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Catalytic Synthesis of High-Molecular Hydrocarbons from Synthesis Gas

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Abstract This study investigates the catalytic synthesis of high molecular weight synthetic hydrocarbons, ranging from pentane to nonadecane, using a flow reactor operating in differential mode. The textural properties of catalysts designed for synthesizing these hydrocarbons from synthesis gas were characterized through nitrogen adsorption and desorption isotherms, employing a Sorbtometer device. Key parameters analyzed include total relative surface area, average particle size, mesopore volume, and pore size distribution, with the BJH method used to determine the micropore and mesopore volumes. The surface morphology of the catalysts was further examined through scanning electron microscopy and light scattering microscopy. The primary objective of this research is to elucidate the kinetic principles governing the synthesis of high molecular weight hydrocarbons from synthesis gas.

Keywords Syngas, High molecular weight hydrocarbons, Catalyst, Kinetic principles, Textural properties

1. Introduction

In order to improve the lifestyle of people around the world, the sudden increase in energy demand in the development of industry and transportation sectors causes some problems [1]. Developing alternative and environmentally friendly ways to produce liquid fuels is a difficult and demanding task. Fischer-Tropsch (FT) synthesis and similar technologies such as gas-to-liquids (GTL), coal-to-liquids (CTL) and biomass-to-liquids (BTL) are alternative methods to replace oil. They can make a positive contribution to the world's energy security and supply. Although the FT process was discovered about a century ago, it is an attractive and alternative source of environmentally friendly liquid hydrocarbon fuels with sulfur-free and aromatic compounds. Fischer-Tropsch synthesis is a catalytic reaction in which syngas (a mixture of CO and H_2) is converted into liquid hydrocarbon fuel [2,3]. FT synthesis has received much attention worldwide in both industrial and scientific fields, with the catalyst being the heart of the process. Catalyst composition and operating parameters such as temperature, pressure, etc. play an important role in FT synthesis, catalyst activity, and product distribution. Thus, it is essential to develop FT catalysts with high activity, selectivity, and stability. Among the intermediate metals, ruthenium, nickel, cobalt, and iron are the most commonly used metals for FT catalyst. Cobalt is an active metal for the synthesis of higher chain hydrocarbons due to its high activity and selectivity to paraffin, low water-gas exchange (WGS) activity, low CO_2-selectivity and less oxygen generation [4-6]. Many researchers have studied the effect of different oxide supports on the activity/selectivity of CO catalysts [7,8]. Al_2O_3, SiO_2, and TiO_2 are the most commonly used materials for Co catalysts, in Co/Al2O3 catalyst, due to the limited reducibility of Co and strong interaction between Co [9-11]. Promoters such as Pd, Pt, Re, and Ru are added to Co catalysts, which promote better Co reduction with strong interactions with nonmetals other than Al_2O_3 [12,13]. Many researchers believe that the Pd promoter improves the adsorption sites of the Co catalyst and thereby increases the rate of hydrogenation in FT synthesis [14-16].

In general, the FTS process involves two main reactions and several side reactions. The synthesis of long-chain paraffin and olefins consists of two main reactions [17-21]:

$$nCO + (2n+1)H_2 \rightarrow C_nH_{2n} + (2+n)H_2O, \quad \Delta H_{298}^0 = -165 kJ/mol \tag{1}$$

$$nCO + 2nH_2 \rightarrow C_nH_{2n} + nH_2O, \quad \Delta H_{298}^0 = -152 kJ/mol \tag{2}$$

$H_2:CO$ when the ratio is high or when hydrogenation is strong, catalysts such as cobalt and nickel are used, and reaction (1) becomes dominant. Unlike; on the other hand, when the H_2:CO ratio is low or hydrogenation catalysts

