

Oxidation of Natural Gas to Dimethyl Ether at Presence of Catalytic System



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Submission: June 01, 2017; Published: August 04, 2017

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Introduction

The product dimethyl ether (DME) at the International Congress in Detroit was noted as environmentally friendly diesel fuel of the 21st century [1]. Diesel fuels obtained by distillation of oil contain various sulfur-, nitrogen-organic toxic substances that degrade the environment and pollute the atmosphere of the environment. In turn, DME does not contain these toxic substances and the problem consists in the development of the catalyst conducting process in one stage with the formation of dimethyl ether. In recent decades, the scientists in this field have been researching the development of a catalyst for the oxidation of methane to dimethyl ether (DME). As is known, in the catalytic oxidation of methane, the main product is methanol [2-4]. And in this respect, the direct oxidation of methane to DME requires the development of an appropriate catalyst system. In addition, in [5-7] it is noted that DME has a number of unique properties that allow it to almost completely displace traditional diesel fuel - especially in large world megacities. Emissions of a diesel engine operating on DME are 6 times lower in accordance with the EURO-4 standard for emissions of carbon monoxide (CO), 4 times less for emissions of toxic hydrocarbons, 4 times less for emissions of particulate matter (soot and soot of diesel engines) and 20% less emissions of nitrogen oxides (NOx). The use of DME will make it possible to radically solve the problem of the so-called cold start of diesel engines - while working on DME, the diesel can freely start even at minus 50 °C. Note that the cold climatic zone creates certain difficulties in the cold start of diesel engines. Along with this, it should be noted that DME as a chemical is much more harmless than the same diesel fuel and gasoline and is used as filler in aerosol packages of household chemicals that have zero ozone-depletion potential (substitute for banned freons). Thus, the production of DME from methane is expedient for which a suitable oxidation catalyst is needed.

For the preparation of catalysts, chloride salts of transition metals of copper and silver were applied with various adsorbents: γ -Al₂O₃, V₂O₅, SiO₂, aluminum phosphate, etc. Active masses such as copper and silver were calculated from the concentration of their aqueous solutions which were impregnated into adsorbents for 6 hours with stirring at temperature about 80-90 °C. The non-adsorbed part of the salts was determined by optical methods. The active mass varied from 1.0 to 10.0% with varying atomic ratios of copper and silver the paste mass separated from the liquid, was dried in air to a dry state and subjected to a thermal treatment for 4 hours (140 °C) and 6 hours at temperature of 380-400 °C. The specific surface area of the obtained mass was determined by the BET method. Reaction products and reagents were determined by gas chromatography using Dani Master GC. The velocity of the argon gas carrier is 50 cm³ / min, the detection is ionization-flame. The reaction of oxidation of methane by air was carried out in flow reactor. To carry out the oxidation of natural gas, account was taken of the flammability limits of complex combustible gases that do not contain ballast impurities, which are determined by the additivity rule:

$$L_r = (r_1 + \dots r_n) / (r_1 / l_1 + \dots r_n / l_n)$$

where L_r - is the lower or upper limit of the flammability of a complex gas in a gas-air or gas-oxygen mixture, vol. %; $r_1 \dots r_n$ is the content of individual components in a complex gas, vol. %; $r_1 + \dots r_n = 100\%$; $l_1, \dots l_n$ - the lower or upper limits of the flammability of individual components in a gas-air or gas-oxygen mixture according to reference data.

In the experiments the partial pressure of methane was changed from 0.2 MPa to 1.0 MPa at a total gas-air mixture pressure of 2.0 MPa. The reaction was carried out in the

temperature range from 200 to 300 °C. To carry out a one-stage process of converting natural gas to dimethyl ether, different substrates from the deposits of Southern Kazakhstan were taken. A flow-through installation was used to study the conversion of methanol to DME.

The products were analyzed by chromatography and chromatography-mass spectrometry. As shown by the experimental data for the oxidation of methane in DME by 5.0% Cu-Ag (1: 1) / γ -Al₂O₃ modified with vanadium oxide 4, 0%. The partial pressure of CH₄ was changed from 0.2 to 1.0MPa at total pressure of system 2.0 MPa. The yield of DME was 4.6% at conversion 24%.

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DOI: 10.19080/RAPSCI.2017.02.555596

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