Hydrocarbons conversion to syngas in inert porous media combustion

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ABSTRACT

Experimental combustion in inert porous media of Liquefied Petroleum Gas (LPG), Butane, Propane, Diesel Fuel and Heavy Fuel Oil (HFO) is presented to assess the syngas production within different stages of refinement of fuels, considering light gases up to a residual oil. The porous media is composed by a fixed bed of alumina spheres and temperatures and gas composition were acquired while stable combustion waves under rich fuel conditions were produced. The results of this study showed that it is possible to partially oxidize HFO producing up to a 10% of H_2 and CO in the products for the equivalence ratio of $\phi = 1.3$, with a superior performance than any other fuel tested in this work. The behavior of the recorded temperature results showed good agreement with the available literature. Upstream regimes of propagation of the combustion wave are observed for the experiments with liquid fuels, while gaseous fuels presented downstream regimes for $\phi > 2$ up to $\phi = 4$. Results suggest that in order to efficiently transform the fuels into syngas, it is convenient to operate the reactor under an upstream regime considering the chemical energy at inlet and outlet. Furthermore, liquid fuels, regardless of its refining level, have the potential to produce syngas in inert porous media in promising volumes.

Introduction

Efficient use of the hydrocarbon resources provided by the planet is a priority. An increase on the awareness on global warming has also increased the regulations over the use of certain energy resources in order to control the emission of pollutants that influence environment and health issues. Although, human activities massively demand the use of energy in several formats depending on the different requirements of the task. Therefore, refineries supply a variety of hydrocarbon fuels that come from petroleum oil that are suitable for almost every requirement. Nevertheless, these

refining processes leave a residual, which is not desired since it carries unwanted elements that are harmful when used. Furthermore, its use is related mostly to situations where the pollutant emission standards allow its use, such as the combustion in ships or in places where expensive mitigation measures can be taken, which implies a low cost of the fuel. On the other hand, refineries require hydrogen as a part of their key processes such as hydrodesulphuration that is a widely used process to reduce sulfur presence in commercially available hydrocarbon fuels or, hydrocracking, where the heavy hydrocarbon compounds of petroleum are cracked into lighter and more valuable fuels in the presence of hydrogen.

Several processes are available for hydrogen production and one of the most promising is the partial oxidation in inert porous media. Currently, a considerable amount of information on combustion in inert porous media burners is available, having several reports regarding the working basis and design principles of the technology, describing construction materials and porous structures used, characterization of emissions and their control, several modern applications of the combustion technology and innovative approaches have been presented [1–9] mostly for common gas fuels.

This technology eliminates or significantly reduces effects caused by some of the disadvantages of the free flame combustion such as: high pollutant emissions, low energy density, small range of thermal power variation, problems with stability of the flame, strong dependence on the type of fuel and a limited range of variation of the equivalence ratio. Most of these problems are related to the flame and the low thermal conductivity of the gases involved. Therefore, the presence of inert solids that enhance the thermal conductivity at high temperatures is suitable, also considering that the filtration of the gases through the porous boosts mixing and diffusion [1,10].

On the other hand, the partial oxidation of liquid fuels in a porous media is a promising technique for the reforming of hydrocarbon fuels into syngas, especially to obtain hydrogen in fuel-rich conditions [11]. Syngas can be a source of hydrogen for fuel cells, enrichment of conventional fuels or it can be used to produce high quality fuels from low energy content or neutral carbon dioxide sources. Liquid fuels have a highly developed infrastructure, considering the extraction, refining and distribution and also, being easy to manipulate and store providing high energy densities. Pastore and Mastorakos [12] studied the rich n-heptane and diesel combustion in two-layer porous media in stable operation for syngas production. Considering equivalence ratio $\phi = 2$ for different thermal input values, the results for n-heptane showed that the reformer can produce syngas with concentrations of 12.6% and 15% of H_2 and CO respectively, about 80% of the corresponding values expected from thermodynamic equilibrium at the sample temperature. Diesel reforming tests were performed at a higher preheat temperature and with a different ceramic foam for the reforming stage. The reformate analysis showed a reforming efficiency up to 77.6% at P = 9 kW and $\phi = 2$, with H_2 and CO concentrations of 15.2% and 19.1%, respectively. Subsequently, they reported the rich combustion of n-heptane, diesel, kerosene and biodiesel EMR to produce syngas suitable for the cleaning and separation stages for fuel

cell application or the enrichment of conventional burners. Rich combustion waves were stabilized in a two-layer inert porous medium combustor and a range of ϕ and porous materials were reported. n-heptane was successfully reformed up to $\phi = 3$, reaching a conversion efficiency (based on the lower heating value of H_2 and CO over the fuel input) up to 75% for a packed bed of alumina beads. Also, diesel, kerosene and bio-diesel reformed to syngas in a zirconia foam burner showing conversion efficiencies over 60% [13].

