Microwave methane plasma pyrolysis and catalysis for the generation of turquoise hydrogen and solid carbon

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Abstract: We report on the performance of a microwave plasma pyrolysis setup for creating turquoise hydrogen and solid carbon. Characterization of product composition analysis, temperature measurements, and solid carbon analysis for varying operating conditions, notably forward and reverse vortex confinements of the core plasma, shows that conversion increases significantly with operation using a reverse vortex flow.

1. Introduction

Hydrogen is proposed as a carbon-free energy carrier aligned with the new hydrogen economy [1]. Steam methane reforming is currently the main pathway for producing hydrogen, but the accompanying CO_2 production is undesirable. Electrolysis, a CO_2 free method, uses H_2O as source of hydrogen, but is to date significantly more expensive. Methane pyrolysis separates the hydrogen from the carbon in methane by heat. Microwave plasma pyrolysis, where the gas is directly heated by an electric energy source, using green electricity, is an obvious candidate for carbon-free H_2 production.

The produced (solid) carbon can improve the profitability of the methane pyrolysis process, becoming more valuable with an increased degree of order in its structure. Optimizing the selectivity for hydrogen and solid carbon in the affluent is a goal of this work, to this end downstream catalysts are utilized to further dehydrogenate hydrocarbons downstream.

2. Methods

In this work, we used a custom-built plasma reactor equipped with a 3kW magnetron. A Raman laser gas analyzer and gas chromatograph are used to determine the composition along the gas stream. Imaging (fig 1), optical emission spectroscopy and pyrometry are used to analyze the plasma region inside the waveguide. Abel inversion of the C_2 species spectra in the plasma region and black body radiation of the solid carbon particles surrounding this region result in spatially resolved temperature profiles. Plasma core temperatures of up to 6000K and 500K/mm gradients are measured. Temperature profiles, gas mixing and residence times, which are in the order of 10ms, determine the reaction rates of methane pyrolysis. Solid carbon particles are filtered out of the gas stream by a diesel particulate filter before the hot gas interacts with the catalyst to further de-hydrogenate the hydrocarbons and increase the hydrogen atom selectivity, which lies around 75-80% irrespective of the varied conditions and without the use of a catalysts. The solid carbon quality is analyzed by surface Raman spectroscopy and TEM imaging.

3. Results and Discussion

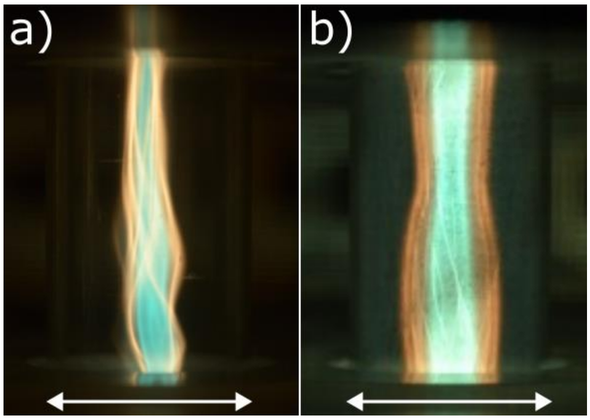

Fig. 1. Color CCD images from inside the waveguide of a) reverse and b) forward vortex confined methane:argon (1:10) plasmas operating at 2.5 kW and 30 slm. The arrows indicate the 30 mm OD of the quartz tube.

As part of the flow optimization, forward (FV) and reverse vortex (RV) configurations for confining the plasma to the center of the quartz tube inside the waveguide are analyzed. The results show a large increase in conversion for the RV case for similar operating conditions. In addition, the impact of mixing choke diameter (8-20mm), inlet swirl orifice diameter (0.5-1mm), total flow rate (10-50slm, combined with the orifice diameters this gives tangential swirl inlet velocities between 20-200m/s), microwave power (0.5-3kW) and gas inlet composition (1-80% CH_4) is also assessed. Axially inserting a tungsten rod for the RV enables operation in a similar low gas flow region as the FV, without it the weak vortex causes filament instability and jitter, resulting in plasma extinction. The thermodynamic performance (conversion, hydrogen yield/production efficiency, fuel upgrading/de-carbonizing efficiency) of the reactor for various configurations and operating conditions will be reported.

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References
[1] Diab et al. Int. Journal of Hydrogen Energy, 47, 61, (2022) 25831-25848