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Steam-water reforming of methane using a AC plasmatron of megawatt power

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This paper presents the results of a study of steam-water conversion of methane using a three-phase AC plasmatron of megawatt power operating on methane. The process was carried out with the simultaneous supply of methane to the arc channels of the plasmatron and water to the plasmatron chamber. It was established that the studied methane decomposition process occurs according to a two-stage scheme: methane pyrolysis in the arc channels in the first stage, followed by steam-water conversion together with water supplied to the plasmatron chamber in the second stage. Data on the influence of the ratio of methane and water consumption on the concentration of pyrolysis products were obtained. The plasmatron operating modes during the study varied within the following ranges: arc power from 1.32 to 1.61 MW, methane flow rate from 43 to 57.5 g/s, and water flow rate from 0 to 67 g/s.

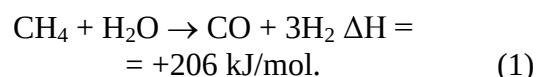
Keywords: plasmatron, electric arc, hydrogen production, methane pyrolysis, methane conversion, synthesis gas.

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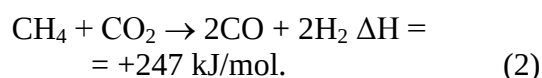
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Introduction

Methane steam-water reforming is one of the methods for producing synthesis gas, which serves as an intermediate feedstock in many chemical processes. There are several methods for its production from natural gas: steam catalytic reforming, CO₂ reforming, and partial oxidation [1–3]. During steam reforming, methane reacts with steam under a pressure of 0.3–2.5 MPa in the presence of a catalyst and produces synthesis gas. Reaction (1) is endothermic, the energy required to carry out the reaction should be obtained from an external source. Currently, steam reforming of methane is the main industrial method of producing hydrogen:



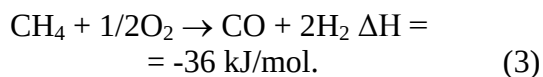
During CO₂ reforming, methane reacts with carbon dioxide to form synthesis gas with an increased content of carbon monoxide. This method has recently attracted considerable interest due to the simultaneous use and reduction of emissions of two common and undesirable greenhouse gases CH₄ and CO₂.



As a result of this process, synthesis gas is produced with a molar H₂/CO ratio close to 1, which is optimal for subsequent Fischer–Tropsch synthesis.

Both reactions (1) and (2) are highly endothermic, requiring energy supply.

Partial oxidation of methane is also an attractive alternative method for producing synthesis gas [3]. The reaction is exothermic, reducing the energy consumption of the reforming process.



The non-plasma technologies considered above have limitations related to the need for catalysts, which gradually deactivate during the process at high temperatures due to agglomeration and carbon deposition.

Unlike these traditional technologies, plasma reforming has a number of important advantages, in particular, high quality of the gas produced and ease of control of its composition, compactness, fast response time, compatibility with a wide range of hydrocarbons and elimination of problems with catalysts. Moreover, plasma technologies are environmentally friendly processes because they do not emit greenhouse gases.

Plasma technologies are based on the use of non-equilibrium plasma or thermal plasma. Corona discharge [4], microwave discharge [5], sliding arcs [6], barrier discharge [7], and glow discharge [8] are used as non-equilibrium plasma. In this case, active particles are formed that participate in the chemical mechanism, however, a break in the chemical process chain at the endothermic stages is extremely likely. Therefore, these methods are very energy-consuming and can only be used in conjunction with catalysts.

In higher-performance devices, thermal plasma of carbon dioxide [9, 10] and water vapor [11] is used. In this case, the conversion rate and selectivity are determined by the process temperature. Significant methanation under the action of water vapor and carbon dioxide occurs at high temperatures (above 1,100 °C) [12]. At the same time, higher temperatures lead to increased selectivity towards acetylene. The economic features of this method are determined by the service life of the electrode assembly, productivity and efficiency of using electrical energy [13].

