# Effect of dilution by Exhaust Gases on the flame reactivity

F. Halter, F. Foucher, L. Landry, C. Mounaïm-Rousselle

LME - Polytech'Orléans, 45071 ORLEANS Cedex 2

# Introduction

Exhaust gases recirculation presents an interesting potential to reduce  $NO_x$  emissions. Indeed, the reintroduction of burnt gases in the fresh gases dilutes the mixture and allows a decrease of the combustion temperature, one of the main criterions of  $NO_x$  formation. However, the presence in the mixture of  $CO_2$  and  $N_2$ , the two main components of burnt gases, affects the reactivity of the flame. Their heat capacity at constant pressure is different from the air's one. The knowledge of this dilution effect on the flame reactivity has motivated our research. Indeed this parameter is a key for a better understanding of the different phenomena occurring inside the engine. Moreover, experimental information are of fundamental importance for developing and testing theoretical and numerical models of laminar flame propagation for multi-component fuels. Iso-octane is the most representative mono-composed carburant of gasoline. As a consequence, it is of primary importance to know the effect of dilution on this fuel. To our knowledge, only few results are available in the literature concerning the impact of dilution in the reactivity of iso-octane / air flame [1, 2]. Some results can be found for other carburant such as methane [3, 4, 5, 6], propane [7], n-decane [8], n-butane [9] or for a real gasoline [10].

This paper presents experimental measurements of laminar burning velocities of diluted methane/air and isooctane/air mixtures. Three different diluents are used:  $N_2$ ,  $CO_2$  and a mixture of both. All measurements are performed at initial conditions of 0.1 MPa and 300K. The results are obtained using a spherical combustion chamber and a shadowgraphic system. Experimental results are compared with results from literature when available and with computations done with different kinematic schemes.

# **Experimental setup**

The experimental set-up used is composed of a stainless steel cylindrical combustion chamber, two tungsten electrodes (dia. 2 mm) linked to a high voltage source, an Ar-Ion laser and a high speed camera. Optical accesses into the chamber are provided by four windows. The combustible mixture is prepared directly in the chamber by adding the fuels and the air at appropriate partial pressures to reach the total initial pressure (0.1 MPa). For experiments with iso-octane, the correct liquid quantity of fuel is weighted with a high precision balance (four significant digits after the gram). This quantity is fully injected in the chamber by a gasoline injector. To facilitate the vaporisation, a fan is located inside the chamber. Then the chamber is filled with the rest of the mixture (air and diluents). The fan mixes all the gases. Finally, the mixture is left at rest to avoid any perturbation during the flame propagation. Knowing that the saturated vapour pressure of iso-octane at 300K is of 5860 Pa, we could study gaseous stoichiometric mixture of iso-octane / air almost till 0.3 Mpa.

The visualisation of the flame is obtained by the classical shadowgraphy method. The parallel light is created by two plano-convex lenses (25 mm and 1000 mm focal lengths respectively). The shadowgraphs are recorded using a high speed video camera (Photron APX) operating at 6000 frames per second with an exposure time of 20  $\mu$ s. The temporal evolution of the expanding spherical flame is imaged and analyzed. Fundamental laminar burning velocities are obtained in taking into account both the effects of the expansion factor and of the stretch [11]. All experimental value of laminar burning velocity reported in this paper is the average of three identical experiments.

### Results

#### Results for methane

The first step was to evaluate the evolution of the laminar burning velocity for different equivalence ratio conditions. Very good agreement was obtained with literature [11]. Then, we investigated the dilution impact on stoichiometric  $CH_4$ /air flames.

Several authors started to evaluate the potential of diluted methane/air flames [3, 4, 5, 6]. Stone et al. [5] obtained laminar burning velocity of methane/air/diluents mixtures for variation in unburnt gas temperature (within 293 to 454 K) and pressures (within the range of 0.5 to 10.4 bar). Their measurements were performed under microgravity conditions. Carbon dioxide, nitrogen and a 15% carbon dioxide / 85% nitrogen mixture were used as diluents. The comparison with their results for N<sub>2</sub> and CO<sub>2</sub> dilution is biased by the fact that they consider a dilution of the fuel and not of the global mixture (air + fuel). As a consequence, their equivalence ratio is changed. In our case, the equivalence ratio is kept constant equal to stoichiometry.

