Powerful high-voltage AC plasma torches for plasma-chemical applications

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\textbf{Abstract.} The paper deals with a series of high-voltage AC plasma torches with power up to 500 kW with rod and hollow electrodes. The life time of continuous operation of electrodes reaches 1000 hours. The application of these plasma torches for air and steam-air reforming of methane is studied. The obtained results are compared with autothermal air reforming. The minimum specific power inputs for air and steam-air reforming of methane were 2.5 MJ/kg of methane 12.5 MJ/kg of methane, respectively.

1. Introduction

Natural gas is the most promising raw material for the production of hydrogen and synthesis gas for various chemical applications. In the world, the main amount of hydrogen is used to produce ammonia, and the main method for hydrogen production is steam catalytic reforming and partial oxidation:

\begin{align*}
\text{CH}_4+\text{H}_2\text{O} &= \text{CO}+3\text{H}_2 \\
\text{CH}_4+0.5\text{O}_2 &= \text{CO}+2\text{H}_2
\end{align*}

The second reaction flows with a low heat release, which can not cover the heat loss of the reactor, therefore part of the methane is burned [1]. That increases the amount of carbon dioxide and reduces the yield of hydrogen.

Plasma reforming of natural gas is a new methane conversion method. For this purpose, different types of discharges are used: DBD [2], glow discharge [3], corona discharge [4], gliding arc [5], air [6] and steam plasma torches [7]. The relatively low capacity of the plants is the main disadvantage of the presented methods. Nitrogen is ballast at air reforming of methane and it must be removed for the production of pure hydrogen. However, in the production of ammonia, nitrogen acts as a reagent, so after removal of nitrogen oxides, the synthesis gas can be used for ammonia production. The disadvantage of this method is low hydrogen content in the produced synthesis gas.
The paper deals with the air plasma reforming of methane using a high-power high-voltage AC plasma torch. The consumed electric power is used for heating the components to the reaction temperature.

2. Plasma torches

Two plasma torches are considered in the work: powerful air plasma torch and steam-air plasma torch. Figure 1 shows an air electric arc three-phase AC plasma torch with hollow cylindrical electrodes with power up to 500 kW [8].

![Figure 1 Air three-phase AC plasma torch with hollow cylindrical electrodes.](image)

The plasma torch has three cylindrical discharge channels, the plasma-forming gas is supplied tangentially, providing stabilization of the arcs in the axial region of the discharge channels, as well as providing circular motion of the arc spots along the inner surface of the hollow electrodes. Solenoids are installed on the electrodes in order to increase the speed of arc spot motion and to provide less erosion [9]. The arcs are ignited by high voltage of the idling power supply (10 kV) between electrodes and the walls of the discharge channels. The thermal efficiency of the plasma torch is about 90 - 95%, which is also significantly higher than in most DC plasma torches (60-80%). Figure 2 shows the dependence of the thermal power of the plasma torch (taking into account the thermal efficiency) on the air flow rate.

![Figure 2 Dependence of thermal power of three-phase AC plasma torch with hollow electrodes on the air flow (current ~ 83 - 86 A).](image)

High power of the presented plasma torch is provided due to high arc voltage drop (to 3 kV) at a relatively low current (to 100 A). It takes place due to the burning of long alternating current arcs (up
to 200 cm) in the plasma torch [10]. This type of plasma torches has the potential to increase power up to 1 MW or more.

Figure 3 shows a schematic of steam-air three-phase AC plasma torch [11].

![Figure 3 Steam-air three-phase AC plasma torch](image)

Figure 3 Steam-air three-phase AC plasma torch. 1 - electrode; 2 - arc channel; 3 - electric arc; 4 - supply of shielding gas (air); 5 - supply of plasma-forming gas (steam); 6 - axis of optical measurements.

The presented plasma torch also has three discharge channels, in which rod copper water-cooled electrodes are installed. Plasma-forming medium is steam and air. Air is used as a shielding gas and supplied tangentially to the electrode area. Steam with temperature of ~ 200 °C is tangentially fed into each channel in the arc burning zone, thus ensuring their axial stabilization.

Figure 4 shows the dependence of the thermal power of the steam-air three-phase AC plasma torch on the air flow rate for various electrical currents. Thermal efficiency of the plasma torch is 94 - 95.5%.

![Figure 4 Dependence of thermal power](image)

Figure 4 Dependence of the thermal power of steam-air three-phase AC plasma torch on the air flow rate at various electrical currents. Steam flow rate is 3.7 g/s.

As can be seen from the graph, the power is significantly dependent on the protective air flow rate. For homogeneous plasma-forming media, an increase in flow rate leads to an increase in the arc voltage drop. However, in the case of steam and air mixture, as the protective air flow rate increases with a constant steam flow rate, the arc voltage drop decreases. Steam plasma has significantly lower electrical conductivity than air [12]. Therefore, the electrical conductivity of the arc increases, and the voltage drop is reduced by reducing the mass concentration of steam in the plasma-forming mixture. Investigation of erosion allows it possible to estimate the lifetime value of continuous
operation of electrodes, taking into account their mass (up to 300 hours). This plasma torch can provide a flexible regulation of the power input and the composition of the plasma-forming medium.

3. Estimation

Calculations were performed in the approximation of thermodynamic equilibrium using Chemical Workbench 3.5 program. Autothermal reforming of methane by air without heat input, allothermic methane reforming by air plasma at 1500 K, allothermal reforming by plasma of a mixture of steam and air were studied. In all cases, the adiabatic regime was considered. The composition of the gasification agent for steam-air plasma reforming is: H₂O - 83.33% mass, N₂ - 12.58% mass, O₂ - 3.86% mass, Ar - 0.22% mass, CO₂ - 0.01% mass. Calculations were carried out for 1 kg of methane, the gasifying agent flow rate was varied in three modes: pyrolysis, partial oxidation, combustion. In the pyrolysis regime, part of the methane was converted to graphite, in the partial oxidation regime, methane was converted to hydrogen and carbon monoxide, in the burning regime methane was converted to water and carbon dioxide.