Experimental conditions and their analysis

In the present work, the AC plasmatron with megawatt power "Zvezda" was used as a source of plasma heating of methane and water (Fig. 1). The plasmatron design features and the description of operation with various gases are given in [14–19]. In this work, methane was supplied to the arc channels (electrodes), pos. 1 in Fig. 1, and water was supplied to the plasmatron chamber, pos. 2 in Fig. 1. To enable operation with methane and use the plasmatron in plasma-chemical technologies, the plasmatron design was modified to avoid negative effects associated with the possible deposition of carbon in the insulator area and methane injection points into the arc channels [20–22]. Towards the camera, pos. 2 in Fig. 1, through the nozzle, pos. 7 in Fig. 1, the adapter is attached, pos. 8 in Fig. 1, designed to increase the residence time of pyrolysis products. Gas is extracted at the end of the mouthpiece for chromatographic analysis from the channel, pos. 10 in Fig. 1. The gas composition of the pyrolysis products was measured using a Crystallux 4000M chromatograph, with argon as the gas carrier. Pyrolysis products flow through the adapter nozzle, pos. 9 in Fig. 1, into the test box and burn up in the atmosphere, as shown in Fig. 2. The total volume of the chamber and the adapter is 2.8 liters, and the total residence time of the pyrolysis products is ~2.5 ms.

The described scheme for supplying plasma-supporting gas into the arc channels and liquid into the chamber was previously used [14] for plasma gasification of a coal-water slurry (CVS) of the composition 40% H₂O, 60% C with a plasmatron power of 680 kW, a plasma-supporting gas flow rate of 97 g/s, and CVS flow rate of 70 g/s.

In the present work, the power of the plasma torch varied in the range from 1.32 to 1.61 MW, and the efficiency measured by heat loss to the plasma torch cooling system was $\eta = 85\%$.

Table 1 contains the test parameters. In all experiments except No. 7, water was supplied to the chamber. Specific energy deposition to methane and water (Table. 1), was calculated as follows:

$$N_{sp} = N_d \cdot \eta / (G_{\text{CH}_4} + G_{\text{H}_2\text{O}}),$$

where G_{CH_4} , $G_{\text{H}_2\text{O}}$ are methane and water flow rate; N_d is the arc power.

The pressure in the plasmatron chamber P_{ch} varied in the range from 0.21 to 0.25 MPa and was determined by both the flow rates of methane and water, as well as the diameters of the adapter outlet nozzles d_n and the plasmatron chamber d_c .

Table 2 contains the results of the measurements of the volumetric fractions of upyrolysis products and the mass fractions μ obtained by calculation. The calculation of mass fractions was based on the results of volumetric

fractions measurements and on the condition that the number of carbon, hydrogen and oxygen atoms in a mixture of methane and water with a total flow of $G_{CH_4} + G_{H_2O}$ is preserved. The fraction of water that reacted was determined using the volumetric fractions of CO in the pyrolysis products. It was assumed that the rest of the unreacted water was discharged with a plume. The last column of Table 2 shows v_{H_2}/v_{CO} , the ratio of the volumetric fractions of H_2 and CO.

