

Mild Combustion of Surrogate Liquid Fuels

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1. INTRODUCTION

Recently significant efforts have been made in order to obtain high thermal efficiencies in high temperature combustion processes without the adverse effect of, in particular, high NO_x emissions caused by high temperature flames [1]. It was found that a massive exhaust gas recirculation coupled with a preheating of the combustion air generates relatively low flame temperatures. If the combustion air is diluted with a large amount of recirculated exhausts, the oxygen concentration in the main reaction region of the furnace is reduced to a lower concentration with respect to the case of undiluted air, allowing for a better control of the reactants kinetic and furnace average temperature. This combustion technology is commonly defined as flameless [2] or mild combustion [3,4].

The possibility to reach mild combustion conditions using gaseous fuels as methane or ethane [5-6], as well as mixtures of gaseous hydrocarbons and hydrogen [4,7], have been extensively studied; on the other hand, much less information are available concerning the sustainability of mild combustion for liquids and solids [8].

For this reason, an experimental study has been performed in a laboratory-scale burner in order to evaluate the sustainability of mild combustion for liquid hydrocarbons, such as surrogate fuels, using a two-nozzle feeding system. Therefore, the influence of different parameters, such as the oxygen excess, the air preheating temperature and the combustion gas recirculation ratio (K_V), on the stability of mild combustion and pollutants formation, has been investigated.

2. EXPERIMENTAL SECTION

In mild combustion processes, the burner geometry and the configuration of the nozzles for fuel and combustion air play a key-role allowing for obtaining high turbulence and a strong recirculation of the exhausts inside the furnace, that are requirements to reach mild combustion conditions. A burner, designed and used in previous mild combustion studies on gaseous fuels [5], and provided by a single high-velocity jet nozzle has been used in this work. The burner, a flow rate control section for the reactants feeding, and a sampling and measurement section for the detection of temperatures and exhaust gases compositions constitute the experimental apparatus. The laboratory-scale burner is a vertical quartz tube made by two sections: the combustion air preheating zone (filled by quartz pellets to enhance heat exchange), and the combustion chamber (height=350mm, ID=50mm). The air preheating is performed in the lower section of the equipment by an electric oven, while another oven prevents heat losses from the combustion chamber.

The combustion chamber bottom is provided by a gaseous reactants feeding system, showed in fig. 1a, into which fuel is fed perpendicularly through a capillary pipe. It is possible to notice the positions of other reactants inlets for the incoming flow rates of primary air (A1),

inert gas (N_2) and for the secondary air flow rate (A2). This single high-velocity nozzle configuration have been successfully used to reach mild combustion conditions using gaseous fuels such as methane or ethane [5] and mixtures of gaseous hydrocarbons and hydrogen [4,7]. Because of possible pyrolysis phenomena, a liquid fuel cannot be however fed through the capillary pipe used for the gaseous fuel and the burner cannot be directly operated from the beginning of a run using the liquid fuel. This one is fed only when the burner reaches stable mild conditions using the gaseous fuel (methane or ethane), as showed in fig.1b. Therefore, the liquid fuel is fed by means of a peristaltic pump and injected within the combustion chamber as a fine and homogeneous spray by a water-cooled (at 60°C) plain jet airblast atomizer; a stream of nitrogen, at a constant flow rate of 2 NI/min, is used to realize the liquid fuel atomization. Concerning the liquid droplets, an SMD of about $35\ \mu\text{m}$ has been estimated for the fuels here investigated.

