



Effect of temperature on soot inception in highly controlled counterflow ethylene diffusion flames

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ARTICLE INFO

Article history:

Received 25 October 2017

Revised 17 December 2017

Accepted 1 February 2018

Keywords:

Soot
Diffusion flames
Nucleation
Optical diagnostics

ABSTRACT

Soot volume fraction and dispersion index measurements were performed by multi-color pyrometry in a series of highly controlled ethylene counterflow diffusion flames in which the peak temperature was varied in a 300 K range, while maintaining constant stoichiometric mixture fraction and global strain rate. In so doing, the temperature-convective time history remained nearly constant, when the temperature was normalized with respect to its peak value. Therefore, the peak temperature was isolated as the key controlling parameter. We observed two distinct trends in the soot measurements: a high-temperature, high activation energy process near the flame front, for temperatures larger than ~ 1600 K, that can be rationalized with conventional nucleation and surface growth models; and a low temperature zero-activation process that is hypothesized as dimerization (homogenous nucleation) of aromatics. The first process is accompanied initially by large values of the dispersion index, denoting small C/H ratios, followed by a rapid decrease towards a plateau, as soot carbonization occurs. The second process is accompanied by a sharp increase in the dispersion index, consistent with a new inception process that is well distinct from that in the high temperature region. In one of the flames studied the soot measurements supplement an extensive set of measurements of gaseous species from a previous study, thereby offering a comprehensive database for modelers.

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1. Introduction

The study of soot formation in diffusion flames has been a major focus of the combustion community for several decades. Reducing soot emission in engines and furnaces was originally motivated by health concerns associated with the inhalation of particulate and by the need to address performance constraints in engines. More recently, the need has been made more acute by the anticipated impact of soot on climate change.

To condition the combustion environment and/or using additives to minimize soot emission, the understanding of the soot formation process must require the quantification of the evolution of the parent molecule into soot precursors and eventually particulate. Yet, with the exception of a small number of studies (e.g., [1,2]), most soot studies in diffusion flames have been preoccupied with the late steps of this evolution, with measurement of key quantities like volume fraction and average particle size of relatively mature soot (e.g., [3]), bypassing the chemical evolution from parent molecule to precursor species. This deficiency is largely mo-

tivated by an incompatibility of diagnostics, since the application of analytical chemistry tools for a proper chemical characterization requires gas sampling from flames using microprobes that invariably tend to clog in the presence of soot. There have been ingenious approaches to prevent it [4], but usually at the cost of using bulky probes and losing spatial resolution. Despite some recent progress in elucidating the soot inception process, as reviewed in [5,6], there is still a substantial gap in its understanding. Addressing such a gap requires quantitative diagnostics, adequate spatial resolution and, inevitably, a light sooting load [7–11].

As to the combustion environment of choice, for the chemical kinetic characterization of these flames/reactors, one needs to remove the fluid mechanic indeterminacy associated with turbulence, and the time-dependence of the engine behavior associated with in cylinder compression/expansion, which significantly alters the thermodynamic environment in which combustion takes place. Also in this case, this need is motivated by the reliance on analytical chemistry tools that are incompatible with highly time-dependent, turbulent, phenomena. Therefore, steady laminar environments with simple fluid mechanics and complex kinetics are favored.

Our group has been studying soot formation in various types of laminar flames [8–11]. The most significant innovations of our

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approach include: (a) a remarkable control of the *temperature-time history* to soot by judiciously choosing the feed stream composition and flow rates to evidence unequivocally cause-effect relationships in the path to soot formation and its dependence on temperature and pressure; and (b) the application of a criterion for accurate gas sampling from these flames for subsequent Gas Chromatograph and Mass Spectrometer (GC/MS) analysis, to monitor the evolution from one species to the next in the growth process. For the latter, the use of microprobes scaled on the basis of the pressure-dependent flame thickness and corrections to the species profile to account for minor displacements of the flame position caused by the insertion of the probe are critical to ensure good agreement of major products and some critical species, like benzene, between experiments and models [9–11]. As a result, intrusive measurements were performed without sacrificing spatial resolution.

In previous studies, we reported on the chemical characterization of such flames with measurements of key intermediates and the quantification of species as large as three-ring aromatics [9–11]. A first goal of the present investigation is to complement those studies in diffusion flames with soot measurements and provide as complete a database as possible with well-established diagnostic tools. A second goal is to extend the investigation to different flames in which the peak temperature is varied by a few hundred degrees to study the dependence of soot formation on temperature. Several studies in the literature focused on temperature effect on soot by using, for example, different diluents as inert gases [12–15], but none reported it in as highly controlled an environment in terms of temperature-time history, as the flames of the present study. A third goal of this study is to rely on the partial validation of a chemistry model, with respect to critical intermediates and growth species such as benzene and acetylene, to assess if changes in soot volume fraction with temperature can be rationalized within the framework of existing models of soot formation and growth [16–18].