An experimental and numerical study regarding the conversion of liquid heptane to syngas through combustion in porous media was presented by Dixon et al. [14], showing that at a constant inlet velocity, hydrogen production is enhanced by increasing ϕ, whereas hydrogen conversion efficiency reaches a peak value around $\phi = 3$. Tests at $\phi = 2.5$ showed that the conversion efficiency increases with the reactants' inlet velocity and values higher than 80% are obtained for the highest tested velocity of 80 cm/s. Similar trends for carbon monoxide conversion and energy efficiencies are observed, where peak values exceed 90% and 80%, respectively. Also, the results indicate favorable conditions for fuel reforming for $2.5 \leq \phi \leq 3.5$, having that the inlet velocity has a significant effect on the performance of non-catalytic fuel reforming. Substantial efficiency gains are observed for increased inlet velocities, which are attributed to cause an increase in the temperature of the reactor.

Non-catalytic conversion of jet fuel and butanol to syngas by transient filtration combustion over a variety of ϕ and inlet velocities were described by Smith et al. [15]. In particular, the results of the total energy conversion efficiency for jet fuel showed that it increases with ϕ up to a peak of 61% at $\phi = 3.15$. For butanol, the total energy conversion efficiency determined experimentally followed the equilibrium value until $\phi > 3$; for this condition, experimental conversion efficiency continue increasing up to $\phi = 3.5$, where values higher than 100% were found. For that reason, an unbalance in soot deposition and consumption at the time of exhaust gas sampling is suspected.

The thermal partial oxidation (TPOX) of diesel fuel for the production of syngas, considering different values of excess air ratio, was studied theoretically and experimentally by Al. Hamamre et al. [16]. The processing of fuel, previous to inlet to the porous media reformer, includes atomization, evaporation, and formation of a homogeneous fuel-air mixture. It is noted that the preparation of the fuel-air mixture is a major factor that influences the quality of the reforming process. The porous media was a ZrO_2 foam of 10 ppi. The results in the porous-based TPOX burner showed that a practical operating condition can be achieved at $\phi = 2.22–2.5$. The maximum H_2 and CO concentrations were achieved at $\phi = 2.22$, observing that for lower ϕ values the syngas concentrations decreased due to the formation of CO_2, and for ϕ values higher than 2.22, higher concentrations of CH_4 and C_2H_2 were detected, and consequently, the concentrations of H_2 and CO decreased. For 873.15 K air preheating temperature the H_2 and CO molar fractions were 16.0% and 18.5%, respectively [16].

Taking in consideration that several fuels with different characteristics are commercially available, mostly coming from the derivatives of oil distillation and refining and the limited volume of literature available on liquid fuels reforming

in porous media, this work assesses the conversion efficiency of gaseous and liquid hydrocarbon fuels to syngas using partial oxidation in inert porous media on different operational conditions for the following fuels: Liquefied Petroleum Gas (50% butane, 50% propane), butane, propane, diesel fuel and fuel oil No. 6. This assessment aims to provide information regarding the level of refining of the source of hydrogen and carbon monoxide produced by a contrast of the results of the gaseous lighter fractions of oil with the heavy residual hydrocarbon mixture remaining of the refining.

Experimental apparatus and procedure

Since the nature of liquid and gaseous fuels is particular, handling and usage of each kind is different. Therefore, different burners have to be considered for this work's assessment. For this reason, gaseous and liquid fuel experimental setups are presented in different sections; regardless of having similar experimental procedures.

