



Sooting structure of a premixed toluene-doped methane flame

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ABSTRACT

The structure of a sooting premixed methane/oxygen flame doped with toluene ($C/O=0.66$) was studied by means of sampling and chemical and spectroscopic analysis of gas and condensed phases. It was found that the addition of toluene in small amounts (0.8 mol%) to the methane flame ($C/O=0.6$) used as reference did not affect the maximum temperature level and the distribution of the main combustion products and C1–C4 hydrocarbons. The main differences observed regarded the higher yield of soot and C5–C6 hydrocarbons as cyclopentadiene and benzene. Some insights on soot structure were obtained by means of spectroscopic tools, i.e. Raman and UV–Visible spectroscopy. Spectral features of toluene-doped soot indicated that toluene doping not only enhances soot production, but also changes its structural properties in terms of aromatic character of soot particles. These features could be of concern about the use of practical fuels rich in aromatic components possibly leading to higher environmental and health impact of aromatic-derived soot in respect to aliphatic-derived soot.

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1. Introduction

Aromatic hydrocarbons are important as additive or components of transportation fuels as gasoline, kerosene and diesel fuels because of their resistance to knocking in turn related to the aromatic ring stability and resistance to oxidation [1,2]. However, aromatics are much more prone to form soot and are considered the main precursors in soot formation [3]. Toluene is the most representative aromatic component of gasolines and a good surrogate of aromatics contained in diesel oils, thereby it is regarded as a primary reference fuel whose chemistry has to be studied in combustion conditions. It is also important to study the toluene combustion as involving the chemistry of benzyl which is an important intermediate in flames of alkylated aromatics leading to polycyclic aromatic hydrocarbons (PAH) [4], and eventually to soot. Most of the experimental work on toluene chemistry concerns pyrolysis in flow reactors and oxidation in a jet stirred reactor [5–8]. Furthermore, some works on toluene chemistry in flames have been carried out in flames of pure toluene at low pressure [4,9–13] and in diffusion flames burning toluene added to aliphatic hydrocarbons as ethylene [14,15], methane [16–18] and heptane [19].

The present work reports the study of the sooting structure of a premixed fuel-rich flame of methane doped with relatively low amount of toluene (0.8 mol%) to avoid thermal perturbations to

the methane flame which has been previously characterized in detail [20–22]. The chemical environment in which soot is formed has been investigated by the detailed analysis of gaseous and condensed phases sampled along the axis of the toluene-doped flame. Particular attention has been given to the structural analysis of the toluene-doped soot in comparison to soot formed in methane flames to study the effects of toluene doping on soot nanostructure, which could be expected on the basis of recent works on the microstructural analysis of soot formed from aliphatic and aromatic hydrocarbon fuels [21–28].

2. Experimental

The toluene-doped premixed flame was produced on a McKenna (Holthuis & Associates) water-cooled burner by adding small amounts (0.8 mol%) of toluene prevaporized to a previously studied methane/oxygen flame [20,21]. The cold gas velocity of the toluene-doped flame (5 cm/s) was equal to that of the methane flame (54.55 CH₄/45.45 O₂ mol.%), hereafter named baseline methane flame. To avoid toluene condensation, the cooling temperature of the burner was kept at 70 °C, largely above the dew point evaluated (0 °C) for the feed mixture (0.8 toluene/54.1 methane/45.1 oxygen mol%).

Flame temperature was measured using a fast-response silica-coated fine wire Pt/Pt–13%Rh thermocouple: 100 μm wires are jointed to 25 μm wires, which are used to realize a very small bead size of about 50 μm. The mechanical strength of the larger wires avoided any vibrations. The temperature profile measured

