



Soot volume fraction measurement in low-pressure methane flames by combining laser-induced incandescence and cavity ring-down spectroscopy: Effect of pressure on soot formation

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Abstract

Soot volume fraction (f_v) profiles are recorded in low-pressure methane/oxygen/nitrogen flat flames using laser-induced incandescence (LII). Experiments are performed from 20 to 28 kPa in flames having the same equivalence ratio (2.32). Calibration is performed by cavity ring-down spectroscopy (CRDS) and indicates a very weak soot volume fraction (0.066 ppb at 21.33 kPa and 0.8 ppb at 26.66 kPa in the burnt gases). Soot volume fraction is found to increase continuously after a given distance above the burner (HAB) and tends to level off in the burnt gases. The reaction time resolution available in low-pressure flames makes it possible to examine the early steps of soot formation. The variation of the LII signal with laser energy before the LII “plateau” region is much weaker at the beginning of soot formation than after a given reaction time. The LII time decays are nearly constant within the first millimetres, whereas an increase in the decay, correlated with the growth of the primary soot particle, is observed later. The growth of soot volume fraction is then analysed by considering the variation of the derivative function df_v/dt with f_v . Three regimes having respectively a positive slope, a constant slope, and a negative slope are observed and are interpreted with respect to the soot inception process. Finally, a very important sensitivity of f_v with pressure P (at 30 mm HAB) is observed, leading to a power law, $f_v = KP^{11}$, confirmed by extinction measurements (by CRDS). The observed dependence of f_v with pressure could be a result of the prominence of the early soot inception process in the investigated low-pressure flames.

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1. Introduction

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The understanding of soot formation in flames has been considerably improved in recent decades thanks to the large number of experimental and modelling

works undertaken on that subject. It is now well established that polycyclic aromatics hydrocarbons (PAHs) largely contribute to the formation of the primary soot particles [1,2] and that the soot surface growth can reasonably be represented by the HACA mechanism [3]. Most of the experimental studies have been performed at atmospheric pressure and have detailed the structure of sooting flames by providing the profiles of a number of gaseous soot precursors and soot particles (e.g., [4,5]). Studies on the local soot growth and oxidation rate in diffusion [6] or premixed [7] flames have also been reported.

Prior investigations performed in low-pressure [8] and atmospheric [9] flames have largely contributed to the understanding of the mass growth of soot in premixed flames. Experiments were conducted by combining extinction and light scattering, leading to volume fractions of soot (f_v), particle number densities, and particle sizes along the reaction zone, which can be a few millimetres wide at low pressure. By probing the soot volume fraction along the distance above the burner, it was shown that the growth of the soot volume fraction follows a first-order rate equation,

$$\frac{df_v}{dt} = k_{SG}(f_v^\infty - f_v), \quad (1)$$

where f_v^∞ is the final soot volume fraction reached at long reaction time and k_{SG} is an apparent first-order rate constant.

Values of k_{SG} typically range from 100 to 200 s⁻¹. The axial soot profile can be well described by means of Eq. (1) except at incipient soot formation as explicitly indicated in [8].

To date, the detailed processes involved in the early stages of soot formation are not well understood. Recent works in atmospheric pressure flames have highlighted the contribution of nanoparticles. Those important findings rely on analysis of the peculiarities of the fluorescence spectra recorded in the selected flame zone [10,11]. Recent improvements in particle sizing techniques have made it possible to obtain size distributions of those very small nanoparticles [12–14]. Concerning the detection of primary soot particle in the narrow zone where soot inception proceeds, recent advances in laser diagnostics allow that zone to be investigated. Low-pressure premixed flames, in which the reaction zone is a few millimetres wide and is well separated from the burner surface, offer an ideal experimental support for such investigation. Surprisingly, laser-induced incandescence (LII) has not been applied in low-pressure flames except in [15,16]. Most of the other reported studies are based on extinction measurements, which are limited in terms of sensitivity.

In this work we present soot volume fraction measurements obtained in low-pressure flames of methane with a particular interest for the soot inception zone. Methane has been selected because of its important use in domestic and industrial burners. However, while chemical mechanisms of methane oxidation even under rich conditions have been the subject of numerous efforts, very few have reported on soot formation except at atmospheric pressure or above 1 bar [7,17,18]. Because the propensity of methane to produce soot is less than that of the more commonly studied fuels (acetylene or ethylene), experimental strategies had to be selected to obtain a satisfying signal-to-noise ratio. The required sensitivity was achieved by combining LII calibrated by cavity ring-down spectroscopy (CRDS). Here, the behaviour of the LII process was first studied under low-pressure conditions to determine the best experimental procedure. Then the axial soot profile was obtained by LII calibrated by CRDS and analysed in terms of surface growth of soot in the inception zone. Finally, the variation in the soot volume fraction with pressure (from 20 to 28 kPa) was studied in various methane flames of constant initial composition and compared to the corresponding studies reported at higher pressures.

2. Experiment

2.1. The burner

Experiments were undertaken in low-pressure (from 20 to 28 kPa) laminar premixed flames of methane/oxygen/nitrogen stabilised on a movable vertical McKenna burner (manufactured by Holthuis Inc.) having a 6-cm sintered bronze plug. The plug is encircled by another one through which nitrogen was flowed, thus forming a protective shroud that reduces mixing of any recirculation of burned gases (and soot) in the low-pressure chamber. The pressure was kept constant with an accuracy of ± 0.5 Torr (66.6 Pa) thanks to a motorised regulation valve connected to the pressure transducer. This control was indispensable regarding the huge sensitivity of soot formation with pressure as detailed later.

To prevent soot deposit and water condensation on the windows under sooting flame conditions, a nitrogen circulation protected the windows and the flanges supporting the windows were heated.

The flow rates and the pressure range were selected to ensure (1) a good stability of the flame and (2) a measurable incandescence signal. The nitrogen dilution was reduced compared to air to improve the flame stability. The cold mixture composition was

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