – | – | – | – | – | 3,6 | 96,3 | – |
| | 500 | 5 | – | 0,1 | – | – | 0,2 | 3,6 | 96,0 | – |
| | 550 | 7 | 0,1 | 0,6 | 0,3 | 0,4 | 1,3 | 3,0 | 94,2 | 0,1 |
| | 600 | 23 | 0,4 | 3,8 | 2,1 | 3,6 | 8,6 | 3,3 | 78,0 | 0,3 |

Note: T – Temperature of reaction; X – Conversion

Figure 1 shows the comparative characteristics of the catalytic activity of the studied catalysts during the conversion of n-butane to $C_2$-$C_4$ olefin hydrocarbons at 600 ° C and a Weight Hourly Space Velocity (WHSV) of 250 $h^{-1}$. It can be seen that the 1% $Pd/\gamma$-$Al_2O_3$ has the highest activity and selectivity towards olefins. The n-butane conversion and the yield of olefin hydrocarbons on this sample are 45 and 31.0%, respectively. An increase in the concentration of palladium deposited on $\gamma$-$Al_2O_3$ leads to a decrease in catalytic activity and selectivity. The lowest activity in the process of n-butane dehydrogenation is shown for catalysts prepared on silica gel support. Moreover, the concentration of introduced palladium does not significantly affect the catalytic properties of samples based on $SiO_2$.

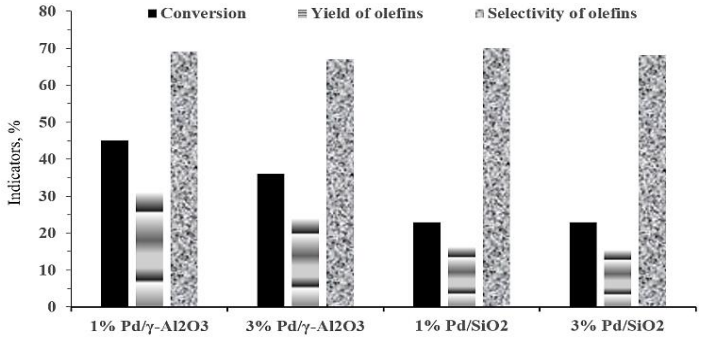


Figure 1 – Comparative activity of palladium-containing supported catalysts (T = 600 °C)

Table 5 shows the elemental composition at the selected sites for all Pd-containing catalysts. It can be seen that the samples supported on $\gamma$-$Al_2O_3$ are characterized by a uniform distribution of palladium with an average content of 1.23% for the 1% $Pd/\gamma$-$Al_2O_3$ sample and 3.18% for the 3% $Pd/\gamma$-$Al_2O_3$. The 1% $Pd/SiO_2$ catalyst is also presented in Table 5, loading of palladium is evenly distributed on the catalyst surface. Its average content is 0.86%, which is slightly less than the calculated amount of palladium. For the 3% $Pd/SiO_2$ catalyst, the elemental composition of three sections was also studied, the obtained data indicate that palladium on silica gel is not distributed evenly. The palladium content reaches 3.30% in some spots, while in others it is 2.11%. The average palladium content was found to be 2.58%, which is lower than the nominal value.

This heterogeneity in palladium distribution may be attributed to limitations of the impregnation procedure, local variations in the porosity of the $SiO_2$ support, and possible agglomeration of Pd species during drying and calcination. Such non-uniform distribution of the active metal may affect the local catalytic activity and overall performance of the catalyst.

Table 5 – Elemental composition for the selected sections of the catalysts $Pd/\gamma$-$Al_2O_3$

| Section | Loading of Pd | | | |
|---|---|---|---|---|
| | 1% $Pd/\gamma$-$Al_2O_3$ | 3% $Pd/\gamma$-$Al_2O_3$ | 1% $Pd/SiO_2$ | 3% $Pd/SiO_2$ |
| 1 | 1,14 | 3,29 | 0,94 | 3,30 |
| 2 | 1,28 | 3,08 | 0,84 | 2,32 |
| 3 | 1,26 | 3,18 | 0,81 | 2,11 |
| average | 1,23 | 3,18 | 0,86 | 2,58 |

