

Characterization of soot particles and their precursors in diffusion flame of methane by in-situ laser diagnostics and ex-situ laser desorption / ionization / mass spectrometry

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Soot and their precursors (gaseous polycyclic aromatic compounds, PAHs) were mapped by non-intrusive laser diagnostics to better understand their oxidation and formation mechanisms in the studied flames. In parallel, on-line samplings coupled to laser based desorption/ionization following by mass spectrometry are dedicated to the chemical characterization of the PAH's structures.

A CH₄ laminar-diffusion flame was chosen so as to have a stratified distribution of soot particles along the flame axis. The flame has been stabilized on a McKenna hybrid burner. The porous plug of this burner contains a central tube allowing the injection of methane diluted into nitrogen in order to stabilize a 15 cm height diffusion flame. Across the porous, the burner was fed by air. Quartz windows placed around the flame protect it from external air movements and allow in situ optical diagnostics into the flame.

In this work, different laser sources have been used to detect either the Laser Induced Incandescence (LII) from soot or the Laser Induced Fluorescence (LIF) from fluorescent species like Poly-Aromatics Hydrocarbons (PAH). Experiments were performed at 1064, 532, and 266 nm using the 2nd and 4th harmonics of an Nd:YAG laser and visible excitations wavelengths from 488 to 658 nm were generated by an OPO laser pumped by the third harmonic of the YAG. The signals were detected at right angle with respect to the laser propagation. Emission was focused using a set of lenses at the entrance of a spectrometer with a 150 g/mm 700 nm blazed grating. Spectra were recorded using an intensified CCD camera placed at the exit of the spectrometer. The detection was performed with a gate-width of 50 ns started when the first photons of the laser are emitted (prompt detection). The spectrometer has been calibrated using spectral lines from a mercury emission lamp. The spectral response of the detection setup has been corrected using an integrating sphere which approximates well the emission behaviour of a blackbody and allows working with different sets of temperature up to 3220 K. The optical setup and collection device arrangements were the same for the calibration procedure and for the detection of LII and LIF spectra.

The soot volume fraction mapping (Fig. 1) was obtained using LII technique at 1064 nm (Lemaire R., 2013).

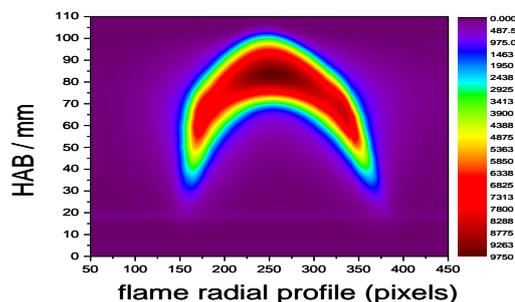


Figure 1. Soot mapping using LII at 1064 nm.

First, for LIF measurements, low laser energies were adjusted to avoid interference with LII signal emitted by soot. LIF mapping using visible excitation wavelengths from 488 to 658 nm were performed to detect the presence of fluorescent species in the flame centre line.

Secondly, for higher laser energies allowing the heating of soot, using only one laser beam (at 266 nm or 532 nm), LII signal measured in the near infrared spectral region serves to map only the soot. LIF signal is extracted taking advantage of the spectral dependence of LII and LIF emission from visible to near infrared. The 266 nm excitation leads to the detection of lighter PAHs and 532 nm to the heavier PAHs. The both approaches at low and high laser energies are compared.

In parallel, the soot and gaseous PAHs are sampled by a microprobe in the flame, diluted in an inert carrier gas and deposited onto a cold substrate directly under vacuum. The frozen samples are then analyzed by mass spectrometry coupled to laser desorption and laser ionization (Faccineto A., 2015) in order to obtain the size distributions of the different molecular families even at trace levels.

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