Interaction of methane with thermal carbon dioxide plasma obtained in the AC plasma torch

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Abstract. The article discusses a new method for producing synthesis gas using an ac electric arc. The relationship between the electrical parameters and the energy characteristics of the process was established: with an increase in carbon dioxide concentration in the electrode zone, the hydrogen concentration increases. This leads to an increase in the plasma torch power, which can be used to adjust the chemical process.

1. Introduction
Hydrogen is one of the main products of the chemical industry. It is used to obtain a wide range of organic and inorganic substances: ammonia, methanol, alcohols. The main methods for producing hydrogen are catalytic reforming of natural gas, partial oxidation of natural gas, gasification of solid fuels, electrolysis of water, and waste treatment (landfill gas, gasification, waste fermentation). The most common method is steam reforming of methane, since in this case, a relatively pure synthesis gas with a high molar ratio of H₂/CO is formed. However, this method has significant drawbacks. 1 – it is necessary to obtain water vapor with a high temperature (more than 800°C). While a large amount of steam remains in the reaction products. 2 – the catalyst used is subjected to considerable thermal stress and is poisoned over time [1].

Another major global challenge is increasing greenhouse gas emissions. In this case, the main greenhouse gas is carbon dioxide. It can also be used for methane reforming:

$$\text{CO}_2 + \text{CH}_4 = 2\text{CO} + 2\text{H}_2$$

The presented reaction is strongly endothermic (260.5 kJ/mol and 16.28 MJ/kg of methane), therefore additional energy is needed for its implementation. For this purpose, electrical discharges can be used. Such methods can be divided into two types: equilibrium plasma and non-equilibrium plasma. Non-equilibrium plasma based methods are widely studied: corona discharge, DBD, microwave discharge, glow discharge, gliding arc [2–6]. The main feature of this method is the relatively low temperature of a chemical reaction. At the same time, energy
is spent on the formation of active chemical particles, which significantly accelerate chemical reactions of reforming.

There are also many scientific articles on the application of thermal plasma obtained using dc plasma torches, alternating current and microwave plasma torches.

The article [7] discusses the use of a microwave plasma torch for methane reforming with carbon dioxide. The plasma torch power was 6 kW, the frequency was 2.45 GHz. Flow rates of carbon dioxide and methane were 0.491 and 0.179 g/s, respectively, the molar ratio of CH₄/CO₂ = 1. It was possible to obtain the conversion of methane and carbon dioxide 68.4 and 96.8%, respectively; energy efficiency was 49.01 MJ/kg of converted methane.

Hrabovsky et al. suggested methane reforming in a direct current plasma torch with arc vortex stabilization [8]. The methane flow rate was 0.9–1.8 g/s, the plasma torch power was 88–136 kW. The conversion of methane and carbon dioxide reached 100 and 93%, respectively; energy efficiency was 81.2 MJ/kg of converted methane. Similar results were obtained with steam reforming of natural gas. In this case, liquid water was used to stabilize the arc.

With the use of an ac plasma torch, it was managed to perform the H₂O-CO₂ reforming of methane [9]. While the molar ratio of H₂/CO was 2.1. The experimental facility consisted of a plasma torch with a power of about 120 kW and the plasma-chemical reactor, the selectivity for hydrogen and carbon monoxide was about 100%.

In a plasma-chemical reactor with gliding arcs and a Ni/Al₂O₃ catalyst, conversions were 15% for methane and 18% for carbon dioxide were [10]. Facility power was 190 W. At the same time, energy consumption amounted to 25.84 MJ/kg of converted methane. Such low energy consumption due to the relatively low conversion of raw materials.

Despite a significant amount of research on plasma carbon dioxide reforming of methane, there are no data on the relationship between the electrical parameters of the plasma torch and the composition and properties of synthesis gas. This article discusses the plasma torch ac operating on a mixture of methane and carbon dioxide.

2. Experimental part
The experimental facility (figure 1) is consisted of an ac plasma torch (figure 2), a lined reactor, a power supply system (not shown in the figure 1), a reagent supply system, and synthesis gas composition measurement. The plasma torch consists of three electric arc channels with rod electrodes [11]. In each electric arc channel there are two gas inputs: the electrode zone and

![Figure 1. Plasma test facility. 1 – plasma torch; 2 – plasma reactor; 3 – thermocouples; 4 – additional plasma torch; 5 – synthesis gas output; 6 – sampler.](image-url)
the arc zone. This allows using virtually any gases and vapors, including steam [12], carbon tetrachloride [13]. The plasma torch is powered from the ac power source described in more detail in [14]. No-load voltage is 10 kV, current is 50 A (for this electrodes type).

