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Syngas Fuel Production from Carbonaceous Feedstocks Using Hybrid Porous Media

Mario Toledo Torres and Nicolás Ripoll Kameid

Abstract

During the last years, hybrid porous media reactors have been developed aiming to partially oxidize solid and gaseous fuels to produce reducing gases. The gases produced are mainly composed of hydrogen (H_2) and carbon monoxide, among other products of gasification. This hybrid process combines inert porous media (IPM) combustion and gasification of solid fuels by replacing a fraction of the inert solid volume with a solid fuel. The gaseous mixture is produced from carbon-rich reactants exposed to the high temperatures of filtration combustion. Experimental results from different solid fuels (coal, biomass, and others) and gaseous fuels (natural gas (NG), propane, and others) are presented, with detailed analysis of high temperatures (between 900 and 1800 K), velocities, and product gas composition of the combustion waves, which is able to produce $[H_2]/[CO]$ ratios from 0.2 to 10.

Keywords: hydrogen production, solid and gaseous fuels, porous media, hybrid filtration combustion

1. Introduction

Hydrogen (H_2) and syngas production technology development has been concentrating most of current efforts toward more efficient and responsible use of fossil carbonaceous feedstocks [1, 2]. Moreover, these technologies can utilize energy more efficiently, supply ultraclean fuels, eliminate pollutant emissions at end-use systems, and significantly reduce greenhouse gases emissions, particularly carbon dioxide (CO_2) [2]. H_2 and syngas are currently mostly produced by steam reforming, partial oxidation, and autothermic reforming which is also known as oxidative steam reforming [2]. For example, H_2 is mainly produced by steam methane reforming (SMR), a process that inherently releases huge amounts of greenhouse gases. The primary energy sources to produce hydrogen are hydrocarbon feedstocks (methane, oil, and coal) with 96% of the supply, while the rest (4%) is attained through water electrolysis [3]. However, in past years, it has been challenging to properly forecast the availability of hydrocarbon feedstocks, which in turn adds to its uncertainty as a main feedstock in the H_2 production chain. Therefore, the development of novel techniques aimed to diversify H_2 and syngas production presents itself as highly necessary, where the gasification of biomass, for example, poses as a promising effort to significantly compete against fossil feedstocks [4, 5], with a carbon-neutral alternative.

Several applications, processes, and configurations have been developed to thermochemically transform solid fuels into a gaseous fuel through a process called gasification. This process consists on the transformation of solid substances that contain carbon such as biomass, coal, or waste into a combustible gaseous product in the presence of air, water steam ($\text{H}_2\text{O}_{(g)}$), oxygen (O_2), CO_2 , or a mixture of these gasifying agents. The conversion of these substances occurs at high temperatures ($\sim 800^\circ\text{C}$) and moderated pressures (from atmospheric pressure of up to 70 barg) [6–8]. The conventional gasifiers are classified based on the type of bed and direction of the gas flow [9]; the description of their functioning principles and main features can be extensively found in technical literature [10]. In particular, biomass gasification differs from coal gasification, mainly because biomass is a carbon-neutral and sustainable energy source and because biomass is more reactive and features a higher volatile content than coal, which results in a lower gasification temperature. This reduces heat loss, undesired emissions, and material problems associated with high temperatures. Biomass also has a low sulfur content, which results in less SO_x emissions, but due to its high alkali contents, like sodium and potassium, slagging and fouling are common problems in biomass gasification equipment [11]. There are several studies regarding solid fuel gasification, such as the results reported by [12–14]. On the other hand, disadvantages of catalytic gasification include increased material costs for the catalyst (often rare metals), as well as diminishing catalyst performance over time. The relative difficulty in reclaiming and recycling the catalyst can also be a disadvantage [8].