**Discussion**

The summarized data on n-butane conversion over the studied catalysts, together with their elemental composition and specific surface area, are given in Table 6. As can be seen, the actual palladium content is slightly higher than the nominal value on γ-$Al_2O_3$, whereas in contrast, for silica gel samples, measured Pd concentration is lower than the calculated amount. This difference may be attributed to the special physicochemical properties of the supports. γ-$Al_2O_3$ contains a significant number of hydroxyl groups and coordinatively unsaturated $Al^{3+}$ sites. As a result, palladium may be anchored more firmly to the surface of the support. Such interactions may conserve palladium and minimize metal loss during drying, calcination, and reduction steps. In addition, strong fixation of Pd species within the alumina surface layer may contribute to the slightly increased measured metal content. On the other hand, silica gel is chemically more inert and exhibits weaker metal-support interactions. Palladium precursor species may be less strongly bound to the $SiO_2$ surface, leading to partial metal loss during preparation steps. Furthermore, differences in pore structure and surface chemistry may result in non-uniform metal distribution, which could explain the lower experimentally determined palladium content compared to the nominal value.

Catalysts supported on γ-$Al_2O_3$ exhibit a relatively lower specific surface area compared to silica gel-based samples. For instance, the 1% Pd/γ-$Al_2O_3$ catalyst possesses a surface area of 232 $m^2/g$, which is 123 $m^2/g$ lower than that of the corresponding 1% Pd/$SiO_2$ sample. Despite the lower surface area, the 1% Pd/γ-$Al_2O_3$ catalyst demonstrates the highest catalytic performance in the conversion of *n*-butane to $C_2$-$C_4$ olefins. At 600 °C, it achieves an olefin yield of 31.0% at an *n*-butane conversion of 45%. This suggests that the reaction is influenced more by metal-support interactions and electronic effects than simply by surface area. The enhanced performance of Pd/γ-$Al_2O_3$ can be attributed to stronger metal-support interactions and the acid-base properties of alumina. In addition, the presence of moderate Lewis acidity on γ-$Al_2O_3$ may promote β-H elimination and facilitate olefin desorption, thereby enhancing selectivity toward $C_2$-$C_4$ olefins while suppressing deep cracking and coke formation. In contrast, silica-supported catalysts, despite their higher surface area, exhibit weaker metal-support interactions, which can result in less stable active sites and slightly lower catalytic efficiency. The lower activity observed for the 3% Pd samples may be explained by the formation of larger palladium particles. As the particles grow, the number of well-dispersed active sites decreases, which can negatively affect selective dehydrogenation. The long-term stability and resistance to catalyst deactivation were not studied in detail in the present work, since the main focus was on the effect of the support type and palladium loading on the activity and selectivity in n-butane conversion. In addition, only trace amounts of aromatic compounds were detected, while heavy liquid products were not observed. This may suggest that the formation of coke precursors under the studied conditions was limited. However, a more reliable evaluation of catalyst stability requires longer catalytic tests together with characterization of the spent catalysts.

Despite the formation of by-products, the process shows relatively high efficiency toward the formation of valuable $C_2$-$C_4$ olefins. In particular, the 1% Pd/γ-$Al_2O_3$ catalyst provided 45% n-butane conversion and a 31% olefin yield at 600 °C. At the same time, the by-products formed during the reaction, including hydrogen and light alkanes, may also be considered useful for petrochemical and refining applications. It should also be noted that in industrial practice unreacted n-butane can be recycled back into the process, which makes it possible to increase the overall conversion of the feedstock while maintaining sufficiently high selectivity toward olefins.

