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Syngas production by non-catalytic reforming of biogas with steam addition under filtration combustion mode

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ABSTRACT

Biogas conversion to syngas (mainly H₂ and CO) is considered an upgrade method that yields a fuel with a higher energy density. Studies on syngas production were conducted on an inert porous media reactor under a filtration combustion mode of biogas with steam addition, as a non-catalytic method for biogas valorization. The reactor was operated under a constant filtration velocity of 34.4 cm/s, equivalence ratio of 2.0, and biogas concentration of 60 vol% Natural Gas/40 vol% CO₂, while the steam to carbon ratio (S/C) was varied between 0.0 and 2.0. Total volumetric flow remained constant at 7 L/min. Combustion wave temperature and propagation rate, product gas composition, reactants conversion as well as H₂ and CO selectivity were measured as a function of S/C ratio. Chromatographic parameters, method validation and measurement uncertainty were developed and optimized. It was observed that S/C ratio of 2.0 gave optimal results under studied conditions for biogas conversion, leading to maximum concentrations of 10.34 vol% H₂, 9.98 vol% CO and highest thermal efficiency of 64.2% associated with a modified EROI of 46.3%, which considered energy consumption for steam supply. Conclusions indicated that the increment of the steam co-fed with the reactants favored the non-catalytic conversion of biogas and thus resulted in an effective fuel upgrading.

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Introduction

Synthesis gas (SG) or syngas is defined as a gas mixture that contains hydrogen (H₂) and carbon monoxide (CO) as main combustible components. Raw SG mostly inevitably contains also considerable amounts of carbon dioxide (CO₂), water

(H₂O) and smaller amounts of other compounds such as ammonia (NH₃), hydrogen sulfide (H₂S), hydrogen chloride (HCl) and tar [1]. SG is an important building block in the chemical, oil and energy industries due to its applications: (a) as a feedstock for the production of several chemicals such as hydrogen, ammonia, methanol, and Fischer-Tropsch

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products [2]; (b) as a fuel in a gas turbine to produce electricity [3]; (c) as a cell fuel for mobile sources [4]; (d) as an electricity supplier through solid oxide fuel cells [5]; (e) as carrier for energy recovery from waste [6,7]; and (f) as a primary fuel.

SG is mainly used as a chemical building block rather than a fuel, representing nowadays a US\$ 50 billion market for an annual production of 40 Mt [8]. Recent reports show that global energy consumption grew, on average, 1.8% per annum in the 2005–2015 period and only 1.0% on 2016, showing a below average growth rate. However, renewable energy grew a 14.4% on 2016, clearly stating a world movement towards sustainable ways to power our society [9]. In that context hydrogen, as an important component of SG, could help to satisfy the world energy demand for cleaner fuels. H_2 is considered the fuel of the future. It contains more energy per unit mass than any other fuel and also it generates minimum emissions after being burned.

Fundamentally, there are three different methods for SG generation: (i) thermochemical processes like steam reforming (STR), auto-thermal reforming (ATR), thermal partial oxidation (TPOX) and biomass gasification; (ii) electrochemical techniques such as high temperature co-electrolysis of H_2O and CO_2 [10–12]; and (iii) chemical conversion of hydrogen sulfide and ammonia [13].

In the other hand, biomass are considered as any material of biological origin, excluding fossil fuels and peat, that contains a chemical store available for conversion to a wide range of energy carriers [14]. It is one of the most abundant and versatile energy sources in the world, and biogas, as a result of biochemical conversion of wet biomass, has the capacity to transform organic waste into valuable products that are converted into both economic income and social and environmental benefits. Biogas is produced on an industrial scale by anaerobic digestion, this is achieved through bacterial fermentation of biomass or as a result of organic source processing. The gas produced by this fermentation process is mainly composed of methane (CH_4 : 45–70%) and carbon dioxide (CO_2 : 30–45%). Because of a higher concentration of methane in its composition, biogas is considered an important source of renewable methane when obtained from waste management processes. Biogas, as a carbon-neutral alternative fuel, has a heating value directly related to its methane content, thus higher CO_2 concentration decreases its energy potential resulting in a less competitive option against fossil fuels. Depending on the nature of the primary feedstock and its formation conditions, biogas also contains traces of other gases, such as hydrogen sulfide (H_2S), ammonia (NH_3), hydrogen (H_2), nitrogen (N_2), oxygen (O_2), and water vapor (H_2O) [15]. Final composition is fully dependent on the composition of the organic material being digested, the specific bio-organism inside the reactor and ambient conditions. As this is a biological process, there are several variables that affect the decomposition path, e.g. temperature regime, retention time, nutrient content (C/N ratio), pH, humidity, as well as others [16].

Biogas can be utilized in various applications, most of them related to production of heat, mechanical power and electricity at an industrial level, as well as power generation, cooking, heating and cooling in distributed renewable energy

applications. Another interesting approach is the thermal upgrading of the available biogas through a TPOX process in order to obtain SG, being the super-adiabatic regime achieved in inert porous media (IPM) reactors an option worth exploring [17]. Combustion in IPM, also known as filtration combustion, is the process of propagation of a chemical exothermic reaction zone in a chemically inert solid under filtration of a gaseous mixture of a fuel and an oxidizer [18–20]. Super-adiabatic partial oxidation techniques allows the attainment of operating temperatures significantly in excess of the adiabatic combustion temperature of the incoming gases [21]. Under this operational conditions, thermochemical conversion of fuel into syngas can be achieved with no need of catalyst activity. Combustion of biogas in IPM combine two mechanisms for syngas production: partial oxidation and dry reforming of methane. Also, it has been proven that the presence of steam as a gasifier agent can enhance methane conversion levels and hydrogen yields by means of steam reforming [22–24]. Products such as H_2 , CO , C_2H_2 , etc., are generated when oxygen concentration is insufficient, leading to a rich and ultra-rich mixture regime [21].

In this work, a non-catalytic method for SG production was developed using filtration combustion of biogas with steam addition. To monitor efficiently the SG production and composition, analytical procedures with the required quality levels for measurements were developed.

Materials and methods

Experimental apparatus

Experiments on filtration combustion of synthetic biogas mixed with air and steam were conducted using the setup schematically shown in Fig. 1. The apparatus consisted of a combustion tube filled with a porous medium, composed of 6 mm solid alumina spheres yielding a porosity of 40%; fuel, air and steam supply system; temperature acquisition system, and gas emission analyzer. The reactor had an internal diameter of 39 mm, wall thickness of 2 mm, and length of 290 mm and was made of quartz. To prevent heat losses and achieve quasi-uniform temperature profiles, the inner surface of the reactor was covered with a 3 mm layer of high-temperature insulation (Luyangwool), and an additional 24 mm thick layer was implemented on the external diameter of the reactor.

Fuel supply was taken directly from local distribution of natural gas with an average molar percent composition of 97% CH_4 /3% C_2H_6 , carbon dioxide was provided from a cylinder with 99.9 mol% concentration (LindeGas), while air at 0.5 barg was provided by a compressor (Mohrfohl, model L-05D) under room conditions of 19 ± 1 °C and atmospheric pressure.

Flow rates for both the fuel and air supply were set using two thermal mass flow controllers (Aalborg, GFC17 – GFC37). Steam supply was given by an in-house built small-scale boiler equipped with two electric resistors of 1.5 kWe each, both connected to a variable autotransformer. A calibration curve enabled quantitative control over the steam supply rate within a range of 1 L/min to 58 L/min at atmospheric pressure.

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