Challenges and artifacts of probing high-pressure counterflow laminar diffusion flames

Lorenzo Figura, Francesco Carbone, Alessandro Gomez *

Yale Center for Combustion Studies, Department of Mechanical Engineering and Materials Science, Yale University, New Haven, CT 06520-8286, USA

Abstract

An experimental study was conducted on two $C_2H_4/O_2/N_2$ laminar diffusion flames with identical compositions and strain rate, but operated at 0.29 MPa and 2.5 MPa, respectively, to assess pressure effects on the onset of soot formation. The low-pressure flame was permanently blue, whereas the high-pressure one was under conditions of incipient sooting. Both flames had nominally identical temperature–time history. Using gas sampling through quartz microprobes followed by GC/MS analysis and addressing potential artifacts associated with the diagnostic intrusiveness and probe-induced chemistry, we demonstrated that the flame structure was resolved even at the highest pressures, as shown by the good agreement with respect to major species between the experimental results and a one-dimensional computational model of the flame with detailed chemistry and transport. The increase in soot propensity correlates with an increase in the concentration of soot precursors like the aromatics and acetylene. Comparison with model predictions shows that the concentration of aromatics is systematically overpredicted. An ancillary computational study to estimate the sensitivity to changes in the velocity boundary conditions showed that minor species like the aromatics are sensitive to these details under certain conditions, because of changes in the temperature–time history that affects predominantly the slow chemistry.
© 2014 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

Keywords: Counterflow diffusion flames; Soot; Aromatics; High pressure

1. Introduction

Soot formation in flames and combustion systems remains a long-standing research problem despite considerable progress with the advent of advanced diagnostic techniques. One of the key unresolved issues is soot nucleation, that is, the molecular growth by chemical processes from gaseous phase to condensed phase that results in particle inception. Understanding this process is critical, since it determines the nucleation sites on which surface growth occurs, which, in competition with oxidation, determines the ultimate soot loading in a flame and particulate emission [1]. The challenges in tackling this task grow dramatically by operating at the high pressures that are

* Corresponding author. Address: Department of Mechanical Engineering and Materials Science, Yale University, P.O. Box 208286, New Haven, CT 06520-8286. Fax: +1 203 432 7654.
E-mail address: alessandro.gomez@yale.edu (A. Gomez).

typical of most engines, since difficulties in implementing appropriate diagnostics are compounded by challenges to adapt them to the high-pressure chamber in which experiments are conducted. As a result, there is a dearth of experimental studies on soot in high-pressure flames [2–8], and, to the best of our knowledge, none with a focus on soot inception.

We report here on the chemical speciation of two high-pressure flames, with emphasis on major species and soot precursors such as the aromatics. To retain adequate resolution even at high pressures we use microprobes to extract gaseous samples for subsequent analysis. Special attention is given to experimental challenges and potential artifacts in resolving the flame structure with adequate spatial resolution, even at the highest pressure, addressing the issue of probe intrusiveness in gas sampling and estimating the effects of probe-induced chemistry.

The focus is on counterflow flames in which we attempt to maintain constant the temperature–time history by operating at constant strain rate, composition and nearly constant peak temperature, regardless of pressure. We define the temperature–time history $T(t)$ as the time-dependent temperature field experienced by a fluid parcel moving from the fuel nozzle to the stagnation plane with the flow convective velocity. With a properly designed experimental system, the flames are essentially one-dimensional, which allows for their computational modeling with commercial software incorporating even very complex chemistry mechanisms, including molecular growth to polyaromatics and soot. In comparing experiments and model results, we discovered a hitherto unanticipated sensitivity of some of the species to the velocity boundary conditions and assess its impact on the validation of the chemistry model. A comprehensive investigation on the chemistry of high-pressure flames as they approach incipiently sooting conditions was reported elsewhere [9].

2. Experimental and computational approach

The burner is enclosed in a high-pressure vessel designed to operate up to 4 MPa, as described in [10]. As shown in Fig. 1, a converging-nozzle combustor is used, with two identical opposed converging nozzles with an area ratio of 12, and a final inner diameter of 6.35 mm. Two conical ducts concentric with each nozzle are used to produce a smooth, annular shroud flow protecting the flame from external perturbations. Fuel is issued from the bottom nozzle, oxidizer from the top. This configuration produces a well-behaved flat laminar flame up to at least $Re_D = 2000$. An image of a 2.5 MPa flame is shown in Fig. 1. Gas is sampled from the flames through silica
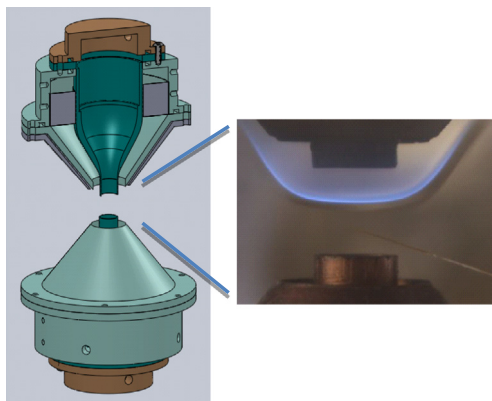

Fig. 1. View of the counterflow combustor. Inset: 2.5 MPa flame with the tilted sampling probe visible on the right.

capillary tubes (Polymicro) of different sizes, depending on pressure. The sampling probe is connected to the GC–MS system with line and internal volumes heated above 150 °C to prevent condensation of heavy species. The chemical analysis is performed with an Agilent GC–MS system, as detailed in [9,11], using a flame ionization detector and a mass spectrometer for the quantitation of CO, CO_2, aliphatics up to decane and aromatics up to indene. Uncertainty in the measurements of major species is $\pm 5\%$. For heavier ($>C_3$) species the uncertainty is estimated to be $\pm 20\%$ if mole fraction is above 0.05 ppm, because of the complexity of the liquid calibration procedure. Below 0.05 ppm, the uncertainty is estimated to be $\pm 50\%$, due to the additional signal integration error associated with the very low ion count. Calibration details are in [9,11] and in the Supplemental Material (SM).

After pressurizing the chamber with nitrogen through an auxiliary line, ignition takes place via a glow coil that is subsequently removed from the combustion zone. The system is thermally stabilized for 30 min, after which the probe tip is positioned near the flame axis and a motorized vertical stage displaces the combustor allowing for the scan of the entire flame structure. Sampling starts by drawing gas through the silica capillary tube into the sample storage volume that is kept under vacuum (<1 mbar), injecting it into the analyzer and flushing the system before moving to the next location, which takes on the order of 12 min. The analysis is run on the fly while the probe is kept in idle state until the next sample is drawn. A complete flame scan lasts eight hours.

Numerical results are obtained with the Chemkin OPPDIFF solver by Reaction Design [12] with the chemistry mechanism in [13], with detailed transport accounting for Soret effect