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## Development of a robust and efficient biogas processor for hydrogen production. Part 2: Experimental campaign

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### ABSTRACT

In this study, a robust and efficient decentralized fuel processor based on the direct autothermal reforming (ATR) of biogas with a nominal production rate of 50 Nm<sup>3</sup>/h of hydrogen and a plant efficiency of about 65% was developed and tested. The ATR unit is composed of a structured catalyst support for the biogas reforming close coupled to a catalytic wall-flow filter to retain eventual soot particles.

The performance of the conventional random foam and homogeneous lattice supports structures for the production of hydrogen from the ATR reaction was investigated. 15–0.05 wt%-Ni-Rh/MgAl<sub>2</sub>O<sub>4</sub>-SiSiC structured catalyst and LiFeO<sub>2</sub>-SiC monolith were selected for the conversion of biogas to hydrogen and for the syngas post-treatment process, respectively. For all the experiments, a model synthetic biogas was used and the catalytic activities were evaluated in three different experimental facilities: lab bench, pilot test rig and demonstration plant. High methane conversions (>95%) and hydrogen yields (>1.8) reached in the lab bench were also achieved in the pilot and demonstration plant operating at different GHSV.

Results of duration test using a foam coupled to the filter has demonstrated that the pre-commercial processor is reliable while offering a satisfactory reproducibility and

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negligible pressure drop. A thermodynamic equilibrium and a cold gas efficiency of 90% were reached for an inlet temperature of 500 °C, O/C: 1.1 and S/C: 2.0, as predicted with the Aspen simulation.

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## Introduction

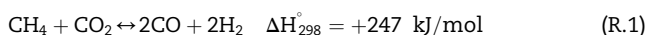
Hydrogen is one of the most promising fuel for energy and transport applications, providing solutions to environmental and economic challenges [1–3]. Energy carriers are used to move, store and deliver energy in a form that can be easily used. Hydrogen is a clean energy carrier similar to electricity [4,5]. Furthermore, hydrogen is a flexible and scalable energy vector which can be used for short- and longer-term energy storage. Moreover, hydrogen has high energy capacity, with the largest amount of energy per mass unit than any other known substance (121,000 kJ/kg) [6]. Green hydrogen can be used in fuel cells to produce power and heat cleanly and efficiently in a wide range of transportation and power generation in stationary and portable applications [6].

Nowadays, the most cost-effective way to produce hydrogen is from the steam reforming (SR) of fossil resources, such as natural gas and coal [6–10]. Environmental pollution, global warming, and the limited supply are the main disadvantages of fossil fuels [11,12]. Then, due to environmental concerns, hydrogen production from renewable sources such as solar, wind, biomass and biogas are needed [3].

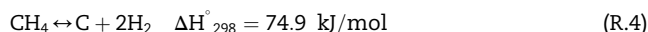
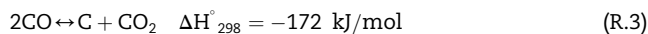
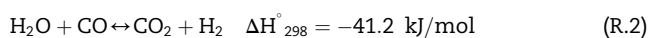
Hydrogen production from biogas is a highly promising energy carrier in the sustainable energetic scenario of reduced greenhouse gas emissions [6,8,10]. Biogas is produced from the anaerobic digestion of organic residues from several origins, typically, it is essentially composed of methane (CH<sub>4</sub>, around 55–70%) and carbon dioxide (CO<sub>2</sub>, around 30–45%), but also contains trace gases of ammonia (NH<sub>3</sub>), hydrogen (H<sub>2</sub>), nitrogen (N<sub>2</sub>), vapor water (H<sub>2</sub>O), hydrogen sulfide (H<sub>2</sub>S) and oxygen (O<sub>2</sub>) [6,13]. Biogas can be directly used as a combustible; however, the combustion process of biogas to generate heat has a low efficiency, because the humidity and CO<sub>2</sub> content decrease the calorific value of the fuel [14–18]. A promising possibility is to produce H<sub>2</sub> by reforming of biogas, which can be then supplied to fuel cell as proposed in the present work. In a catalytic reforming process, the presence of CO<sub>2</sub> and humidity in the biogas are advantageous for converting biogas into H<sub>2</sub> via dry and steam reforming reactions.

Hydrogen can be produced from biogas reforming through four different technologies, dry reforming (DR), steam reforming (SR), catalytic partial oxidation (CPOX), and auto thermal reforming (ATR).

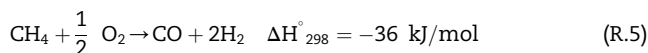
DR of methane is an endothermic reaction (R.1) and should be conducted at high temperatures (800–900 °C) to produce syngas (CO and H<sub>2</sub>) yielding a H<sub>2</sub>/CO ratio close to one. This value is considered to be the most appropriate for Fischer–Tropsch and other synthesis reactions for the production of liquid hydrocarbons [19].



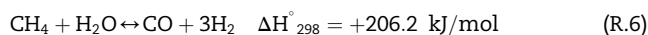
According to the literature there are parallel reactions such as the reverse water-gas shift reaction (R.2 reverse) and carbon forming reactions, Boudouard reaction (decomposition of CO) (R.3) and decomposition of CH<sub>4</sub> (R.4), which modify the equilibrium conversion of CO<sub>2</sub> and CH<sub>4</sub>. So, catalysts are useful in order to decrease the activation energy of the DR reaction and increases the selectivity to H<sub>2</sub> and CO while decreases the selectivity to carbon formation.



In the CPOX process, methane is partially oxidized over a catalyst bed to produce syngas with H<sub>2</sub> to CO ratio close 2, at atmospheric pressure. The reaction (R.5) is moderately exothermic and requiring temperatures between 700 and 900 °C to ensure complete conversion and to reduce soot formation [6]. As agreement in literature, the highly exothermic combustion of methane also take a place at the beginning of the catalyst bed, which results in high reaction temperature increase, which in turn can lead coke formation [13,20].



SR of methane as aforementioned is currently the most widespread and at the same time least expensive process for hydrogen production. SR is also an endothermic reaction (R.6), where the methane reacts with water vapor to produce H<sub>2</sub> and CO in a range of temperature between 650 °C and 850 °C.



H<sub>2</sub>/CO ratio produced in SR is equal to three and commonly steam is introduced in excess in order to shift the water-gas shift equilibrium (R.2) which enables towards the production of an additional amount of H<sub>2</sub> and less CO production at the reactor outlet [6,7].

A syngas production method in large-scale with higher energy efficiency than steam reforming technology is required. Since the endothermic heat due to reforming reactions is supplied from the reactor outside, the reaction rate is limited by the heat transfer [21]. ATR process has recently been the preferred technology for large scale Fischer–Tropsch-plants [21,22].

In the ATR process, the endothermic (SR) and exothermic (CPOX) reactions are coupled. The peculiar feature of ATR lies in the fact that heat is directly provided within the reactor, through partial oxidation of the biogas. This reduces the need of heat exchangers, and increases the flexibility of the plant [23].

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