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Chapter

Non-Catalytic Reforming of Biogas in Porous Media Combustion

Abstract

Mario Toledo Torres

Rich combustion of biogas inside an inert porous media reactor was investigated to evaluate hydrogen and syngas production. Temperature, velocities, and product gas composition of the combustion waves were analysed, while varying its filtration velocity, for a range of equivalence ratios (ϕ) from $\phi = 1.0$ to $\phi = 3.5$. A numerical model based on comprehensive heat transfer and chemical mechanisms was found to be in a good qualitative agreement with experimental data. Partial oxidation products of biogas (H₂ and CO) were dominant on rich combustion. Different gas mixtures of methane and carbon dioxide, which simulated synthetic biogas, and the addition of a varying fraction of water steam were experimentally analysed. It was observed that an increasing steam to carbon ratio (S/C) improved hydrogen and syngas production. The non-catalytic process investigated results in an effective biogas upgrading, and to be essentially higher than under natural gas filtration combustion.

Keywords: biogas reforming, porous media burner, hydrogen, syngas, non-catalytic

1. Introduction

Large-scale production of hydrogen (H_2) is mainly obtained through the thermochemical conversion of methane (CH_4) into H_2 and carbon monoxide (CO), a mixture also known as syngas. The main conversion processes are dry reforming, partial oxidation, steam reforming, and autothermal reforming, all of which typically use fossil fuels as main carbonaceous feedstock, the natural gas being the most widely used. However, current efforts have been focused on the development of sustainable, carbon-neutral alternatives for H_2 production. Hence, biogas upgrading by partial oxidation of CH_4 in the presence of oxygen (O_2) , steam (H_2O) , or carbon dioxide (CO_2) is considered an interesting alternative to produce syngas while reducing GHG emissions to the atmosphere.

Accordingly, research has been focused on improving the syngas production from biogas process efficiency, being the use of fluidized bed reactors a promising real alternative to effectively increase efficiency. However, they have presented a major drawback related to catalytic wearing mainly associated to the elevated temperatures (over 1000 K and up to 1300 K) required for the efficient and cost-effective conversion of biogas into syngas [1–4].

Moreover, the widely reported effects of sintering and coking on the catalyst both responsible for the generation of depositions that cause catalytic deactivation [5–6], observed to occur on high-temperature regimes (over 1100 K), have prevented the commercial development of the technology. Nevertheless, further studies related to upgrading the thermal properties of catalysts used in biogas reforming, and their resistance to the aforementioned phenomena, have been performed, particularly as a countermeasure to the effects of GHG [7–9].

However, since the catalytic approach has proven to be thermally challenging, an alternative non-catalytic method, such as partial oxidation (POX) in inert porous media (IPM), has proven to be an interesting option for high-temperature biogas conversion. The advantage of using a porous matrix to enhance several reaction processes, such as combustion, partial oxidation, steam reforming, and dry reforming, among others, has been extensively studied by numerous researchers, where the use of a chemically inert porous media enables the propagation of an autothermal reacting wave which benefits from the increased heat transfer on the reaction zone due to the solid matrix. Specifically, this enhanced thermal mechanism, mainly attributed to the heat conduction and radiation, and the highly developed inner surface of the porous media being heated by the reaction wave, acts as a heat recirculation mechanism which distributes the thermal energy up- and downstream of the reacting zone, thus preheating the fresh mixture and homogenising the reaction temperature across the reacting wave. Furthermore, the existence of multiple flow paths for the filtered gas increases its diffusion and heat transfer with the solid phase. Finally, filtration combustion has shown to increase the operational ranges of free-flame combustion on a wide range of filtration velocities, equivalence ratios, and power loads.

When combusting gaseous fuels in porous media, steady and transient systems are the two approaches commonly employed [10–16]. The first approach is widely used in radiant burners and surface combustor-heaters, where the combustion wave maintains its position due to an equilibrium of the heat transfer mechanisms. While the transient operation considers a reaction wave travelling on an upstream or downstream fashion through the porous media, the direction of wave propagation depends mostly on the physical properties of solid and gas, filtration velocity, temperature, and air excess of the mixture. Combining these parameters, it can be noted that the movement velocities of these waves are much lower than free-flame combustion velocities [13].

