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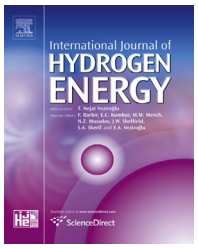

Optimization of hydrogen production by filtration combustion of natural gas by water addition

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ABSTRACT

The effect of adding steam during filtration combustion of natural gas–air mixtures was studied with the aim to evaluate the optimization of hydrogen production. Temperature, velocity, chemical products of combustion waves, and conversion from fuel to H_2 and CO were evaluated in the range of equivalence ratio (φ) from stoichiometric ($\varphi = 1.0$) to $\varphi = 3.0$ and steam content in the mixture from 0% to 39%, at filtration velocities from 12 to 25 cm/s. Numerical simulation was carried out using GRI-MECH 3.0. Results suggest that H_2 and CO concentrations, dominant for rich and ultrarich combustion, are products from partial oxidation and steam natural gas reforming processes. Experimental and numerical results show that hydrogen yield increase with an increase of steam content in the natural gas–air mixtures.

Introduction

The current power crisis is the consequence of the increase in the worldwide consumption of energy [1–3]; therefore, it is indispensable to explore the use of alternative sources of clean, renewable, efficient and sustainable energy. Interest for the production of hydrogen (H_2) has incremented in last years, as it is considered a clean fuel, which generates a minimum of emissions once it has been burned, having more energy per unit of mass than any other fuel. Large scale production of H_2 is carried out by reforming Natural Gas (NG) with steam, which accounts for 70% of production worldwide. However, reforming is an endothermic process that requires high temperatures for the conversion of methane (CH_4), demanding much energy [4,5]. As a consequence, it requires innovation for simpler and efficient processes to produce H_2.

During the last two decades, technology related to partial oxidation in inert porous media (IPM) has been developed, that allows partial oxidation of fuel rich mixtures of fuel–air, generating reduced gases, such as H_2 and carbon monoxide (CO), mixture also known as synthesis gas (syngas). Specifically, partial oxidation in IPM consists of a process of propagation in the zone where the exothermic chemical reaction of a gas occurs along a solid media that is chemically inert [5]. During this process, there is participation of IPM as well as the reactant gas, in which three characteristic zones can be identified within IPM: (a) the first zone, immediately ahead from the combustion front, where all gases mix naturally and they are pre-heated as a result of IPM's release of heat; (b) a

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second, very narrow, zone in which the chemical reaction occur, moving forward or backward at the point of mixture fuel–air, where enthalpy is released in large quantities that is absorbed by IPM. A regeneration of heat is produced in the system, with an excess of heat in the chemical reaction zone and slow increase in temperature at the front, which could surpass adiabatic temperature; (c) a third zone, located beyond the front, having the combustion products, which actively interchange heat with IPM. These systems are characterized by the presence of two dynamic fronts; heat generation and high temperatures. Resulting from the selection of the appropriate geometrical parameters, dynamic and thermal fluids, both fronts can be superimposed, intensifying one another [4–9]. Various works in literature have reported that partial oxidation could reach super-adiabatic temperatures in rich and ultra-rich mixtures fuel–air [10–17].

Currently, there are mathematical models available in the literature that describe the combustion in IPM with available reaction mechanisms, e.g., for CH_4 exist models that describe combustion mechanisms, such as GRI 3.0 [18], Leeds mechanism [19], from the National Institute for Science and Technology (NIST) [20], and Konnov mechanism [21]. All of which have been verified using stoichiometric conditions or lean fuel mixtures [22].

Toledo et al. [9] reported a theoretical (using GRI 3.0 mechanism) and experimental investigation on the partial oxidation of CH_4, ethane (C_2H_6), and propane (C_3H_8), in the range of equivalence ratio (φ) of 1.0 and 2.5. Results indicated a correlation between experimental and simulated data; furthermore, increasing the equivalence ratio increases H_2 and syngas production.

