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Microwave plasma-based method of hydrogen production via combined steam reforming of methane

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ABSTRACT

This paper presents a contribution to the development of microwave plasma-based technology for hydrogen production. The efficiency of hydrogen production via combined steam reforming (i.e. with the addition of CO_2 and water vapour) of methane in a waveguide-supplied metal cylinder-based microwave plasma source (MPS) was for the first time tested experimentally. The operating parameters were: microwave frequency of 2.45 GHz, maximum absorbed microwave power of 6 kW, and working gas (methane + CO_2 + water vapour) flow rates up to 9000 NL/h. The tested parameters of hydrogen production efficiency were: the hydrogen production rate, energy yield, methane conversion degree, and hydrogen volume concentration in the outlet gas. It was proven that using the microwave system, the plasma steam reforming of methane can be run stably at high gas flow rates (several thousand NL/h). By optimizing the process input parameters, i.e. the absorbed microwave power, working gas composition and flow rate, an energy yield of hydrogen production of 42.9 g(H_2)/kWh could be achieved. The test showed that the microwave plasma method presented in this paper can also be used efficiently for reforming other gaseous and liquid compounds. In this paper, a new plasma hybrid system for H_2 production is also presented.

1. Introduction

Clean energy and alternative energy have become major areas of research worldwide for sustainable energy development [1]. With circumstances of global warming and diminishing fossil fuel reserves, the newly developed energy must simultaneously meet the requirements of being environmentally-friendly and renewable. Owing to these restrictions, hydrogen is one of the most promising alternative energy carriers [2]. Therefore, developing and implementing systems for hydrogen production and storage are of high importance. The importance of hydrogen for the future energy economy is listed in the energy development strategy of many developed countries (e.g. the European roadmap for hydrogen and fuel cells of the European Commission [3] and the US Department of Energy "National hydrogen energy roadmap" [4]). According to the European Commission, in the medium to long term (beyond 2020), the main focus should be on developing and implementing systems for hydrogen production from renewable electricity and biomass. According to the U.S. Department of Energy (DOE), the distributed hydrogen production, i.e. production of hydrogen at points of use, may be the most viable approach for introducing hydrogen as an energy carrier because it does not require a substantial transport and delivery infrastructure or large capital investments as those needed for large central production plants. In this case, such technologies as natural gas reforming, electrolysis, reforming of ethanol and methanol (both from biomass) are pursued. The 2020 targets of the DOE regarding hydrogen production in a distributed scale are lower production costs and higher production efficiency.

Recently, another technology has been proposed for hydrogen production [5–27]. This technology uses thermal and non-thermal plasmas. In thermal plasmas (e.g. arc plasma), all the charged species (electrons and ions) and neutral species (atoms, molecules and radicals) are almost in thermodynamic equilibrium. This means that the temperatures of the species are the same. Typical gas temperatures of thermal plasmas are several thousand K. On the

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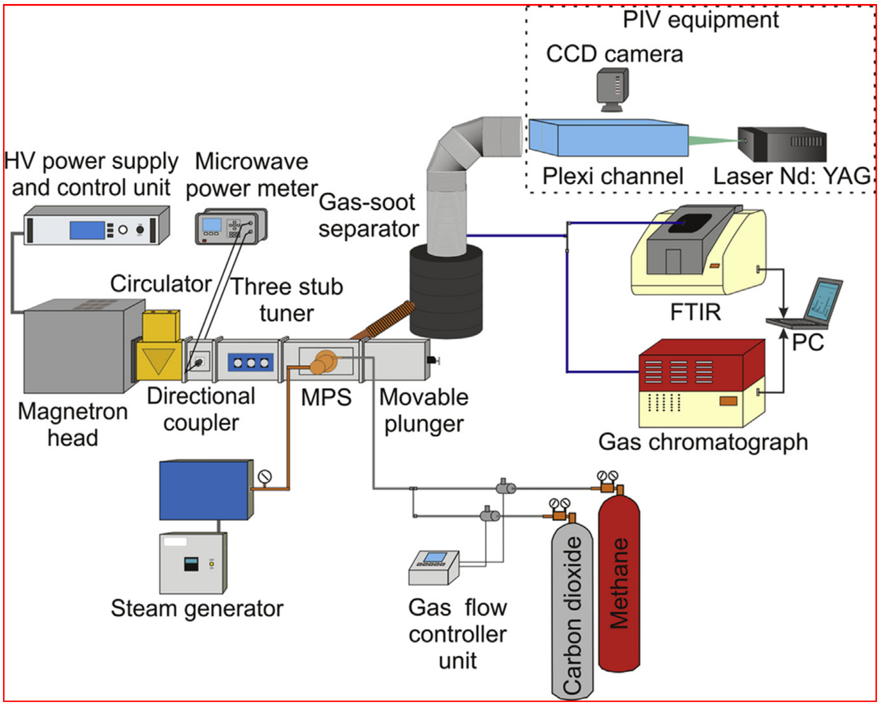