A transient operation mode is characterised by concentrating or diluting the heat released from the chemical reaction while travelling in a downstream or upstream direction, respectively; thus the reaction front can reach a temperature considerably different from the free-flame adiabatic flame temperature. This phenomenon is mainly attributed to the reaction chemistry and the heat transfer mechanisms. Under a counterflow configuration, the downstream propagation results in superadiabatic reaction waves which effectively increase the conventional free-flame flammability limits for both ultra-lean and ultrarich operation modes.

Superadiabatic filtration combustion of rich and ultrarich mixtures allows the stable operation of both partial oxidation and thermal cracking of hydrocarbons. This technology for hydrogen or syngas production uses an IPM [17–20]. The fuels used in porous combustion systems are basically of gaseous form due to fluidity, volumetric capacity, and shorter mixing length scale [21–28]. Accordingly, this chapter presents the numerical and experimental results obtained for different biogas compositions (CH_4 and CO_2) on syngas production by filtration combustion in an IPM reactor.

2. Numerical model

Experimental temperature measurements showed a minimal radial gradient; thus a one-dimensional numerical simulation was considered adequate for representing the reaction wave inside the porous media. A volume-averaged model [29] was used to solve the two-temperature mathematical model proposed to describe the filtration combustion under isobaric, stationary, and one-dimensional conditions. The combustion wave propagation rate was considered to be at least three orders of magnitude smaller than the filtration velocity of the gaseous mixture. The two-temperature approximation was formulated in order to describe a fully developed stationary reaction wave in a coordinate system moving with the reaction zone [19, 25, 30]. Therefore, this model describes both solid and gaseous phases through their fluid dynamics and heat transfer interactions.

Continuity equation.

$$\frac{\partial(\dot{M})}{\partial x} = \frac{\partial(\epsilon \rho v)}{\partial x} = 0 \tag{1}$$

Gas phase energy equation.

$$c_{p}\rho_{g}v\frac{\partial T_{g}}{\partial x} = \frac{\partial}{\partial x}\left(\left[k_{g} + (c_{p}\rho_{g})D_{ax}\right]\frac{\partial T_{g}}{\partial x}\right) - \sum_{k}\left(\dot{\omega}_{k}h_{k}W_{k} - \rho_{k}Y_{k}V_{k}c_{pk}\frac{\partial T_{g}}{\partial x}\right) - \frac{h_{v}}{\epsilon}(T_{g} - T_{s}) \quad (2)$$

Solid phase energy equation.

$$-(1-\varepsilon)c_{s}\rho_{s}u\frac{\partial T_{s}}{\partial x} = \frac{\partial}{\partial x}\left([k_{e}+k_{R}]\frac{\partial T_{g}}{\partial x}\right) - \beta(T_{s}-T_{0}) - h_{v}(T_{g}-T_{s})$$
(3)

Species conservation equation.

$$\rho_{g}v\frac{dY_{k}}{dx} + \frac{d}{dx}(\rho_{g}Y_{k}V_{k}) = \dot{\omega}_{k}W_{k}$$

$$(4)$$

Boundary conditions chosen for the inlet and outlet of the reactor consider thermal equilibrium between gas and solid phases.

Inlet:	x = 0 ,	$T_0 = T_g = T_s$,	$Y = Y_{k,0}$
Outlet:	$\frac{\partial Y_k}{\partial x}=0 \ , \label{eq:eq:expansion}$	$T_{\rm s} = T_{\rm g} = T ,$	$\frac{\partial T}{\partial x} = \beta (T - T_0) / (\epsilon c_p \rho_g v - (1 - \epsilon) c_s \rho_s u)$