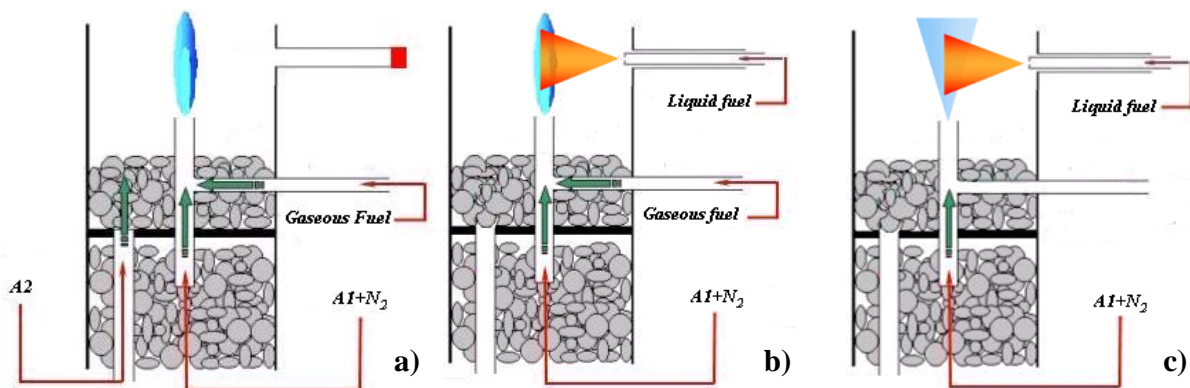


Fig.1 Reactants feeding system: (a) single-nozzle gaseous fuel feed; (b) dual-nozzle liquid and gaseous fuels feed; (c) dual-nozzle liquid fuel feed.

The transition from the gas to the liquid fuel mild combustion (fig. 1c) is then reached reducing progressively the gas flow rate (say CH_4) fed to the burner nozzle while increasing the liquid flow rate into the atomizer. This means also a transition from a single nozzle feeding system designed for gaseous fuels (fig. 1a), to a dual nozzle feeding system with the liquid fuel (fig. 1c), where only the combustion air is fed through the burner nozzle, while the liquid is fed through the lateral atomizer. The two jets interact perpendicularly and mix to each other in a high turbulence region.

Exhaust have been sampled, dried and analyzed with two on-line instruments, namely a gas analyzer Horiba PG-250 (for NO_x , O_2 , CO and CO_2 measurement) and a GC-FID Perkin Elmer Clarus 500 (for C1-C6 unburnt hydrocarbons measurement); maximum deviations of about 1 ppm and 2 ppm from the mean value have been estimated for NO_x and CO concentrations, respectively. C1-C6 hydrocarbons concentration can give a rough indication of highly toxic pollutants production since they are PAH and soot precursors. Moreover, three thermocouples have been located at different heights of the combustion chamber to detect temperatures inside the furnace.

As previously mentioned, the combustion gas recirculation, the dilution ratio inside the combustion chamber, K_V , has a paramount role determining mild combustion working conditions. As a consequence of several numerical simulations performed with a general-purpose code for computational fluid dynamics [5] and accounting also for both internal and

external recycle as well as the presence of a secondary air inlet, which flow rate is lower than the entrained one, the K_V can be computed as:

$$K_V = \frac{R - S/A}{1 + S/A} + \frac{(I/A) \cdot (R+1)}{(1 + F/A) \cdot (1 + S/A)} \quad (1)$$

where A and S are primary and secondary air, respectively, I is the inert gas flow rate (nitrogen in this case), F the fuel flow rate and R the maximum value of the recirculation factor imposed by the main jet of the burner; R can be estimated as the ratio between the flow rate of recirculated exhausts and the inlet jet flow rate. As expected, when neither secondary air nor inert gas are fed to the laboratory-scale burner, the previous relation leads to $K_V = R$. Assuming an incoming high velocity jet of the reactants [5], it is possible to highlight that R values do not change significantly in the investigated velocity range resulting in a maximum value of R equal to about 5; this value is reached at about 5.5 cm from the bottom nozzle tip, where the lateral spray-jet has been located. Therefore, in the dual nozzle setup, with the atomizer for the liquid fuel, this jet enters practically to the centre of the combustion chamber, interacting with the incoming air jet where the maximum aerodynamic recirculation is created. Mild conditions can be practically identified by both the flame disappearing and the reduction of temperature gradients inside the furnace, that means a reduction of NO_x emissions; moreover, in a previous study [7], a clean mild condition has been also defined, considering as upper threshold levels for the pollutants $\text{NO}_x < 30$ ppmv and $\text{CO} < 50$ ppmv.