In general terms, gasification as a process still requires further optimizations to enhance its energy efficiency by overcoming the main aforementioned challenges, such as tar production and moisture content of the biomass. Although new technologies have been developed as effective ways to utilize even toxic and wet biomass for power generation [15] and conventional techniques have been proven to provide a feasible option to reform solid fuels, there are still limitations on the characteristics of the fuel that restrict the use to certain feedstocks. Fixed bed gasifiers may work with solid fuels containing up to 50% of humidity, while fluidized bed gasifiers can work with solid fuels with up to 60% of humidity in the most advanced developments [16]. The products obtained in the different configurations of gasifier devices are mainly composed of H_2 , CO , CO_2 , H_2O , N_2 , heavier hydrocarbons ($\text{C}_2\text{--C}_6$), ashes, tar, oils, and small solid carbon particles, among others. Finally, the main disadvantages of conventional autothermic gasification technology are related to the production of undesired species such as particulate matter, tar, and char. The emissions of these species are highly associated to the operational parameters of the process such as temperature, pressure, time and heating speed, solid fuel particle size, and residence time [17]. For these reasons, researchers have studied the technology of inert porous media (IPM) combustion detecting many important advantages, such as low pollutant emissions, high thermal stability, increased reaction temperature due to its internal heat recirculation, and extended flammability limits, among others [18–21].

The main objective of this chapter is to present the use of IPM technology for achieving high-temperature gasification of solid fuel in a hybrid porous media reactor.

2. Hybrid filtration combustion for solid fuels

IPM is a thermochemical process proven to be a feasible option to address current global requirements for cleaner energy sources and processes [22]. This technology is known to be able to produce H_2 from several feedstocks and allows the direct use of liquid and gaseous fuels that interact with an inert solid matrix.

The common approaches to use the technology are the stationary and transient configurations.

A relation between IPM combustion and solid fuel gasification converges into hybrid filtration combustion (HFC), a process that combines the properties of the aforesaid processes by replacing a fraction of the inert solid's volume with a solid fuel. In this case, a reaction wave is produced by a flow that can contain hot air, $\text{H}_2\text{O}(\text{g})$, or a gaseous fuel-air mixture that propagates along the reactor reforming the solid fuel inside within a wide-power-range, high-efficiency, high energy concentration per unit of volume and stable combustion over a wide range of equivalence ratios [23]. Several experimental studies on HFC for syngas and H_2 production have been conducted [24–35], showing that the technology presents a strong and feasible option for syngas production from gaseous and solid fuels in a batch configuration.

In [24] three types of algae were analyzed, showing that an increase of volume algae fraction in the hybrid bed and an increase of moisture content in the algae used increased both combustion temperature and hydrogen yields. Different gasifying agents were used on experiments with biomass pellets and alumina spheres using equal volumetric fractions [25]. While operating with natural gas (NG), the combustion wave temperature increased only using insignis pine, whereas the usage of cereal plantation residuals enhanced the syngas production. Using steam, the combustion wave temperature presented a slight decrease as the steam presence increased. In the case of natural gas in a porous medium composed of coal and alumina particles [26], the flame temperature decreased with an increase of coal fraction, and hydrogen and carbon monoxide were dominant partial oxidation products. Further experimental studies [29, 31] consistently reported that hydrogen and carbon monoxide are dominant partial oxidation products for atmospheric hybrid combustion waves.

Industrial applications of HFC have been successfully implemented in Northern-European countries, such as Finland and Russia, where two reactors capable of processing up to 15,000 ton/yr of municipal solid wastes (MSW) were engineered by the IPCP-RAS (Russia) and developed by Europrofile Ltd. in Lappeenranta (Finland) and Moscow (Russia) [36, 37].

