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Atmospheric pressure microwave plasma source for hydrogen production

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ABSTRACT

Nowadays hydrogen is considered as a clean energy carrier and fuel of the future. That is why the interest in production and storage of hydrogen is still increasing. One of the promising technology is using microwave plasma for hydrogen production. In this study we propose two types of an atmospheric pressure microwave plasma source (MPS) for hydrogen production via methane conversion. The first one was a nozzleless waveguide-supplied coaxial-line-based. The second one was a nozzleless waveguide-supplied metal-cylinder-based. They can be operated with microwave frequency of 2.45 GHz and power up to a few kW with a high gas flow rates (up to several thousands l/h). We present experimental results concerning electrical properties of the MPS, plasma visualization, spectroscopic diagnostics and hydrogen production. The experiment was carried out with methane flow rate up to 12,000 l/h. An additional nitrogen or carbon dioxide swirl flow was used. The absorbed microwave power was up to 5000 W. Our experiments show that MPSs presented in this paper have a high potential for hydrogen production via hydrocarbon conversion.

1. Introduction

Development of plasma sources based on the microwave technique are of increasing interest from industrial point of view. Microwave sustained plasma has been widely investigated [1–5] and found practical applications in various fields, like gas purification [6], decomposition of gaseous pollutants [7], sterilization, food preservation, living cells treatment [8], surface modification [9,10] etc. Recently, one of the promising application is using microwave plasma for hydrogen production [11–13]. Hydrogen is more and more attractive as an efficient and environmental friendly source of energy. It is considered as a promising fuel of the future. According to a European roadmap for hydrogen and fuel cells published by European Commission in "Hydrogen Energy and Fuel Cells: A vision of our future" [14] in the medium to long term (it means beyond 2020) the main attention should be paid for developing and implementing systems for hydrogen production from renewable electricity and biomass. At the same time the research and development of other carbon-free hydrogen sources, such as solar thermal and advanced nuclear should be continued.

Hydrogen can be produced from different kind of feedstock like fossil fuels, water, biomass. Recently, in industry methane or natural gas reforming is widely used to obtain hydrogen or synthesis gas, which are utilized, for example as source materials for the production of raw chemicals (e.g. methanol and ammonia), as well as hydrogenation agents in oil refinery and reducing gases in steel industry. There are a few different methane based methods of hydrogen production like:

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pyrolysis, steam reforming, dry reforming, partial oxidation, auto-thermal reforming. They process through the following reactions:

$CH_4 \rightarrow C + 2H_2$ (pyrolysis)

$CH_4 + H_2O \rightarrow CO + 3H_2$ (steam reforming)

$CH_4 + CO_2 \rightarrow 2CO + 2H_2$ (dry reforming)

$CH_4 + 0.5O_2 \rightarrow CO + 2H_2$ (partial oxidation)

$2CH_4 + O_2 + CO_2 \rightarrow 3CO + 3H_2 + H_2O$ (auto-thermal reforming)

$4CH_4 + O_2 + 2H_2O \rightarrow 4CO + 10H_2$ (auto-thermal reforming)

The advantage of methane pyrolysis is that it produces almost pure hydrogen, however, often accompanied with C_2H_2 production. Other benefit of methane pyrolysis is the lack of CO_2 emissions. The method disadvantage is production of undesirable carbon which forces the use of the gas-soot separators and filters. It is also expensive due to high energy consumption. Steam reforming and partial oxidation are performed at high temperature (700–900 °C) and in the presence of catalysts. The former method, based on the reaction of water with methane, produces hydrogen and carbon monoxide. The dry reforming uses reaction of carbon dioxide with methane resulting in hydrogen and carbon monoxide. A review of different technologies concerning hydrogen production is published in Ref. [15].

In this paper we propose a method for production of hydrogen via methane conversion in the atmospheric-pressure plasma generated by the nozzleless waveguide-supplied coaxial-line-based MPS and by the nozzleless waveguide-supplied metal-cylinder-based MPS [16]. It was already shown that microwave plasma can be successfully used in hydrogen production by decomposition of methanol and ethanol [12], methane and tetrafluoroethane [7,17,18] and water [19]. The MPSs presented in this paper can be operated at frequency of 2.45 GHz with microwave power up to a few kW and a high gas flow rates (up to several thousands l/h) at atmospheric pressure. The plasma in the form of a flame is generated inside a quartz tube. To assess the usefulness of our MPS for the hydrogen production the following experimental works were undertaken. At the beginning, to determine the efficiency of microwave power transfer to the plasma and stability of MPS operation, the tuning characteristics of the MPS were measured for different work conditions. Next, the photos of methane plasma flame were taken. The spectroscopy measurements in the meaning of optical emission spectroscopy were carried out to determine the gas temperature. Finally, the conversion of methane was investigated as a function absorbed microwave power and gas flow rate. The hydrogen production rate and energy efficiency were calculated from the experimental results.

2. MPS and experimental setup

In this study two types of MPS were used for investigations of hydrogen production via methane conversion in an atmospheric pressure microwave plasma. The first one was the nozzleless waveguide-supplied coaxial-line-based MPS, and the second was the nozzleless waveguide-supplied metal-cylinder-based MPS.

Schematic view of the nozzleless waveguide-supplied coaxial-line-based MPS is shown in Fig. 1. The MPS was based on a standard WR 340 rectangular waveguide with a section of reduced-height preceded by a tapered section. The tapered section assured smooth transition from the standard waveguide dimensions to the waveguide of reduced height. There was an inner cylinder electrode of outer diameter of 20 mm which penetrated the MPS through circular gaps on the axis of the waveguide wide wall and protruded below bottom waveguide wall. The inner cylindrical electrode protruding length d influences the plasma operation and stability. The end part of the inner cylindrical electrode could be changed. During the experiment, the end parts made of brass and tungsten were used. The inner cylindrical electrode was surrounded by a quartz tube. Both the inner cylindrical electrode and the quartz tube were surrounded by outer cylindrical electrode on the outside of the waveguide. The inner diameters of the quartz tube and outer cylindrical electrode were 36 mm and 47 mm, respectively. The inner cylindrical electrode and the outer cylindrical electrode formed a coaxial line at a distance l. The distance l of the coaxial line plays the crucial role on the plasma generator tuning characteristics. The working gas was injected axially to the plasma by the inner cylindrical electrode. An additional gas was introduced to the plasma by four gas ducts which formed a swirl flow inside the quartz tube. The swirl concentrated near the quartz cylinder wall, stabilized plasma generation and protected the quartz tube wall from overheating. The plasma in the form of a flame was generated inside a quartz tube above the end part of the inner cylindrical electrode. The plasma could be observed through viewing and visualization windows. The initiation of the plasma was done without any admixture of noble gases.

In Fig. 2 the schematic view of the nozzleless waveguide-supplied metal-cylinder-based MPS is shown. Its design is similar to the coaxial-type MPS described above but there are a few very important differences. First one is that there is no internal electrode thus there is no axial gas flow. Secondly, there is no any section of reduced height. The internal dimensions of the MPS are the same like the internal dimensions of the WR 340 standard waveguide (86.4 × 43.2 mm). The operating gas is introduced into the plasma via four gas ducts forming a swirl.

The overall diagram of the experimental setup is shown in Fig. 3. It consists of a microwave generator system, microwave power measuring system, gas supply and flow control, plasma generator, gas analysis system and optical emission spectroscopy (OES) system. The microwave generator system uses a high voltage power supply with a control unit and a magnetron head equipped with a circulator. Circulator protects the magnetron head against damages caused by the reflected microwave power. The system generates the