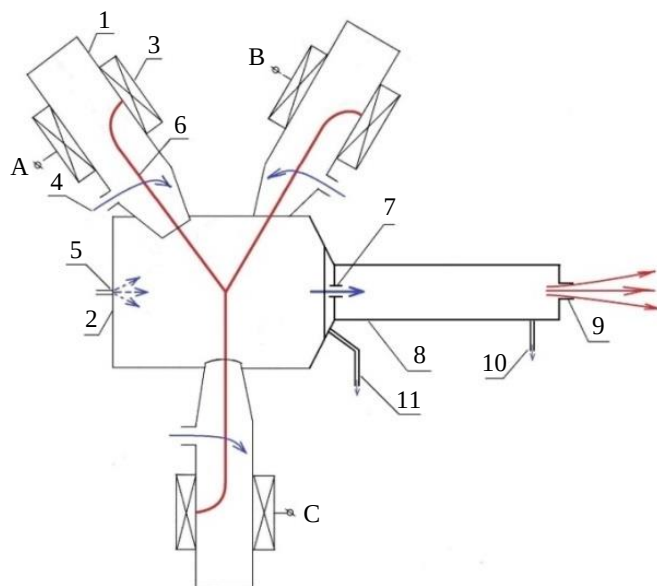


Fig. 1. The plasmatron "Zvezda" scheme: 1 – electrode; 2 – chamber; 3 – magnetic coils; 4 – CH_4 supply to electrodes; 5 – H_2O supply to the plasmatron chamber; 6 – electric arc; 7 – chamber outlet nozzle; 8 – adapter; 9 – nozzle at the adapter outlet; 10, 11 – sampling for chromatographic analysis

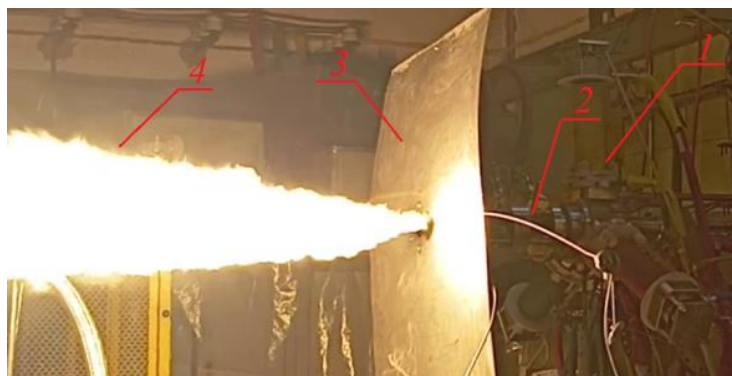


Fig. 2. Photo of the plasmatron start-up: 1 – plasmatron; 2 – adapter; 3 – protective shield; 4 – combustion of pyrolysis products in the atmosphere

Figure 3 shows the dependence of the pyrolysis products volumetric fractions on the inlet ratio of G_{H_2O}/G_{CH_4} flow rates. As can be seen, the hydrogen concentration decreases from 82% to 74% as the flow rate of the supplied water increases. In turn, the CO concentration also decreases, but more significantly, from 15.8% to 10%. There is a significant increase in acetylene concentration as water flow rate increases: from 1% to 8%, the percentage of ethylene and unreacted methane also increases. Water was not

detected in the gaseous pyrolysis products due to its condensation in the pipeline connecting the sampling device to the chromatograph. The ratio of volumetric fractions v_{H_2}/v_{CO} increases from 5.2 to 7.3 with increasing water flow into the plasmatron chamber (Curve 6 in Fig. 3).

The water and carbon concentrations were guided by the dependences of the mass fraction distributions presented in Table 2 and Figure 4, which show the flow rates of pyrolysis products directly in g/s. At the minimum water supply

$G_{H_2O}/G_{CH_4} = 0.55$, the mass fraction of water in the pyrolysis gas is minimal and amounts to 2.6%, the remaining water participated in the reaction. At the same time, the methane conversion rate is more than 99%. As G_{H_2O}/G_{CH_4} increases, the concentration of water in the pyrolysis gas increases to 48% by weight. Figure 4 also shows the dependence of the

equilibrium temperatures in the chamber Te (curve 8), obtained by calculation [23] using data on specific energy consumption N_{sp} . It can be seen that an increase in G_{H_2O}/G_{CH_4} at the inlet leads to a decrease in Te , while a decrease in the content of CO and C is accompanied by an increase in the content of acetylene by more than 4 times.