Gaseous fuels

The experiments on filtration combustion with LPG, butane and propane were performed in the reactor shown in Fig. 1.
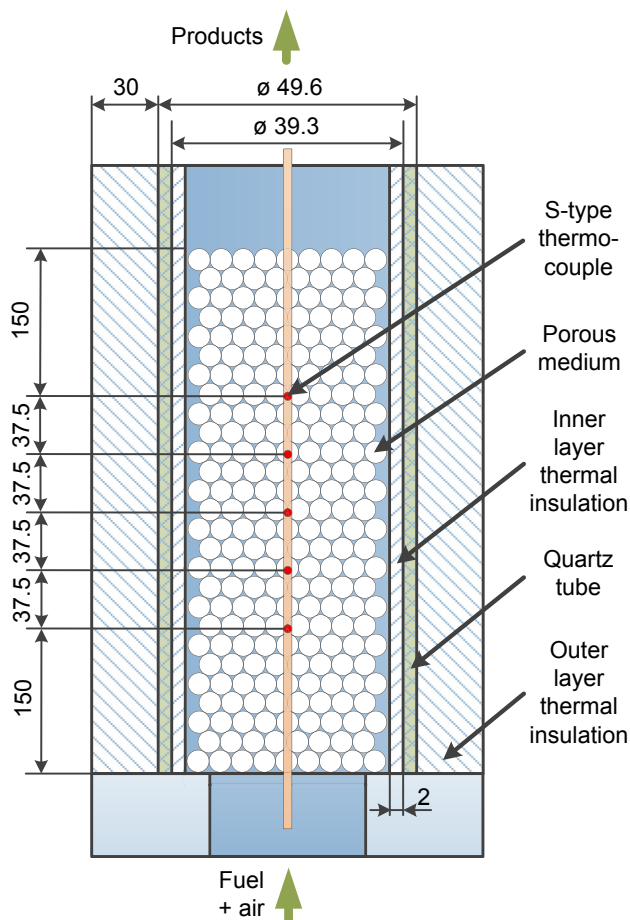

Fig. 1 – **Experimental setup for gaseous fuels partial oxidation (dimensions in mm).**

The apparatus consists on a quartz tube ($39.3 \times 49.6 \times 450$ mm, ID $\times$ OD $\times$ length) having a porous medium; fuel and air supply system, temperature measurement system, and gaseous products analyzer. The inner surface of the tube was covered with a 2 mm layer of Fiberfrax insulation. To prevent heat losses and achieve quasi-uniform temperature profiles, additional 30 mm thick high-temperature insulation was applied on the external diameter of the reactor. A packed bed of solid Al_2O_3 spheres (6.0 mm D) was used as a porous medium presenting a void porosity of 44%. The fuel mixtures of LPG, butane and propane with air were prepared by a continuous flow method where the fuel and air flows were controlled and supplied using a set of MKS mass flow controllers (Model 1179A, MKS Inc.). To ensure a uniform gas composition, the reactants were premixed in a mixing chamber and the gases were introduced in the reactor through a distribution grid at the bottom of the reactor. The outlet of the reactor was open to the atmosphere. During the experiments, an upstream propagating combustion wave was initiated at the reactor's exit using a lighter and temperatures were recorded. Running axially in the center of the quartz tube, a ceramic shell (5 mm OD, having six bores of 0.8 mm D) contains five S-type (platinum/rhodium) thermocouples, providing temperature information of the combustion wave, which is known to be a close approximation to the temperature of the alumina spheres; reactants' temperature is expected to be higher; temperature experimental acquirement error is estimated at 50 K. System diagnostics were required to assess the temperature profile in the reactor and the chemical composition of the output gases having an OMB DAQ-56 data acquisition system to read and record the temperatures. The thermocouple junctions were equally spaced along the length of the shell (Fig. 1). Chemical composition of the output gases was characterized using a Perkin Elmer (Clarus 500) Gas Chromatograph, with the same detection method and devices described in the work of Araya et al. [10]. The chromatographic samples were obtained when the combustion wave was passing through the central thermocouple. Samples of gaseous fuels were acquired by an extraction through a ceramic tube slightly inserted into the porous medium at the exit of the reactor. The error associated to the GC measurements is estimated through the relative standard deviation for each set of samples; using this value and considering a confidence level of 95%, the maximum relative error obtained was 10%. It must be also mentioned that the samples were water cooled.

All experiments with gaseous fuels were developed at a constant filtration velocity of $u_f = 0.31$ m/s.

Liquid fuels

Two reactors are considered for this section, one for each liquid fuel. The schematic of the HFO burner is shown in Fig. 2 and consists on a steel tube (6″ Sch 40, 600 mm L), partially filled with a fixed bed of alumina spheres (6.0 mm D). For diesel, the diameter and length of the steel tube were 65 mm and 300 mm (Fig. 3), respectively, having a medium of 6 mm alumina spheres. A LPG-air combustion wave was used prior to liquid fuels injection and it was burned in rich mixture conditions (a high temperature and quick propagation of the combustion wave was needed, which was obtained with
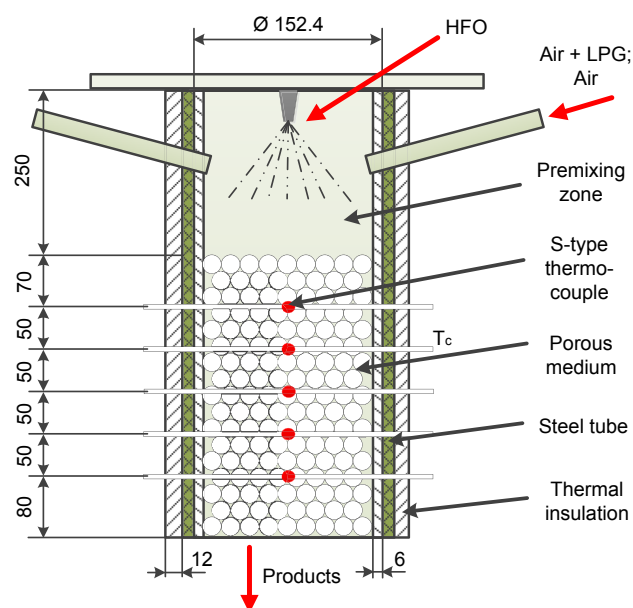
Fig. 2 – Experimental setup for heavy fuel oil partial oxidation (dimensions in mm).

