



International Journal for Innovative Engineering and Management Research

A Peer Reviewed Open Access International Journal

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IJIEMR Transactions, online available on 8th Feb 2021. Link

[:http://www.ijiemr.org/downloads.php?vol=Volume-10&issue=ISSUE-02](http://www.ijiemr.org/downloads.php?vol=Volume-10&issue=ISSUE-02)

DOI: 10.48047/IJIEMR/V10/I02/04

Title **PRODUCTION OF SYNTHESIS GAS BY CONVERSION OF METHANE IN PLASMA OF WATER VAPOR AND CARBON DIOXIDE**

Volume 10, Issue 02, Pages: 18-22.

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PRODUCTION OF SYNTHESIS GAS BY CONVERSION OF METHANE IN PLASMA OF WATER VAPOR AND CARBON DIOXIDE

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Annotation: widespread in the chemical industry are catalytic methods of methane conversion in the presence of water vapor.

Keywords: methane, synthesis gas, natural gas, liquid fuels, plasma of water vapor.

A brief description of the experimental setup and the results obtained on the conversion of methane in the plasma of water vapor and carbon dioxide are presented. At a mass flow rate of H₂O ~ 3 g / s and CO₂ ~ 3 g / s, the CH₄ flow rate varied from 2.5 to 3.7 g / s. The energy consumption was 29–42 MJ per 1 kg of CH₄. The H₂ / CO ratio in the resulting synthesis gas was 2.2–2.4. The CH₄ conversion was 90.8–99.8%. The content of H₂ and CO in the synthesis gas was ~ 95%.

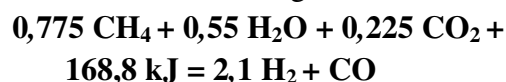
The process of converting methane into synthesis gas is of great practical importance. The reason for this is the storage and transportation problems of gas fuel [1], as well as the existing imbalance in prices for natural gas and liquid fuel. In addition, when oil is extracted from small wells, there is a problem of processing associated gas [2].

At the moment, catalytic methods of methane conversion in the presence of water vapor [3], as well as partial oxidation with oxygen [4], have become widespread in the chemical industry. As a result, synthesis gas (a mixture of CO and H₂) is obtained, which is used in the synthesis of liquid organic substances on catalysts. The highest demands on the quality of synthesis gas are imposed in the Fischer-Tropsch and methanol synthesis processes.

One of the promising methods for obtaining synthesis gas with a given H₂ / CO

ratio can be the use of plasma, which allows one to achieve higher degrees of methane conversion with lower energy costs [5]. The authors previously studied the process of methane conversion by steam plasma for the production of hydrogen [6], however, the synthesis of liquid fuels from associated gas in remote oil fields is of greater practical importance. This work is devoted to the conversion of methane by the plasma of a mixture of water vapor and carbon dioxide.

For the Fischer - Tropsch process on cobalt catalysts, the stoichiometric H₂/CO ratio in the synthesis gas should be ~ 2.1. Then, at stoichiometric conversion, the gross reaction can be written in the following form:



The minimum possible amount of energy consumption at standard atmospheric pressure would be ~ 13.58 MJ per 1 kg of methane. According to the thermodynamic equilibrium of the above system, to prevent the formation of carbon, it is necessary to spend at least ~ 20.2 MJ / kg, which corresponds to the heat content of the plasma ~ 12.7 MJ / kg (~ 3209 K). However, kinetic estimates show that at such an energy consumption in the range of industrial conversion rates of ~ 20 200–76 400

h - 1, the methane conversion depth is ~ 87.3–85.7%, and the selectivities for hydrogen and carbon monoxide are only ~ 76.5–74.4 and ~ 35.8–34.1%, respectively.

A higher level of conversion is achieved at higher energy costs, temperatures and specific oxidant costs. For example, at a heat content of a vapor-carbon dioxide plasma of ~ 15 MJ / kg (~ 3332 K), the proportions of reagents CH₄: H₂O: CO₂ = 0.775: 0.605: 0.2475, and a space velocity of ~ 22 600–66 900 h⁻¹, the degree of methane conversion is ~ 97–96.3 %, and the selectivities for H₂ and CO are ~ 97.1–94.8 and ~ 89.0–80.8%, respectively. The volume-average temperature of the process is 1984–1946 K. The purpose of the experiments on plasma methane conversion described below is to show that such parameters are attainable.