Fig. 1. Sketch of the experimental setup for hydrogen production via combined steam reforming of methane.

other hand, the non-thermal plasmas (e.g. corona discharge, dielectric barrier discharge (DBD) and surface discharge plasmas) are in the thermodynamic non-equilibrium, in which the electron temperature (several tens of thousands of K) is much higher than the temperatures of the other species mentioned above (i.e. ions and neutrals). The plasmas can be generated at a lower or higher (usually atmospheric) pressure of the working gas. Although for the lower pressure plasmas, such as the radio frequency (RF) or microwave (MW) plasma, an efficient conversion of hydrocarbons with high hydrogen selectivity can be achieved, the low hydrogen production rate and the extra energy requirement for vacuum vessels restrict the practical use of the low pressure method.

The atmospheric-pressure plasmas are very attractive for gas treatment because, firstly, the plasma gas temperatures can be relatively high (10^3–10^4 K), and secondly, the plasmas are a source of very active ions, radicals and high-energy electrons, which enhance the chemical reaction rates thus enabling expensive and impurity vulnerable catalysts to be avoided. When steam is used as the plasma supporting gas, reductive and oxidative radicals such as H, OH, and O are produced in the plasma, enabling the plasma to be more effective for processing various hydrogen-abundant compounds, among them gaseous (methane, natural gas, biogas, etc.) and liquid (gasoline, heavy oils, alcohols, etc.) fuels.

Various atmospheric-pressure plasmas have been used for hydrogen production on an experimental scale in the laboratory. They were plasmas of a gliding arc [5–8], dielectric barrier discharge [9–11], corona discharge [12–14], and microwave discharge [15–27]. This technology uses plasmas for reforming gaseous (e.g. methane, natural gas), liquid (e.g. ethanol, water) and gasified solid (e.g. coal, charcoal) compounds containing hydrogen. They can originate both from fossil fuels and biomass.

The microwave plasma sources (MPSs) at atmospheric pressure stand out from other plasma sources offering significant advantages. The role of microwaves is to supply the energy to the plasma generator to create the plasma in gases. MPSs are considered to be more efficient in energy transfer [28]. When properly designed, a high efficiency (almost 100%) of microwave power transfer from the microwave generator to the plasma can be achieved. Since microwave plasma is operated by electricity, a fast response time of high density of electrons and active species, such as ions and free radicals can be achieved. Owing to the electrodeless operation, a high purity plasma can be obtained using microwaves. As a result of this, the use of microwave plasma for gas processing like gas purification and abatement of gaseous pollutants is attractive. The microwave plasma capability, its high reactivity, as well as its high energy density, results in compactness of the plasma reactors. Various designs of MPSs fed through microwave waveguides [29,30], coaxial line components [31,32] and micro-strip lines [33,34] have been developed.

In this paper, a methane-based method is presented for the production of hydrogen using atmospheric pressure plasma generated by a waveguide-supplied metal cylinder-based MPS described in Refs. [17,35]. The waveguide MPS can generate the microwave plasma of a power of several kW. Compared to other MPSs, it enables the processing of large volumes (several thousand NL/h) of different gases, also mixtures of CH_4, CO_2 and water vapour. The waveguide-supplied metal cylinder-based MPS is versatile and offers various processing operations such as pyrolysis, dry reforming, and combined steam reforming that is used for hydrogen production. However, in the case of methane pyrolysis, the soot produced limits the more long-term stable operation of the MPS [35]. The other reforming processes, i.e. the dry reforming and combined steam reforming of methane have been expected to reduce the soot production. Our previous experimental results of microwave plasma dry reforming of methane confirmed this supposition [17]. The objective of this paper was to test the usefulness of the microwave plasma combined steam reforming of methane in mixtures of CH_4, CO_2 and water vapour for sootless and efficient production of hydrogen, using the same MPS as in Ref. [17]. To our knowledge, the microwave plasma combined steam reforming of methane in mixtures of CH_4, CO_2 and water vapour has not yet been carried out.