$\phi = 1.2$ and a filtration velocity of $u_f = 0.27$ m/s). For the experiments made with diesel, the LPG combustion was performed until a maximum temperature in central thermocouple is observed, while for HFO, until the
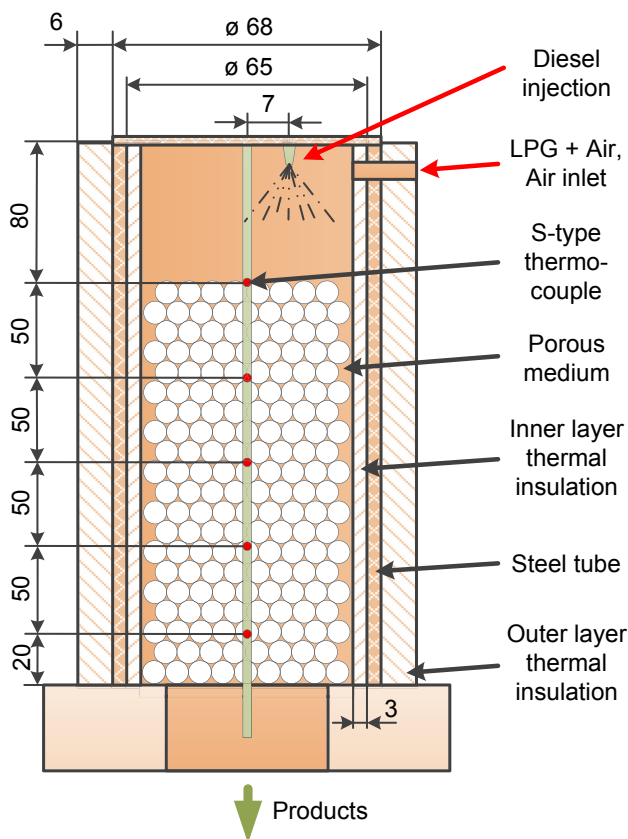

Fig. 3 – Experimental setup for diesel fuel partial oxidation (dimensions in mm).

thermocouple labeled T_c reached a maximum. Temperature data can be instantly observed during the experiments, noting that a maximum temperature showed in a thermocouple signal indicated that the combustion wave was passing through its position. In these cases, the temperature data instantly observed provided information about the position of the combustion wave. When the combustion wave reached the mentioned position, LPG supply was discontinued and liquid fuel was injected in order to acquire the data for the combustion process, i.e. temperature and product gases. The LPG supply was controlled by an Aalborg mass flow controller (GFC 17). HFO required heating before being pumped prior to its pulverization in a nozzle inlet due to its high viscosity. Diesel did not require heating and it was properly pumped and atomized at room temperature. Atomization of diesel was obtained with a nozzle with a 30° angle. HFO inlet system consisted in a tank provided with an electric resistance that heats the fuel (up to 130 °C) before pumping it to a 45° nozzle. Temperature data acquirement was made using five S-type thermocouples and an OMB DAQ 54 acquisition module; signals were processed by the Personal DaqViewPlus Software (OMEGA Engineering Inc. USA). The thermocouples were held inside a ceramic rod (5 mm D) with six bores (0.8 mm D each). The composition of the exhaust gases was determined with the same Gas Chromatograph and procedure as in gaseous fuels.

The experiments with diesel considered a constant u_f of 0.20 m/s, performing experiments at $\phi = 1.0$, 1.4 and 1.9, while the experiments with HFO were performed at filtration velocities that varied from 0.19 m/s ($\phi = 1.3$) to 0.33 m/s ($\phi = 1.8$), considering that the noozle and pumping system correctly atomize the fuel under certain volumetric flows, therefore, the change of ϕ is made through changes in the air flow (which implies a change of filtration velocity). It should be noted that tests were performed under different conditions and some unstable combustion wave was obtained for HFO. Additionally, the experiments with LPG were performed at $\phi = 1.0$, 1.5, 2.5, 3.0, 3.5 and 4.0. Butane's equivalence ratios considered were 1.5, 2.5, 3.0, 3.5 and 4.0 while propane's were: 1.0, 1.5, 2.5, 3.0, 3.5 and 4.0.