According to this criterion it is possible to build, for each fuel, usual T vs. K_V diagrams which represent the influence of K_V on the combustion characteristics as shown, for the sake of example, in fig. 2. This diagram reports five main areas: a mild clean combustion region (B), where mild conditions can be easily maintained without any significant pollutant emission; a mixed zone (A), where both low-emissions and mild conditions can be achieved depending on the experimental conditions, such as a selection of a suitable combustion air preheating temperature; a thermal NO_x region, where mild conditions can be sustained but thermal NO_x production becomes significant; a no combustion (or extinction) zone and a traditional flame combustion region.

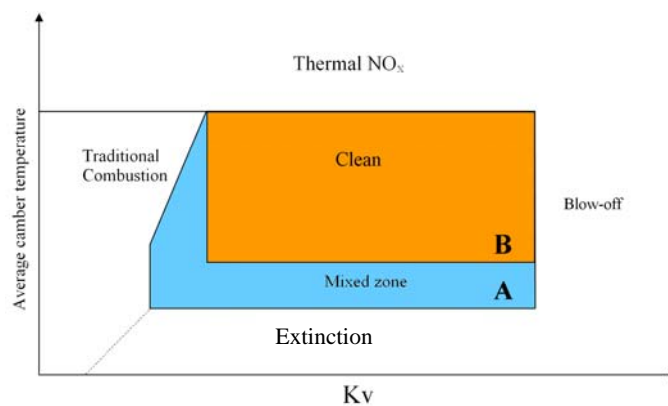


Fig. 2 T vs. K_V diagram sketching boundaries and peculiar regions identified in this work.

3. RESULTS AND DISCUSSION

Using the aforementioned two-nozzle feeding system, the possibility to use a mild combustion burner with fuels constituted by a mixture of several liquid hydrocarbons (surrogate fuels) has been tested, in order to simulate the behavior of a real fuel used in traditional combustion systems. Initially, the addition of various liquid hydrocarbons was done gradually by adding each class of hydrocarbons to a mixture of n-octane:i-octane 60:40% vol.; it constituted a reference fuel to evaluate the effects of the addition of other compounds to the mix.

First of all, a n-paraffin with a higher molecular weight, such as n-decane, was added to the octane isomers mixtures. Mild combustion can be again sustained but a slight worsening of CO emissions, especially increasing the n-decane amount, has been obtained (fig. 3a); this results also in a moderate temperature and consequently NO_x increase, caused by the higher thermal input of n-decane containing fuels.

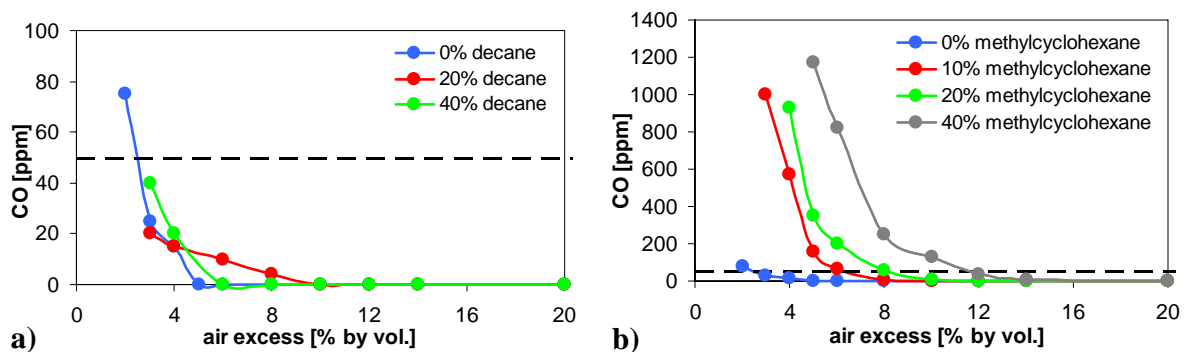


Fig. 3 CO emissions detected for a) fuels with different contents of n-decane ($K_v=5.7$); b) fuels with different contents of methylcyclohexane ($K_v=5.7$); dashed line represents the CO clean mild threshold.