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with such a thermocouple system can be considered quite unperturbed and a reliable reference for adjusting the axial coordinates of intrusive sampling data. A fast-insertion procedure was used to avoid massive soot deposition on the thermocouple bead and the consequent change of the bead size and of the emissivity of the thermocouple junction. Temperature was also corrected for the radiative losses [29]. The uncertainty of the measured temperature was estimated to be as high as 100 K. By means of in-situ laser measurements, the extinction coefficient (defined as $-\ln(I/I_0)/L$, where I_0 is the incident monochromatic radiation intensity, I is the emerging radiation intensity and L is the flame thickness) was measured using a 5 mW He/Ne laser ($\lambda = 632.8$ nm). Gaseous and condensed phases were sampled from the flames by a stainless steel, water-cooled, isokinetic probe with a conical tip (i.d. 2 mm, o.d. 3 mm, the outer diameter increases from 3 to 6 mm with a cone angle of 60°). Stable gases (O_2 , CO , CO_2 , H_2 , C_1 – C_6 hydrocarbons and toluene) were analyzed by on-line gas chromatographic analysis. C_1 – C_6 hydrocarbons and toluene were analyzed on a HP5890A gas chromatograph equipped with a 7515 Chrompack Al_2O_3/KCl capillary column and a flame ionization detector (FID). O_2 , CO , CO_2 , H_2 were analyzed by using a 8700 Altech coaxial column and a thermal conductivity detector (TCD) on a HP5700A gas chromatograph. The gas sampling at low height above the burner (HAB) was performed removing the ice trap in order to avoid the condensation of unburned toluene before of the gas sampling valve. The data were obtained as average of measurements repeated thrice. Condensed phases collected in an ice-cooled trap and on a teflon filter placed along the sampling line were extracted with dichloromethane (DCM) to separate the organic carbon soluble in DCM, named DCM-extract, from soot. Both DCM-extract and soot were dried and weighed to determine their concentration in flame. To get enough amounts of condensed phases and to estimate the reproducibility of the measurements, each sampling point was repeated at least three times and the samples were added together for further analysis. The uncertainty of the measured concentrations due both to the sampling and analytical procedure was about 25% at low HAB within the main reaction zone, less than 10% up to 10 mm HAB. The uncertainty increases again up to 20% after 10–12 mm due to the oscillations of the flame tail approaching the stabilizer plate. Twenty-four polycyclic aromatic hydrocarbons (PAH) with molecular masses up to 300 u (Acenaphthylene, Acenaphthene, Anthracene, Benz[a]anthracene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo(ghi)fluoranthene, Benzo[ghi]perylene, Benzo[a]pyrene, Benzo[e]pyrene, Biphenyle, Biphenylene, Chrysene, 4H-Cyclopenta(def)phenantrene, Coronene, Dibenz[a,h]anthracene, Fluorene, Fluoranthene, Indeno[1,2,3-cd]pyrene, Naphthalene, Phenanthrene, 2-phenyl-naphthalene, Perylene, Pyrene) were detected and quantified by gas chromatography/mass spectrometry (GC-MS) of the DCM-extract by a HP5890 gas chromatograph coupled with a HP5975 mass spectrometer equipped with an HP-5MS crosslinked 5% PhMe siloxane $30\text{ m} \times 0.25\text{ mm} \times 0.25\text{ }\mu\text{m}$ film thickness column. UV-Visible absorption spectra of soot and DCM-extract suspended in N-methyl-2-pyrrolidinone (NMP) (concentration = 10 mg/l) were measured in a 1-cm quartz cell by using an UV-Visible spectrophotometer (HP8453). Raman spectra of soot were measured directly on the filter in the range of 900 – 3400 cm^{-1} (Raman shift) by means of a Horiba XploRA Raman microscope system with an excitation wavelength of 532 nm. To minimize the possibility of structural damages due to the thermal decomposition induced by the laser, the power of the excitation laser beam was reduced to about 0.1 mW. Hydrogen and carbon content of soot and carbon standard materials were measured by a Leco CHN628 elemental analyzer. FT-IR spectra of soot were obtained with a Nicolet iS10 spectrophotometer in the range of 600 – 3400 cm^{-1} on dispersions prepared by mixing and grinding

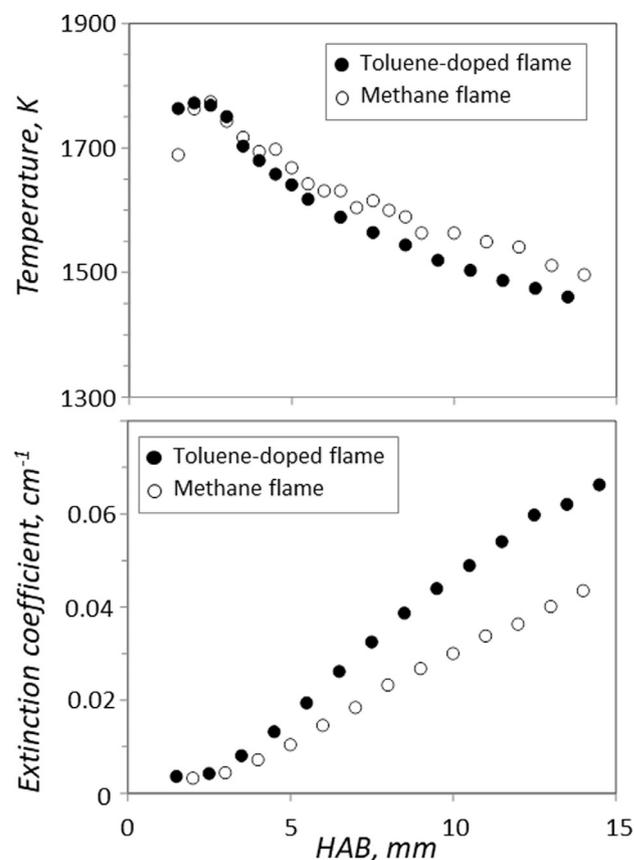


Fig. 1. Axial profiles of temperature (upper part) and in-situ laser extinction coefficient measured at 632 nm (lower part) measured in the toluene-doped (full symbols) and in the baseline (open symbols) methane flames.

the carbon samples in KBr pellets at a fixed concentration (0.25 wt. %) [30].

3. Results and discussion

3.1. Flame structure

The thermal field and sooting structure of the toluene-doped flame and baseline methane flame [20–22] are represented in Fig. 1 that reports the axial profiles of the temperature and in-situ laser extinction coefficient, measured in the visible ($\lambda = 632.8$ nm). It can be noticed that the temperature steeply increases to reach the same maximum value (1780 K) around 2 mm HAB in both flames, demonstrating that there is no effect of toluene addition on the thermal field in the early oxidation region. Downstream of the temperature peak, the steeper temperature decrease in the toluene-doped flame is symptomatic of a larger soot formation causing higher radiative losses and the consequent flame temperature reduction. The higher soot formation along the toluene-doped flame axis is confirmed by the much higher laser extinction coefficient values measured in the visible (lower panel of Fig. 1), which is directly associated to soot absorption.

The axial concentration profiles of gaseous and condensed phases measured respectively, by on-line gas chromatography and batch analysis, give an overview of the chemical structure of the toluene-doped flame. To account for the probe interference on the thermal and fluid-dynamic flame fields, the concentration profiles have been shifted upstream of few millimeters, namely 2.4 mm, evaluated by placing the maximum of CO concentration in correspondence of the flame position (height) where the temperature reaches the maximum value [29].

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