Results and discussion

Temperature

The results of the temperature of the combustion wave acquired are shown in Fig. 4. Since the experiments provide information only for the several equivalence ratios mentioned before, a polynomial fit is used to describe the tendencies in the studied range. Since the temperatures indicated by the thermocouples have a rapid response, the direction of the combustion wave propagation can be easily noted. For this reason, as a reference in Fig. 4, three lines indicate the ϕ that separates the different regimes of propagation (i.e. ϕ at which the propagation velocity of the combustion wave is zero) and presents a steady regime. It was found that butane's combustion wave did not propagated at $\phi = 1.82$, LPG at $\phi = 1.92$ and propane at $\phi = 1.77$, which is close to value reported by
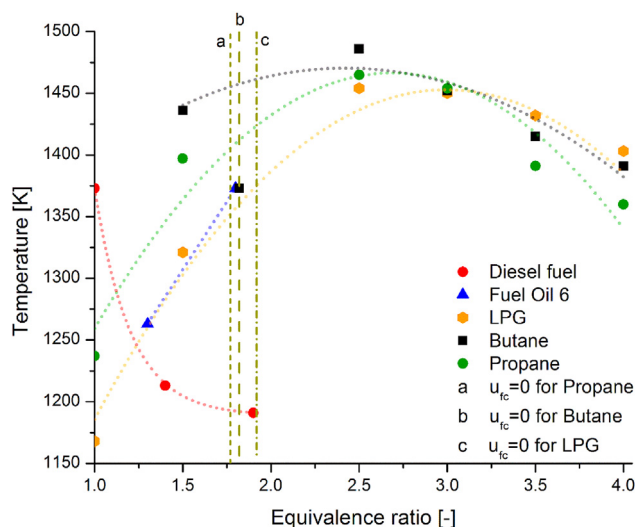
Fig. 4 – Temperature of combustion as a function of the equivalence ratio.

Toledo et al. [9]. Liquid fuels showed an upstream regime for the ϕ considered.

For the gaseous fuels, the temperatures acquired for LPG showed a maximum of 1454 K at $\phi = 2.5$ and it changes from 1168 K at $\phi = 1.0$ to 1403 K at $\phi = 4.0$. Butane's temperature results also show a maximum of 1486 K at $\phi = 2.5$ and varies from 1436 K at $\phi = 1.5$ to 1391 K at $\phi = 4.0$. In the case of propane, a maximum of 1465 K is observed at $\phi = 2.5$ and the temperature varies from 1237 K at $\phi = 1.0$ to 1360 K at $\phi = 4.0$. The gaseous fuels show a similar behavior among them, presenting a maximum, which suggests a change in the chemical kinetics mechanisms considering the presence of other intermediate species with endothermic reactions during a steam reforming step. The differences may be caused by the prior predominance of the combustion of one specie in the LPG's butane and propane mixture considering the flammability limits and their characteristic in chemical kinetics description during combustion [17,18].

On the other hand, regarding liquid fuels results, HFO's experiments show an increase on the temperature as ϕ increases from 1263 K to 1373 K in the ϕ range considered, following a similar behavior as gaseous fuels for that range. A decrease on combustion temperature from 1373 K to 1191 K as ϕ increased was observed in diesel results, presenting similar behavior to liquid n-heptane combustion in porous media [14], acknowing the use of n-heptane as an important component of diesel surrogates. Regarding these results-considering the similarities that can be found between HFO and diesel-, different tendencies are obtained, which may be related to the increase of the filtration velocity performed in HFO test as ϕ increased (diesel tests ad a constant filtration velocity).

It is important to mention that in the experiments with HFO, a carbonaceous material is observed to be formed in the surface of the alumina spheres after the experiments, producing an agglomeration that difficult further filtration of the fuel and phase changes, which inherently influences the chemical conversion of the reactants. A combustion wave of a

lean mixture of natural gas and air can regenerate the porous medium.