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such as iron are used, reaction (2) dominates. Depending on the operating conditions and the nature of the catalysts, in addition to the two main reactions, the methanization reaction (3), the reaction of the formation of oxygen compounds of hydrocarbons (4) and the reaction of CO2 formation (5) can occur:

$$CO+3H_2 \rightarrow CH_4+H_2O \tag{3}$$

$$nCO+2nH_2 \rightarrow C_nH_{2n}+(n-1)H_2O \tag{4}$$

$$2nCO+nH_2 \rightarrow C_nH_{2n}+nCO_2 \tag{5}$$

The most active catalysts of the Fischer-Tropsch process are iron, cobalt, nickel and ruthenium. These catalysts ensure the maximum rate of chemical and physical adsorption of syngas components under synthesis conditions. Currently, all operating Fischer-Tropsch synthesis plants use only iron catalysts. ThO_2 and K_2O compounds are used as promoters. These promoters are particularly effective in combination with iron catalysts. Al_2O_3, SiO_2, polystyrene, high-silicon zeolites [22-28], etc. are usually used as carriers in Fischer-Tropsch synthesis, oil and gas processing.

2. Experimental Part

Catalytic properties of catalysts in the synthesis of hydrocarbons from CO and H2 were investigated in a flow isothermal reactor filled with 30 cm^3 of quartz (catalyst layer 10 cm^3) under the following optimal conditions: temperature range 280-300℃, 0.5 MPa and volumetric gas flow rate (GHT) 1000 h^{-1}. Balanced experiments were carried out for at least 150 hours, the composition of the incoming and outgoing gas was analyzed, and the amount of hydrocarbons and water of reaction obtained was recorded.

The activity of hydrocarbon synthesis catalysts was evaluated according to the following indicators: CO conversion, selectivity for hydrocarbons and productivity. The calculation error did not exceed 2.5%. CO conversion was calculated according to the following formula:

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$$X_{CO} \frac{V_{inC(CO)_{in}} - V_{outC(CO)_{out}}}{V_{inC(CO)_{in}}} \cdot 100\% \tag{6}$$

here - V_{in}, V_{out} - gas consumption at the entrance to the reactor and the exit from the reactor, dm^3/hour;

$C(CO)_{in}, C(CO)_{out}$ - concentration of CO at the inlet and outlet of the reactor, unit percentage.

Methane selectivity was calculated according to the following formula:

$$S_{CH4} = \frac{V(CO)_{CH_4}}{V(CO)_{con}} \cdot 100\% \tag{7}$$

here - $V(CO)_{CH_4}$ - the volume of CO used to produce methane, dm^3;

$V(CO)_{con}$ - volume of CO converted to methane, dm^3.

The selectivity for C5+ hydrocarbons was calculated using the formula:

$$S_{C5+} = \frac{V(CO)_{C5+}}{V(CO)_{con}} + 100\% \tag{8}$$

Here $V(CO)_{C5+}$ - S_{C5+} the volume of CO used to form hydrocarbons dm^3.

S_{C5+} - For hydrocarbons, productivity was developed according to the following formula:

$$G_{C5+} = \frac{m_{C5+}}{V_{cat}} \tag{9}$$

Here a m_{C5+} - mass of hydrocarbons, kg;

V_{cat} – catalyst volume, m^3;

τ - time, hour.

2.1. Determination of the Composition of Synthesis Products

The composition of the gaseous synthesis products was analyzed by gas adsorption chromatography and two columns with an active Hysep R phase and NaX molecular sieves in a Crystall 5000 (Chromatek, Russia) chromatograph equipped with a thermal conductivity detector. The assay mode is temperature programmable with a heating rate of 8℃/min.

The composition of C5+ hydrocarbons was determined by the capillary gas-liquid chroma-mass spectrometric method on a gas chromatograph (Agilent, USA) equipped with an MSD 5975C mass-selective detector.

