

Naphthalene Measurements in Methane Non-Sooting and Sooting Flames by using Laser Jet Cooled Fluorescence

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Polycyclic Aromatic Hydrocarbons (PAH) are considered as the main species involved in the soot formation processes. We recently developed a new spectroscopic method based on the LIF technique to quantitatively measure PAH after their extraction from the flame by a microprobe and their cooling inside a expanded free jet

Introduction

The detection of PAH in flames requires very sensitive techniques and a particular task concerns the measurement of these species in sooting flames in presence of soot particles.

It is known that PAH highlight interesting properties for laser diagnostics experiments (high absorption coefficients and fluorescence efficiency) which could enable to measure them even at low concentrations. However, the use of laser based techniques to detect them in flames is largely compromised due to the broadband and unresolved features of the spectra of PAH under flames conditions. That is why the measurements of PAH are usually done by sampling methods (gas chromatography, mass spectrometry, high performance liquid chromatography...) after extraction of the species from the flame.

In this poster, we present first the principles and potentials of the new spectroscopic method we developed for PAH measurements as well as the experimental set-up. We report also some quantitative profiles of naphthalene which have been measured within different methane non-sooting and sooting flames (equivalence ratio from 1.82 to 2.32). The effect of the pressure on the naphthalene formation is also presented (from 140 to 200 torr). From these measurements, it appears that, whereas the formation of naphthalene is nearly linearly sensitive with the pressure, a strong dependence on the equivalence ratio is highlighted, well fitted by an exponential law as already observed at atmospheric pressure.

Theory

The new setup [1] we developed for PAH measurements is based on the extraction of the species from a low pressure burner via a microprobe which are then directly sent into an analysis chamber where they are cooled down to about a few dozens Kelvin within a free jet

expansion. Under the jet conditions, that is, a temperature around 90 Kelvin and a pressure near 10^{-3} torr, the PAH spectra become structured (because only the first energetic levels of the molecule remain sufficiently populated to lead to electronic transitions), allowing their selective detection by LIF. The analysis chamber can also be connected to a reservoir filled with known concentrations of the studied species. Due to the very low pressure inside the jet, LIF signals are independent from quenching variations. Therefore, the intensities of the LIF signals corresponding to a specific transition of the probed PAH can be accurately calibrated with exactly the same optical arrangement.

Experimental

The set-up is based on the use of a low pressure Mc Kenna burner (6 cm diameter) enabling to stabilise premixed flat flames between 140-200 torr (within the scope of the work). Species are extracted from the flame by a microprobe centred and fixed above the burner. The burner being vertically moveable, gases can be extracted from different heights above the flame. Sampling species are directly and continuously sent into an analysis chamber (where the pressure is maintained around 10^{-3} torr) through a nozzle with 1 mm diameter aperture in order to create a supersonic jet. In this jet, PAH can be selectively analysed by LIF thanks to the spectral simplification that occurs under the jet temperature and pressure condition of the jet. A schematic representation of the setup is displayed in figure 1.

The calibration is realised by using a reservoir with a known concentration of gaseous naphthalene diluted in N_2 . The line joining this reservoir to the analysis chamber is coupled to another one within which pure

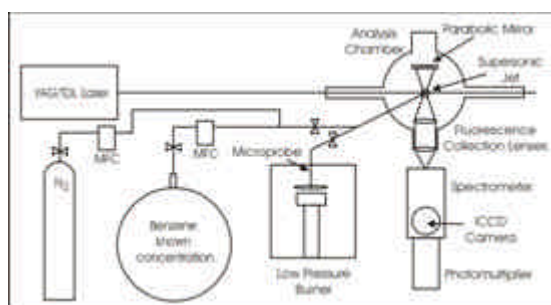


Figure 1 : Experimental Setup

N_2 can be sent in order to generate different concentrations of naphthalene inside the analysis chamber. Calibration curves are generally realized after the recording of a profile in order to be exactly in the same condition in terms of wavelength, energy, optical and collection setup so as to minimize the uncertainties of the calibration procedure. By this procedure we determined a sensitivity of a few ppb for the setup.

Results

Profiles we obtained clearly highlight 3 different regions in the flame. A first one where naphthalene is rapidly formed (until 6-8 mm according to the flame conditions, i.e. as long as there is O_2 present in significant concentration). Then a second zone, where there is rapidly no O_2 left and where PAH are consumed corresponding to the beginning of the soot formation region. And a third one where naphthalene increases again in the postflame when soot have finished their formation.

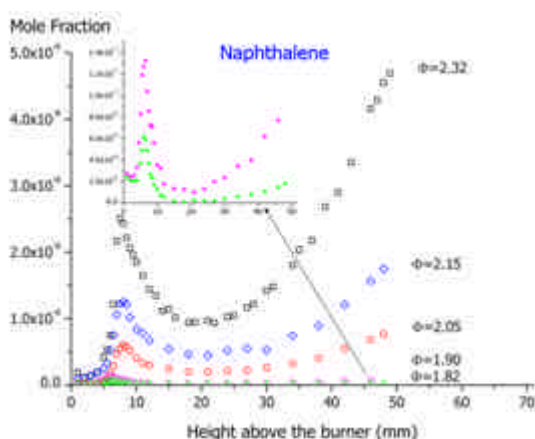


Figure 2 : Naphthalene Mole Fraction Profiles ($P=200$ torr)

Pressure effects :

A linear evolution of the mole fraction of first peak of the profile vs the pressure (140 – 200 Torr) is observed.

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Moreover, the decrease of naphthalene is more important with the pressure decrease and the re-increase of its concentration in the postflame is also more noticeable as the pressure increases

Equivalence ratio effects:

First, the concentration maximum of the first peak is extremely sensitive to the equivalence ratio. If we plot the evolution of the maximum mole fraction values (X_{max}) vs the equivalence ratio (Φ) under a logarithm scale, according to the empirical law $X_{max}=A.(\Phi)^n$ [2], it highlights two different behaviours in the naphthalene formation according to the equivalence ratio range corresponding to different sooting conditions. For sooting flames ($\Phi=2.05-2.41$), we find a value of n_i around 11 (i.e. similar to atmospheric pressure) while for nearly or non-sooting flames ($\Phi=1.82-2.05$), n_i is about 19. It is also observed that this last re-increase of the naphthalene mole fraction is not so marked for the less sooting flames, especially for 1.82 and 1.90 where no soot have been detected by laser induced incandescence (LII). Whereas the mole fraction of naphthalene at 50 mm reaches a value superior to the one reached at the peak for the richest flames (2.05-2.32), it seems to tend toward a mole fraction threshold lowest than the peak value for the poorest ones (1.82-1.90). Eventually, profiles are slightly shifted towards the burner for the lowest equivalence ratio flames.

Conclusions

Thank to this new sensitive method, we have been able to measure naphthalene profiles within different sooting and non-sooting methane flames. Dependence to the pressure and the equivalence ratio concerning the formation of this naphthalene is clearly highlighted. Pyrene is the next challenging species to be measured by this new method.

References

- [1] X. Mercier et al, Appl. Phys. B 91, 387-395 (2008)
- [2] T.R. Melton et al., Proc. Comb. Inst., 27, 1631-1637 (1998)