H_2 and CO production

The results obtained regarding H_2 and CO concentrations are shown in Figs. 5 and 6, respectively. Starting equivalence ratios above 1.0 complete combustion cannot be achieved due to insufficient oxygen content of the mixtures. Regarding the gaseous fuels, the experiments with LPG show that H_2 concentration increases for $1.0 \leq \phi \leq 4.0$ as ϕ increased, changing from 0% to 3.75%. On the other hand, the concentration of CO shows slight changes under rich conditions but a major increase is observed in the downstream regime, varying from 0% to 16.46% in the studied range. In the case of butane, a minimum of H_2 is observed at $\phi = 2.5$, showing a variation from 4.3% to 5.1% within $1.5 \leq \phi \leq 4.0$. Furthermore, a maximum of CO (14.1%) is obtained at $\phi = 3.0$ and the concentrations vary from 10.8% to 11.0% in the extremes of the ϕ range considered.

Literature provides results of propane combustion in a similar porous media reactor [9] at $u_f = 0.12$ m/s, showing good agreement in the tendencies for CO concentrations, where the concentration increases with an increase on ϕ, having a variation from 0% ($\phi = 1.0$) to 5.4% ($\phi = 4.0$) in this case. However, the H_2 concentrations show a constant behavior that it is different to the increase trend obtained for Toledo et al. [9]. Contrast with these results indicate that the H_2 production is sensitive to a change in filtration velocity or an increase in the residence time in locations with the highest combustion temperatures. A 0% is observed at $\phi = 1.0$ while 3.7% of H_2 for $\phi > 1.5$.

For the experiments with liquid fuels; considering diesel results, the concentration of H_2 presents a slight difference from 0% to 2.9–3.0% as ϕ increased within $1.0 \leq \phi \leq 1.9$. CO production is constant at 0–0.5%. For HFO, a very slight increase on H_2 concentrations is acquired by an increase of ϕ, changing from 10.0% ($\phi = 1.3$) to 10.9% ($\phi = 1.8$) while CO concentration decreased from 15.9% ($\phi = 1.3$) to 1.1% ($\phi = 1.8$)
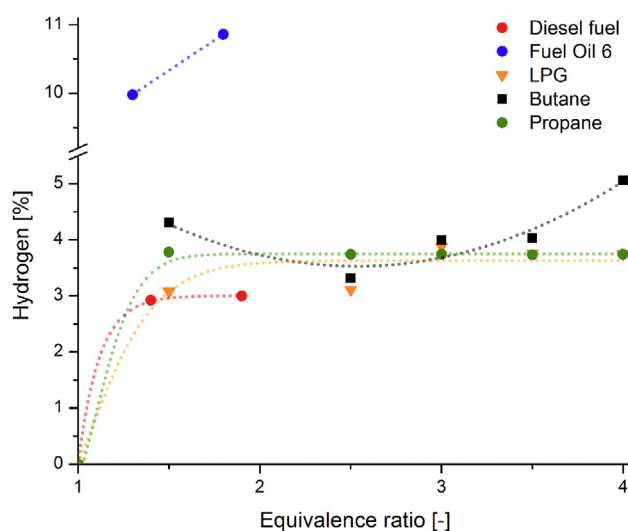

Fig. 5 – Hydrogen volumetric concentration in the products of combustion as a function of the equivalence ratio.
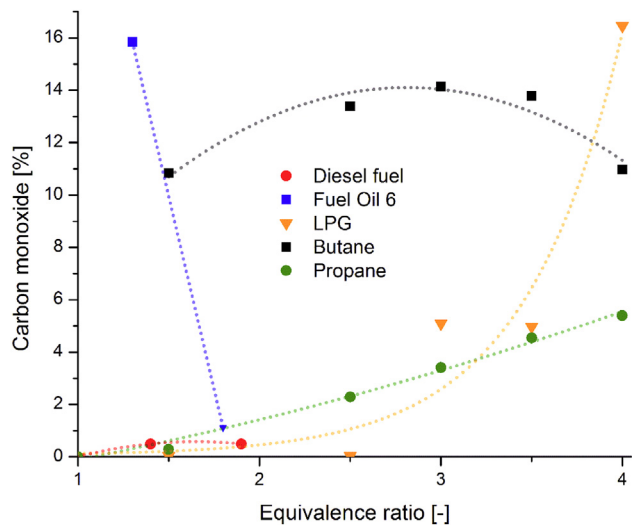
Fig. 6 – **Carbon monoxide volumetric concentration in the products of combustion as a function of the equivalence ratio.**

Fig. 7 – **Efficiency of the conversion to energy in syngas as a function of the equivalence ratio.**

which can be related to the formation of the already mentioned carbonaceous material in the surface of the porous medium, accumulating carbon in the medium instead of partially oxidizing it.

