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KINETICS OF THE CATALYTIC DEHYDROAROMATIZATION OF METHANE

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Аннотация

В этой работе изучено влияние различных факторов на конверсии метана, селективность и выход продуктов реакции дегидроароматизации метана. Катализатор дегидроароматизации метана 5%Mo/Бентонит был приготовлен методом твердофазного синтеза путем смешения бентонита с MoO_3 и последующего прокаливания и активации.

Ключевые слова: Метан, дегидроароматизация, катализатор, цеолит, бентонит.

Abstract. In this work, the effect of various factors on methane conversion, the selectivity and yield of the products of the methane dehydroaromatization reaction were studied. The methane dehydroaromatization catalyst 5% Mo/Bentonite was prepared by solid-phase synthesis by mixing bentonite with MoO_3 and subsequent calcination and activation.

Keywords: Methane, dehydroaromatization, catalyst, zeolite, bentonite.

Introduction

A number of research works are currently underway to produce motor fuels and aromatic hydrocarbons in the presence of zeolite catalysts from natural and associated petroleum gases. The main component of the composition of natural gas is methane, thermodynamically stable and resistant to the action of many reagents. Direct synthesis based on methane is very difficult, but its products easily react and are more active than methane. The reaction proceeds on zeolite-containing catalysts. In this process, the oxidizing agent is not involved [1-4].

Modern technology is being developed using alternative energy sources and is gradually replacing existing crude oil resources. Natural gas is one of the more promising alternatives that fills the gap between fossil fuels and renewable technology. Because, recent advances in the discovery and recovery of technology have contributed to an increase in excess and a decrease in the price of natural gas,

which increases interest in the use of natural gas. New advances in C1 chemistry and technology expand its utilization of both raw materials for liquid fuels and alternative raw materials for the petrochemical industry.

The conversion of methane to more valuable products is one of the most interesting and complex issues in the field of natural gas exploitation. The catalytic conversion of methane takes place in a direct or indirect way [5-7].

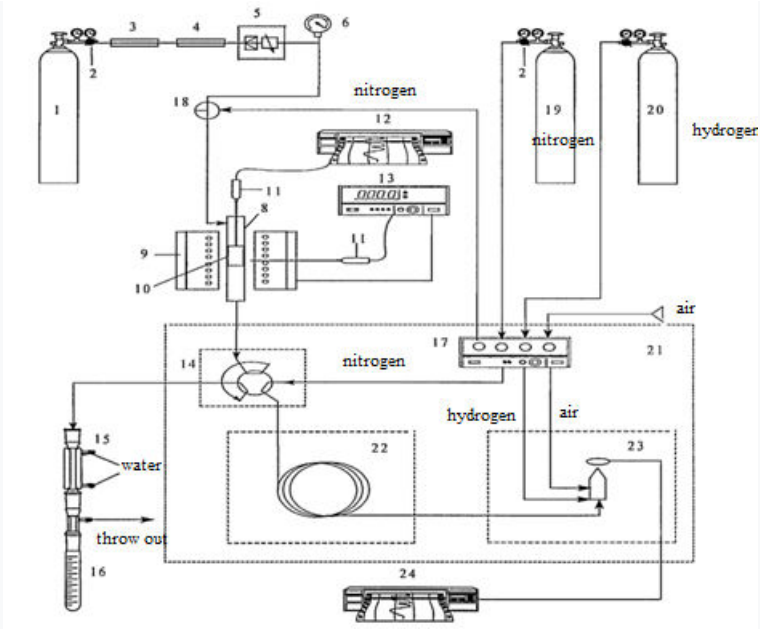
Experimental

An analysis of the literature data showed that the bulk of the MTB process studies were carried out on high-silica zeolite type ZSM-5 and its modified forms in the temperature range 650-750 ℃ and at normal atmospheric pressure.

In this work, a zeolite of the penthasyl structure of the ZSM-5 type was used as a catalyst for the process under study. Mo/ZSM-5 catalysts were prepared by dry mixing the zeolite with MoO_3 and subsequent calcination, activation in a stream of nitrogen. The studies were carried out in a flow reactor in the temperature range 650-750 ℃, at a normal pressure of 0.1 MPa, the volumetric feed rate was 500 h^{-1}. The methodology for catalytic testing was carried out in the work.

Methane conversion on a zeolite catalyst proceeds with the formation of gaseous and liquid products. An analysis of the composition of gaseous products showed the presence of H_2, CH_4, ethane and ethylene. The composition of the liquid products of the process contains benzene, toluene, xylene and naphthalene, which are the target products. Based on the obtained experimental data, an analysis of the kinetic laws of conversion to aromatic hydrocarbons under various conditions of the process is carried out.

The products were analyzed on a Chromatek-Crystal 5000.2 series chromatograph with an NR-1 (DB-1) flame ionization detector 50m×0.21mm×0.5 μm.