Although the present study is focused on atmospheric pressure flames, the ultimate goal is to extend it to pressures on the order of several MPa of relevance to engines. To that end, the selected flames were configured in counterflow which is well suited to high pressure conditions [9]. An additional constraint was to use a small burner to keep cost manageable and size of the apparatus within the spatial constraints of an instrumented bench top.

As optical diagnostic for soot measurements, we used multi-color pyrometry with a digital camera [19]. This technique is well-suited to the selected flames because: (i) it is non-intrusive; (ii) contrary to laser-based alternatives, it is minimally affected by the pronounced temperature/density gradients with attending steering of the laser beam, even at the high pressures that will be the focus of future investigations; and (iii) it can provide information on the soot optical properties and, indirectly, a qualitative assessment of the soot elemental composition. In fact, if the local temperature is known and above some characteristic temperature that depends on the camera sensitivity (1200 K in the present case), the measurements provide the dispersion exponent of soot [8]. The latter describes the wavelength dependence of the emissivity (or, equivalently, absorptivity) and provides a rough indication of the soot hydrogen-to-carbon (H/C) ratio.

2. Experimental conditions, diagnostics and modeling

2.1. Boundary conditions, flame selection criteria and key flame properties

Details of the counterflow burner and analytical chemistry microsampling technique are described exhaustively elsewhere [9–11] and are only briefly recalled hereafter. The burner consists of

two identical converging nozzles oriented in a counterflow configuration, with the outlets separated by $L=8$ mm and having an internal diameter of 6.35 mm. Both nozzles are surrounded by conical enclosures to flow shroud nitrogen and minimize external disturbance of the flame.

Calibrated flows of nitrogen/ethylene and of nitrogen/oxygen are fed to the bottom nozzle and top one, respectively, to stabilize diffusion flames at atmospheric pressure. The studied flames are one-dimensional in proximity of the burner axis, as verified by both flame digital imaging and 2-D axisymmetric modeling [10]. The boundary conditions of all the investigated flames are listed in Table 1, including: mass fractions, mass average velocity, the axial velocity and its gradient on the centerline as computed by a two-dimensional model (see Section 2.3). The table also lists: the maximum flame temperature and the integrated production rate of measured soot per unit flame surface. The structure of one particular flame, with figures in bold in Table 1, with maximum temperature $T_{max}=1984$ K, was characterized exhaustively by measuring the mole fraction profiles of H_2 , O_2 , N_2 , CO , CO_2 , and hydrocarbons up to three-ring [10,11]. These measurements were used to validate the computational kinetic model, at least with respect to some important variable in soot formation like acetylene and benzene, and build confidence in its predictive capability for modest departures in peak temperature from the validated conditions. Calibrations of all mass flow controllers are performed prior to each experiment and verified to be within 1% of the full range. As a result, the (95% confidence) uncertainty in the fuel and oxidizer mole fractions is ± 0.0024 and ± 0.0020 , respectively. The mole fraction of the diluent in each stream is evaluated as the complement to unity of the reactant mole fraction. The variability in the mass averaged velocity are taken as the maximum variability of the total volumetric flow rate due to a simultaneous increase/decrease of both mass flow controllers assigned to each stream (i.e., C_2H_4 and N_2 supplying the fuel stream), yielding an uncertainty ± 0.1 cm/s. The uncertainty in the computed temperature as a result of the uncertainties in composition and mass averaged velocity of the two streams is ± 16 K throughout the entire soot forming region.

To study the effect of temperature on soot formation, we perturb flames of constant stoichiometric mixture fraction, $Z_{st} = (1 + sY_{FF}/Y_{OO})^{-1}$, and global strain rate, $a = (V_f + V_{ox})/L$ by varying the mass fraction of ethylene in the fuel stream, Y_{FF} , while readjusting the oxygen value in the oxidizer stream, Y_{OO} , as listed in Table 1. Mass averaged velocities of fuel, V_f , and oxidizer, V_{ox} , streams are selected to ensure momentum flux balance ($\rho_f V_f^2 = \rho_{ox} V_{ox}^2$) and are approximately constant for the investigated flames, because changes in stream composition are minor and the nozzle separation distance, L , is constant. The flames investigated in this study have peak temperatures spanning a range of nearly 300 K, in increments of 50 K, bracketing the maximum value of the fully characterized flame ($T_{max}=1984$ K).

The constancy of Z_{st} and a ensures that the position of the flame relative to the stagnation plane is unchanged and that the flame width is approximately constant. Perturbing the flame maximum temperature, T_{max} , with these two restrictions is very useful to isolate the effect of the perturbation on the kinetic of soot inception. In fact, as discussed in [11], the temperature-convective time history remains nearly constant, when the temperature is normalized with respect to its peak value. Therefore, the peak temperature is isolated as the key controlling parameter, as desirable in high activation energy processes leading to soot formation. Furthermore, as regards to diffusion effects, the characteristic diffusion length $\delta \approx \sqrt{D/a}$, where D is an effective diffusivity and a is the strain rate, is constant because of the constancy of the strain rate, except for changes in diffusivity because of temperature changes. Such changes are estimated to be within 20% in the soot laden

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