Efficiency

The conversion efficiency for each experiment (with gaseous and liquid fuels), based on the lower calorific value of the syngas (only H_2 and CO) produced was evaluated by the following relation [12]:

$$\eta = \frac{\dot{m}_{H_2} \cdot LHV_{H_2} + \dot{m}_{CO} \cdot LHV_{CO}}{\dot{m}_{fuel} \cdot LHV_{fuel}} \qquad (1)$$

where η represents the conversion to syngas efficiency, $\dot{m}_i$ is the mass flow of the specie i and LHV_i is the lower heating value of specie i.

This efficiency provides information regarding the net energy available as syngas (chemical energy) related to the energy fed by the fuel.

The syngas conversion efficiency from the considered fuels within the equivalence ratio range studied is shown in Fig. 7. It is noted that in each case, an increase on ϕ produces a decrease on the efficiency, except in the case of propane, where efficiency remains constant. Diesel showed an increase and a decrease from 0 to 5.8% to 4.4% within $1.0 \leq \phi \leq 1.9$. In HFO results, the efficiency dropped from 65.7% to 15.9% for $1.3 \leq \phi \leq 1.8$. LPG's efficiency showed an increase from 0% at $\phi = 1.0$ to 16.3% at $\phi = 4.0$. Butane's results show that the efficiency decreases by an increase of ϕ, from 29.1% to 12.1% for $1.5 \leq \phi \leq 4.0$. On the other hand, propane results do not show any significant variation with the change of ϕ, presenting an efficiency that increases from 0% ($\phi = 1.0$) to varying between 6.7% and 7.4% within the range $1.5 \leq \phi \leq 4$. An increase in the equivalence ratio implies an increase of the presence of a variety of intermediate combustion products -beside H_2 and
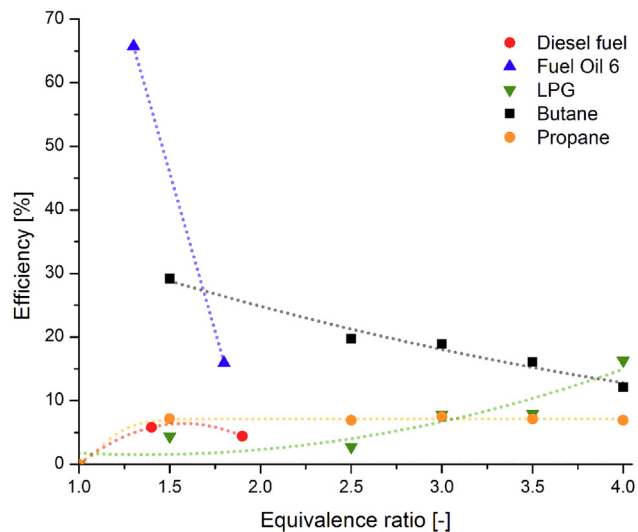
CO- and therefore, the conversion efficiency to syngas decreases, as other heavier and complex intermediate products gain predominance. Considering the magnitude of the values, LPG presents results similar to propane, having significant differences with butane, suggesting that regarding syngas conversion, propane's chemical mechanisms are dominant over butane's.

Conversion to H_2 and CO

The conversion of the different fuels to H_2 and CO is shown in Figs. 8 and 9, respectively. The conversion of liquid fuels is calculated using a common surrogate specie. Diesel was compared with n-heptane, while HFO is compared with n-dodecane since it has a molecular weight similar to the lightest fractions of HFO [19].
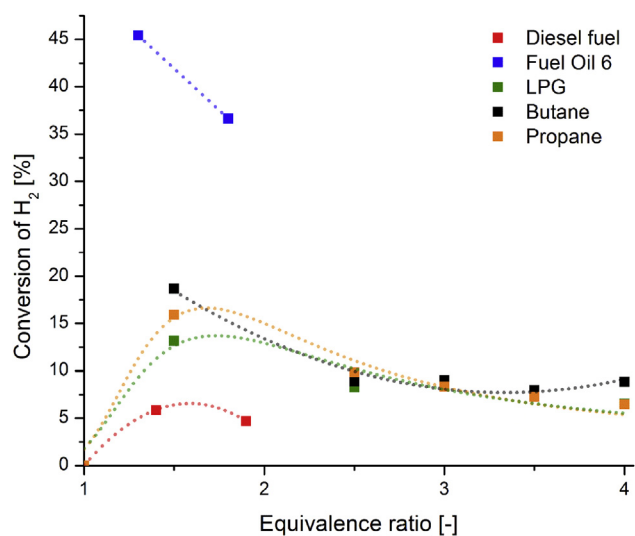