The schematic diagram of the pilot installation is shown in Fig. 1.

1-source of natural gas; 2,5-reducers; 3,4- adsorbers with silica gel; 6-monometer; 8-reactor; 9-oven; 10-catalyst; 11-thermocouple; 12- indicator of temperature; 13-temperature control; 14 heating batcher-cock; 15-condenser; 16-receiver; 17-unit preparation of gas; 18-tap; 19-nitrogen container; 20- hydrogen container; 21-chromatographer; 22- chromatographic column; 23 - detector; 24-recorder.

During the experiments, technological parameters such as:

- consumption of incoming nitrogen to purge the reactor with the catalyst;

- methane consumption in cm^3/min;

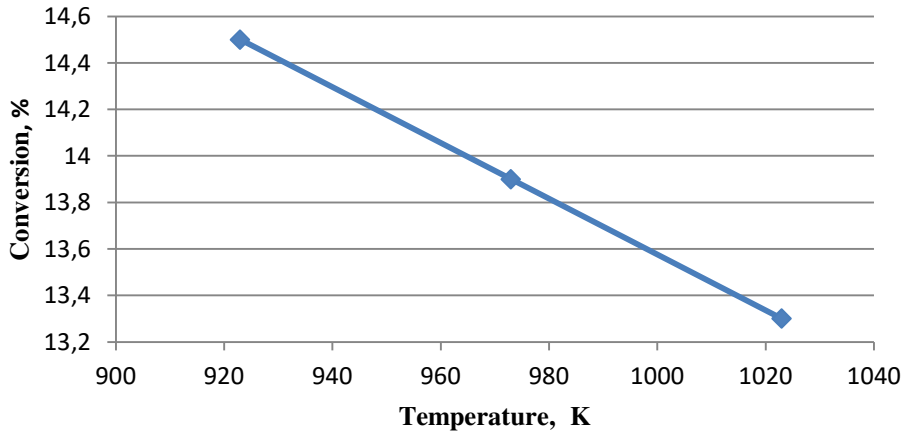
- temperature (℃);

Results and discussion

The efficiency of the process of conversion of methane to benzene was determined by the following indicators of methane conversion, selectivity by reaction products and yield of reaction products.

The results of a study of methane to aromatics are presented in Figure 2.

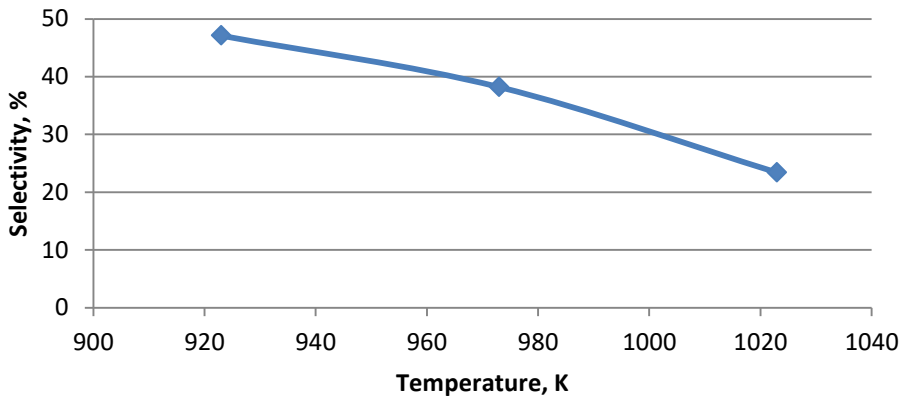
Fig. 2. Effect of temperature on conversion of methane over 5% Mo/Bentonite catalyst.



Thus, in the temperature range 650-750 ℃ methane conversion reaches a maximum at 650 ℃, a further increase in temperature negatively affects the degree of conversion, which decreases from 14.5 to 12.5 mass. % at 650 and 750℃, respectively.

Figure 2 shows the temperature dependence of the content of aromatic hydrocarbons in the composition of products. With increasing temperature from 650 to 750 ℃, benzene selectivity decreases from 47.1 to 23.4 mass. %, and a further increase in temperature leads to the formation of coke and compact products.

Fig.3. Effect of temperature on benzene selectivity over 5% Mo/Bentonite catalyst.



Conclusions

As a result of the studies, the effectiveness of the zeolite structure of the penthasyl type ZSM-5 in the reaction of the conversion of methane to aromatic

hydrocarbons was established. The maximum values of methane conversion, benzene content and yield were 14.5, 57.6 and 12.9% of the mass. respectively. The study of the main process parameters showed that the temperature range of 650-750 ℃ and a pressure of 1 atm are optimal. Thus, as a result of the study, the main principles of the process of the conversion of methane to aromatics on zeolite structure penthasyl type ZSM-5 in the studied range of basic parameters were established.

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