Oscillatory behavior in simple hydrocarbons combustion: on the influence of the operating parameters

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Introduction

Despite much progress in the application of new combustion concepts to practical systems, such as homogeneous low-temperature combustion processes, there remain unresolved issues on the fuel oxidation chemistry and on the stabilization of the oxidation process. For instance, HCCI engines [1] can suffer of instabilities, which may cause loss of process efficiency. Therefore, the exploitation of system critical parameters on combustion properties is mandatory to promote the use of non-conventional fuels and new combustion technologies.
Given this background, the objectives of the current work were to investigate the methane combustion stability under well-stirred flowing conditions, at conditions relevant to diluted combustion systems. The temperature oscillations features are analyzed in relation to (i) mixture stoichiometry and (ii) the nature of the bath gases (He, N_2 or CO_2).

Experimental and numerical tools

In this work two Jet Stirred Flow Reactors (JSFR) were used: the former is available at the Laboratoire Réactions et Génie de Procédés, Université de Lorraine, CNRS, while the latter is at the Institute for Research on Combustion, Napoli. Hereafter they will be referred as the "Nancy" and the "Napoli" reactors.
The detailed description of the reactors is provided elsewhere [1, 2]. The two reactors can work in different ranges of system external parameters (such as mixture stoichiometry and composition, residence time), thus the simultaneous use of the two systems allows for the exploitation of the methane oxidation process in a large range of working conditions.
The oxidation process is monitored by an unshielded type R thermocouple (0.2 mm bead size) inserted in the reactors with very short signal acquisition rate to resolve in time the oscillatory behavior.
Numerical analyses were carried out using the OpenSMOKE++ [3] PSR code to understand the chemical nature of oscillations and their features (amplitude, frequency) with respect to system parameters. Three detailed kinetic mechanisms GRI 3.0 [4], the NUI [5] and the POLIMI [6] were initially tested to check their validity against experimental data obtained in this work.

Results and discussion

The experimental tests were carried out for fuel lean to rich mixtures in a wide range of inlet temperatures, at atmospheric pressure. The bath gas was changed along with the mixture dilution level. The experimental conditions are summarized in Tab. 1.

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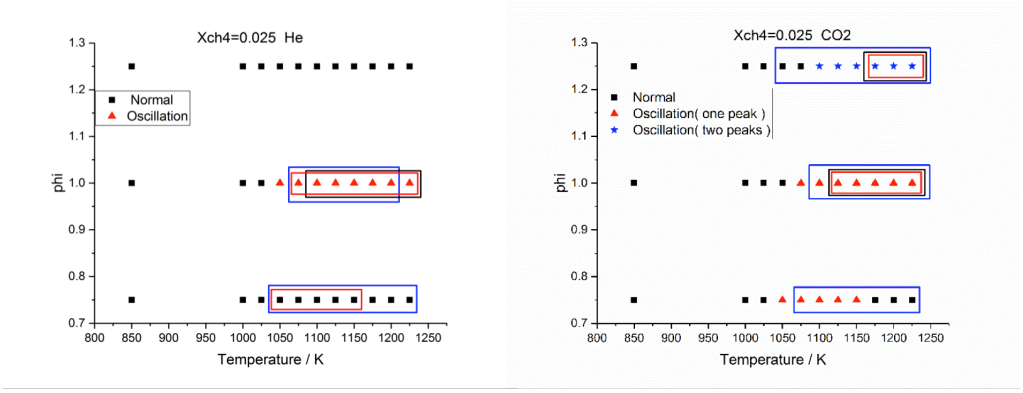
The experimental results where reassumed in T_{in}-Φ maps reported Figs. 1 and 2. Symbols represent experimental data. Different symbols identify different combustion behaviors: black squares represent stable combustion, red triangles periodic oscillations while blue asterisks represent double oscillations.