2.2. Texture Characteristics of Catalysts

Textural characteristics of catalysts designed for the production of high molecular synthetic, pentane to nonadecane hydrocarbons from synthesis gas were determined on the basis of absorption and desorption isotherms of nitrogen in the Sorbtometer device, based on total specific surface area, average particle size, mesopore size, and pore size distribution. The volume of micropores and mesopores was determined by the BJH method. The surface-to-surface ratio was calculated by the Brunauer-Emmett-Taylor (BET method):

$$G = \frac{G_m Cp / p_s}{(1 - p / p_s)[1 + (C - 1) p / p_s]} \tag{10}$$

Kelvin's equation is used to find the pore radius.

$$\ln = \frac{P_\gamma}{P_5} - \frac{2V_m \sigma}{rRT} \tag{11}$$

The radius of the pores of the catalyst designed for the production of high molecular weight synthetic hydrocarbons from pentane to nonadecane from synthesis gas was determined by the absorption and desorption isotherms of benzene vapours:

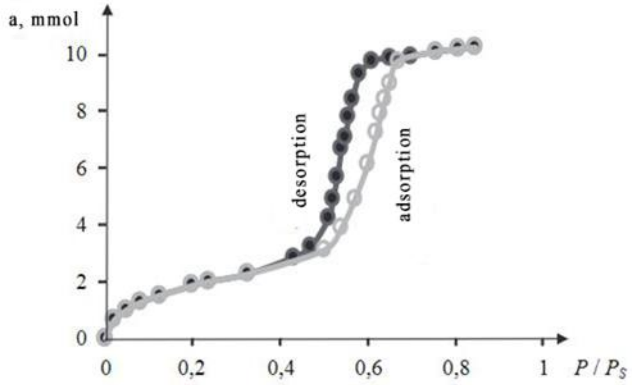

Figure 1. Absorption and desorption isotherms of benzene vapors

The dynamic light scattering method was used to measure the particle size. This method is distinguished from other methods by its expressiveness and can be used to determine the main laws in the formation of particles.

2.3. Surface Morphology of Catalysts Designed for the Production of High Molecular Weight Synthetic Hydrocarbons from Pentane to Nonadecane from Syngas

The morphology of the surface of the catalysts used in synthesis gas, i.e. in synthesis gas, for obtaining high molecular synthetic hydrocarbons from synthesis gas, from pentane to nonadecane, in oxidized and reduced forms, by scanning electron and radiation microscopy method was studied.

According to light microscopy data, changes in the composition of catalysts designed for the synthesis of high-molecular-weight synthetic hydrocarbons from pentane to nonadecane do not affect the distribution of cobalt particles (Fig. 2), the size of the initial gas and high-molecular synthetic hydrocarbons from hydrogen, from pentane to nonadecane Co- Fe-B-Zr/HSZ Synthesis-gas high molecular synthetic, from pentane. The average value of cobalt particles in the catalyst designed for the production of hydrocarbons up to nonadecane corresponds to the size of 8.3 nm, which is 3-13 nm.

3. Results and Discussion

3.1. The Effect of the Method of Preparing a Catalyst for the Production of High-Molecular Hydrocarbons from Synthesis Gas with High Catalytic Activity on the Reaction Yield in the Synthesis of High-Molecular Liquid Synthetic Hydrocarbons

Preparation of catalysts designed for obtaining high molecular weight hydrocarbons from synthesis gas with high catalytic activity selected for obtaining high molecular weight liquid synthetic hydrocarbons from syngas can be carried out in one or more steps. In the latter case, the active ingredients were introduced to the surface of the carrier by successive absorption with aqueous solutions of the respective salts. In our experiments on the effect of the catalyst activity enhancer for obtaining high molecular weight hydrocarbons from synthesis gas with high catalytic activity on the activity of the catalyst intended for obtaining high molecular weight hydrocarbons from synthesis gas with high catalytic activity selected for obtaining high molecular weight liquid synthetic hydrocarbons from synthesis gas the activity of a catalyst designed to obtain high molecular weight hydrocarbons from syngas with high catalytic activity zirconium by three consecutive absorptions of samples prepared with the introduction of a raising agent. Such a methodology does not create difficulties for laboratory tests, for which small amounts of the catalyst intended for obtaining high molecular weight hydrocarbons from synthesis gas with high catalytic activity are required.