4. Results and discussion

Figures 5, 6 show the dependence of equilibrium composition of the products of the reforming reaction of methane from the specific air flow rate. As can be seen from figure 5, the content of solid products (graphite) is observed up to 5 kg/kg of air. This value corresponds to the maximum content of hydrogen (33.2% mol.) and carbon monoxide (15.4% mol.). Further, water, oxygen and nitrogen oxides content is increasing as a result of methane burning. Figure 6 illustrates the growth of the adiabatic temperature from the specific air flow rate. With the increase in the specific air flow rate, the temperature increases continuously, however, at the transition from methane reforming to combustion, the rate of temperature growth increases. The temperature drop after 16.5 kg/kg is explained by the excess of air for complete methane combustion.

Figures 7 and 8 show calculation results of air plasma methane reforming. The process temperature is assumed constant (1500K) for simplification. As can be seen from figure 7, there are no solid products (graphite) in the reaction products at specific air flow rate of 4.2 kg/kg. The limiting content of hydrogen and carbon monoxide is 40 and 20% mol., respectively. Further increase in the specific air flow rate leads to increase in the concentration of nitrogen and water in the reaction products. Figure 8 shows the dependence of the plasma enthalpy on the specific flow rate of the air plasma. The calculated value of the required plasma enthalpy is 2.5 MJ/kg. For the air plasma torch discussed above, the characteristic plasma enthalpy is 3.63-5.09 MJ/kg, which taking into account the thermal losses in the plasma-chemical reactor is sufficient for the chemical process.
Figure 7 Dependence of the composition of allothermic methane reforming products on the air plasma flow rate

At steam-air plasma methane reforming (Figure 9), the hydrogen content in reaction products (71% mol.) is significantly higher than at autothermal reforming and air plasma reforming. This is due to steam methane reforming (equation 1). The plasma enthalpy at a given ratio of air and steam (1:5) reaches 12.5 MJ/kg. This value is slightly less than the specified value (18 MJ/kg, figure 10). Specific plasma flow rate of 1.47 kg/kg and the following composition of synthesis gas: H₂ - 63.9% mol., CO - 20.0% mol., H₂O - 11.5% mol., N₂ - 3.2% mol., CO₂ - 1.4% mol. will correspond to this enthalpy.

Figure 8 Dependence of the required enthalpy of plasma on the air plasma flow rate

The obtained results show that in the steam-air reforming of methane, the nitrogen content is the lowest, which under certain conditions will make it possible to obtain a mixture of hydrogen and nitrogen for the production of ammonia. It is important that the autothermal process has low stability (flame failure is possible). A plasma torch application eliminates this problem and the synthesis gas with different H₂/CO ratios (from 2 to 3.5) can be obtained.

Figure 9 Dependence of the composition of allothermic methane reforming products on the steam-air plasma flow rate

Figure 10 Dependence of the required enthalpy of plasma on the steam-air plasma flow rate

5. Conclusion

Air reforming is characterized by low power inputs, but the produced synthesis gas contains a lot of ballast nitrogen, which hampers further chemical processing. When energy is supplied by plasma torches, the yield of hydrogen can be increased, which will allow synthesis gas production with different H₂/CO ratios. In addition, the plasma torch provides a stable flame front increasing the chemical facility stability.
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6. References

[1] Loukou A, Mendes M A A, Frenzel I, Pereira J M C, Ray S, Pereira J C F and Trimis D 2017 *International Journal of Hydrogen Energy* **42** Issue 1 652–663

[2] Khoja A H, Tahir M and Amin N A S 2017 *Energy Conversion and Management* **144** 262–274

[3] Cheng D, Zhu X, Ben Y, He F, Cui L, Liu C 2006 *Catalysis Today* **115** Issues 1–4 205–210

[4] Nguyen H H, Nasonova A, Nah I W and Kim K-S 2015 *Journal of Industrial and Engineering Chemistry* **32** 58–62

[5] Rafiq M H, Jakobsen H A and Hustad J E 2012 *Fuel Processing Technology* **101** 44–57

[6] Jo S, Lee D H and Song Y-H 2013 *International Journal of Hydrogen Energy* **38** Issue 31 13643–13648

[7] Czylkowski D, Hrycak B, Jasiński M, Dors M and Mizeraczyk J 2016 *Energy* **113** 653–661

[8] Surov A V, Popov S D, Popov V E, Subbotin D I, Serba E O, Spodobin V A, Nakonechny Gh V and Pavlov A V 2017 *Fuel* **203** Issue 1 1007–1014

[9] Surov A V, Popov S D, Serba E O, Nakonechny G V, Spodobin V A, Ovchinnikov R V, Kumkova I I. and Shabalin S A 2012 *Journal of Physics: Conference Series* **406** 012007.

[10] Rutberg Ph G, Popov S D, Surov A V, Serba E O, Nakonechny Gh V, Spodobin V A, Pavlov A V and, Surov A V 2012 *Journal of Physics: Conference Series* **406** 012028

[11] Rutberg Ph G., Kuznetsov V A, Serba E O, Popov S D, Surov A V, Nakonechny Gh V, and Nikonov A V 2013 *Applied Energy* **108** 505 – 514.

[12] Rutberg F G, Kuznetsov V A, Serba E O, Nakonechnyi G V, Nikonov A V, Popov S D, Surov A V 2013 *High Temperature* **51** Issue 5 608–614.