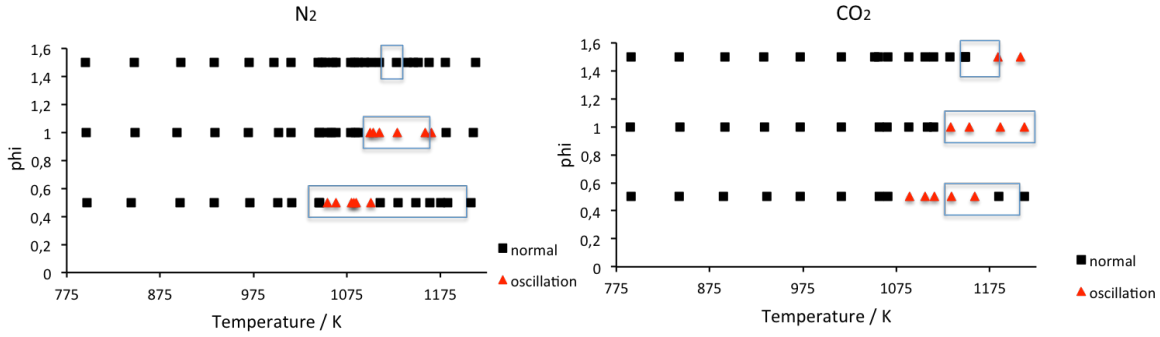
Conditions	**Range**
Inlet temperature	800-1225 K
Equivalence ratio (Φ)	0.5-1.5
Diluents	He, N_2, CO_2
CH_4 mole fraction	0.01-0.05
Residence time (τ)	0.5, 2 s

Tab. 1 Experimental conditions studied in the JSFRs for CH_4 mixtures. P = 1.1 atm when τ = 0.5 s (Napoli) and P = 1.05 atm when τ = 2 s (Nancy).



(a) (b)

Fig. 1 Comparison between experimental (symbols (Nancy)) and simulated (rectangles) data at τ = 2 s: (a) Black rectangles: GRI 3.0; blue rectangles: POLIMI; red rectangles: NUI.

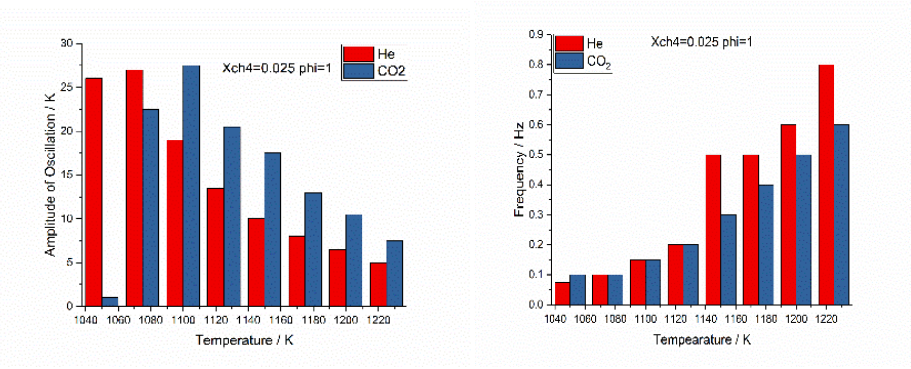


(a) (b)

Fig. 2 Comparison between experimental (symbols (Napoli)) and simulated (rectangles) data at τ = 0.5 s: (a) Blue rectangles: POLIMI.

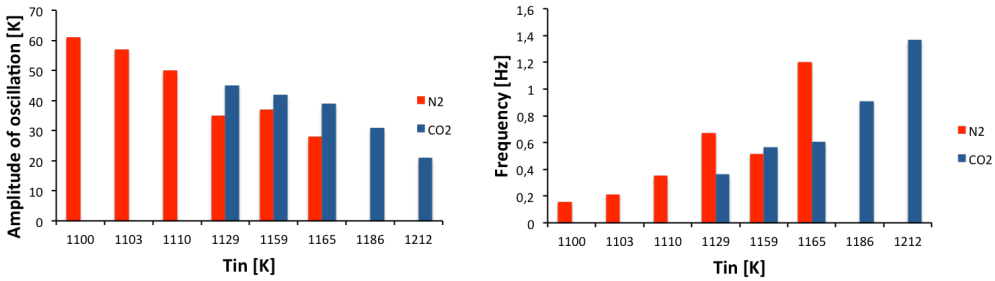
Under He and N_2 bath gas conditions, the dynamic behaviors do not occur under rich conditions (Figs. 1a and 2a), while for CO_2 diluted mixtures, the temperature oscillations appear for all the mixture equivalence ratios investigated (Figs. 1b and 2b). The onset of temperature oscillation gradually decreases with decreasing the equivalence ratio.