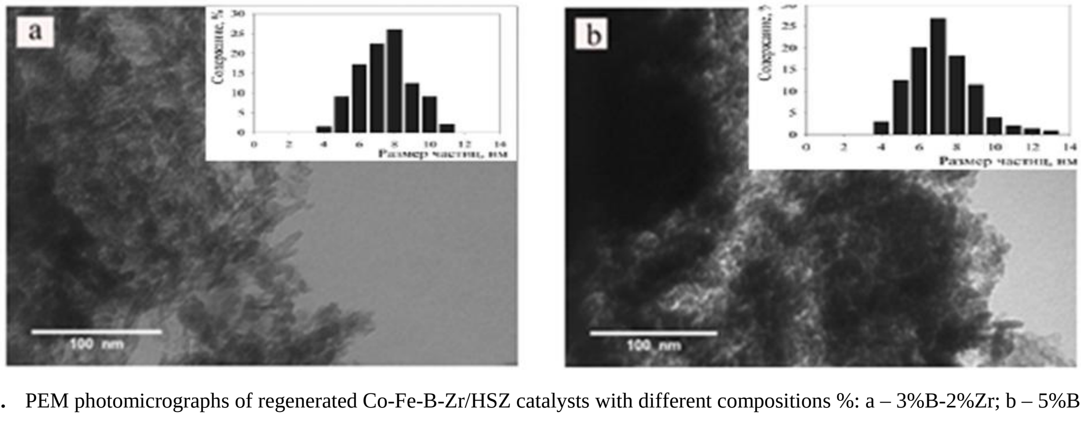

Figure 2. PEM photomicrographs of regenerated Co-Fe-B-Zr/HSZ catalysts with different compositions %: a – 3%B-2%Zr; b – 5%B-0.5%Zr

Tests were conducted under the following optimal conditions: pressure 0.1 MPa, volume velocity 100 h^{-1}. The results were compared with the sample values prepared in 3 dilutions.

As can be seen in Fig. 3, the sample prepared in 3-fold digestion was more active in the entire temperature range - the greenhouse gas conversion in it was 5-10% higher than that of the other two samples. In this case, the samples prepared 1 and 2 times were almost indistinguishable from each other.

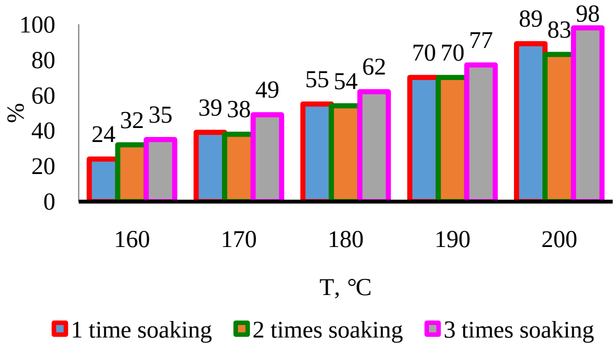

Figure 3. Dependence of heat gas conversion on synthesis temperature

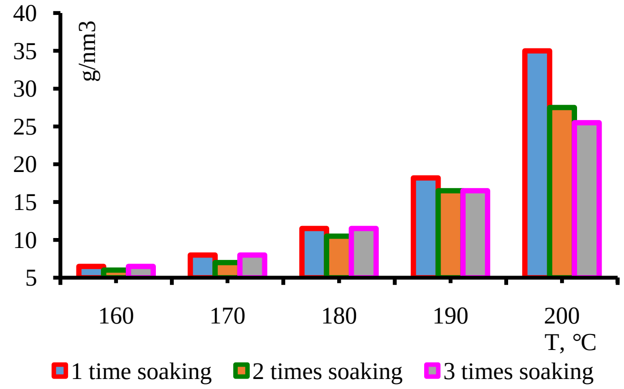

Figure 4. Dependence of methane yield on synthesis temperature

Temperature dependences of methane yield for all three samples are shown in Fig. 4. Methane production in the lower temperature range was not significantly different for all three samples. Thus, the yield of methane was 1 g/m3 at 160℃ and 5-6 g/m^3 at 180℃. Only synthesis gas, i.e. production of high-molecular liquid synthetic hydrocarbons from synthesis gas, at 200℃ temperature, methane production in the sample prepared in 1 digester differed significantly: in it, methane yield was 30 g/m3 compared to 20-22 g/m^3 for the remaining samples.