Table 1

Experimental setting

Experiment No.	G_{CH_4} , g/s	G_{H_2O} , g/s	G_{H_2O}/G_{CH_4}	N_d , kW	N_{sp} , MJ/kg	d_c , mm	d_H , mm	P_k , MPa
1	43.28	24	0.555	1.61	20.62	30	32	0.216
2	44.99	36	0.800	1.52	16.17	30	32	0.235
3	44.08	36	0.817	1.49	15.97	30	32	0.233
4	47.2	66	1.398	1.32	10.02	30	32	0.246
5	46.95	67	1.427	1.35	10.21	30	28	0.241
6	46.92	67	1.428	1.64	12.39	30	28	0.238
7	57.54	0	0	1.384	20.67	30	60	0.182

Table 2

Composition of pyrolysis products

Experiment No.		H ₂	C ₂ H ₄	C ₂ H ₂	CH ₄	CO	C	H ₂ O	v_{H_2}/v_{CO}
1	v , %	82.18	0.48	1.02	0.49	15.83	0	0	5.19
	μ , %	19.07	1.56	3.08	0.91	51.43	21.35	2.61	
2	v , %	77.82	0.45	5.17	1.95	14.62	0	0	5.32
	μ , %	14.77	1.2	12.75	2.96	38.84	10.01	19.48	
3	v , %	78.87	0.597	5.23	1.06	14.24	0	0	5.53
	μ , %	14.83	1.57	12.78	1.59	37.51	10.87	20.84	
4	v , %	74.68	0.65	8.12	5.78	10.77	0	0	6.93
	μ , %	9.17	1.12	12.94	5.68	18.52	6.17	46.39	
5	v , %	74.99	0.91	7.89	6.12	10.07	0	0	7.44
	μ , %	8.89	1.51	12.17	5.81	16.72	6.85	48.05	
6	v , %	74.61	0.77	8.12	6.3	10.20	0	0	7.31
	μ , %	8.86	1.28	12.54	5.99	16.97	6.46	47.91	
7	v , %	77.61	0.97	12.29	9.13	0	0	0	—
	μ , %	17.62	3.08	36.28	16.59	0.000	26.42	0.000	

Using the dependence of Te on G_{H_2O}/G_{CH_4} shown in Fig. 4, calculations of v_{H_2O}/v_{CO} were performed in the equilibrium approximation using [23] (curve 7 in Fig. 3). As can be seen, the nature of the curve obtained as a result of calculations differs significantly from the experimental one: an increase in G_{H_2O}/G_{CH_4} leads

to a decrease in v_{H_2}/v_{CO} to ~ 3.2 , while in the experiment there is an increase in v_{H_2}/v_{CO} (curve 6 in Fig. 3). This is due to the processes non-equilibrium in a chamber. This is confirmed by the results obtained without supplying water to the chamber (line 7 in Tables 1 and 2). In this case, the composition was measured directly before entry into the adapter through the existing

gas sampling channel, item 11 in Fig. 1. As can be seen, in this case, the maximum concentration occurs for acetylene, which indicates the processes non-equilibrium due to the short residence time. As a result, when water is supplied to the chamber from the nozzle, it interacts with both undecomposed methane (16.6% by weight) and acetylene (36.3% by weight). The interaction of acetylene with water has been sufficiently studied at low temperatures

(the Kuchеров reaction). For high temperatures, without catalysts, the kinetic process has been poorly studied. However, the equilibrium calculation shows that, for example, at $G_{H_2O}/G_{C_2H_2} \sim 1$ and a temperature of 1,500 K, the pyrolysis products consist of H_2 , CO , C in a molar fractions ratio of 4.4 : 2.5 : 1, in the absence of C_2H_2 . This indirectly confirms the two-stage nature of methane steam-water reforming in the structure under consideration.