Fig 3 and 4 show the oscillations amplitude and frequency for different inlet temperature T_{in}, in the two sets of experiments.



(a) (b)

Fig. 3 Oscillation amplitude (a) and frequency (b) as a function of reactor temperature at Φ = 1 in different bath gases with X_{CH4} = 0.025 and τ = 2 s (Nancy).



(a) (b)

Fig. 4 Oscillation amplitude (a) and frequency (b) as a function of reactor temperature at Φ = 1 in different bath gases with X_{CH4} = 0.033 and τ = 0.5 s (Napoli).

It is possible to note that as T_{in} increases oscillations amplitude decreases while frequency increases. In both the datasets, CO_2 postpones the onset of temperature oscillations to higher inlet temperature with a higher amplitude and lower frequency with respect to He and N_2.
In Fig. 5 several combustion regimes are identified in relation to the working temperatures of the reactors datasets for the stoichiometric mixture, thus "no ignition", "slow-steady", oscillations and "high temperature-steady" regimes are reported. Also data for a stoichiometric propane/O_2 mixtures diluted in N_2 and CO_2 are included to show some general features of the analyzed behavior. The use of the reactor temperatures allows a comparison of results independently from the different operating conditions (such as the residence time, the reactor adiabaticity, fuels). It is very interesting to note that the onset of the oscillatory behavior occurs at the same working temperature (about 1100 K), thus suggesting the some kinetics controlling such a phenomenology. On the contrary, the end of oscillations depends on system operating conditions.
Numerical simulations were realized to verify the capacity of detailed kinetic schemes to reproduce the dynamic phenomenology. The comparison between experimental data and numerical results are presented in Figs. 1 and 2 (a)-(b), where the rectangle identifies the operating conditions where temperature oscillations were numerically identified. The black rectangle is relative to numerical data obtained with GRI 3.0; the red rectangle to NUI and the blue one to POLIMI. All the detailed kinetic schemes are able to reproduce temperature oscilla-

tions, but the POLIMI one well replicates the inlet temperature range where oscillation are experimentally detected, thus has been used to perform further analyses to characterize the effect of system external parameters on the oscillatory behavior.
The numerical analysis has suggested that oscillations originate from the competition between several kinetic pathways promoted under diluted conditions. Hydrogen radicals branching/recombination reactions and methyl radical oxidation/recombination reactions lead to system instabilities.

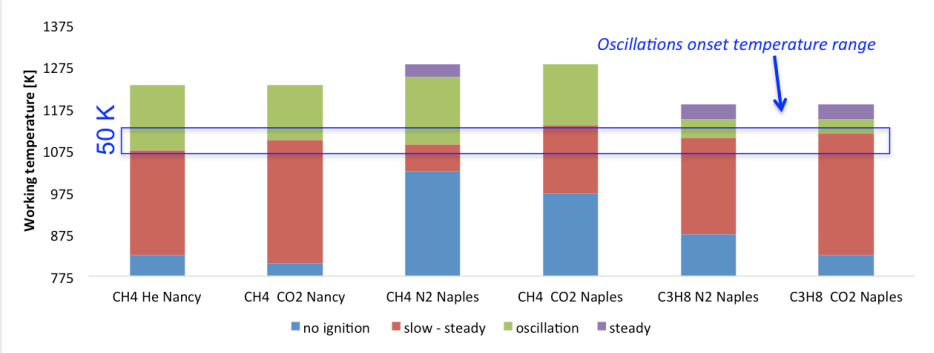


Fig. 5 Working temperature ranges for the different regimes established in different bath gases and fuels with Φ = 1.

Conclusion

Two JSFR were used to characterize the instabilities of methane/O_2 mixture diluted with several bath gases as a function of system external parameters. The study suggests that the onset of temperature oscillations occurs at a noticeable reactor working temperature, thus defining an invariant behavior with respect to system operating conditions.

Acknowledgments: The work leading to this intervention has received funding from the European Union H2020 (H2020-SPIRE-04-2016) under grant agreement n°723706 and from the COST Action CM1404 “Chemistry of smart energy carriers and technologies”.

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