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CATALYST PRODUCTION TECHNOLOGY FOR METHANE OXYCONDENSATION REACTION

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Abstract

The article presents the method of preparation of the catalyst for the synthesis of C_2-hydrocarbons was changed and various d-element salts were added to it as promoters. The catalyst was prepared by two methods: precipitation and absorption. As well as, formal-kinetic analysis of the chemical and physical stages occurring in the synthesis of the catalyst was carried out in a non-isothermal mode by the derivatographic method according to the known equations were given.

Keywords: *catalyst, methane, GTL, method*, *hydrocarbon, temperature*

Introduction

As is known, adiabatic-type reactors are widely used for catalytic processes, the selectivity of the processes changes little over a wide temperature range. The advantage of such reactors is their low metal capacity of 2–3 t/m^3 (cat) and the ease and cheapness of reactor construction. The disadvantage is that the reactions occur with a large heat effect and the selectivity is not uniform over the catalyst bed. As we noted above, when the methane: oxygen ratio is 5, the adiabatic heat of heating of the gas mixture is 575 °C. Taking into account the accepted upper approximation, the methane oxycondensation reaction can be carried out in a 4–5-stage device. Under these conditions, the process is considered an effective method (Togaev, 2025).

For Fischer-Tropsch synthesis, rare metals from Group VIII of the Periodic Table of D. I. Mendeleev can be used as promoters to increase the activity of highly catalytically active cobalt catalysts intended for the production of liquid hydrocarbons from natural gas (Rakhimov, 2024). These metals have the ability to dissociate hydrogen, as a result of which, when added to highly catalytically active cobalt catalysts intended for the production of liquid hydrocarbons from natural gas, the reduction of cobalt oxides occurs more easily. This leads to a decrease in the temperature and a significant increase in the activity of these catalytic systems. Thus, the introduction of 0.1–0.7% Pd or Ru into the highly catalytically active catalysts for the production of liquid hydrocarbons from natural gas based on 10% Co/Al_2O_3 and 10% Co/CuO_2 allowed to increase the CO conversion (by 10%) and the yield of C_5^+ hydrocarbons (from 55 to 95. –97 g/m^3) compared to the highly catalytically active catalyst for the production of liquid hydrocarbons from unpromoted natural gas (Yuldashev, 2024). The highly catalytically active cobalt-ruthenium catalysts for the production of liquid hydrocarbons from natural gas were characterized by high selectivity to the products formed as a result of the liquid reaction (more than 80%). In addition, cobalt, which contains rare metals, has a longer service life, providing catalysts with high catalytic activity for the production of liquid hydrocarbons from natural gas (Kuybokarov, 2024).

Materials and Methods

The method of preparation of the catalyst for the synthesis of C_2-hydrocarbons was changed and various d-element salts were added to it as promoters. The catalyst was prepared by two methods: precipitation and absorption.

In the precipitation method, aqueous solutions of manganese acetate and sodium molybdate were used to prepare the catalyst. The carrier (carrier) – silicon oxide – was prepared by adding sulfuric acid to an aqueous solution of sodium silicate. The precipitate was filtered, dried at 130 °C and calcined in a calcining furnace for 5 hours at 800–1100 °C. In the absorption method, sulfuric acid was first added to an aqueous solution of sodium silicate. The precipitate was filtered, dried at 130 ° C and calcined at 800–1300 °C. Then aqueous solutions of manganese acetate and sodium molybdate salts were added. After 3 hours, the obtained catalyst was dried and calcined at 800–130 °C. Then, the required amount of zirconyl nitrate solution was added to the obtained mass. Then, the catalyst was filtered, dried and calcined in the above sequence.

Catalyst production technology in industry and characteristics of starting materials.

The cobalt-iron-nickel-zirconium/SiO_2 catalyst is in the form of dark brown cylindrical granules with dimensions d = 2.5–3.0, h =3.0÷3.5 mm.

Composition, % mass: 20%Co-15%Fe--10%-Ni-1%ZrO2/YKC

Co_2O_3–20.0
Fe_2O_3–15.0
Ni_2O_3–5.0
ZrO_2–1.0
SiO_2–59.00

Mechanical strength, not less than 3 MPa.

Operating time before regeneration, hours, not less than 108.

