Introduction. Methanol is considered one of the most important products of organic synthesis. It is widely used as a solvent, semi-finished product in the production of other organic products (formaldehyde, methyl methacrylate, methyamine, acetic acid, urea, etc.). Also, recently in the microbiological synthesis of proteins, methanol is widely used as raw material, an energy source, and also for the synthesis of a component of motor fuel-methyltrettbutylefir (an effective anti-detonator). Currently, methanol ranks 7-8 among other organic synthesis products in terms of production volume.

Methods of processing methane: first, synthesis-production of gas and production of chemical products based on it; second, oxidation of methane (natural gas) - production of ethylene; third, direct catalytic oxidation of methane(natural gas)up to products containing oxygen. Let's focus on three alternative methods of producing methane-conversion and synthesis-gas.

1) Conversion with water vapour:

\[
CH_4 + H_2O \leftrightarrow CO + 3H_2;
\]

\[
\Delta H_{298} = 206 \text{ kJ/mol};
\]

\[
T = 700-900^\circ C;
\]

\[
cat : Ni
\]

Conducting steam reforming of hydrocarbons has the following disadvantages: the high cost of superheated steam; formation of excess CO₂. The synthesis of H₂: CO = 3: 1 is suitable for the synthesis of gaseous ammonia and not suitable for the synthesis of methanol, acetic acid and hydrocarbons by the Fischer-Tropsch method.

2) Partial oxidation with oxygen:

\[
CH_4 + 1/2O_2 \leftrightarrow CO + 2H_2
\]

\[
\Delta H_{298} = -35.6 \text{kJ/mol}
\]

The disadvantages of oxygen conversion include the high cost of oxygen content; the formation of excessive amounts of CO₂.

3) Carbonate conversion:

\[
CH_4 + CO_2 \leftrightarrow 2CO + 2H_2
\]

\[
\Delta H_{298} = 247 \text{ kJ/mol}
\]
Conversion with carbonate creates the following problems: high exothermicity of the process; coking catalyst:

\[ CH_4 \leftrightarrow C + 2H_2 \quad \Delta H_{298} = -83.3 \text{ kJ/mol} \]

Conversion with carbonate has not yet been commercialized due to the lack of a long-term stable catalyst but is important in terms of CO\textsubscript{2} loss. Methane synthesis gas and conversion with carbonate is a promising method for obtaining initial reagents for hydrocarbon production by the Fischer-Tropsch method.

Carbonate conversion of methane is also a promising method with the simultaneous use of two different greenhouse gases, which has significant environmental and economic implications. Another advantage of this method is that the process of carbonate conversion of methane is carried out at normal atmospheric pressure (0.1 MPa) at a temperature of 650-800 °C \[4\].

**Hydrocarbons are produced from synthesis gas in two stages:**

\[
CO + 2H_2 \rightarrow CH_2OH \quad \Delta H = -90.73 \text{kJ/mol} \\
CO + 3H_2 \rightarrow CH_3OH + H_2O \quad \Delta H = -49.53 \text{kJ/mol}
\]

Also dimethyl ether receive in reaction 3CO + 3H\textsubscript{2} \rightleftharpoons CH\textsubscript{3}OCH\textsubscript{3} + CO\textsubscript{2}.

In the next step, methanol is processed into gasoline or lower olefins. The process consists of three consecutive steps:

At present, the reaction of producing hydrocarbons using the mutual conversion of carbon dioxide and water vapour at normal atmospheric pressure is of great interest to scientists. Schematically, this process can be represented by the following reaction equation:

\[
\begin{align*}
CH_4 + CO_2 & \rightarrow 2CO + 2H_2 \\
CO + 3H_2 & \rightarrow CH_4 + H_2O \\
2CO + 5H_2 & \rightarrow C_2H_6 + 2H_2O \\
CO_2 + H_2 & \rightarrow CO + H_2O
\end{align*}
\]

In general

\[ nMe + 3CO_2 + 5H_2O \rightarrow CH_4 + C_2H_6 + nMeO \]

The implementation of this reaction is important not only from an energy point of view but also from an environmental point of view. In the chemical industry, which is used as a synthetic gaseous feedstock, two groups are isolated. The first group includes only processes aimed at hydrogen production. It is aimed at producing ammonia and hydrogen. The second direction deals with the production of gas and hydrogen in different proportions (Table 1).

