

# RESEARCH OF THE PROCESS OF ETHYLENE PRODUCTION FROM METHANE

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**Abstract.** The article describes the methods of methane processing, the production of synthesis gas from methane and the production of chemical products based on it, the methods and applications of methane oxycondensation to ethylene, and the direct catalytic oxidation of methane to oxygen-containing products. The kinetic equations of the reaction, as well as the essence of the process and methods for its improvement, are analyzed.

**Keywords:** methane, oxygen, methanol, acetic acid, hydrocarbon, catalyst, mechanism, coking, conversion.

Before carrying out kinetic studies, it is advisable to obtain information on the equilibrium composition of the mixture of substances formed during the oxycondensation of methane. Such information is very useful for determining the stage of the reaction and explaining the kinetic laws. Thermodynamic analysis of some technically important reactions involving methane has been carried out by a number of scientists. These studies mainly concern reactions such as dehydrogenation, oxidation and chlorination of methane. In these studies, the values of Gibbs energies for the above-mentioned reactions were calculated at different temperatures. They did not carry out a complete thermodynamic analysis of the reactions. There is absolutely no information in the scientific literature on the equilibrium yields of olefins formed during the interaction of methane with molecular oxygen. This does not allow us to even approximately estimate the forms in which methane used as the starting material can be converted into other substances in the reactions. Therefore, there was a need to study the equilibrium composition of methane oxycondensation reaction products. Analysis of data in the literature shows that the oxycondensation reaction of methane goes through the following chemical changes:

- 1)  $2\text{CH}_4 + \text{O}_2 = \text{C}_2\text{H}_4 + 2\text{H}_2\text{O}$
- 2)  $2\text{CH}_4 + \text{O}_2 = \text{C}_2\text{H}_2 + 2\text{H}_2\text{O} + \text{H}_2$
- 3)  $2\text{CH}_4 + 1/2\text{O}_2 = \text{C}_2\text{H}_6 + \text{H}_2\text{O}$
- 4)  $\text{C}_2\text{H}_6 + \text{O}_2 = \text{C}_2\text{H}_2 + 2\text{H}_2\text{O}$
- 5)  $\text{CH}_4 = \text{C} + 2\text{H}_2$
- 6)  $\text{CH}_4 + 2\text{O}_2 = \text{CO}_2 + 2\text{H}_2\text{O}$
- 7)  $\text{CH}_4 + \text{O}_2 = \text{CO} + 2\text{H}_2$

To determine the equilibrium compositions of the substances in the given reactions, we used the  $\lg K_p$  values of the components given in the literature. The  $\lg K_p$  values were calculated using the extrapolation method for temperatures of 1100, 1200 and 1300 K.

It can be seen from the above reaction equations that the main reactions are parallel, except for the reactions of ethane formation from methane and acetylene formation from it. The  $\lg K_p$  values for parallel reactions were calculated using the following equation:

$$\lg K_{p(\text{reac})} = \sum \lg K_{p(\text{reac. prod.})} - \sum \lg K_{p(\text{starting material})}$$

The method recommended by Kandiner and Brenkley was used to determine the equilibrium concentrations of the products of the methane oxycondensation reaction. The essence of the method is that the composition of the equilibrium mixtures is the result of several reactions occurring simultaneously, and their composition can be calculated very simply and accurately using specially selected "base" components. According to the Kandiner and Brenkley method, the amount of components is equal to the maximum possible matrix level.

Calculations were continued until the molar amounts of the initial and final "base" components reached a sufficient level of accuracy. The equilibrium composition of the components was calculated using a program written in the BASIC algorithmic language for personal computers at a methane:oxygen ratio of 1:2, with a degree of methane conversion to products of 86%.

The calculation results are presented in Table 1.

**Table-1**

**Equilibrium composition of oxycondensation reaction components at different temperatures (in moles)**

Temperature, K	Components, (mol)			
	$n_5 - C_2H_4$	$n_6 - C_2H_2$	$n_7 - C_2H_6$	$n_8 - C$
1000	0,150	0,050	0,065	0,080
1100	0,200	0,035	0,030	0,100
1200	0,175	0,030	0,010	0,250

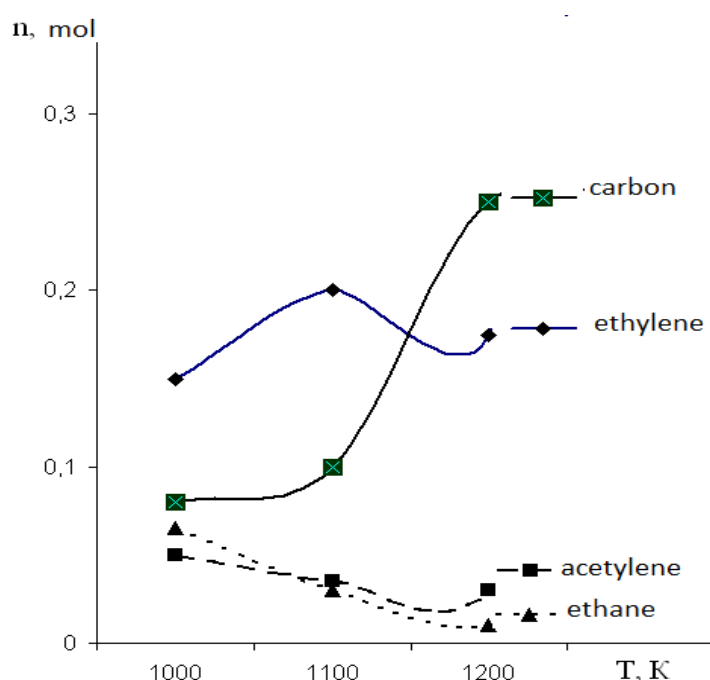
Based on the results of Table 1, a graph of the temperature dependence of mole fractions was constructed (Figure 1) and the degree of methane conversion into various products was calculated (Table 1).