The yield of ethane, propane, butane and ethylene, propene, butenes did not differ nearly for all three samples of the catalyst designed for obtaining high molecular weight hydrocarbons from synthesis gas with high catalytic activity. At 160℃, ethane, propane, butane and ethylene, propene, butenes were detected in residual amounts in the exhaust gas. As the temperature increased, the yield gradually increased and reached 10-13 g/m^3 at 200℃ (Figure 5).

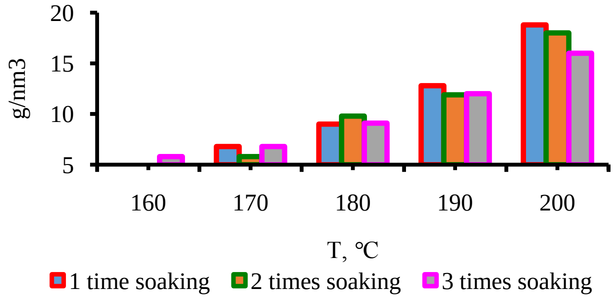

Figure 5. Dependence of the yield of ethane, propane, butane and ethylene, propene, butenes on synthesis temperature

Figure 6 shows the dependence of the yield of liquid synthetic hydrocarbons on the temperature of the process of obtaining high molecular liquid synthetic hydrocarbons from heat gas and hydrogen. As a result of experimental studies, it was found that the yield of liquid synthetic hydrocarbons increased with increasing temperature in obtaining high molecular liquid synthetic hydrocarbons from carbon dioxide and hydrogen and reached a maximum at 190℃. 136, 140, 136, 140, It was 145 g/m^3.

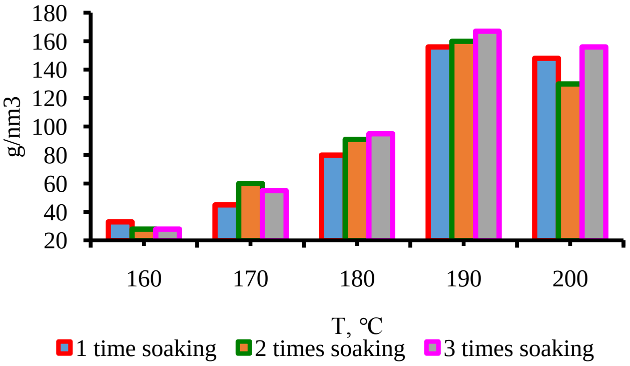

Figure 6. Dependence of yield of liquid synthetic hydrocarbons on synthesis temperature

The selectivity for the formation of methane in the production of high molecular liquid synthetic hydrocarbons from carbon dioxide and hydrogen remained almost unchanged, at the level of 6-7% (Table 1).

Table 1. Effect of the selected catalyst preparation method for the production of high molecular weight liquid synthetic hydrocarbons from 20%Co-20%Fe-5%B-1.5%Zr/HSZ synthesis gas on the synthesis performance

Number of wins method	T, °C	K_{CO}>%	Yield, g/m^3				S_{CH4}, %	Spentane and higher molecular liquid synthetic hydrocarbons,%
			CH_4	C_2-C_4	C_{5+}	CO_2		
1win times	190	74	14	10	139	16	8	86
2times	190	75	12	9	141	14	7	87
3 times	190	81	11	9	147	13	6	88

Table 2. Effect of the preparation method of the selected catalyst for the production of high molecular weight liquid synthetic hydrocarbons from 20%Co-20%Fe-5%B-1.5%Zr/HSZ synthesis gas on the composition of the catalyst

Number of wins	Composition of liquid synthetic hydrocarbons.%			Content of saturated carbohydrates, %		
	olefin	n-alkane	i-alkane	C_5-C_{10}	C_{11}-C_{18}	C_{19+}
1 time	9	60	35	64	36	8
2 times	9	64	31	59	35	11
3 times	9	76	19	42	44	19

Table 3. Effect of catalyst activity enhancer content in the selected catalyst for production of high molecular weight liquid synthetic hydrocarbons from UCC synthesis gas on hydrocarbon synthesis parameters