Table 6 – The conversion of n-butane on palladium-containing catalysts (T = 600 °C) and their specific surface area

| Catalyst | Actual content of Pd, % | Specific surface area, $m^2/g$ | Conversion, % | Yield of olefins, % |
|---|---|---|---|---|
| 1% Pd/γ-$Al_2O_3$ | 1,23 | 232 | 45 | 31,0 |
| 3% Pd/γ-$Al_2O_3$ | 3,18 | 221 | 36 | 23,9 |
| 1% Pd/$SiO_2$ | 0,86 | 355 | 23 | 16,3 |
| 3% Pd/$SiO_2$ | 2,58 | 322 | 23 | 15,5 |

**Conclusion**

The obtained results show that palladium catalysts can effectively convert n-butane into $C_2$-$C_4$ olefins. Their performance depends strongly on both the palladium loading and the type of

support. The 1% Pd catalysts demonstrate better activity and selectivity than the 3% Pd samples, mainly due to improved metal dispersion and a higher number of accessible active sites, while higher loading promotes particle aggregation and side reactions. Among the studied systems, Pd/γ-$Al_2O_3$ exhibits the best catalytic behavior, which can be attributed to stronger metal-support interactions and the contribution of surface acid-base sites to C-H bond activation. Overall, the 1% Pd/γ-$Al_2O_3$ catalyst provides the most balanced combination of activity and selectivity and appears promising for the efficient utilization of light alkanes as feedstock for petrochemical production. The obtained results contribute to the understanding of structure-performance relationships in Pd-based dehydrogenation catalysts and may serve as a basis for rational design of selective systems for light alkane.

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**Д.Б. Каримова[1,2*], А.А. Восмериков[2], С.Р. Конуспаев[3], Л.Н. Восмерикова[2], А.В. Восмериков[2]**

[1]Әл-Фараби атындағы Қазақ Ұлттық университеті,
050040, Алматы, Қазақстан, аль-Фараби даңғылы, 71
[2]РҒА СБ Мұнай химиясы институты,
634055, Томск, Ресей, Академиялық даңғылы, 4
[3]Ә.Б. Бектұров атындағы Химия ғылымдары институты,
050010, Алматы, Қазақстан, Ш.Улиханов көшесі,106
*e-mail:dana.tleugabylova@mail.ru


**Pd/γ-$Al_2O_3$ ЖӘНЕ Pd/ $SiO_2$ КАТАЛИЗАТОРЫ ҚАТЫСЫНДА Қ-БҰТАННЫҢ ТӨМЕНГІ ОЛЕФИНДЕРГЕ АЙНАЛУЫН ЗЕРТТЕУ**

*Мақалада н-бутанның палладийді тасымалдағышқа отырғызылған катализаторларында 400-600 °С температура аралығында, стационарлық жағдайда, атмосфералық қысымда, газдың көлемдік жылдамдығы 250 сағат$^{-1}$ н-бутанның өзгерісі келтірілген. Катализаторларды тасымалдағыштың ылғал сыйымдығы негізінде сіңіру әдісімен даярланды, палладий концентрациясы 1 мен 3%, тасымалдағыш ретінде γ-$Al_2O_3$ және $SiO_2$ (силикагель) алынды. Катализаторлар эксперимент алдында реакторда сутекпен тотықсыздандырылды. Н-бутан дегидрлеуімен бірге крекинг, изомерлену, ыдырау реакциялары жүреді, бұл түзілген олефиндермен (этилен, пропилен, бутен-1, цис- және транс-бутены-2) мен алкандармен (метан, этан, пропан, изобутан) дәлелденеді. Н-бутан изобутанға изомерленді, ал басқа реакциялардың көлемі аз. Контакт газдың құрамындағы анықталған сутектің көлемі дегидрлеу мен ыдырау өнімдеріне эквивалентті. Реакция өнімдерінің құрамында конденсацияланбаған сұйық пентан мен ароматты қосылыстардың ізі табылады. Катализаторлардың меншікті беті БЭТ әдісімен анықталып, СЭМ-ЭДС арқылы бетінің химиялық құрамы анықталды.*

***Түйін сөздер:** катализатор, палладий, алюминий оксиді, кремний оксиді, дегидрлеу, алкандар, қ-бутан, олефиндер.*

**Д.Б. Каримова[1*], А.А. Восмериков[2], С.Р. Конуспаев[3], Л.Н. Восмерикова[2], А.В. Восмериков[2]**
[1]Казахский национальный университет имени аль-Фараби,
050040, Алматы, Казахстан, проспект аль-Фараби, 71,
[2]Институт Химии Нефти СО РАН,
634055, Томск, Россия, проспект Академический, 4
[3]Институт химических наук имени А.Б. Бектурова,
050010, Алматы, Казахстан, улица Ш.Уалиханова,106
*e-mail:dana.tleugabylova@mail.ru.