An analytical solution of Eqs. (1)–(4) was used to impose the boundary conditions of the reaction wave temperature at the outlet. This is achieved by assuming the aforementioned conditions for the outlet and adding that $\dot{\omega}_k = 0$ $h_v = (6\epsilon/d^2)Nuk_g$ and that radiation can be neglected at the outlet since it is far from the reaction zone. Thus, calculation for temperature and species in the reaction wave can be computed for a finite and a well-defined spatial domain. The convective heat transfer coefficient (h_v) was taken from [19] as $h_v = (6\epsilon/d^2)Nuk_g$, whereas the correlation for Nu was considered as $Nu = 2 + 1.1Re^{0.6}Pr_g^{1/3}$ as presented by Wakao and Kaguei [31]. A radiant conductivity model taken from [29] was used to include the effect of radiation, where $k_R = 4F\sigma T_s^3$ with F being the radiation exchange factor. This has to be modelled for each material since it is dependent upon its thermal conductivity and emissivity. For the solid phase, composed of 5.6 mm in diameter alumina spheres, values from 0.3 to 0.6 are used. Effective thermal conductivity of the packed bed and its porosity were estimated as $k_e = 0.005 k_s$ and $\varepsilon = 0.4$. The tortuosity of the porous media and its porosity contributes to flow irregularities which affect the effective mass diffusion of species in the gas phase; this phenomenon is described by an axial gas dispersion coefficient, $D_{ax} = 0.5dv$ [31]. Dispersion coefficients for both thermal and mass diffusivities are considered to be equal according to a heat/mass transfer analogy. As presented by Henneke and Ellzey [30], a sum of molecular diffusion and dispersion is used to represent the effective diffusion.

Finally, all thermophysical properties from the solid phases, such as thermal conductivity, heat capacity, and radiative properties, were obtained from openly available technical reports [32] and verified against the technical specifications from the ceramic manufacturer (Coors, Inc.).

Chemical kinetics of the process was modelled through the implementation of the GRI 3.0 [33] chemical kinetics mechanism, which includes NO_X chemistry, alongside the CHEMKIN [34] package. Even though this mechanism is not designed specifically to simulate reactions of ultra-lean nor ultrarich mixtures, it is considered as an acceptable first approach to understand the possible reaction mechanisms occurring under these conditions.

The calculations were performed for a given value of the filtration (interstitial) velocity, and the implemented numerical algorithm in the modified PREMIX [35] code was used to find the wave propagation velocity.

3. Syngas production by filtration combustion

In this section, numerical and experimental results are showed for wave velocities, combustion temperature, and H_2 and CO concentrations for rich combustion of biogas in porous media. Additionally, results on rich combustion of natural gas in porous media are presented.

The combustion system is shown in **Figure 1** and consists on a cylindrical quartz tube. This tube is filled with a bed of alumina spheres (Al_2O_3) with an average diameter of 5.6 mm, yielding a porosity of \sim 40%. The inner and outer surfaces of the tube were covered with a 2 and 25 mm thick Kaowool insulation material, respectively. System diagnostics were required to assess the temperature profile in the reactor along with emission concentrations in the product gases. The axial temperature distribution of the reactor was acquired using S-type thermocouples. These thermocouples were housed in a long multibore ceramic shell; therefore measured temperatures were considered to be very close to the temperatures of the solid phase. Temperature measurements were digitised using a data acquisition module and transferred to a PC. The reaction wave propagation rates were obtained from thermocouple measurements over time and the known distance of the thermocouples. The concentrations of H_2 , CO, CH_4 , and CO_2 in the product gases were measured using a gas chromatograph fitted with a thermal conductivity detector (TCD), while gas samples were acquired through an alumina tube immersed at the end of the reactor. To avoid the effect of external air vortices on the composition, the probe was inserted 20 mm into the packed bed.



Figure 1. *Schematic of the experimental setup.*

The methane, carbon dioxide, air and steam mixture flowed continuously through the quartz tube, and the reactant concentrations were controlled using Aalborg mass flow controllers. A temperature experimental measurement error was estimated at 50 K, which is considered mainly as a radial error. The reaction wave velocity measurement error was estimated at 10%, and the chemical sampling accuracy was considered close to 10%. The experimental uncertainties were defined based on the accuracy of the laboratory equipment and the repeatability of the experimental data.

3.1 Combustion wave temperature and propagation rate

One objective of the numerical simulation was to clarify the effect of interfacial heat transfer. Calculations performed with the same initial conditions in two-temperature approximations are presented in **Figure 2** for natural gas-air and biogas-air mixtures. Thermal non-equilibrium between gas and solid phases needs to be applied for consideration of propagation waves. Downstream, upstream, and standing waves were observed for natural gas-air and biogas-air mixtures, mainly depending on the equivalence ratio. Stable combustion of rich and ultrarich mixtures was observed experimentally for the region of equivalence ratios studied.