Composition Zr, %	find°C	K_{CO}, %	Yield, g/m³•h				S_{CH_4}, %	Spentane and higher molecular liquid synthetic hydrocarbons,%
			CH_4	C_2-C_4	C_{5+}	CO_2		
1	200	72	21	20	101	14	15	72
3	190	76	14	9	138	14	8	87
5	200	69	18	21	105	10	13	75

When the number of digestions decreases, the content of isosaturated hydrocarbons formed in the catalyst almost doubles and reaches 33% in a sample prepared for one digestion (Table 2). At the same time, there are no changes in the function of the catalyst for obtaining high molecular weight synthetic hydrocarbons from syngas with high catalytic activity.

Thus, simplifying the preparation procedure will slightly reduce the catalyst activity, the yield of the target liquid synthetic hydrocarbons, and the selectivity of their formation. The average molecular mass of hydrocarbons also decreases. The selectivity of methane formation remains unchanged. Reduction of the number of absorptions 20%Co-20%Fe-5% B-1.5%Zr/HSZ selected catalyst for obtaining high molecular weight synthetic hydrocarbons from synthesis gas with high catalytic activity, its surface acidity despite the absence of changes, increases the isomer capacity.

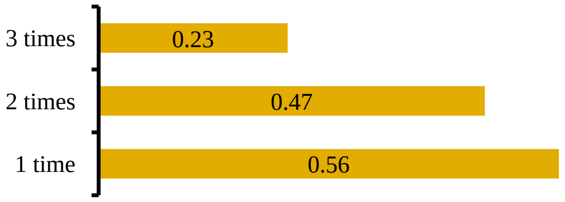

Figure 7. Effect of amount of ingestion on iso/n-unsaturated carbohydrate ratio

3.2. Effect of B-Zr Content in the Catalyst Selected for the Production of High Molecular Weight Liquid Synthetic Hydrocarbons from 20%Co-20%Fe-5% B-1.5%Zr/HSZ Synthesis gas on Hydrocarbon Synthesis

The material additive composition of the catalyst designed to obtain high molecular hydrocarbons from synthesis gas with high catalytic activity, which increases the activity, affects the synthesis of high molecular synthetic hydrocarbons from carbon dioxide and hydrogen, from pentane to nonadecane. The sequence of unloading the components also has a significant impact on the process. 5%B-1%Zr/HSZ for the highest activity of the catalyst designed to obtain high molecular weight synthetic hydrocarbons from synthesis gas with high catalytic activity with the presence of a substance that increases the activity of the catalyst intended for The amount of the substance increasing the activity of the catalyst intended for obtaining high molecular hydrocarbons from synthesis gas with high catalytic activity affects the activity of the catalyst intended for obtaining high molecular hydrocarbons from synthesis gas with high catalytic activity selected for obtaining high molecular liquid synthetic hydrocarbons from synthesis gas. Thus, the sample containing 1% Zr and 5% B was more active. It achieved ~20% higher conversion compared to two other syngas high molecular weight liquid synthetic hydrocarbon catalysts with high catalytic activity selected for synthesis gas high molecular weight hydrocarbons.

Tables 3, and 4 show performance indicators of catalysts designed for obtaining high molecular weight hydrocarbons from synthesis gas with high catalytic activity selected for obtaining high molecular weight liquid synthetic hydrocarbons from synthesis gas. Thus, the selectivity for C5+ in the optimum Zr (3%) content was 85% against 74-74% in the 0.5% content of catalyst 2 and 0.5%, which is intended for the production of high molecular hydrocarbons from syngas with high catalytic activity (3- table).