3. Experimental results by hybrid filtration combustion

Figure 1 shows the experimental setup generally used in hybrid porous media reactors. The filtration combustion system consists of a tube, usually made of quartz, filled with uniformly mixed aleatory ceramic spheres and solid fuel particles. To compensate for the different thermal expansion rates of the packed bed and tube, the inside diameter of the tube is covered with an insulation blanket (ceramic fiber). Heat losses due to conduction through the tube wall are minimized with an additional insulation layer covering the outside of the tube. Air, fuel, and/or steam, metered using mass flow controllers, are premixed before entering the reactor and introduced into the reactor from its bottom. The upstream or downstream propagating combustion wave is ignited using a lighter at the reactor exit or reactor bottom. System diagnostics are required to assess the temperature profile in the reactor and the chemical composition of the output gases. The axial temperature distribution of the reactor is measured by thermocouples. These thermocouples are housed in a multi-bored ceramic shell. A data acquisition system is used to read and record the temperatures. The digital conversion of the resultant analog signals is performed with a data acquisition board. Finally, the chemical composition of the flue gases is measured using a gas chromatograph.

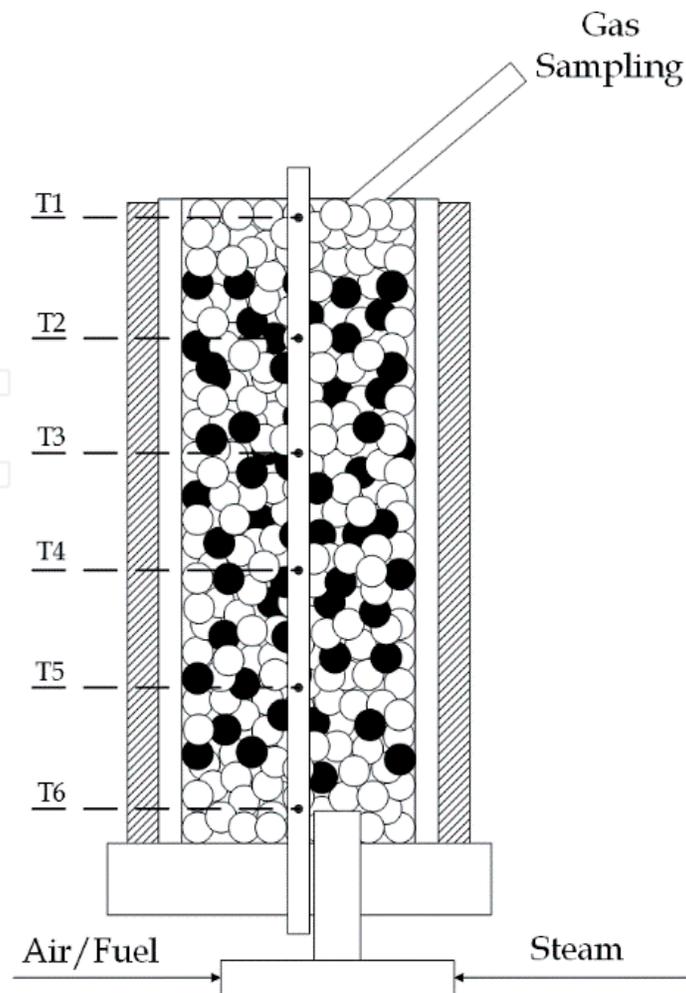


Figure 1.
Typical experimental setup.

In **Figure 2**, the experimental results for measured combustion temperatures and propagation rates, and hydrogen and carbon monoxide concentrations, in the form of the ratio $[H_2]/[CO]$, are displayed for a range of equivalence ratio (ϕ) of $0.3 < \phi < 2.6$. This particular graph displays profiles belonging to three different gaseous and solid fuel mixtures in hybrid process as natural gas and wood, propane and polyethylene, and butane and wood. All experiments were conducted on a hybrid bed composed of 50% of solid fuel and 50% inert alumina spheres.