For natural air mixtures (**Figure 2A**), upstream wave propagation was observed for the range of equivalence from stoichiometric to 1.7. The velocity of the wave decreases with an increase of the natural gas concentration, approaching zero at 1.7. A stationary combustion wave is formed under these conditions. With further increase of the natural gas content, the regime of propagation changes towards a downstream direction. This regime is observed for the range of equivalence ratios from 1.7 to 3.5, where the speed increases with the natural gas content. It was found experimentally that the maximum combustion temperature remains almost constant (Ts = 1529 K) throughout the rich region and is practically independent of the natural gas content. These combustion temperatures are attributed to the changes of



(A)



(B)

Figure 2.

Combustion temperatures and wave velocities for natural gas-air mixtures (A) and biogas (60% CH₄/40% CO_2)-air mixtures (B) varying the equivalence ratio from stoichiometry ($\varphi = 1.0$) to $\varphi = 3.5$.

combustion chemistry, because all other governing parameters such as flow rates, porous body properties, and heat content are similar.

For biogas-air mixtures, the combustion in porous media shows similar behaviour (**Figure 2B**). Upstream wave propagation is observed for the range of equivalence from stoichiometric to 1.5. A standing combustion wave was formed at 1.5. The regime of downstream propagation was observed for the range of equivalence ratios from 1.5 to 3.5. It was found experimentally that the maximum combustion temperature remains almost constant (Ts = 1491 K) throughout the rich region and was practically independent of the biogas content.

3.2 Combustion products

Starting from equivalence ratios higher than 1.0, complete combustion of natural gas and biogas will not be achieved because of the low oxygen content in the mixture. Consequently, concentrations of CO_2 and H_2O decrease, while partial oxidation products such as H_2 and CO increase their presence in the product gases (**Figure 3**).

Hydrogen and carbon monoxide concentration on the reaction waves of natural gas and biogas showed a direct relation to an increase of equivalence ratio. Unburned methane was detected in the gaseous products starting from $\phi \approx 1.3$. Its concentration grew with an increasing equivalence ratio. Natural gas and biogas thermochemical processing could be characterised as fuel reformation or cracking rather than combustion.

In comparison, product concentration had a similar behaviour for natural gas and biogas. H_2 and CH_4 concentrations are highest for natural gas and CO_2 and CO for biogas, in the range of equivalence ratios studied, which is consistent with the previous results, under similar conditions, as reported by [10, 16, 20].

3.3 Biogas composition

Biogas is obtained from the anaerobic digestion of wet biomass which is a relevant component of most urban residual wastes, as well as industrial food and



Figure 3. *Composition of chemical products as a function of equivalence ratio for rich and ultrarich mixtures.*

agricultural waste. It is known as a gaseous admixture mainly composed of methane (40-65% v/v) and carbon dioxide (35-55% v/v) with traces of hydrogen sulphide (0.1-3.0% v/v), moisture, and other trace contaminants.

This section shows the experimental results of combustion temperatures and syngas production from filtration combustion of synthetic biogas-air mixtures, using different compositions of CH₄ and CO₂ for equivalence ratio of $\varphi = 1.5$ and $\varphi = 2.0$. The experimental temperatures reached presented slight differences (<53 K) between φ = 1.5 and φ = 2.0, for the tested biogas-air mixtures (Figure 4A). The maximum combustion temperatures experimentally found were 1564 and 1563 K for 55:45 using $\varphi = 1.5$ and $\varphi = 2.0$, respectively. A temperature decrease of 31 K (ϕ = 1.5) and 8 K (ϕ = 2.0) was observed with an increment of the CO_2 content in the biogas mixtures from 100:0 to 40:60. These peaks in the temperature profile could be associated with the partial oxidation of the CH₄ component of the biogas (exothermic reaction), while the decline in the temperature profile corresponds to the dry reforming of biogas (endothermic reaction). Also, the different behaviours of temperature profile are attributed to the changes of combustion chemistry, CO₂ presence, and filtration velocity, since all other parameters such as heat content (CH₄ flow rates), porosity of inert media, geometry, and dimensions of the reactor were kept constant. The effect of decreasing the filtration velocity while reducing the CO₂ content in the synthetic biogas could be responsible of the similarity between the peak temperatures recorded for all tested conditions, since it is well known that an increasing filtration velocity is responsible of enhancing the diffusion inside the reactor due to larger turbulence inside the pores of the solid matrix, whereas the peak temperature recorded while operating with a biogas composition of 55:45 using $\varphi = 1.5$ and $\varphi = 2.0$ could be attributed to a more intense role of the exothermic reactions in comparison to the decreasing filtration velocity.