Then, n-decane has been replaced, by adding to the surrogate fuel different cycloalkanes, such as cyclohexane, methylcyclohexane and decaline, respectively. Once again clean mild conditions have been attained, evidencing the capacity of the system to overcome possible problems due to the fuel composition. However, the results obtained using these three cycloalkanes were very similar and showed a significant worsening of CO emissions when the cycloalkane content is increased (fig. 3b).

In particular, in the absence of methylcyclohexane, the CO mild clean threshold limit has been respected until a 3% O₂ excess, while in its presence the limit has been exceeded for 6, 8 and 11% O₂, using mixtures with 10, 20 and 40% of methylcyclohexane, respectively. This is probably due to higher difficulties in breaking the cyclic structure of the cycloalkanes compared to the rupture of a simple linear structure. Regarding NO_x emissions, there were no substantial differences in the tests performed with mixtures containing cyclohexane (or methylcyclohexane) and the reference mixture constituted by octane isomers. Similar average furnace temperatures have been also measured. On the other hand, for mixtures containing decaline an increase in NO_x emissions has been evidenced, particularly increasing its content within the surrogate fuel. This phenomenon is undoubtedly due to higher temperatures reached in the combustion chamber because decaline is characterized by a higher calorific value (6277 kJ/mol) with respect to octane (5074 kJ/mol).

In fig. 4a NO_x emissions and exhaust temperatures have been shown, as a function of the air excess, for a dilution ratio $K_v=2.7$. With regard to CO emissions, even in tests conducted at

low K_v , measured trends were found to be similar to those recorded at $K_v=5.7$, so an increase of the cycloalkane content within the surrogate fuel produced an increase in CO emissions. However, these tests evidenced that CO emissions can be reduced, at a given air excess, by lowering the dilution ratio of the system (fig. 4b); this dependence from the dilution ratio can be ascribed to the different local oxygen concentration that is higher into the combustion chamber when the exhaust entrainment is reduced, thus influencing the CO formation.

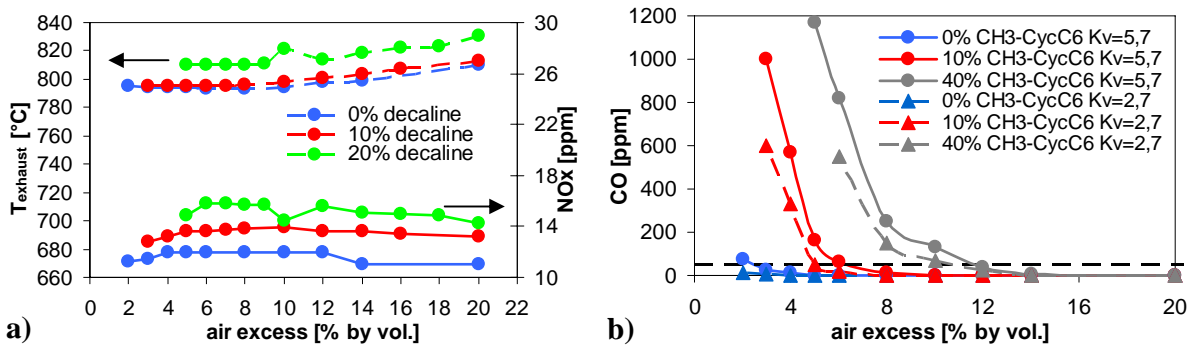


Fig. 4 Exhaust temperatures and NOx emissions measured for fuels containing different amounts of decaline (a); influence of the K_v value on the CO formation for fuels containing different amounts of methylcyclohexane (b).

Then, a more complex fuel mixture has been realized in order to closely reproduce a typical real fuel; in particular, a mixture with the following composition (% by vol.) has been prepared: 30% n-octane, 30% n-dodecane, 20% i-octane, 10% methylcyclohexane, 10% decalina. Different amounts of p-xylene have been later added to this surrogate fuel in order to analyze the effect of the aromatic compounds on the mild combustion sustainability. All these tests have been conducted at a lower preheating temperature of the combustion air with respect to those previously presented; the oven temperature has been reduced from 900 to 800°C. The results for CO emissions are shown in fig. 5a; these trends are similar to those found in experiments carried out with the fuel mixture containing 20 or 40% of methylcyclohexane. This means that the simultaneous presence of different chemical compounds into the surrogate fuel induces a higher CO production.