| Production | CO: H\textsubscript{2} ratio |
|------------|-----------------------------|
| Methanol synthesis | 2:1 |
| Synthesis of the highest alcohols | 2:1 |
| Fischer-Tropsch's synthesis | 2:1 |
| Acetic anhydride | (1+2):1 |
| Ethyl aldehyde | (1+2):1 |
| Ethylene glycol | 1:1 |
| Vinilasetat | 1:1 |

**Table 1**

Synthesis gas is obtained by converting natural gas. There are the following methods of converting natural gas: steam, steam-oxygen; Paro is carbonate. In all the above transformations, the CO: H\textsubscript{2} ratio is higher than 3. For steam transformations, the ratio CO:H\textsubscript{2} and (CO+CO\textsubscript{2}):H\textsubscript{2} is shown in Table 2.
**Table 2**

The ratio of CO:H\(_2\) (CO + CO\(_2\)):H\(_2\) for various conversions.

| Conversion Type                        | CO: H\(_2\) ratio | (CO+CO\(_2\)):H\(_2\) ratio |
|----------------------------------------|--------------------|------------------------------|
| with steam                             | 4÷8,7              | 3,2÷3,6                      |
| with steam-oxygen                      | 3÷3,7              | 2÷2,2                        |
| with steam-air-oxygen                  | 2,9÷3,5            | 2÷2,1                        |
| with steam + steam-oxygen              | 4,3                | 2,8                          |
| with steam + steam-air-oxygen          | 4,2                | 2,7                          |
| with steam-carbonate                   | 3,8                | 3,2                          |

**Experimental part.** Analysis of gases was carried out by a Crystal 2000M chromatograph using thermal conductivity detectors and flame ionization detectors connected in series. Separation of the analysed mixture was carried out on a column filled with Porapak Q at the following temperature-programmable modes: determination of components H\(_2\), CO, CO\(_2\), CH\(_4\) - Т\_колонки = 30°C; 60 °C, Т\_детекторов=150 °C, assay time - 1 min; determination of H\(_2\)O - column temperature rise to 110 °C (heating rate - 40 °C per minute), analysis time - 4 minutes. The rate of the reaction mixture, reaction products, argon carrier gas entering the reactor and chromatograph was measured by soap-foam flowmeters. The composition of the reaction mixture was measured after the catalyst reached a steady-state, which was judged by the constancy of chromatographic peak areas. The main indicators of the reaction were the conversion of methane, the consumption rate of methane, carbon dioxide and the formation of carbon monoxide and hydrogen. The quantitative interpretation was performed by absolute calibration using Chromatic Analyst 2.5. The number of moles of reagents and products in the analyzed dose was calculated using regression equations of the type:

\[ n_i = k_i S_i \]

where:
- \( n_i \) – the number of moles of the \( i \)-th substance in the analysed dose;
- \( S_i \) – area of the \( i \)-th peak, mV * min;
- \( k_i \) – correction coefficient for the \( i \)-th substance, mol/min * mV.

Reagent consumption rates \((W_i, \text{ mol}/(\text{h} \times \text{ha. f.}))\) were calculated using the formula:

\[ \omega_i = \frac{(n_i^o - n_i)W}{Vm_{aph}} \]

where:
- \( n_i \) - the amount of the \( i \)-th product in the gas phase, mol;
- \( n_i^o \) - the initial amount of the \( i \)-th product in the reaction mixture, mol;
- \( W \) - volume velocity of the reaction mixture, l/h;
- \( V \) is the volume of the chromatograph loop, 0.208•10^{-3} l;
- \( m \) is the mass of the active (metal) phase of the catalyst, g.

The content of products in the gas phase (the rate of formation of mol/(h * ha. f.)) was determined by the formula:

\[ W_i = \frac{n_i W}{V_{aph}} \]
CH$_4$ and CO$_2$ conversions ($\alpha_i$) were calculated using the formula:

$$\alpha_i = \frac{n_{i}^o - n_{i}}{n_{i}^o} \cdot 100\%$$

$n_i$ – the amount of CH$_4$ or CO$_2$ in the gas phase, mol,
$n_{i0}$-the initial amount of the i-th product in the reaction mixture, mol.