Figure 4B illustrates the hydrogen and carbon monoxide yields for the biogasair mixtures in the inert bed. The maximum hydrogen yields recorded were 17.68 and 15.30% for $\varphi = 1.5$ and $\varphi = 2.0$, respectively, using 100:0 (natural gas-air mixtures). On the other hand, the maximum peak of hydrogen yields using biogasair mixtures (at 55:45) were 23.34 and 20.40% to $\varphi = 1.5$ and $\varphi = 2.0$, respectively, before gradually declining with the biogas mixture of 40:60.

Similar results have been previously reported by Zeng et al. [36] while operating an inert porous media reactor in a stationary regime with a $50:50 \text{ CH}_4/\text{CO}_2$ ratio and a filtration velocity of 25.6 cm/s.

3.4 Steam addition

Variations on the steam to carbon (S/C) ratio from 0.0 to 2.0 were performed under constant values of filtration velocity (34.4 cm/s), an equivalence ratio of φ = 2.0, and biogas composition (60:40 CH₄/CO₂).

Figure 5A depicts peak operational temperatures, reported as the mean value of maximum temperatures recorded inside the reactor, for several S/C ratios. A decreasing temperature in the reaction zone due to an increasing S/C fraction, which went from 1543 K at baseline conditions to 1501 K at an S/C ratio of 2.0, could be associated to an increased contribution of endothermic reactions in the thermochemical conversion of the mixture. However, since biogas is mainly composed by CH₄ and CO₂, the filtration combustion mode studied could be considered as a trireforming process where thermal partial oxidation (TPOX), dry reforming (DRR), and steam reforming (STR) simultaneously interact with CH₄. Thus, this process can be considered as a non-catalytic alternative for biogas valorisation. Regarding the reaction wave propagation rates, **Figure 5B** shows the values computed for each experimental run, which for all experiments propagated in a downstream direction with no relevant variations.



Figure 4.

Combustion temperatures (A) and hydrogen and carbon monoxide yield (B) for equivalence ratio of $\varphi = 1.5$ and $\varphi = 2.0$, varying biogas composition and filtration velocities.



Figure 5.

Combustion temperatures (A), wave propagation rate (B), thermal efficiency, and EROI (C) varying steam to carbon (S/C) ratio. Energy evaluation parameters.

Figure 5C presents the computed values for thermal efficiencies and a simplified energy return on investment (EROI) of the process as a function of the S/C ratio. Overall, both values were positively affected by an increasing presence of steam in the reactants, reaching their maximum values at an S/C fraction of 2.0. In particular, the peak thermal efficiency was accounted as 64.2% which represents an upgrade of 69% when compared to baseline conditions, while the EROI, where both the initial heating value of the biogas and the energy required to supply the steam were considered, reached a maximum of 46.3%, which corresponded to an effective increase of 22% relative to baseline conditions. Therefore, an increasing steam

presence in the mixture is considered to favour the effective thermochemical conversion of biogas by means of a non-catalytic filtration combustion reactor.

4. Conclusion

In this chapter numerical and experimental results were presented for filtration combustion of rich biogas-air mixtures, in comparison with methane-air mixtures. Predictions of a numerical model, based on the two-temperature approximation and multistep gas phase combustion mechanism (GRI 3.0), are in good qualitative agreement with experimental data, including combustion temperatures and wave velocities.

Applications for the reforming of biogas fuel with different compositions of methane and carbon dioxide into hydrogen and syngas were presented. Also, some improvement as steam addition to biogas-air mixtures allows higher efficiency for hydrogen and syngas production.

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