CATALYTIC AROMATIZATION OF NATURAL AND PETROLEUM GASES

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Abstract: Hydrocarbon aromatization reaction, discovered in the 80s of the 20th century, is one of the rapidly developing and advancing directions of catalysis. As a result of the research conducted in a short period of time, a large number of effective catalysts were developed, which allowed to obtain C_2 - hydrocarbons with a yield of 30-35%. In this article, problems such as choosing a catalyst for the process of catalytic aromatization of natural and petroleum satellite gases, studying the laws of the reaction, modeling and optimization of the process are highlighted.

Key words: Natural gas, air, petroleum gases, gas condensate, motor fuel, aromatic hydrocarbon, benzene, toluene, xylene.

Introduction. Based on the above, it is urgent to create new methods of production of synthetic substances important for the national economy, as well as waste-free, ecologically clean, high-quality and competitive technologies based on local raw materials and industrial waste.

At the same time, one of the promising ways to choose a catalyst for the process of catalytic aromatization of natural and petroleum gases is the one-step production of C_2 hydrocarbons from methane, which are distinguished by their high economic efficiency.

Research method: Based on the set goals and tasks, the following methods of investigation were used:

1. Thermodynamics.

2. Kinetic.

3. Gas chromatography.

Due to the development of automobile transport, the extensive use of petroleum fuels is causing fuel shortages and environmental problems on a global scale. This requires solving global problems such as the use of non-petroleum fuel products for motor transport, the search for alternative energy sources and their introduction into industry. Among the available non-oil sources of hydrocarbons - coal, natural gas, wood, biomass, etc. Natural gas is the most promising for the production of liquid fuels. Based on the above, the purpose of this work is to obtain export-oriented environmentally friendly liquid fuels that replace imports by catalytic aromatization of natural and petroleum gases, which are the source of local raw materials, and to create



energy and resource-saving technologies. At present, a number of scientific and research works are being carried out on obtaining motor fuels and aromatic hydrocarbons from natural gas, petroleum gases and gas condensates with the participation of zeolite catalysts. In order to obtain liquid fuels from natural and petroleum satellite gases, we conducted experiments in a flow reactor in the range of $350-8500^{\circ}$ C, at normal atmospheric pressure, with a volume velocity of natural gas of 300-1500 h⁻¹. The reaction products were analyzed by gas chromatography. As a research object, natural gas and an artificial mixture with the following composition were used: methane - 40.0%, ethane - 10%; propane - 25.0%, normal and isobutanes - 20.3%, pentanes - 4.7%. Bentonite, a local raw material rich in silicon, was used as the core substance (nositel) for the reaction.

Sol-gel technology is a promising method of catalyst preparation for obtaining motor fuels and aromatic hydrocarbons from methane (natural gas) and petroleum gases. The use of nanocatalysts showing high catalytic activity, selectivity and stability leads to the improvement of the environmental characteristics of processes and technologies in industry, the reduction of harmful waste emitted into the atmosphere, and the creation of environmentally friendly types of alternative energy resources. Catalyst for the direct aromatization reaction of C_2 - C_5 hydrocarbons was prepared by the "Sol-gel" method as follows. Aqueous solutions of Zr(NO₃)₄, ZrO(NO₃)₂ salts were used for sol-gel synthesis of nanosized particles. The required amount of reagents was withdrawn and dissolved in bidistilled water to which gel-forming additives: citric acid and ethylene glycol were added. The resulting mixture was thoroughly stirred at 80°C. Then it was thermally treated at 200°C for 2 hours and at 500-1000°C for 3 hours. Spatial composition of the resulting catalyst was checked by X-ray structural method in "DRON-3M" diffractometer (CuK_{α} - light source). In the presence of bentonite containing 4.0% Mo, the transformation of lower alkanes into aromatic hydrocarbons begins at 400°C, and when the temperature reaches 450-500°C, the expected substances (benzene, toluene, xylene) are formed. Methane and ethane, a small amount of hydrogen and C_2 - C_4 alkenes are formed as by-products. As the temperature increases, the conversion of the initial mixture and the selectivity of the formation of aromatic hydrocarbons increase. When the volumetric rate is increased from 500 to 600 h^{-1} , the conversion of 550°C and $C_2 - C_5$ hydrocarbons and the selectivity of the formation of aromatic hydrocarbons decrease. When the temperature is increased to 550°C, the conversion of lower alkanes and the yield of aromatic hydrocarbons increase. At the same time, the selectivity of cracking products (C1-C2 alkanes) and lower olefins decreases. When the temperature is increased from 550°C, the yield of the reaction and the selectivity of formation of aromatic hydrocarbons decrease.

The influence of various factors on the yield of aromatization reaction of lower alkanes was studied and the following optimal conditions were selected: $T = 550^{\circ}C$,



volumetric rate 500 hours. In the formation of aromatic hydrocarbons, the activity of the catalyst containing 4.0% molybdenum increases when it is modified with various metals. Therefore, we added zinc, gallium, zirconium, manganese, and iron metals to it and studied their catalytic activity. It was found that the production yield of aromatic hydrocarbons from C₂-C₅ hydrocarbons is higher when bentonite is modified with Zr, Zn and Ga. Under the selected optimal conditions, the conversion of the synthetic mixture reaches 48.3%, and the yield of aromatic hydrocarbons reaches 41.6% when 550° C and V_{mixture} = 500 h⁻¹. The selectivity for aromatic hydrocarbons was 86.1%.

Conclusions. Catalytic conversion of methane to aromatic hydrocarbons was almost not observed when we used the selected catalyst for aromatization of natural gas under conditions of $T = 550^{\circ}$ C, volume velocity 500 h⁻¹. In the presence of this catalyst, the influence of some factors on the reaction rate of the catalytic conversion of methane to aromatic hydrocarbons (volumetric velocity, temperature, contact time, height of the catalyst layer, mass ratios of the active components of the catalyst) was studied, and as a result, the following optimal reaction conditions were selected: T = 750°C, volumetric velocity V_{CH4} = 1000 hour⁻¹; the height of the catalyst layer is 10 cm. As a result of research, an optimal catalyst with the following composition was selected: 5.0% Mo, 1.0% Zn, 1.0% Zr. The yield of aromatic hydrocarbons in this catalyst is 40.4% when V_{CH4} = 1000 h⁻¹, T = 750°C.

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