In **Figure 2A**, the combustion temperatures range is from 1,000 to 1,300 K for lean mixtures ($\phi < 1.0$), from 1,100 to 1,200 K in rich mixtures ($1.0 < \phi < 1.65$), and from 1,200 to 1,500 K in ultrarich mixtures ($\phi > 1.3$). The high stable combustion temperatures evidenced in the range of $1,000 < \phi < 1,500$ show that hybrid process feasibility is almost independent of the gaseous and solid fuels for the equivalence ratio experimented. The high overall combustion temperature represents that the reactors provide suitable conditions for the combustion chemistry to convert simultaneously gaseous and solid fuels into synthesis gas. This figure reveals some interesting characteristics of transient filtration combustion waves depicting the regions of wave propagating counter or concurrent to the unburned gas. At $\phi < 0.4$ upstream superadiabatic wave is observed. If more gaseous fuel is added in the mixtures, stable underadiabatic waves are developed. Underadiabatic waves are established from approximately $\phi = 0.4$ to $\phi = 2.4$. Further addition of gaseous fuels develops upstream superadiabatic waves at $\phi > 2.4$.

In **Figure 2B**, the products of combustion comparatively show $[H_2]/[CO]$ ratio. It was observed that lean mixtures with higher oxygen content in the oxidizer stream