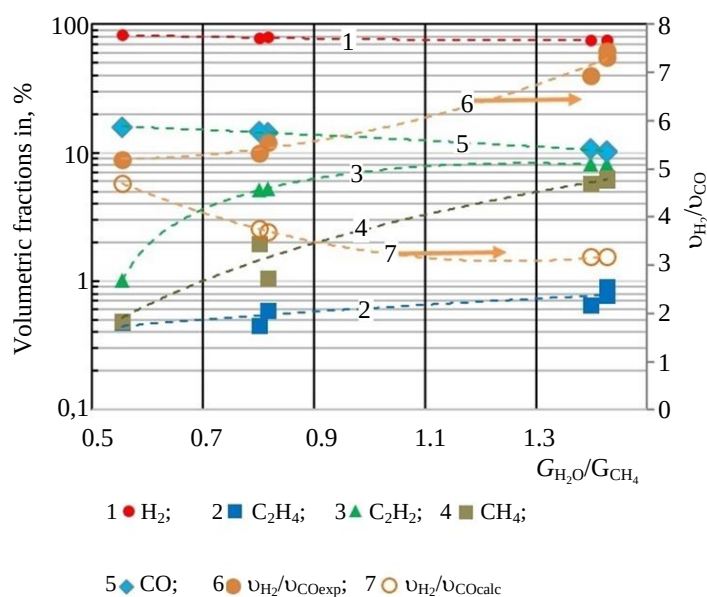


Fig. 3. Dependence of the volumetric fractions of pyrolysis products on the ratio G_{H_2O}/G_{CH_4}

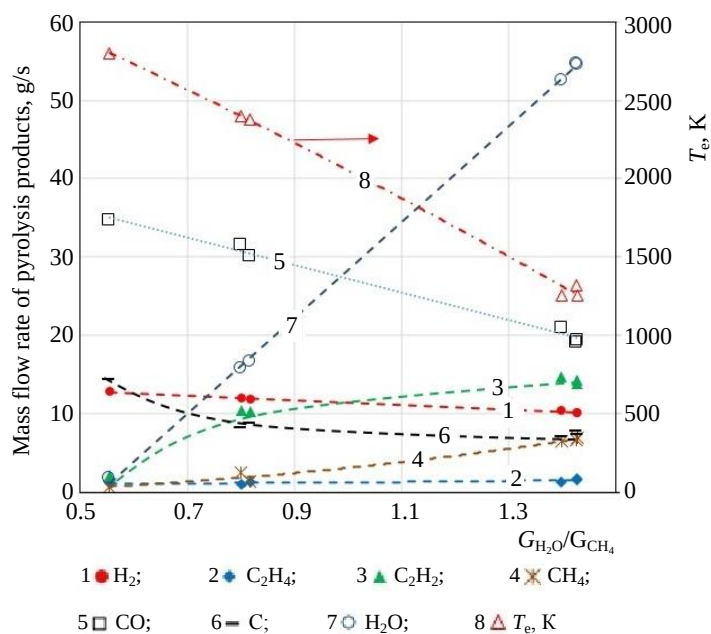


Fig. 4. Dependence of mass flow rates of pyrolysis products on the ratio G_{H_2O}/G_{CH_4}

Conclusion

1. The possibility of autonomous operation on methane of the AC plasmatron with megawatt power, as well as when supplying water to carry out steam–water methane reforming, during which methane is supplied to the arc channels of the plasmatron, and water supply to the plasmatron chamber has been demonstrated. Data on the influence of the methane-to-water flow rate ratio on the concentration of pyrolysis products have been obtained. The operating modes of the plasmatron varied in the following ranges: arc power from 1.32 to 1.61 MW, methane flow rate from 43 to 57.5 g/s, water flow rate from 0 to 67 g/s.

2. It was demonstrated that reducing the water-to-methane flow rate ratio to $G_{H_2O}/G_{CH_4} = 0.55$ results in a minimum $v_{H_2}/v_{CO} \sim$ ratio of 5.2 at a methane pyrolysis degree exceeding 99%. When increasing G_{H_2O}/G_{CH_4} to ~ 1.42 , the ratio of v_{H_2}/v_{CO} increases to ~ 7.3 .

3. It has been established that the investigated processes are significantly influenced by non-equilibrium conditions due to the short residence time of pyrolysis products, and the decomposition process itself proceeds via a two-stage scheme: methane pyrolysis in arc channels – at the first stage, followed by steam–water reforming together with water supplied into the plasmatron chamber – at the second stage.

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