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Full Length Article

Hydrogen-enriched gas production from kerosene using an atmospheric pressure microwave plasma system

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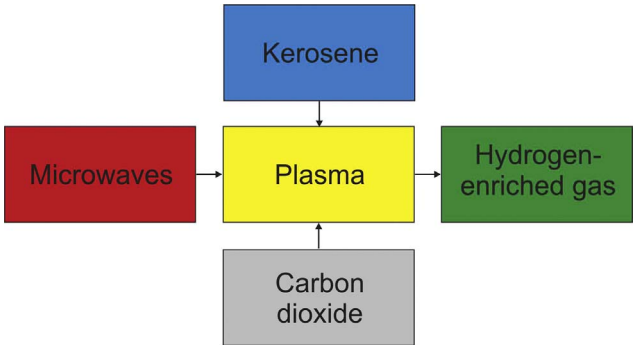
GRAPHICAL ABSTRACT



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ABSTRACT

Growing interest has recently been observed in hydrogen production and storage technologies. Proposed here is a catalyst-free method of hydrogen-enriched gas production by decomposition of kerosene vapour in a carbon dioxide microwave (915 MHz) plasma system operating at atmospheric pressure. The results are given for both experimental and theoretical investigations. The roles analysed included the energy supply to the microwave plasma, the kerosene flow rate and the carbon dioxide flow rate, on the concentrations of gaseous compounds resulting from the kerosene processing. Carbon dioxide, hydrogen, carbon monoxide, methane, acetylene and ethylene all occurred as gaseous by-products. It was observed that 470 NL of hydrogen could be obtained from 1 L of kerosene at an absorbed microwave power of 6 kW. The results confirm the capability of an atmospheric pressure microwave plasma system in decomposing kerosene in the production of hydrogen-enriched gas.

1. Introduction

Hydrogen is considered to be an environmentally clean energy carrier and a promising fuel of the future, and while it is the most abundant element in the Universe it does not exist in its molecular form in nature. However, hydrogen is a constituent of almost all gaseous, liquid and solid state organic compounds. The various types of renewable and non-renewable sources potentially available for hydrogen production include fossil fuels, alcohols, water and biomass. However, hydrogen recovery from these resources does require the application of energy to the production process. Due to its physical and chemical properties, hydrogen storage and handling is technically problematic,

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which is why hydrogen is still not widely used as a fuel. Distributed hydrogen production (production at the point of use) [1–5], on-board hydrogen production [6–10] and hydrogen-enriched gas production (gas mixture of hydrogen with other components) [10–12] all appear to share the potential to avoid these drawbacks. The past few years have seen increased interest in hydrogen-enriched hydrocarbon combustion, both gaseous and liquid. It has been shown that hydrogen-enriched gas supplementation of internal combustion processes could potentially reduce NO_x and CO emissions and reduce fuel consumption, combined with stable combustion flame fronts allowing operation under lean conditions [10,13–19]. Different kinds of technologies have been proposed for on-board hydrogen production, such as a low current plasmatron fuel converters [19], gliding arc plasma reactors [20], and scaled-down autothermal membrane reformers [9], to name but a few. The plasma processing of gases and liquids by microwave discharge is of high interest due to its numerous advantages, one of which being that the plasma is a source of ionised and excited states of atoms, molecules and radicals, with highly reactive properties [21,22]. Due to the high density of the microwave plasma, which involves highly reactive species that enhance the chemical reaction rates, the catalyst-free processing of hydrocarbons becomes possible [22], and thus avoiding the problems related to catalyst deactivation (chemical, mechanical, and thermal) and regeneration [23]. Microwave plasma (915 MHz, 2.45 GHz) achieves a higher density of reactive radicals in comparison with RF plasma (2–60 MHz), and also ensures faster response times. A well designed microwave plasma system with suitably selected operational parameters can achieve almost 100% microwave power coupling efficiency, from microwave generator to the plasma. Another advantage is the possibility of electrodeless operation, resulting in high purity plasma. This is why this paper describes a method of hydrogen-enriched gas production, based on kerosene combined with an atmospheric pressure microwave plasma system. Atmospheric pressure operation, in contrast to systems using a vacuum plasma, simplifies the method and reduces the operational costs. Kerosene is a mixture of liquid hydrocarbons and is widely used in industry (e.g. as jet engine fuel) and in households (e.g. as lighter fuel and heating oil). The plasma reforming of kerosene has been studied both theoretically and experimentally [11,24,25]. We have published the experimental results of microwave plasma kerosene reforming related to hydrogen production in earlier papers [26,27]. In [26] the optical emission spectroscopy study of CO_2 plasma with the addition of kerosene vapour was presented, while in [27] the hydrogen production rate and the energy yield of hydrogen production as the hydrogen production efficiency parameters were evaluated. This paper focuses on the concentrations of gaseous compounds resulting from kerosene processing in a CO_2 plasma as a function of the energy delivered to the microwave plasma, kerosene flow rate and CO_2 flow rate. The results are presented of both the experimental and the theoretical studies.