Table 4. Effect of catalyst Activity enhancer content on liquid synthetic hydrocarbons

Zr, %	olefins	n-alkanes	i-alkanes	C_5-C_{10}	C_{11}-C_{18}	C_{19+}
1	10	62	33	67	31	8
3	9	60	36	63	34	9
5	8	54	41	65	32	8

The amount of unsaturated ethylene series compounds in the catalyst was 8.5-9%, which did not change regardless of the zirconium oxide content. The sample with the highest amount of catalyst activity enhancer, designed to obtain high molecular hydrocarbons from syngas with high catalytic activity, showed the highest ability: the content of isostructural hydrocarbons in the catalyst was 40% (31-33% in two other samples). The group composition of the catalyst was almost independent of the content of the substance that increases the activity of the catalyst designed to obtain high molecular hydrocarbons from synthesis gas with high catalytic activity: the content of the gasoline group was 60-65%, the content of solid saturated hydrocarbons was 7-8% (Table 4).

3.3. Effect of Total Pressure on the Synthesis of High Molecular Weight Synthetic, Pentane to Non-Decane Hydrocarbons from Carbon Dioxide and Hydrogen

20%Co-20%Fe-5%B-1.5%Zr/HSZ is selected by us for obtaining high molecular liquid synthetic hydrocarbons from synthesis gas with high catalytic activity in the catalyst designed for obtaining high molecular hydrocarbons from synthesis gas and hydrogen effect on the synthesis of high molecular weight synthetic hydrocarbons from pentane to nonadecane was studied. Experiments on the synthesis of high molecular liquid synthetic hydrocarbons from carbon dioxide and hydrogen were carried out at a pressure of 0.5-0.6 MPa and a volume velocity of 2000-2500 h-1, raising the temperature from 60 to 190℃. When this temperature was reached, the pressure was increased to 2.0 MPa in 0.5 MPa steps.

An increase in the activity of the catalyst selected for obtaining high molecular weight synthetic liquid hydrocarbons from syngas with high catalytic activity was observed when the pressure was increased - the gas conversion increased from 11% at 0.5 MPa to 19% at 2.0 MPa. The increase in activity was observed with the production of liquid synthetic hydrocarbons. As the pressure increased from 0.5 to 2.0 MPa, the yield increased from 28 to 41 g/m3. Accordingly, the productivity of liquid synthetic hydrocarbons also increased from 55 to 89 kg/m3 cat.h (Figures 8, 9).

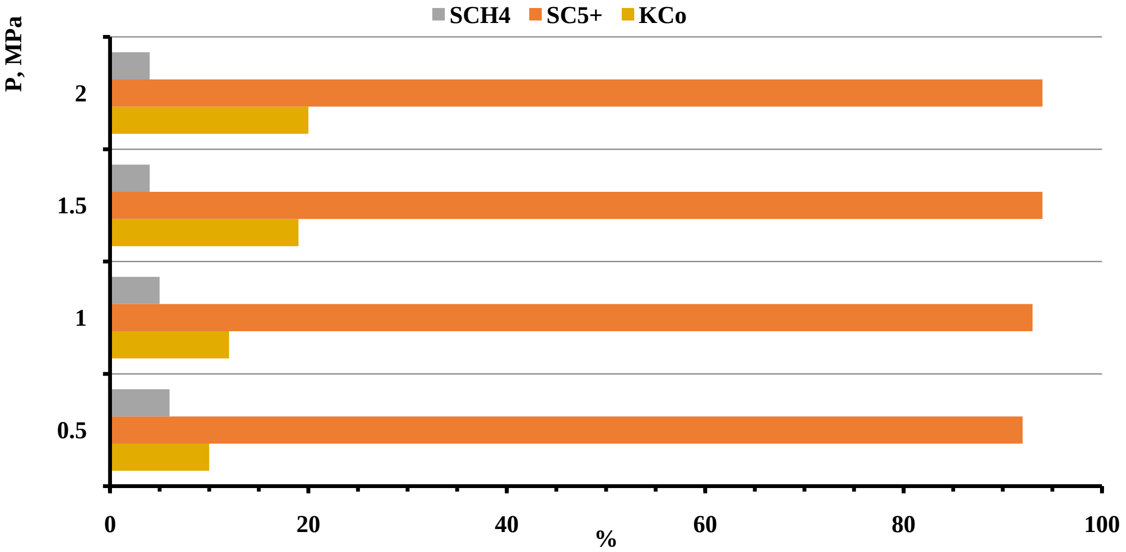

Figure 8. Effect of pressure on the activity and selectivity of a selected catalyst for the production of high molecular weight liquid synthetic hydrocarbons from 20%Co-20%Fe-5%B-1.5%Zr/HSZ synthesis gas. T=190℃, space velocity 2000 h^{-1}