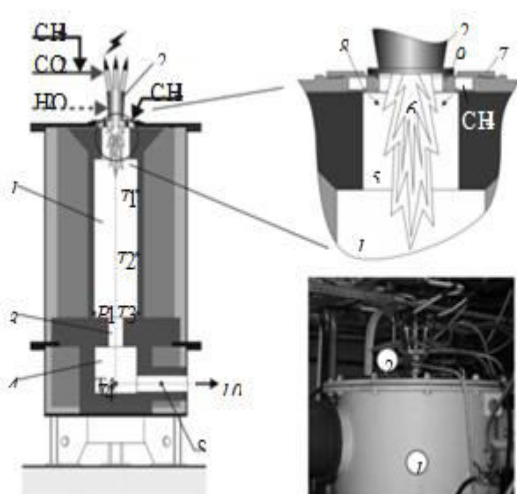


Figure: 1. General view of the experimental setup:

1 - flow reactor; 2 - plasma generator; 3 - aerodynamic pinching; 4 - lower chamber; 5 - mixing chamber; 6 - plasma jet; 7 - device for distributed methane injection; 8 - methane supply hole; 9 - cooling jacket; 10 - synthesis gas outlet. P - place of pressure measurement; T is the place of temperature measurement; S - gas sampling point for composition analysis

The experiments were carried out in a flow reactor (Fig. 1). A three-phase high-

voltage plasma generator with a power of 80–120 kW was used [7–9]. Plasma-forming medium is a mixture of superheated water vapor (200 ° C) and carbon dioxide. Carbon dioxide was fed tangentially into the near-electrode zone. In addition to the flow of carbon dioxide (~ 3 g / s), methane was mixed (up to 1 g/s). Water vapor was fed tangentially into the arc combustion zones and ensured their axial stabilization. When the methane flow rate varied from 0 to 1 g / s, the thermal efficiency of the plasmatron was 93.8–95.9%, the power was 86.6–117.2 kW, the voltage drop across the arc was 990–1360 V, and the arc current was ~ 50A.

The plasma flow from the plasma generator 2 entered the mixing chamber 5 with a diameter and height of 0.2 m, from where it entered the flow-through reactor 1, which is a vertical cylindrical chamber with a diameter of 0.3 m and a height of 1 m, ending in an aerodynamic pinch 3 with a diameter of 0.1 m.

The initial stage of the experiment was the heating of the reaction space by air plasma generators (not shown in Fig. 1) to temperatures close to 1000 ° C, after which the air plasma supply was stopped and the plasmatron 2 was launched. At the first stage, it was operated on a mixture of carbon dioxide and water. steam, the flow rates of which were ~ 3 g / s. In this case, the power of the plasmatron was ~ 80 kW, which made it possible to raise the temperature in the reactor to ~ 1300 ° C. After that, the methane feed began. Methane was supplied in two ways: the first, by mixing plasma-forming carbon dioxide with a flow rate of 0.5–0.8 g / s, and the second, by feeding it into the plasma jet through a distributed input device 7 with a flow rate of 1.7–2.9 g / s. In the course of the experiments, the ratios between the methane flow rates supplied to the plasmatron and to the plasma jet were varied, temperatures and

pressures, media flow rates, and the electrical parameters of the plasmatron were controlled. The pressure P1 was maintained in the range of $-0.2-0$ kPa.

Superheated steam was supplied from a steam generator at a constant flow rate, maintained by a calibrated supply washer. Carbon dioxide (volume fraction of the main substance $> 99.8\%$) and methane ($> 96.5\%$) were supplied from cylinders, and their flow rates were measured with float rotameters with local readings.

To determine the composition of synthesis gas, an MKS Cirrus 300 quadrupole mass spectrometer with a Faraday detector and a Nicolet 380 IR Fourier spectrometer with a single-pass gas cell with an optical path length of 10 cm were used.

The data on the flow rates of the supplied media, the power of the plasmatron, and on the temperatures in the reactor are shown in Fig. 2, and the data on the composition of the resulting synthesis gas are shown in Fig. 3. The table shows the experimental and calculated data of the main parameters of the ongoing process in different modes.