The effects of nitrogen and carbon dioxide addition on  $CH_4/air$  flames are reported in Figure 1. Experiments are represented by symbols and computations using the PREMIX code of the CHEMKIN package and the GRIMech 3.0 [12] as reduced kinetic mechanism by solid lines.



Figure 1 : Effect of N2 and CO2 dilution on the laminar burning velocity of stoichiometric CH4/air flames

We observe a decrease of the laminar burning velocity when the mixture is diluted. This decrease is more important for  $CO_2$  than for  $N_2$ . This result is directly linked to the heat capacity of the different gases. We observe a very good agreement between experiments and computations for both dilutions. For high dilution levels, experiments overestimate simulations. This is certainly due to the difficulty to extract a radius from flame pictures when the propagation velocity is reduced. Indeed, in these critical conditions the flame is largely affected by natural convection effects. As a consequence, the hot burnt gases volume is convected to the top as illustrated in Figure 2.



Figure 2 : Flame picture for a N2 dilution of 24%

Once we have studied the effect of nitrogen and carbon monoxide separately, we can consider a dilution representative of exhaust gases. Elia et al. [3] investigated experimentally the laminar burning velocities of methane/air flames with addition of a mixture of 86% N<sub>2</sub> and 14% CO<sub>2</sub>, in order to simulate the effect of residual gases from exhaust gas recirculation. They used a thermodynamic model to evaluate the laminar burning

velocity from a pressure-time history of the combustion process. Liao et al. [6] achieved an explicit formula of laminar burning velocities for dilute mixtures ( $88\% N_2 + 12\% CO_2$ ). In Ponnusamy et al. [4], the EGR was simulated by a mixture of  $81.5 \% N_2$  and  $18.5 \% CO_2$ .

For technical reasons, the water is not present in the different EGR compositions. However, we decided, in our study, to consider the water volume in the  $CO_2$  content. This approximation is motivated by their close heat capacities (33 J/(mol.K) for H<sub>2</sub>O and 37 J/(mol.K) for H<sub>2</sub>O at 0.1 MPa and 300K). With this hypothesis, the EGR used in our work is simulated by a mixture of 71.6 % N<sub>2</sub> and 28.4 % CO<sub>2</sub>.

Our results and computations are presented in Figure 3.



Figure 3 : Effect of EGR dilution on the laminar burning velocity of stoichiometric CH4/air flames

As expected, we observe a decrease of the laminar burning velocity when EGR is added. Our results exhibit a very good agreement with results from other authors, even if the mixture compositions are slightly different. Results from computations are in very good agreement with our experimental results. We can conclude that the GRImech 3.0 is correctly validated for methane/dilution/air mixtures (for T=300K and P=0.1 MPa). The next step will be to add water in the exhaust gases and also to increase temperature and pressure conditions.

Results for iso-octane

Figure 4-a presents laminar burning velocities obtained for different equivalence ratios and results obtained by other authors [1, 13, 14]. We used the reduced mechanism of Hasse et al. [15] in our computations. Good agreement is obtained with data available in literature and with CHEMKIN computations.



Experimental results and numerical simulations (with the mechanism of Hasse [15]) of diluted stoichiometric mixtures are presented in Figure 4-b. Effects of nitrogen, carbon dioxide and simulated EGR dilutions on laminar burning velocities are investigated. The experimental limit is obtained for a non propagation of the flame front. As for methane mixtures, a larger decrease of the laminar burning velocities is observed when carbon

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dioxide is added (compared to nitrogen dilution). Values obtained for a simulated EGR dilution are located between nitrogen and carbon dioxide values. Experimental results are in good agreement with numerical results.

#### Conclusion

The present work aims at characterizing the effect of dilution on the laminar burning velocity of  $CH_4/air$  and  $C_8H_{18}/air$  flames. Three different diluents are used: nitrogen, carbon monoxide and a mixture representative of exhaust gases (71.6 % N<sub>2</sub> and 28.4 % CO<sub>2</sub>). Experimental results and CHEMKIN computations are presented and compared with literature results, when available. Pressure and temperature conditions are respectively 0.1 MPa and 300K.