## ИССЛЕДОВАНИЕ ПРЕВРАЩЕНИЯ Н-БУТАНА В НИЗШИЕ ОЛЕФИНЫ НА КАТАЛИЗАТОРАХ Pd/γ-$Al_2O_3$ И Pd/$SiO_2$

*Представлены результаты конверсии н-бутана на нанесенных палладиевых катализаторах в стационарном режиме в интервале температуры 400-600 °С, при атмосферном давлении и объемной скорости подачи н-бутана 250 ч$^{-1}$. Катализаторы получены методом пропитки по влагоемкости носителей, в качестве которых использовали γ-$Al_2O_3$ и $SiO_2$ (силикагель), концентрация палладия в них составляла 1 и 3%. Установлено, что наряду с дегидрированием н-бутана параллельно протекают процессы крекинга, приводящие к образованию как олефинов (этилен, пропилен), так и алканов (метан, этан, пропан), изомеризации н-бутана в изобутан и другие реакции. Водород, содержащийся в контактных газах, эквивалентен количеству продуктов дегидрирования и деструкции. В следовых количествах в газовой фазе присутствуют несконденсированные жидкие продукты, такие как пентан и ароматические соединения. Катализаторы исследованы методом СЭМ-ЭДС с определением элементного состава отдельных участков поверхности, а также определена площадь удельной поверхности методом БЭТ.*

***Ключевые слова:** катализатор, палладий, оксид алюминия, оксид кремния, дегидрирование, алканы, н-бутан, олефины.*

### Сведения об авторах

**Дана Бекмырзаевна Каримова*** – докторант, КазНУ им.аль-Фараби, г. Алматы, Казахстан; e-mail: dana.tleugabylova@mail.ru. ORCID: https://orcid.org/0000-0002-7013-7585.

**Антон Александрович Восмериков** – к.х.н., Институт химии нефти СО РАН, г. Томск, Россия; e-mail:antonvosmerikov@gmail.com. ORCID: http://orcid.org/0000-0003-2876-8647.

**Сапаркали Ретаевич Конуспаев** – д.х.н., профессор Института химических наук имени А.Б. Бектурова, г. Алматы, Казахстан; e-mail: srkonuspayev@mail.ru.

**Людмила Николаевна Восмерикова** – д.х.н., Институт химии нефти СО РАН, г. Томск, Россия; e-mail: lkplu@ipc.tsc.ru. ORCID: http://orcid.org/0000-0002-9436-6147.

**Александр Владимирович Восмериков** – д.х.н, профессор, директор Института химии нефти СО РАН г. Томск Россия; e-mail: pika@ipc.tsc.ru. ORCID: http://orcid.org/0000-0002-7569-0902.

### Information about the authors

**Dana Bekmyrzaevna Karimova*** – PhD student, al-Farabi Kazakh National University, Almaty, Kazakhstan; e-mail: dana.tleugabylova@mail.ru. ORCID: https://orcid.org/0000-0002-7013-7585.

**Anton Aleksandrovich Vosmerikov** – Candidate of Chemical Sciences, Institute of petroleum chemistry, SB RAS, Tomsk, Russia; e-mail: antonvosmerikov@gmail.com. ORCID: http://orcid.org/0000-0003-2876-8647.

**Saparcali Retaevich Konuspaev** – Doctor of Chemical Sciences, Bekturov institute of chemical sciences, Almaty, Kazakhstan; e-mail: srkonuspayev@mail.ru.
**Lyudmila Nikolaevna Vosmerikova** – Doctor of Chemical Sciences, Institute of Petroleum Chemistry, SB RAS, Tomsk, Russia; e-mail: lkplu@ipc.tsc.ru. ORCID: http://orcid.org/0000-0002-9436-6147.
**Aleksander Vladimirovich Vosmerikov** – Doctor of Chemical Sciences, Institute of Petroleum Chemistry, SB RAS, Tomsk, Russia; e-mail: pika@ipc.tsc.ru. ORCID: http://orcid.org/0000-0002-7569-0902.