Fig. 8 – **Conversion to hydrogen as a function of the equivalence ratio.**
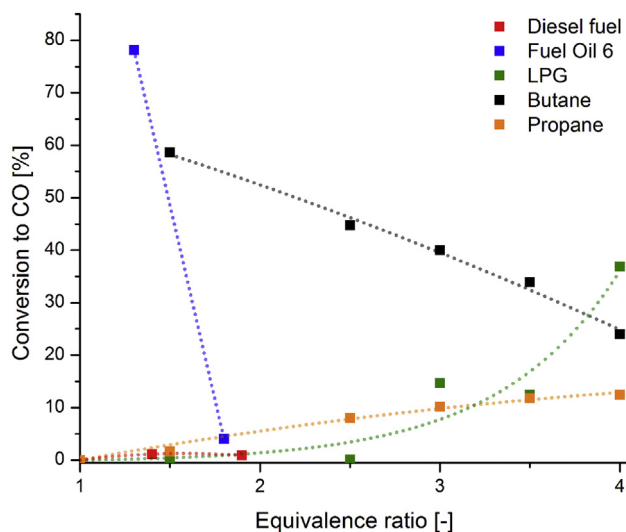
Fig. 9 – Conversion to carbon monoxide as a function of the equivalence ratio.

Considering the conversion to H$_2$, each fuel showed a decrease of the conversion as ϕ increased, which can be related to the production of other species (rather than H$_2$) as oxygen is limited. As could be noted, HFO conversion to H$_2$ presents significantly higher values than the rest of the fuels, having a conversion of up to 45.4%. Also, diesel conversion to H$_2$ decreased as ϕ increased. Maximum values of conversion for LPG, butane and propane are around 15–20% located at $\phi = 1.5$, beyond this point, a decrease can be observed as ϕ is incremented. Regarding the conversion to CO (Fig. 9), HFO results can be highlighted as the most sensitive to a change on the equivalence ratio and filtration velocity, as a dramatic drop can be pointed from 78% ($\phi = 1.3$) to 4% ($\phi = 1.8$). This drop can be related to the carbonaceous material deposited in the porous medium, that is boosted by an increase of ϕ.

Comparing butane and propane, the results at this point indicate that an increase in molecular weight (change of the fuels) allows higher conversions to H$_2$, which agrees with the results of Smith et al. [15]. Nevertheless, when comparing the results of gaseous fuels with diesel, a contrary trend is noted, resulting in lower conversions to H$_2$ with an increase on the molecular weight. On the other hand, CO results show different behavior for each gaseous fuel. Butane showed a steady reduction of the conversion as ϕ increased, while propane and LPG presented an increase. This can be related to the chemical kinetics and the temperature of combustion. Butane showed higher temperatures than propane and LPG at $\phi < 3.0$, although, these temperatures are closer for higher values of ϕ, which can be associated with the behavior of the conversion to CO.

Conclusions

The partial oxidation in inert porous media of several hydrocarbon fuels has been presented in order to assess the syngas production. A remarkable achievement of this work is that syngas was produced from HFO in inert porous media, opening new challenges for the future developments of the porous media technology to use it as a hydrogen source from this residual and low price fuel. In this case, significant volumetric concentrations were acquired, having up to a 10% in the products for slightly rich conditions ($\phi = 1.3$), setting it up as an attractive fuel to produce syngas with a superior performance than any of the others fuels tested in this work. This result also implies that the porous medium can provide the appropriate thermal and fluid dynamic conditions for a stable and steady combustion wave of HFO under certain conditions (fuel preheating) to perfect atomization of the fuel.

The behavior of the temperature results recorded showed good agreement with the available literature. Upstream regimes of propagation of the combustion wave are observed for the experiments with liquid fuels, while gaseous fuels presented upstream and downstream regimes. Normally, to maximize hydrogen and carbon monoxide production and conversion, an option would be to increase the equivalence ratio, nevertheless, considering the results of the efficiency of the conversion to syngas, in order to efficiently use the fuel it is convenient to operate the reactors in an upstream regime, which limits the increase in the equivalence ratio. This behavior applies for the studied fuels except for LPG and propane, where LPG showed an important increase in the efficiency is observed with an increase of ϕ up to 4.0 and propane did not show any significant variation related to ϕ. Observing the results of the gaseous fuels, the propane-butane mixture of LPG allows a direct comparison with the experiments of propane and butane, having differences that show a tendency of the LPG to behave similarly to propane, pointing out that the chemical mechanisms of that component are predominant in the production and conversion to syngas. Results suggest that in order to efficiently transform the fuels into syngas, it is convenient operate the reactor under an upstream regime. Furthermore, the use of liquid fuels—regardless of its refining level-has the potential to produce syngas in significant volumes, but further studies that look to describe the behavior in a wider range of equivalence ratios and that face the difficulties such as the formation of a carbonaceous material over the porous media or the availability of HFO injector that provides a wide range of flows, are required to provide a complete assessment.

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