The flow of carbon dioxide and methane was regulated by a mass flow controllers (MFC). Thermal efficiency of such plasma torches is about 95%.

3. Results and discussion
Previously, it was found that increasing the hydrogen content in the plasma-forming gas increases the power of the plasma torch [12]. This is associated with higher values of heat capacity and thermal conductivity of hydrogen. The plasma torch has a self-adjusting arc, which is ensured by a significant excess of the power of the power source compared to the power of the plasma torch (current source). In this case, any change in power is fully connected with the electrical conductivity of the electric arc. In our experiments, the influence of the share of carbon dioxide flow rate on the plasma torch power was considered. So it can see from table 1, with increasing concentration of carbon dioxide in the electrode zone, the plasma torch power decreases. This is due to the formation of hydrogen by reaction (1). The total amount of each reagent corresponds to the stoichiometric ratio.

To exclude the thermal effect of the reaction, a thermodynamic calculation of plasma reforming was carried out with mass flow rates of CO\textsubscript{2} (6.9 g/s) and CH\textsubscript{4} (2.4 g/s) and the plasma torch thermal power (power taking into account thermal efficiency). Thermodynamic calculations were carried out using the Chemical Work Bench 3.5 program (an adiabatic reactor with constant pressure and energy input). The stoichiometric process described by reaction (1) requires 21.26 MJ/kg of converted methane (under adiabatic conditions). As can be seen from table 2, the composition of the products practically does not change with increasing power. The average temperature of the reaction products increases. In most cases, with a constant composition, an increase in temperature leads to an increase in the power leads to increase in electrical conductivity and decrease a temperature. The conductivity of an electric arc was determined experimentally from information about a voltage drop and an electric current. At the same time, near-electrode processes were not taken into account, since their values are substantially smaller than the total voltage drop the electric arc [11]. It can be seen from table 2 that the electrical conductivity increases. The only factor that can affect this is the volume of the area occupied by hydrogen and its concentration in this area. With an increase in the proportion of carbon dioxide supplied to the electrode zone, this area cannot change. Only the concentration of hydrogen in the electrode region can change.

**Figure 2.** Diagram of the ac plasma torch. 1 – electrodes; 2 – arc channel; 3 – electric arc; 4 – supply of protective gas (the electrode zone); 5 – supply of plasma gas (the arc zone).
Table 1. Experimental parameters.

| No | Flow rate of CH$_4$ to electrode zone (g/s) | Flow rate of CH$_4$ to arc zone (g/s) | Flow rate of CO$_2$ to electrode zone (g/s) | Flow rate of CO$_2$ to arc zone (g/s) (on reaction) | Voltage drop (V) | Electric power (kW) |
|----|---------------------------------|---------------------------------|---------------------------------|---------------------------------|-----------------|--------------------|
| 1  | 1.4                             | 3.2                             | 0.145                           | 3.7                             | 1405            | 123.36             |
| 2  | 1.4                             | 3.3                             | 0.150                           | 3.6                             | 1389            | 120.68             |
| 3  | 1.4                             | 3.5                             | 0.159                           | 3.4                             | 1364            | 119.63             |
| 4  | 1.4                             | 3.6                             | 0.164                           | 3.3                             | 1355            | 118.67             |
| 5  | 1.4                             | 3.7                             | 0.168                           | 3.2                             | 1329            | 116.08             |

Table 2. Calculation of the equilibrium composition and electrical conductivity of the arc.

| No | CO  | H$_2$ | H$_2$O | CO$_2$ | Thermal power (kW) | Average temperature (K) | Arc conductivity (S) |
|----|-----|-------|--------|-------|---------------------|------------------------|----------------------|
| 1  | 50.47 | 48.46  | 0.92  | 0.15  | 117.2               | 3201                   | 0.0364               |
| 2  | 50.47 | 48.47  | 0.91  | 0.15  | 114.6               | 3171                   | 0.0365               |
| 3  | 50.47 | 48.47  | 0.91  | 0.15  | 113.6               | 3159                   | 0.0375               |
| 4  | 50.47 | 48.48  | 0.90  | 0.15  | 112.7               | 3147                   | 0.0377               |
| 5  | 50.48 | 48.48  | 0.89  | 0.14  | 110.3               | 3116                   | 0.0383               |

4. Conclusion
It has been established that the electrical conductivity can serve as a control parameter for determining the hydrogen content in the plasma torch. In this case, inverse relationship is possible (by changing the area of the electric arc), occupied by hydrogen, it can change the plasma torch power. This parameter significantly affects the temperature in the reactor and the entire plasma-chemical process. Therefore, by changing the proportion of carbon dioxide supplied to the electrode zone of the plasma torch, it is possible to change the technological parameters of the plasma-chemical facility for processing methane and carbon dioxide.

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