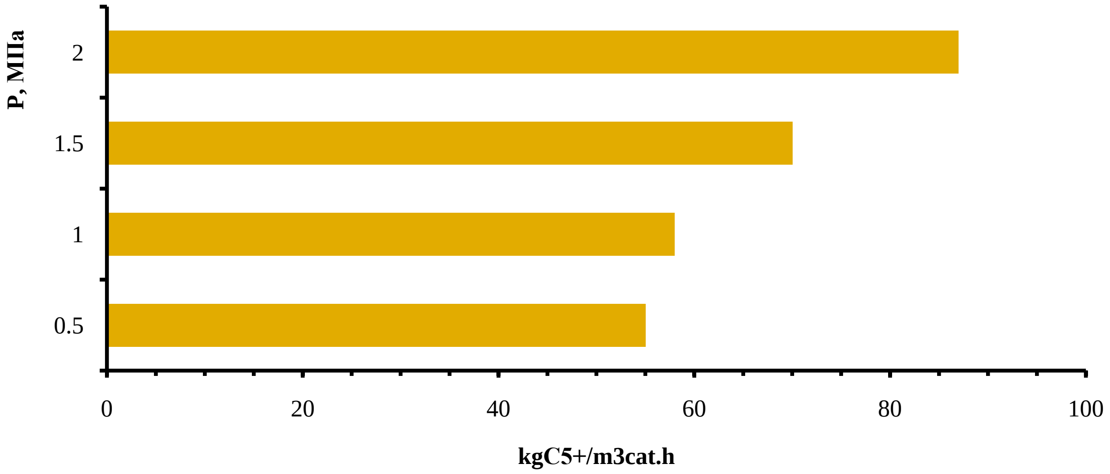

Figure 9. Effect of pressure on the performance of a selected catalyst for the production of high molecular weight liquid synthetic hydrocarbons from 20%Co-20%Fe-5%B-1.5%Zr/HSZ synthesis gas. T=190℃, space velocity 2000 h^{-1}

Table 5. Effect of pressure on the composition of hydrocarbons obtained over a selected catalyst for the production of high molecular weight liquid synthetic hydrocarbons from 20%Co-20%Fe-5%B-1.5%Zr/HSZ synthesis gas

P, atm	LiquidGroup composition of UVs, %			Content of saturated carbohydrates, %			α
	Olefins	n-alkanes	i-alkanes	C_5-C_{10}	C_{11}-C_{18}	C_{19+}	
5	8.4	88.7	6.2	37.5	45.8	28.7	0.93
10	7.5	92.6	6.1	32.4	42.6	32.5	0.95
15	8.4	89.5	6.4	34.8	43.5	29.6	0.93
20	9.2	88.4	6.6	33.4	44.7	27.4	0.92

When the pressure increased from 0.5 to 2 MPa, the selectivity to higher molecular weight liquid synthetic hydrocarbons than pentane increased from 93 to 96% (Fig. 8).

4. Conclusions

The obtained data are placed into certain laws of the effect of pressure on the activity and selectivity of catalysts designed for obtaining high molecular hydrocarbons from synthesis gas with high catalytic activity selected for the synthesis of high molecular liquid synthetic hydrocarbons from synthesis gas.

Thus, the catalytic synthesis of high molecular weight synthetic hydrocarbons from carbon dioxide and hydrogen, from pentane to nonadecane. The process was carried out in a flow reactor operating in differential mode. Textural characteristics of catalysts designed for obtaining high-molecular-weight synthetic hydrocarbons from pentane to nonadecane from syngas were determined. The volume of micropores and mesopores was determined by the BJH method. The morphology of the surface of the catalysts designed for the production of high molecular weight synthetic hydrocarbons from pentane to nonadecane from syngas was studied by scanning electron microscopy and light microscopy.

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