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Proceedings of the Combustion Institute 31 (2007) 841–849

Proceedings
of the
Combustion
Institute

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Molecular species adsorbed on soot particles issued from low sooting methane and acetylene laminar flames: A laser-based experiment

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Abstract

Recent advances in the field of laser desorption/laser ionization mass spectrometry (LD/LI/MS) have renewed interest in these separation methods for fast analysis of chemical species adsorbed on soot particles. These techniques provide mass-separation of the desorbed phase with high selectivity and sensitivity and require very small soot samples. Combining LD/LI/MS with in situ measurements of soot and gaseous species is very promising for a better understanding of the early stage of soot growth in flames. In this work, three lightly sooting laminar jet flames (a methane diffusion flame and two premixed acetylene flames of equivalence ratio (ϕ) = 2.9 and 3.5) were investigated by combining prompt and 50 ns-delayed laser-induced incandescence (LII) for spatially resolved measurements of soot volume fraction (f_v) and laser-induced fluorescence (LIF) of polycyclic aromatic hydrocarbons (PAH). Soot and PAH calibration is performed by two-colour cavity ring-down spectroscopy (CRDS) at 1064 and 532 nm. Soot particles were sampled in the flames and analysed by LD/LI/Time-of-flight-MS. Soot samples are cooled to -170 °C to avoid adsorbed phase sublimation (under high vacuum in the TOF-MS). Our set-up is novel because of its ability to measure very low concentration of soot and PAH together with the ability to identify a large mass range of PAHs adsorbed on soot, especially volatile two-rings and three-rings PAHs. Studied flames exhibited a peak f_v ranging from 15 ppb (acetylene, $\phi = 2.9$) to 470 ppb (acetylene, $\phi = 3.5$). Different mass spectra were found in the three flames, each exhibiting one predominant PAH mass; 202 amu (4-rings) in methane, 178 amu (3-rings) in acetylene, $\phi = 2.9$ and 128 amu (2-rings) in acetylene, $\phi = 3.5$. These variations with flame condition contrasts with other recent studies and is discussed. The other PAH masses ranged from 102 (C_8H_6) to 424 amu ($C_{34}H_{16}$) and are well predicted by the stabilomer grid of Stein and Farr. © 2006 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

Keywords: Soot; Desorption; PAH; Laser; Laminar flame

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1. Introduction

Soot formation is one of the most challenging problems associated with pollutant formation in combustion processes. Polycyclic aromatic hydrocarbons (PAH) are known to play a key role in the soot inception but the chemical process of PAH and soot formation is not yet fully understood [1]. Another challenge lies in the knowledge of chemical species adsorbed on soot particles because of their potential impact on human health.

Soot, PAH and species adsorbed on soot particle are intimately linked through chemical and physical processes. Soot characterisation includes the soot volume fraction f_v , the optical and morphology properties such as primary particle diameter d_p , aggregate size and particle size distributions. To measure all these properties, several optical and physical sampling methods must be combined. Recent papers show that laser-induced incandescence (LII) for soot concentration and d_p determination [2], scanning mobility particle sizer (SMPS) [3] and small-angle neutron scattering (SANS) [4] techniques for size distribution tend to supplant or complement the extinction/scattering techniques [5]. Concerning PAH detection, species selectivity is generally obtained through probe sampling coupled with gas chromatography (GC) analysis, performed on the gas phase and/or on the condensed species [6], or by molecular beam mass spectrometry [7]. In situ PAH diagnostic can be performed by laser-induced fluorescence (LIF). Although LIF mainly arises from a mixture of PAHs (not a specific one), combined knowledge of both soot and total PAH is important for soot chemistry understanding [8]. Large PAHs react first to form soot precursor particles with a typical size of a few nanometers. While several recent papers have been devoted to nanoparticles identification [4,9,10], only a few studies concern the characterisation of the chemical species adsorbed on soot particles. Hepp and Siegmann [11] established that the photoemission yield from soot particles collected in a methane diffusion flame is enhanced in flame zones containing gaseous PAHs, suggesting the contribution of adsorbed PAHs on soot.

Recent advances in the field of laser desorption/laser ionization mass spectrometry have complemented conventional (and long duration) separation methods for PAH analysis [12]. Fourier transform laser microprobe mass spectrometry (FT-LMMS) was applied in a combustion chamber of an incineration plant [13]. Laser desorption/laser ionization time-of-flight mass spectrometry (TOF-MS) was applied on Diesel particulate matter collected during driving cycles [14]. Species adsorbed on soot surface have long been considered as being PAH. But a recent study of Oktem et al. [15] in a premixed ethylene flame

showed that the early stage of particle growth is dominated by aromatics while the later stage shows a greater contribution of aliphatics. The authors used the photoionization aerosol mass spectrometry (PIAMS) technique [16]. This instrument allows direct sampling and analysis of soot particles in near real time by combining 1064-nm laser desorption and VUV ionization. In previous studies, Dobbins et al. [10,17] identified PAH in the 200- to 472-amu range in an ethylene diffusion flame by LMMS using 266-nm radiation. Aliphatic and light aromatic could not be detected because of their evaporation in the vacuum.

As shown above, selectivity, sensitivity and fast analysis of species adsorbed on soot particles are achievable by laser desorption techniques. However, these techniques remain very difficult to implement and need to be validated through additional measurements. In this work, a new set-up has been designed to study several lightly sooting flames. The experimental strategy combines prompt and temporally delayed LII to provide soot volume fraction and PAH LIF profiles. Calibration is obtained by cavity ring-down spectroscopy (CRDS) allowing very lightly sooting flames to be investigated [18]. The PAH adsorbed on soot are analysed by laser desorption/laser ionization/TOF-MS using a set-up initially designed for doped-ice analysis [19]. In this paper, soot concentration, PAH and chemical species adsorbed on soot are examined in a diffusion methane flame and two premixed C_2H_2 flames having a peak soot volume fraction ranging from 15 to 470 ppb.

2. Experimental

2.1. The burner and the soot collector

The piloted jet burner consists of a hybrid Holthuis burner equipped with a central stainless-steel injector of 4 mm inner diameter. The heat from the lean premixed flat flame of methane serves to stabilize a large range of diffusion or premixed jet flames. Perturbations from ambient air are minimised by an air shroud. Three sooting flames are investigated: a laminar methane diffusion flame ($Q_{fuel} = 0.11 \text{ min}^{-1}$) and two premixed C_2H_2 /air flames of equivalence ratio (ϕ) 3.5 ($Q_{fuel} = 0.72 \text{ min}^{-1}$) and 2.9 ($Q_{fuel} = 0.61 \text{ min}^{-1}$).

Soot particles are collected through a collector made of two concentric quartz tubes with holes on the i.d. of the inner tube through which nitrogen is pumped. Fast dilution of samples with nitrogen ensures rapid quenching of chemical reactions. In this study, the collector was placed 50 mm above the burner where it also serves to stabilize the flames. Soot particles are trapped on a porous filter made of borosilicate placed at the top of the collector thus preventing gaseous condensation.

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