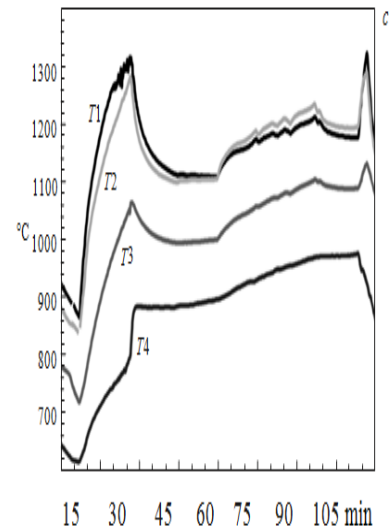
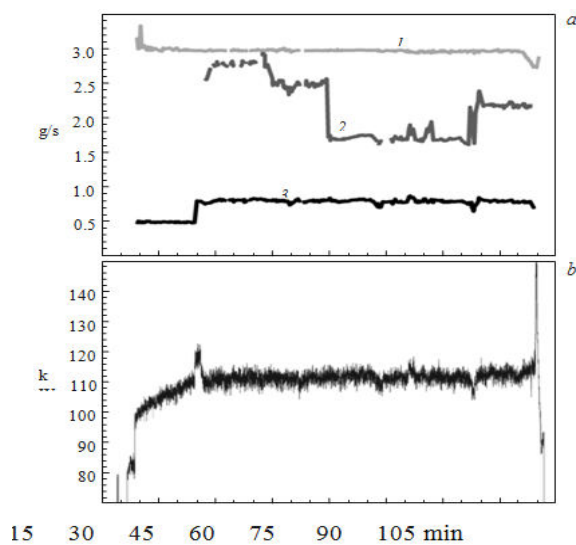


Figure 2. Change of parameters in the course of experiments on plasma conversion of methane:

a - flow rates of the supplied media (1 - CO₂; 2 - CH₄ into the mixing chamber, 3 - CH₄ into the plasmatron); b is the power of the plasmatron; c is the temperature in the reactor.

During the tests, the flow rates of water vapor, carbon dioxide and methane were maintained in 4 different ratios. A part of methane ($\sim 22.6-31.9\%$ of the total consumption) was supplied to the plasmatron and was decomposed during plasma generation. The temperature at the outlet of the plasmatron was ~ 3375 K at a plasma heat content of ~ 16.11 MJ / kg. Together with an excess content of oxidants, this creates conditions for an almost complete conversion of the supplied methane ($\sim 0.82 \pm 0.02$ g / s). The magnitude of the characteristic peak of methane ($m / z = 15$), recorded by the mass spectrometer in this mode, is close in magnitude to the noise signal.

When additional methane is supplied (modes I, II, and IV), the conversion level decreases, which is evident from the composition of the synthesis gas (see Fig. 3). In mode III, the characteristic methane peak also approaches the noise level.

A characteristic feature of the observed process is the presence of methane in the reaction products and the almost complete

absence of acetylene. The acetylene content was measured with an IR Fourier spectrometer and, on average, did not exceed ~ 0.01% in all modes (maximum ~ 0.1%).

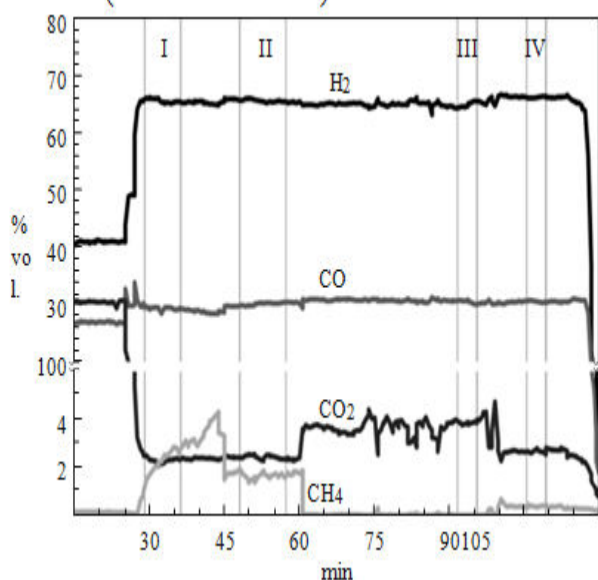


Figure: 3. Changes in the consumption of material flows at the entrance to the plasma-chemical reactor and the composition of the synthesis gas at the exit from it during experiments on plasma conversion of methane.