Methane vapour-carbonate conversion was performed in a flow laboratory device. Experimental studies were carried out with the participation of a 2 cm$^3$ cylindrical reactor a catalyst with a diameter of 2 cm and a length of 20 cm.

**Results and discussion.** Tables 3-4 show the material balance of experiments for the hydrogen-to-hydrogen steam conversion of methane and carbon under various conditions. The material balances of hydrogen and carbon are the most important criteria for the reliability of experiments. It follows from the studies that the largest deviation in carbon is 4.0 ± 0.5%, in hydrogen-2.7 ± 0.5%. These are valid measures.

### Table 3

**Material balance for carbon.**

| Temperature, $^\circ$C | Conversion of methane,% | Carbon was introduced, mol | Carbon was obtained, mol | Absolute error, mol | Relative error, % |
|------------------------|-------------------------|---------------------------|-------------------------|---------------------|------------------|
| 700                    | 78,44                   | 8,41                      | 8,19                    | 0,22                | 2,6              |
| 750                    | 90,35                   | 8,38                      | 8,27                    | 0,11                | 1,3              |
| 800                    | 97,83                   | 7,68                      | 7,52                    | 0,16                | 2,1              |
| 850                    | 99,62                   | 6,98                      | 6,70                    | 0,28                | 4,0              |
| 900                    | 99,96                   | 6,05                      | 5,98                    | 0,07                | 1,2              |

### Table 4

**Material balance for hydrogen.**

| Temperature, $^\circ$C | Conversion of methane,% | Hydrogen was introduced, mol | Hydrogen was obtained, mol | Absolute error, mol | Relative error, % |
|------------------------|-------------------------|-----------------------------|---------------------------|---------------------|------------------|
| 700                    | 78,44                   | 6,99                        | 6,89                      | 0,10                | 1,4              |
| 750                    | 90,35                   | 6,25                        | 6,08                      | 0,17                | 2,7              |
| 800                    | 97,83                   | 6,14                        | 6,01                      | 0,13                | 2,1              |
| 850                    | 99,62                   | 5,65                        | 5,60                      | 0,05                | 0,9              |
| 900                    | 99,96                   | 5,76                        | 5,68                      | 0,08                | 1,4              |

Various factors (CO$_2$:CH$_4$ ratio, temperature, initial space velocity and other factors, the yield of the desired product, process conversion and selectivity, and catalyst activity) were used to determine optimal conditions for the methane-to-carbonate vapour conversion reaction. the influence of promoters has been studied. As the amount of CO$_2$ in the feed increases (except for stoichiometric ratios), the methane conversion varies within 96-99%. As the ratio CO$_2$: CH$_4$ increases, the conversion CO$_2$ decreases to 88%. Based on the results obtained, it can be concluded that the optimal ratio of CO$_2$: CH$_4$ in the carbonate conversion reaction of methane is 1.5. Under these conditions, the maximum yield of CO is 96% and the yield of hydrogen is 92%. The effect of temperature on the rate of steam-carbonate conversion of methane was studied in the range 700-9000 °C with a pitch of 500 °C in the ratio CO$_2$:CH$_4$=1,5.
Based on the results of studies on the effect of temperature on the reaction of methane vapour carbonate conversion, it can be noted that the yield of synthesis gas, the conversion of starting materials and the selectivity of the process are the highest at 820 °C. The maximum hydrogen yield is 96%, the conversion of methane is 99.2%, the selectivity of the components of synthesis gas is 87-98%. The conversion of methane is 98.6% at a space velocity of 1000 h⁻¹. Above this, methane conversion is reduced due to saturation and coking of the active catalyst centre.

The reaction proceeds according to a radical mechanism. Therefore, the determination of the activation energy ($E_a$) of the catalytic methane carbonation reaction in the presence of the catalyst (Ni$_x$O$_y$(Co$_z$O$_m$)$_n$(ZrO$_2$)$_2$(B$_2$O$_3$)$_m$/Al$_2$O$_3$) provides important information for evaluating the efficiency of the catalyst and the reaction in the presence of the catalyst (Ni$_x$O$_y$(Co$_z$O$_m$)$_n$). The activation energy ($E_a$) of the methane vapour carbonate conversion reaction was determined.

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