Characteristics of starting raw materials and semi-finished products

1. Cobalt (II, III) oxide, gray-black crystal, insoluble in water.

Appearance: gray-black crystal;
Molecular formula: Co_3O_4
Molecular mass: 240.80 g/mol
Density: 6.073 g/cm^3
Melting point (°C): 900

2. Iron (II, III) oxide, solid black powder, insoluble in water.

Appearance: solid black powder;
Molecular formula: Fe_3O_4
Molecular mass: 231.533 g/mol
Density: 5.17 g/cm^3
Melting point (°C): 1597

3. Nickel (II, III) oxide, sharp gray or black powder, insoluble in water.

Appearance: solid black powder;
Molecular formula: Ni_3O_4
Molecular mass: 240.08 g/mol
Density: 3.33 g/cm^3
Melting point (°C): 1682

4. SiO_2 is a porous material.

Results and discussion

Production chemistry and physicochemical foundations

Formal-kinetic analysis of the chemical and physical stages occurring in the synthesis of the catalyst was carried out in a non-isothermal mode by the derivatographic method according to the known equations

$$1-\left(1-\alpha\right)^{\frac{1}{n}}=k\tau \qquad (1)$$

$$\lg\left[-ln\left(1-\alpha\right)\right]=nlgkt \qquad (2)$$

$$\left[-\lg\left(1-\pm\right)\right]^{\frac{1}{n}}=k\tau \qquad (3)$$

The Raginsky-Todes-Mashnel equation (1), the so-called “compressible sphere” equation, is suitable for the case when the limiting stage is considered to be the growth stage of the aggregates. The Erofeev-Avrami equation (3) is characteristic for the emergence of multi-stage reaction centers; the Prodt-Tompkins equation is for the autocatalytic course of processes. We obtain equations (5), (6), and (7) by solving the Arrhenius equation (4) and the joint differential equations (1)–(3) together with equation (5) in the form of a linear temperature change.

$$k = k_0 e^{-\frac{E}{RT}} \tag{4}$$

$$\frac{dT}{dr} = g - const \tag{5}$$

$$\ln\left[\frac{1}{(1-\alpha)2/3} \cdot \frac{d\alpha}{dT}\right] = \ln\frac{k}{g} - \frac{E}{RT} \tag{6}$$

$$\ln\left[\frac{1}{\alpha^{n}/1-2} \cdot \frac{d\alpha}{dT}\right] = \ln\frac{k_0}{g} - \frac{E}{RT} \tag{7}$$

$$\ln\left[\frac{1}{\alpha(1-\alpha)} \cdot \frac{d\alpha}{dT}\right] = \ln\frac{k_0}{g} - \frac{E}{RT} \tag{8}$$

Here is the degree of decomposition calculated from the derivative of the TG melting curve, A is a parameter that depends on the mechanism and geometry of the reaction rate of the formation of complexes; E, R are the activation energy and gas constant.

Processing of derivatographic data according to equations (6)–(8) shows that the decomposition of the manganese-molybdenum-sodium-zirconium/SiO_2 catalyst under the influence of heat obeys equation (8).

X-ray studies revealed that the manganese-molybdenum-sodium-zirconium/SiO_2 catalysts exhibit sufficiently high activity.

Catalyst production process flow chart

In the experiments, 50% solutions of salts prepared from $Co(NO_3)_2$; $Fe(NO_3)_2$, $Ni(NO_3)_2$ and $ZrO(NO_3)_2$ were used. The precipitation of the corresponding hydroxides was carried out in a reactor equipped with a stirring device with a volume of 1000 cm^3 at a temperature of 90–110 °C. After precipitation, the suspension was evaporated, the precipitate was additionally dried at 130 °C and calcined in a muffle furnace at temperatures of 350–550 °C.

The study of the samples by thermal analysis methods was carried out on an SDT Q600 derivatograph. Measurements were carried out in a nitrogen atmosphere in the range of 20–700 °C with a scanning rate of 10 °C/min.

Conclusion

It was found that the time for the complete phase change of the mixture at a temperature of 500 °C is not less than 15 hours. It should be noted that the acceleration of the change process by further increasing the temperature is conceivable due to the rapid adhesion of the precipitate particles and a decrease in their catalytic activity. Because increasing the incineration temperature above 500 °C deteriorates the quality of the catalyst, increasing the efficiency of the incineration temperature is possible only by homogenizing the precipitate composition during the deposition stage.

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