For methane/air flames, we used the GRImech 3.0 for our computations. As expected, we observe a decrease of the laminar burning velocity when the mixture is diluted. The highest the heat capacity is, the largest the laminar burning velocity is affected. Computations with CHEMKIN are in very good agreement with our results. For our initial thermodynamics conditions, the validity of the GRImech 3.0 is tested with success. One limit of these experiments is reached for high dilution levels. Indeed, convection effects are no more negligible and affect largely the flame propagation as pointed out in Figure 2. A solution to this problem could be to perform experiments under microgravity conditions, as done by Stone [5]. However, their analysis is based on the pressure evolution (according to Dahoe et de Goey [16], this method is not the most suitable one) and their microgravity is created thanks to a drop of 300mm (which is limited). Results for isooctane/air mixtures are in good agreement with literature and numerical simulations.

#### References

- Y. Huang, C. J. Sung and J. A. Eng, Laminar flame speeds of primary reference fuels and reformer gas mixtures, Combustion and Flame, 139 (2004), pp. 239-251.
- [2] T. W. Ryan and S. S. Lestz, *The laminar burning velocity of isooctane, n-heptane, methanol, methane and propane at elevated temperature and pressures in the presence of a diluent,* SAE technical Paper Series, 800103 (1980).
- [3] M. Elia, M. Ulinski and M. Metghalchi, *Laminar Burning Velocity of Methane–Air–Diluent Mixtures*, Journal of Engineering for Gas Turbines and Power, 123 (2001), pp. 190-196.
- [4] S. Ponnusamy, M. D. Checkel and B. A. Fleck, *Maintaining burning velocity of exhaust-diluted methane/air flames by partial fuel reformulation*, IFRF Combustion Journal, article number 200506 (2005).
- [5] R. Stone, A. Clarke and P. Beckwith, Correlations for the Laminar-Burning Velocity of Methane/Diluent/Air Mixtures Obtained in Free-Fall Experiments, Combustion and Flame, 114 (1998), pp. 546-555.
- S. Y. Liao, D. M. Jiang and Q. Cheng, Determination of laminar burning velocities for natural gas, Fuel, 83 (2004), pp. 1247-1250.
- [7] Z. Zhao, A. Kazakov, J. Li and F. L. Dryer, *The initial temperature and N2 dilution effect on the laminar flame speed of propane/air*, Combustion Science and Technology, 176 (2004).
- [8] Z. Zhao, J. Li, A. Kazakov, F. L. Dryer and P. Zeppieri, Burning velocities and a high-temperature skeletal kinetic model for n-decane, Combustion Science and Technology, 177 (2005).
- [9] Y. Huang, C. J. Sung and J. A. Eng, *Effects of n-butane addition on reformer gas combustion: implications for the potential of using reformer gas for an engine cold start* Proc. Combust. Inst., 29 (2002), pp. 759-765.
- [10] Z. Zhao, J. P. Conley, A. Kazakov and F. L. Dryer, Burning velocities of real gasoline fuel at 353 K and 500 K, SAE, 112 (2003), pp. 2624-2629.
- [11] F. Halter, C. Chauveau, N. Djeballi-Chaumeix and I. Gokalp, Characterization of the effects of pressure and hydrogen concentration on laminar burning velocities of methane-hydrogen-air mixtures, Proc. Combust. Inst., 30 (2005), pp. 201-208.
- [12] G. P. Smith, D. M. Golden, M. Frenklach, N. W. Moriarty, B. Eiteneer, M. Goldenberg, C. T. Bowman, R. K. Hanson, S. Song, W. C. J. Gardiner, V. V. Lissianski and Z. Qin, <u>http://www.me.berkeley.edu/gri\_mech/</u>.
- [13] S. G. Davis and C. K. Law, Laminar flame speeds and oxydation kinetics of iso-octane-air and n-heptane-air flames, Proceeding of the combustion institute, 27 (1998), pp. 521-527.
- [14] M. Metghalchi and J. C. Keck, Burning velocities of mixtures of air with methanol, isooctane, and indolene at high pressure and temperature, Combustion and Flame, 48 (1982), pp. 191-210.
- [15] C. Hasse, M. Bollig, N. Peters and H. A. Dwyer, *Quenching of laminar iso-octane flames at cold walls*, Combustion and Flame, 122 (2000), pp. 117-129.
- [16] A. E. Dahoe and L. P. H. de Goey, On the determination of the laminar burning velocity from vlosed vessel gas explosions, Journal of Loss Prevention in the Process Industries, 16 (2003), pp. 457-478.