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INFLUENCE OF THE TYPE OF SUPPORT ON THE PHYSICOCHEMICAL PROPERTIES OF COBALT CATALYSTS

Abstract. The article presents the results of studies of the physicochemical properties of cobalt catalysts on a zeolite support. For research, BET methods were used. The nature of the influence of the nature of the support on the structural (specific surface area, dispersion of metal cobalt, size of metal crystallites) and chemical (degree of cobalt reduction) properties of catalysts has been established.

Keywords: Catalyst, Fischer-Tropsch, zeolite.

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Introduction. Recently, due to limited oil reserves, the production of valuable chemical products, for example, high-octane motor gasoline, from non-oil raw materials (coal, natural gas, peat, biomass) has become relevant [1]. The synthesis of hydrocarbons from CO and H_2 (Fischer–Tropsch synthesis) in the industry of a number of countries is carried out using cobalt catalysts, which are the most selective catalytic systems with respect to the formation of normal alkanes. Liquid hydrocarbons obtained by this method, unlike petroleum hydrocarbons, do not contain impurities of aromatic, sulfur- and nitrogen-containing compounds.

Conditions and methods of research. In this regard, the processing of gases mixed with oil is becoming one of the most urgent problems. However, water as a fusion product has a major impact on the environment of a synthetic fuel company. Water is an integral product of Fischer-Tropsch synthesis.

Research results. Accumulation of water in the reaction medium leads to oxidation and burning of active metal particles and can cause rapid deactivation of the catalyst [2,3]. One of the ways to overcome the negative effect of water in the Fischer-Tropsch process is to adjust the hydrophilicity/hydrophobicity of the catalyst. At the same time, it is effective to use zeolites as a carrier for catalysts [4,5]. Therefore, it was aimed to study the properties of cobalt catalysts for Fischer-Tropsch synthesis, including the cationic forms of different types of zeolites, in particular, the effect of the type and form of zeolite on the main synthesis parameters.

Composite catalysts for Fischer-Tropsch synthesis consisting of zeolites of various forms were created in the work, and some of their properties were characterized by physico-chemical methods.

In the work, all catalyst samples were prepared by the impregnation method. Conventionally, the catalysts were designated as Co/KA, Co/CaA, Co/NaX. Mass fraction of CO in all catalysts was 20%. The composition of prepared catalysts is shown in Table 1.

Table 1

Composition of prepared catalysts

Title	Zeolite	Si/Al	Method of obtaining	Catalyst composition			
				Co	Zeolite	Met. Al	A linker
Co/KA	KA	1	impregnation method	20	24	40	Other things
Co/CaA	CaA	1					
Co/NaX	NaX	2					

To prepare the carrier, a solution of HNO_3 (64%) and distilled water was prepared in a glass beaker. 2 g of boehmite and 1 g of zeolite were thoroughly mixed in a porcelain beaker, and then the acid solution was slowly added to the mixture. The resulting mixture was thoroughly mixed until a homogeneous gel-like mass was formed. At this time, 4 g of metallic aluminum powder was weighed into a separate container. 1.13 g of TEG and 2.25 g of ethyl alcohol were added to the resulting mixture. Carefully, without applying force, mix so that the aluminum is evenly moistened with a mixture of alcohol and TEG. A mixture of prepared aluminum powders was added to the mixture and thoroughly mixed until a uniform consistency of soft and plastic dough was obtained. Next, tablets with a diameter of 2.5 mm were prepared. To do this, tablets of a homogeneous dough-like consistency were prepared using medical syringes. The prepared tablets were placed in flat containers and air dried for 15 hours to ensure air flow was not obstructed. The tablets, air-dried for 15 hours, were fired in a muffle furnace.

Discussion of research results. The pore volume of the catalysts and their specific surface area were determined from 30 to 100 Å (mesopores). The structural parameters of the porous structure of the catalysts (mesopore volume, specific surface area) were determined by nitrogen sorption using a NOVAVin installation (Quantachrome Instruments) at low temperature. And the size of the macropores of the samples was determined by measuring the moisture content of the granules. Since all samples differ only in the type of zeolite, and the other components are the same, it is possible to evaluate the influence of the nature of the zeolite on the secondary structure of the pore system (mesopore region). In Table 2 shown data characterizing the porous system of the studied catalysts.

Table 2

Data on pore volume of samples

Catalyst	Macropore volume, cm^3/g	Mesopore volume, cm^3/g	Diameter of pores determining the volume of mesopores, Å	Specific surface area, m^2/g
Co/KA	0,410	0,071	67	46
Co/CaA	0,631	0,098	37	93
Co/NaX	0,632	0,087	38	75

It has been shown that when KA zeolites are added to the catalyst, the macropore volume of the resulting samples increases twofold, and when zeolites CaA and NaX are added, the volume increases threefold. Pore sizes of these sizes were determined by pore diameters close to 37–39 Å for both zeolites, except zeolite KA. The introduction of NaX zeolite into the catalyst increased the volume of macropores by 1.8 times, and CaA zeolite – by 1.4 times.

With the introduction of CaA and NaX zeolites, the volume of mesopores increased by 1.3 times, and with the introduction of CA zeolite it decreased by 1.4 times. The mesoporous volume of the Co-NaX catalyst was determined by pores with a diameter of 38 Å, the pores of the Co-KA catalyst – 67 Å, and the Co-CaA pores – 37 Å.

Conclusion. The surface area of cobalt catalysts with zeolites cationized with alkali and alkaline earth metals varies depending on the composition of the zeolites. It is known from the literature that the larger the surface area of the catalyst, the more active it is in the Fischer-Tropsch synthesis [6]. It has been shown that the introduction of zeolite into the composition of a composite cobalt catalyst makes it possible to change the nature of the catalyst, which can be used as a method of controlling the effect of water on the active catalytic surface in Fischer-Tropsch synthesis.

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КОБАЛЬТ КАТАЛИЗАТОРЛАРЫНЫҢ ФИЗИКАЛЫҚ ЖӘНЕ ХИМИЯЛЫҚ ҚАСИЕТТЕРІНЕ ТАСЫМАЛДАУШЫ ТҮРІНІҢ ӘСЕРІ

Аңдатпа. Мақалада цеолиттік тірекке кобальт катализаторларының физикалық-химиялық қасиеттерін зерттеу нәтижелері берілген. Зерттеу үшін BET және IR спектроскопия әдістері қолданылды. Тірек табиғатының катализаторлардың құрылымдық (меншікті бетінің ауданы, металл кобальттың дисперсиясы, металл кристаллиттерінің өлшемі) және химиялық (кобальттың тотықсыздану дәрежесі) қасиеттеріне әсер ету сипаты белгіленді.

Тірек сөздер: катализатор, Фишер-Тропш, цеолит.

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ВЛИЯНИЕ ТИПА НОСИТЕЛЯ НА ФИЗИКО-ХИМИЧЕСКИЕ СВОЙСТВА КОБАЛЬТОВЫХ КАТАЛИЗАТОРОВ

Аннотация. В статье представлены результаты исследования физико-химических свойств кобальтовых катализаторов на цеолитовом носителе. Для исследования использовались методы БЭТ. Установлен характер влияния природы носителя на структурные (удельная поверхность, дисперсность металлического кобальта, размер кристаллитов металла) и химические (степень восстановления кобальта) свойства катализаторов.

Ключевые слова: катализатор, Фишера-Тропша, цеолит.