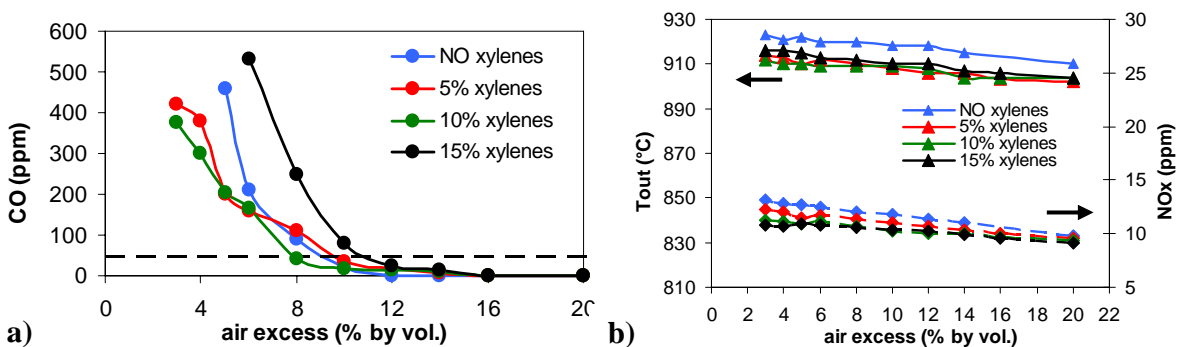


Fig. 5 CO emissions ($K_v=5.7$)(a), exhaust temperatures and NOx emissions ($K_v=2.7$)(b) measured for fuels containing different amounts of xylenes; ; dashed line represents the CO clean mild threshold.

Concerning NO_x emissions, the trends measured in tests performed with xylene-enriched fuels evidenced higher emissions compared to tests carried out with aromatic-free fuels, as shown in fig. 5b. At the same time it is possible to notice an increase of the exhaust temperatures compared to the other tests, which could explain the high NO_x emissions.

In the experiments performed with the xylenes-containing fuels, NO_x emissions reached a maximum value of 24 ppm, which is still lower than the mild clean threshold. Taking into account only the effect of p-xylene, it is possible to notice a progressive temperature reduction that follows the increase of the p-xylene percentage into the surrogate fuel. This is consistent with the fact that p-xylene has a lower heating value with respect of the other compounds contained in the liquid fuel mixture. This temperature reduction does not produce low NO_x emissions as it is clearly visible in fig. 5b. This particular behavior can be explained by a possible contribution of the prompt NO_x mechanism to the overall NO_x formation; in mixtures characterized by a high amount of p-xylene, the presence of phenyl or benzyl radicals could enhance reactions that breaks the N₂ triple bond with the subsequent formation of NO_x emissions.

4. CONCLUSIONS

An experimental study has been performed in a laboratory-scale burner in order to evaluate the sustainability of mild combustion for liquid hydrocarbons, such as surrogate fuels, using a two-nozzle feeding system. Independently of both the composition and the complexity of the investigated fuel mixture, the experimental issues showed that it is always possible to operate the burner in clean mild conditions, also for low dilution ratios ($K_v < 3$). The clean threshold limit values are actually exceeded only in extreme conditions (low excess of the combustion air and particularly low values of the recirculation degree. Moreover, several exhaust analyses, performed by means of a gas chromatograph confirmed the absence of unburnt products within the flue gas for all tests.

These findings support the idea that a two-nozzle mild combustion burner can create a suitable environment for NO_x, PAH and soot depression, allowing the use of a wide range of liquid wastes and low-BTU liquid fuels, even with unsteady composition or coming from different sources, making possible to use them not at the expense of increased emissions.

5. REFERENCES

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