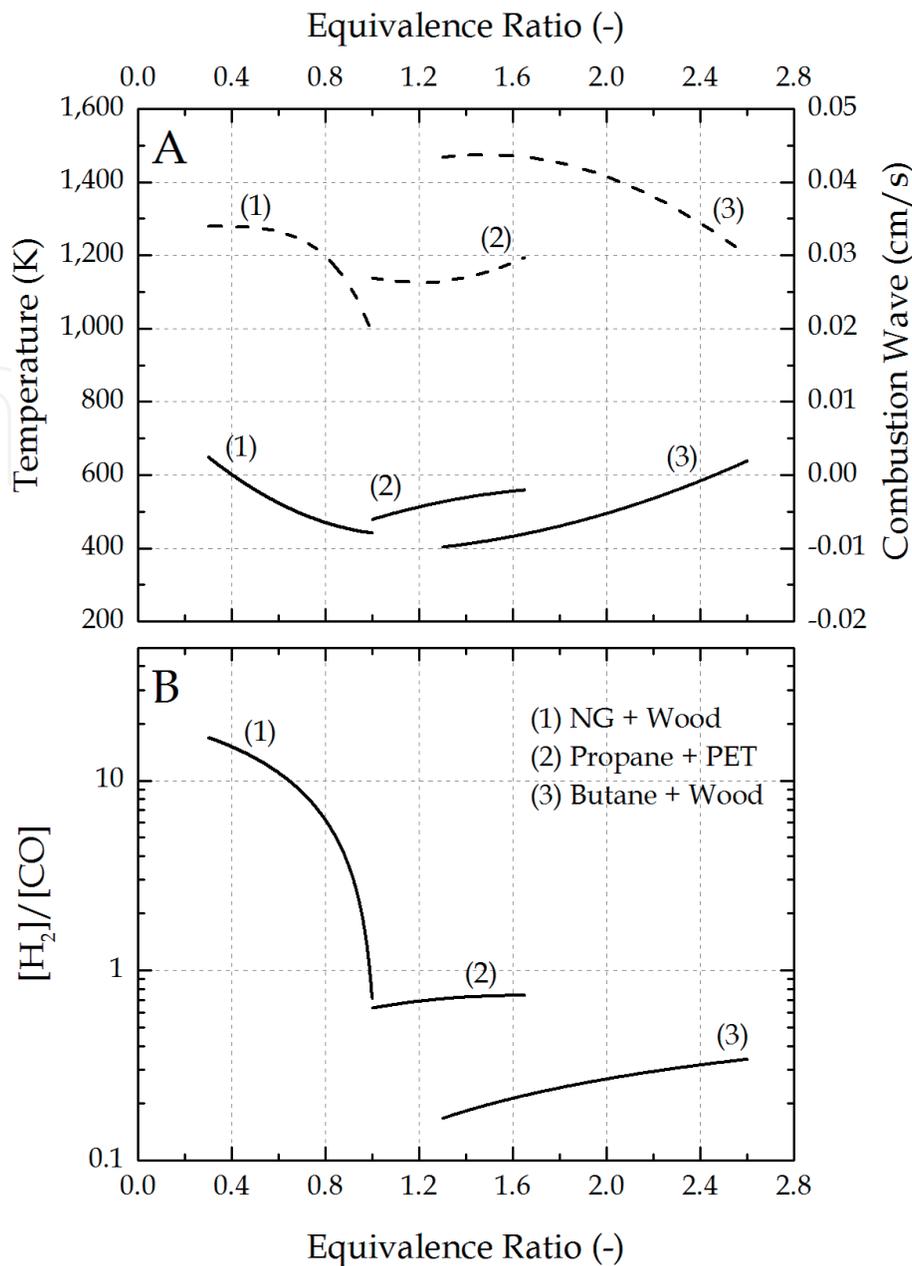


Figure 2. Effect of varying the equivalence ratio on (A) maximum recorded temperatures inside the reactor (dashed lines) and combustion wave propagation rates (solid lines) and (B) $[H_2]/[CO]$ ratios (solid lines), for three different experimental sets: (1) ultralean natural gas and air mixtures with wood pellets [30], (2) rich propane-air mixtures with polyethylene [31], and (3) rich and ultrarich butane-air mixtures with wood pellets [29].

generate higher amounts of $[H_2]/[CO]$ ratios. The overall region of high ratio is between $0.3 < \phi < 1.0$. In rich and ultrarich mixtures, the $[H_2]/[CO]$ ratios are less than 1. Of utmost importance in this chapter is the adequate ratio in the combustion products, since it will determine the usefulness of the obtained syngas in the desired applied process. From lean to rich regimes, the products of combustion are controlled by temperature and residence time. The process can be characterized as gaseous and solid fuel reforming rather than combustion such that a stable thermal process exists with a concentration of oxygen. This type of hybrid system is possible by the unconstrained movement of the combustion zone to recuperate its energy from the porous matrix.

An important operational parameter of the hybrid filtration combustion reactor is the mass fraction of solid fuels in the inert porous matrix. **Figure 3** shows experimental results for temperatures, combustion waves, and $[H_2]/[CO]$ ratios. It was found that the combustion temperature remains higher (900–1,700 K) in all the range