The volumetric rate of the process for regime I is ~ 248 h⁻¹. The volume average temperature for this rate and oxidation stoichiometry is ~ 1560 °C, which is at least ~ 460 °C higher than the temperatures observed in the second chamber of the setup. In other modes, the space velocity is even lower, the excess of the oxidant is higher (at a constant heat content of the plasma) and, accordingly, the volume-average process temperatures are higher. On the whole, this allows us to assume that the main part of the reactions took place in the zone of mixing of plasma and methane flows - before the mixture entered the second chamber, where the decomposition of thermostable hydrocarbons was significantly slowed down due to a decrease in temperature. Therefore, estimates of the volumetric velocity are made from the volume of the mixing chamber.

Basic parameters of the experiment

Parameter	Unit.	Mode			
		I	II	III	IV
Expenses	H ₂ O	2.90 ± 0.10	2.90 ± 0.10	2.90 ± 0.10	2.90 ± 0.10
	CO ₂	2.97 ± 0.01	2.97 ± 0.01	2.96 ± 0.01	2.97 ± 0.01
	CH ₄	3.70 ± 0.03	3.37 ± 0.06	2.56 ± 0.03	3.08 ± 0.04
Energy consumption	MJ/kg	29.0	31.8	42.0	35.3
Выход синтез-газа	m ³ /kg	5.66	5.75	6.13	5.98
Syngas composition	CH ₄	2.31	1.66	0.05	0.42
	H ₂	65.35	65.28	64.92	65.86
	CO ₂	2.31	2.44	3.85	2.67
	CO	29.32	30.07	30.62	30.52
	N ₂	0.52	0.51	0.48	0.49
	O ₂	0.17	0.03	0.07	0.03
Lower calorific value of synthesis gas	MJ/m ³	10.61	10.47	9.98	10.18
Conversion rate CH ₄	%	90.81	93.31	99.78	98.22
Volumetric velocity	hour ⁻¹	2950	2683	2044	2454

According to mass spectrometry data, the nitrogen concentration in the synthesis gas fluctuated significantly and was mainly lower than the values corresponding to the balance of elements. This was primarily caused by the influence of methane and internal instrument noise on the m / z = 14 peak. This was most pronounced in modes I, II, and IV, and in mode III the measured values were close to those obtained from the balance.

The mass average density of the plasma outflowing from the plasmatron was ~ 0.05 kg / m³, and the density of methane introduced into the mixing chamber was ~ 0.68 kg / m³. The ratio of the volumetric plasma flux (~ 131.2 l / s) to the methane flux is ~ 31.1–50.8, depending on the regime. Considering this and the large temperature difference between the plasma and methane (supplied at room temperature), a high mixing rate of the flows can be expected. According to estimates for regime I, under such conditions, the synthesis gas should contain CH₄ ~ 0.33%, C₂H₂ ~ 0.88%. However, due to insufficient thermal insulation in the mixing chamber, heat losses can be significant, which explains the increased content of CH₄.

The data obtained indicate the possibility of increasing the volumetric rate of the process without reducing its efficiency (including by introducing a larger amount of methane into the plasmatron).

As a result of experiments in long-term operation, a high degree of methane conversion into synthesis gas (90.8–99.8%) was achieved for the first time in modes close to stoichiometric, at a low level of energy consumption (29–42 MJ / kg), which is significantly better than similar indicators of known plasma processes. The resulting synthesis gas consisted of more than 95% vol. from CO and H₂ with a molar ratio H₂/CO of 2.2-2.4. The plant productivity was 9.2-13.3 kg / h for methane, and 56.4-75.3 m³/h for synthesis gas.

The main advantage of this plasma process is the ability to control the composition of the products and obtain synthesis gas, practically free of ballast impurities, with different H₂/CO ratios. The relative simplicity of this plasma technology already at this stage makes it possible to predict its large-scale implementation for the production of a wide range of liquid organic substances, including motor fuels, in optimal conditions.

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