Section 2 of this paper presents the microwave plasma system for hydrogen-enriched gas production from kerosene including a waveguide-based microwave plasma source (MPS). Section 3 presents the experimental results, while Section 4 (Numerical modelling the Thermodynamic Equilibrium Reactor (TER)) describes the model, the results of the gas composition simulations and the sensitivities of all calculated species to the reactions used in the model. Finally, Section 5 contains a summary and the conclusions.

2. Experimental method

The experimental method of hydrogen-enriched gas production is based on the decomposition of kerosene vapour in a carbon dioxide microwave plasma, at atmospheric pressure. A schematic representation of the system is shown in Fig. 1, which consists of a 915 MHz microwave generator equipped with a circulator and water load, bi-directional coupler with dual channel power meter, three-stub tuner, microwave plasma source (MPS), adjustable plunger, kerosene tank, vaporizer supply and control unit, induction vaporizer, CO_2 compressed gas tank, gas flow control of the thermal mass flow type, gas-soot separator and SRI 8010C and Shimadzu 2014 gas chromatographs for inlet and outlet gas composition analyses. All the system waveguide components are based on a WR 975 rectangular waveguide, of internal dimensions 247.65 × 123.825 mm.

In the waveguide-based MPS (Fig. 2), the microwave power was delivered to the field-shaping structure in the form of a reduced height section preceded and followed by tapered sections. The reduced height section ensured higher electric field intensity in the plasma forming region, while the tapered sections gave a smooth transition from the standard WR 975 waveguide dimensions to the reduced height waveguide. The dielectric (quartz) tube, of inner diameter 26 mm, outer diameter 30 mm and length 1000 mm, was positioned centrally in the reduced height section and vertically in relation to the waveguide wide walls. Outside the waveguide the quartz tube was surrounded by an outer metal cylinder. The carbon dioxide (Air Liquide Polska), of a purity greater than or equal to 99.8 (% vol.), in the quartz tube was supplied by four inlets, ensuring the formation of a swirl gas flow through the tube. Such a method of introducing gas into the quartz tube stabilizes the plasma and protected the tube wall against overheating. Kerosene (lamp oil) of a chemical composition given by the supplier (Dorex, Poland, Table 1) was injected axially into the quartz tube. To initiate plasma generation inside the quartz tube, an igniter in the form of a movable metal pin was used. After plasma ignition, the igniter was withdrawn from the plasma region.

The experimental study was performed with a CO_2 flow rate of 2700 NL/h, and a kerosene flow rate between 0.4 kg/h and 1.2 kg/h. The kerosene vapour temperature before entering the microwave plasma was set at 400 °C, with the absorbed microwave power being varied between 4 kW and 6 W. In our experimental investigations the total outlet gas flow rate was determined from the mass balance of conversion products. As it was earlier checked results obtained in such a way are in a good correlation with the total gas flow rate of the outlet gas measured using the Particle Image Velocimetry (PIV) technique. The application of PIV technique for the total outlet gas flow rate determination was reported by us in [28]. To determine the volume concentration of the gaseous plasma components in the outlet gas each outlet gas sample was analyzed three times. The error in determining the volume concentration of the gaseous plasma components in the outlet gas did not exceed 5%. To confirm reproducibility of the achieved experimental results selected experimental measurements were performed few times for statistics. Results obtained that way confirm good repeatability of the experimental results. The study was performed without the aid of any catalyst.

3. Experimental results

Composition of kerosene used in our experiments is presented in Table 1. We assumed that kerosene may be represented solely by n-dodecane, thus all chromatographic measurements and further calculations as well as numerical modelling used n-dodecane as kerosene. Initial composition of gas before plasma processing was: CO_2 95.5% and kerosene (represented by n-dodecane) 4.5%.

The relationship between the volume concentrations of the gaseous plasma components in the outlet gas and the absorbed microwave power is shown in Figs. 3 and 4. For the results in Fig. 3 the measurements were carried out at a CO_2 flow rate of 2700 NL/h, kerosene flow rate of 0.4 kg/h and absorbed microwave power between 4 and 5 kW. In the case of Fig. 4 the measurements were carried out at a CO_2 flow rate of 2700 NL/h, kerosene flow rate of 0.8 kg/h and absorbed microwave power between 4 and 6 kW. The following gaseous by-products were registered in the outlet gas volume: hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), acetylene (C_2H_2), and ethylene (C_2H_4). It was observed that the products CO_2, CO and H_2 had the highest volume concentrations, while those of