Conversion of CH_4 to H_2 during a process of partial oxidation of the mixtures CH_4–air in IPM requires optimization. This conversion is dependent upon mixture of the gases used, specifically, the relation fuel-oxidant. Bingue et al. [23] reported an increment in the content of H_2 in the combustion products by using a constant equivalence ratio and increasing the amount of oxidant (oxygen). Dobrego et al. [12] numerically optimized the process of conversion from CH_4 to H_2 by adding steam during partial oxidation of CH_4 in IPM, using mixtures CH_4–air and a reciprocating flow burner. Its results propose the design of new porous media reactors that consider combustion process with steam addition.

The objective of this research is to increase the production of H_2 in IPM reactors by investigating the theoretical and experimental partial oxidation of NG–air mixtures with steam addition. We use the numerical model of Toledo et al. [9] under PREMIX program. Temperature profiles, velocity of flame propagation, and analysis of combustion products (H_2, CO, CH_4 and CO_2) are presented and discussed.

Reactor design

Partial oxidation of NG–air mixtures (NG with 96% of CH_4) was carried out in a lab-scale reactor (Fig. 1). Experimental equipment consisted of: IPM reactor, injection system for fuel, air and steam; system for temperature measurements; and equipment to analyze concentration of gases produced by combustion. Reactor is a quartz tube 35 cm length by 40 mm OD by 2 mm wall thickness, packed with alumina (Al_2O_3) spheres 5.5 mm in diameter and having 40% porosity. Fiberfrax insulation was provided to the outside of the unit to minimize heat loss and to obtain stable temperature profiles.

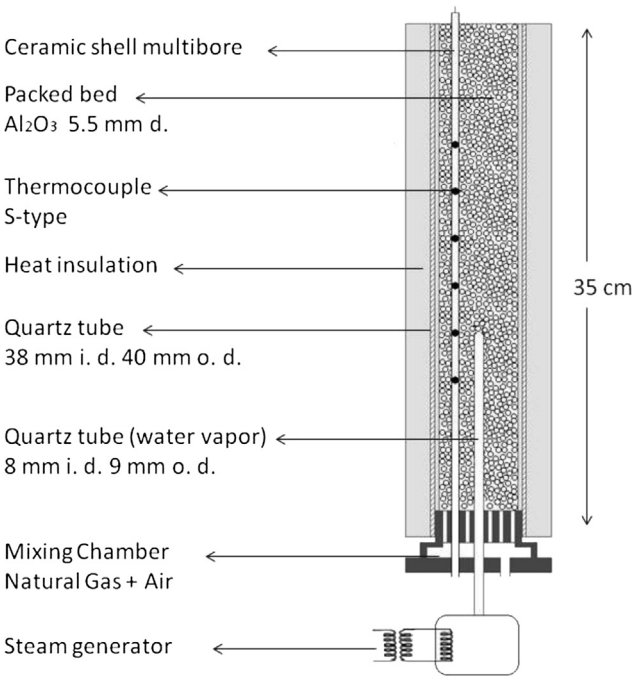

Fig. 1 – Schematic of the experimental setup.

Ceramic rods (0.5 cm diameter) were used as thermocouple shields and placed along the length of the reactors; these rods have 6 side-ports (0.08 cm diameter) and S-type thermocouples (platinum–rhodium, Omega) were inserted longitudinally inside the ceramic rod. Voltage measurements from thermocouple were converted to temperature readings by using module DAQ 54 and DaqView software, also from Omega, and captured in a PC. Each thermocouple (T1 to T6, from top to reactor base) was 4 cm apart, leaving 8 cm of distance between T6 and the base of the reactor. Experimental error associated with temperature measures and flame velocities were estimated as 50 K and 10% respectively.

Air is fed by using an industrial air compressor (Qualitas) and regulated with a flow control unit (Aalborg); NG was fed and regulated with a flow controller from Aalborg. Both streams were mixed before entering the reactor. The main component for the generation of steam was to incorporate an electrical resistance (132 Ω) that was assembled inside the unit and attached to a variable voltage controller. A K-type thermocouple and a pressure gauge were installed for monitoring purposes. Steam flow was previously calibrated by mass balance, using a condenser, a stopwatch, and a graduated cylinder.

During the experiments, the upstream propagating combustion wave of NG–air mixtures was initiated at the reactor exit. The upstream propagation was recorded. As the wave reached the quartz tube of water vapor (Fig. 1), the steam flow was injected. Then, the downstream propagation was recorded. As the wave reached the reactor exit, the flame was