**Авторлар туралы мәліметтер**
**Дана Бекмырзаевна Каримова*** – докторант, аль-Фараби ҚазҰУ,Алматы ,Қазақстан; e-mail: dana.tleugabylova@mail.ru. ORCID: https://orcid.org/0000-0002-7013-7585.
**Антон Александрович Восмериков** – Химия ғылымдарының кандидаты, РҒА мұнай химиясы институты, Томск, Ресей; e-mail: antonvosmerikov@gmail.com. ORCID: http://orcid.org/0000-0003-2876-8647.
**Сапарғали Ретаевич Қоныспаев** – Химия ғылымдарының докторы, А.Б. Бектұрова атындағы химия ғылымдары институтының профессоры, Алматы қаласы, Қазақстан Республикасы; e-mail: srkonuspayev@mail.ru.
**Людмила Николаевна Восмерикова** – Химия ғылымдарының докторы,РҒА мұнай химиясы институты, Томск қаласы, Ресей; e-mail: lkplu@ipc.tsc.ru. ORCID: http://orcid.org/0000-0002-9436-6147.
**Александр Владимирович Восмериков** – Химия ғылымдарының докторы, профессор, РҒА мұнай химиясы институтының директоры, Томск қаласы, Ресей; e-mail: pika@ipc.tsc.ru. ORCID: http://orcid.org/0000-0002-7569-0902.

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**Р.Н. Ажигулова[1,2], Б.Х. Мусабаева[3], А.К. Жусупова[1,2], А.Темирова[1], А. Абылайхан[1,2]***
[1]Жану проблемалары институты,
Қазақстан Республикасы, Алматы қ., Бөгенбай батыр көшесі, 172
[2]Әл-Фараби атындағы Қазақ ұлттық университеті,
050040, Қазақстан Республикасы, Алматы қаласы, Әл-Фараби даңғылы, 71
[3]Астана халықаралық университеті,
Қазақстан Республикасы, Астана қаласы, Қабанбай батыр даңғылы, 8
*e-mail: akerkeabylaikhan2@gmail.com

# ТАУ-КЕН ЖӘНЕ МЕТАЛЛУРГИЯ КЕШЕНІНІҢ ТЕХНОГЕНДІК ҚАЛДЫҚТАРЫН БИОЛОГИЯЛЫҚ ШАЙМАЛАУ ӘДІСІМЕН ӨҢДЕУ

***Аңдатпа.*** *Бұл мақалада Қазақстан Республикасының тау-кен металлургия кешенінің техногендік қалдықтары түсті және сирек кездесетін металдардың перспективалы екінші реттік көзі ретінде қарастырылады. Зерттеу нысаны ретінде Балқаш байыту фабрикасының мыс кендерін байыту қалдықтары алынды, олар силикатты матрицамен (Si ≈ 36 %) және Cu (≈0,32 %) мен Zn (≈0,77 %) қалдық мөлшерлерінің болуымен сипатталады. ICP-OES әдісімен олардың химиялық, гранулометриялық және физика-химиялық құрамына кешенді талдау жүргізілді. Шикізатты дайындау процестері, оның ішінде ұсақтау, майдалау және жіктеу зерттелді, сондай-ақ майдалау режимдерінің оңтайландырылуы жүргізілді. Бөлшек өлшемін <40 мкм дейін азайту шаймалау тиімділігін арттырудың негізгі факторы екені көрсетілді. Микрофлюидтік бөлу мен биошаймалауды қамтитын интеграцияланған қайта өңдеу технологиясы ұсынылды. Микрофлюидтік бөлу процесі мақсатты фракцияда мыстың 0,52-0,58 %-ға дейін селективті концентрленуін және 70-75% деңгейінде алынуын қамтамасыз ететіні анықталды. Оңтайлы жағдайларда (pH 1,8-1,9; T ≈ 31-32°C) жүргізілген биологиялық шаймалау нәтижесінде металдарды бөліп алу дәрежесі Cu 60-63 %, Zn – 55-58 % дейін жететіні көрсетілді. Алынған нәтижелер*