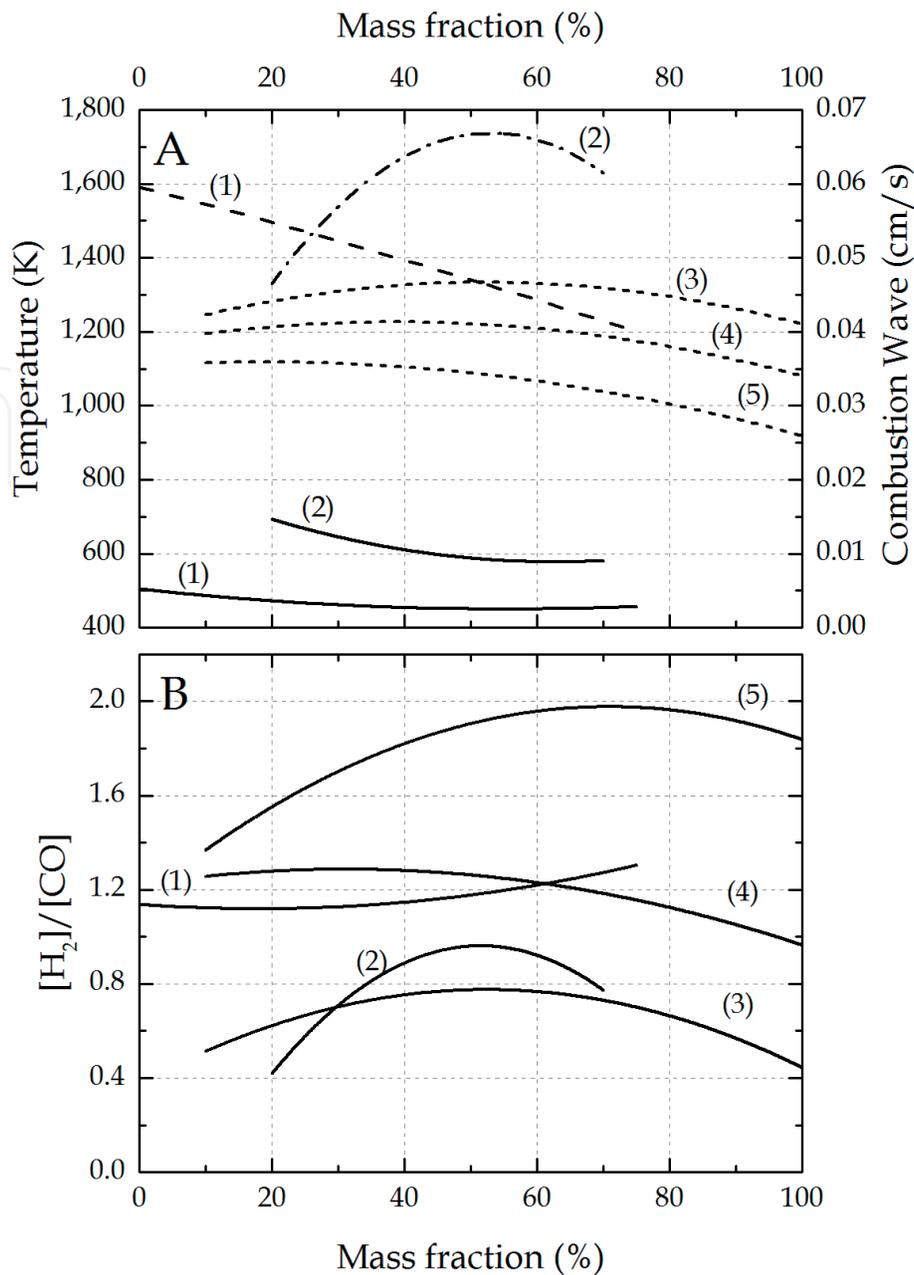


Figure 3. Effect of varying the mass fraction of different carbonaceous fuels in the porous matrix on the (A) peak recorded temperatures (dashed, dotted, and dash-dotted lines) and combustion wave propagation rate (solid lines) and (B) $[H_2]/[CO]$ ratios for three different experimental sets: (1) ultrarich natural gas and air mixture with bituminous coal [26], (2) steam-air mixtures with bituminous coal [23], and (3) charcoal with different fractions of $[H_2O]/[O_2]$ [38]: (1) 1.0; (2) 2.5; and (3) 4.5.

of mass fraction presented. In high mass fraction ($> 60\%$), temperature decreases because heat is necessary to convert solid fuels into syngas. The combustion waves for (1) and (2) were found to be totally superadiabatic over the whole range of mass fraction tested. With different gaseous and solid fuels, the $[H_2]/[CO]$ ratios formed are between 0.4 and 2.0, in all the range of mass fraction reported (**Figure 3B**).

4. Conclusion

In this chapter, experimental results were presented for hybrid filtration combustion of different gaseous and solid fuels. Results are focused in combustion temperatures, waves, and $[H_2]/[CO]$ ratios with varying equivalence ratio and mass fraction of solid fuels in the inert porous matrix.

Depending on operational parameters, the range of high combustion temperature is between 900 and 1,800 K. Considering that the $[H_2]/[CO]$ ratios are from 0.2 to 10, the applications for the hybrid reactor will depend on the use of this ratio in the next applied process.

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