Figure 1 shows that in the range of 1000–1100 K, the ethylene production yield increases from 0.15 mole fraction to 0.2 mole fraction, reaches a maximum at 1100 K, decreases slightly (by 0.02 mole fraction) in the range of 1150–1200 K, and begins to increase again from 1200 K.

**Table 2**

**Rate of conversion of methane to products at different temperatures**

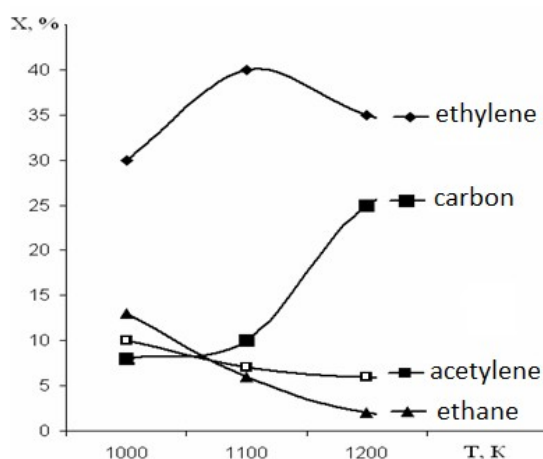
Temperature, K	Rate of conversion of methane to products, (%)			
	C <sub>2</sub> H <sub>4</sub>	C <sub>2</sub> H <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>	C
1000	30	10	13	8
1100	40	7	6	10
1200	35	6	2	25



**Figure 1. Dependence of the yield of subordinate components on temperature.**

The products of ethane and acetylene formation decrease from 0.6 and 0.5 mole fractions, respectively, in the range of 1000 -1150 K, reaching a minimum at 1180 K and starting from 1200 K, they begin to increase slowly again. At the same time, a significant part of methane is converted into carbon in the range of 1100 K to 1200 K.

Based on the results of Table 2, a graph of the degree of conversion of methane into products versus temperature was constructed (Fig. 2).



**Figure 2. Temperature dependence of the degree of methane conversion to products.**

As can be seen from Figure 2, there is an exponential relationship between the reaction yield of ethylene and temperature, which reaches a maximum at 1100 K. This indicates that the process in the given temperature range obeys the Arrhenius equation.

Based on the above, and assuming that the sorption process obeys the Langmuir isotherm, the following Langmuir-Hinshelwood-type equations were proposed as kinetic equations for the catalytic oxycondensation reaction of methane:

$$W = \frac{k \cdot K_{CH_4} \cdot P_{CH_4} \cdot K_{O_2} \cdot P_{O_2}}{(1 + K_{CH_4} \cdot P_{CH_4} + K_{O_2} \cdot P_{O_2})} \quad (1)$$

$$W = \frac{k \cdot K_{CH_4} \cdot P_{CH_4} \cdot K_{O_2} \cdot P_{O_2}}{(1 + K_{CH_4} \cdot P_{CH_4} + K_{O_2} \cdot P_{O_2})^2} \quad (2)$$

$$W = \frac{k \cdot K_{CH_4} \cdot P_{CH_4} \cdot K_{O_2} \cdot P_{O_2}}{K_{CH_4} \cdot P_{CH_4} + K_{O_2} \cdot P_{O_2}} \quad (3)$$

It is necessary to check to what extent the selected equations (1-3) satisfy the experimental kinetic laws obtained under the conditions of a differential reactor. Based on the given equations and experimental results, the parameters of the kinetic equations should be determined by the minimum deviation of the sum of the squares of the experimental values from the theoretically calculated value. Since the proposed equations are nonlinear, their solution can be mathematically infinite. From a kinetic point of view, the solution of these equations is found by checking the adequacy of the equations based on the experimental results obtained in the ranges of changes in the kinetic parameters. Thus, the solutions of the proposed kinetic equations, the reaction rate constant  $k$ , and the values of the adsorption coefficients of oxygen and methane ( ) are found based on the values of the partial pressures of oxygen and methane ( ) and the experimental reaction rate ( $W$ ). The program "NELIN-NEUTZ" written in the BASIC algorithmic language was used to solve the problem. The adequacy of the equations was checked based

on the value of the root mean square deviation ( $s$ ) of the difference between the experimentally obtained and theoretical calculation results.

The root mean square deviation of the reaction rates found on the basis of the theoretically calculated and experimentally obtained values of the constants and coefficients in the proposed kinetic equations ranges from 2.0% to 26.0%. If the root mean square deviation is analyzed according to these equations, its value according to equation (1) is up to 21.23%, and according to equation (3) it is up to 25.7%. Thus, the values of the constants and coefficients calculated according to equations (2) and (3) deviate from the adequacy, and therefore do not fully satisfy the kinetic process. The results with high adequacy are constants and coefficients calculated based on equation (2), so it was selected for further calculations. The mean square deviation of this equation does not exceed 5% and it adequately satisfies the kinetic parameters of the process within the given range of variation.

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