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Soot formation in counterflow non-premixed ethylene flames at elevated pressures

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ABSTRACT

Quantitative soot volume fraction measurements were conducted in a counterflow non-premixed flame configuration using ethylene/nitrogen as the fuel stream, oxygen/nitrogen as the oxidizer stream, and a pressure range of 1–8 atm. The laser-induced incandescence technique, calibrated using the light extinction method, was used to measure the soot volume fraction distributions. The variations of soot formation along the centerline of the counterflow flame with pressure were compared by keeping the density-weighted strain rate constant. Maintaining a constant density-weighted strain rate allows the overall flame thickness, as well as the reactant mass fluxes entering the flame, to remain unchanged for all pressures. As such, the effect of pressure on soot chemistry can be isolated from the effect of convective-diffusive transport. Based on the measured soot volume profiles, the soot layer thickness variation with pressure was determined. It was found that when keeping the density-weighted strain rate constant, the soot layer thickness remains similar over the pressure range investigated. However, the soot layer thickness was seen to decrease with increasing pressure when holding the strain rate fixed. In addition, the effects of fuel mole fraction and oxygen mole fraction on soot formation were investigated. Furthermore, the pressure scaling factors of soot formation under varying mixture conditions were deduced from experimental measurements. A literature gas-phase reaction mechanism including polycyclic aromatic hydrocarbon (PAH) chemistry up to pyrene was also used to simulate the experimental counterflow flames. The pressure effect on PAH formation was presented and discussed.

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1. Introduction

Most practical combustion devices operate at elevated pressures to enhance thermodynamic efficiency and net power, allowing for a reduction in their physical size. Concomitantly, the variation of pollutant emissions with pressure also needs to be taken into account. For instance, it has been reported that soot formation increases exponentially with pressure [1–5]. Soot is known to adversely affect the lifetime of combustion devices, as well as pose health related concerns to humans. Consequently, it is important to examine soot formation at elevated pressures, with the ultimate goal being able to control and reduce soot formation [6].

There have been various soot models developed to study soot formation at elevated pressures. Since a comprehensive understanding of soot formation processes, even at atmospheric pressure, is still lacking, the accuracy of these models remains limited for various applications. Additional information under physically relevant conditions, such as sooting flame studies at elevated

pressures, is necessary to improve these soot models. In the past, studies of soot formation at elevated pressures have been conducted in laminar non-premixed [1–5,7–11] and premixed [12–14] flames. The objectives of these studies were to express the relationship that soot volume fraction is proportional to pressure as P^n , where n is the pressure scaling factor. Lee and Na [1], using two-color pyrometry, measured soot concentrations in coflow non-premixed flames for pressures of 1–4 atm. For the non-premixed flames of [1], the oxidizer stream consisted of air, while the fuel stream consisted of either pure ethylene or a binary mixture of propane with ethylene. Based on the maximum local soot volume fraction data reported in [1], a square dependence on pressure, i.e., $n = 2$, was inferred by Bento et al. [2]. Flower and Bowman [3] used a coflow laminar non-premixed ethylene flame at an elevated pressure range of 1–10 atm to measure the soot volume fraction using the line-of-sight light extinction (LE) diagnostic method, and reported a pressure scaling factor of $n=1.2 \pm 0.1$. McCrain and Roberts [4] investigated the formation of soot at elevated pressures up to 2.5 MPa in methane–air coflow non-premixed flames and up to 16 atm in ethylene–air coflow non-premixed flames. It was observed from laser-induced incandescence (LII) images that the flames got narrower with increasing pressure, and that the local

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peak soot volume fraction was found to scale with pressure for the flames. McCrain and Roberts [4] reported that the local peak soot volume fraction scaled with pressure according to $n = 1.2$ for methane–air flames, and according to $n = 1.7$ for ethylene–air flames. Furthermore, the path-integrated soot volume fraction, which was obtained using the LE technique, was found to scale with pressure according to $n = 1.0$ and $n = 1.2$ for methane–air and ethylene–air flames, respectively [4]. Hence, the pressure dependence of soot volume fraction based on the local maximum is different than that determined from the integrated value [4]. Guo et al. [7] reported a similar pressure scaling factor of $n = 1.2$ for maximum annularly integrated soot volume fraction in coflow ethylene non-premixed flames over a pressure range of 1–8 atm. Radially-resolved soot concentration and soot temperature measurements have been reported by Thomson et al. [8] in laminar non-premixed coflow methane flames for pressures up to 40 atm. Using the soot emission spectroscopy and line-of-sight attenuation techniques, Thomson et al. [8] obtained a square pressure scaling factor, i.e., $n = 2$. Zhou et al. [10,11] conducted LII soot volume fraction measurements in laminar non-premixed coflow *n*-heptane flames over a pressure range of 1–3 bar. The integrated soot volume fraction (i.e., integrating over the entire area of a two-dimensional flame) suggested pressure exponents of $n = 3.4 \pm 0.3$ for prompt gating measurements [10] and $n = 1.38 \pm 0.32$ for delayed gating measurements [11], respectively. It is also reported in [11] that the distribution areas of the soot become slightly thinner toward the central area of the flame with increasing pressure (1–3 bar).

Most of these literature studies at elevated pressures have been conducted in coflow non-premixed flame conditions. However, these flame configurations are two-dimensional and require significant computational resources to accurately model them. In contrast, a counterflow flame configuration is quasi-one-dimensional in nature, facilitating simulations with detailed chemistry and soot model [15]. Recently, Amin and Roberts [16] reported a pressure scaling factor for maximum soot volume fraction of $n = 3.45$ in counterflow non-premixed ethylene flames for a pressure range of 2–5 atm when the global strain rate was kept constant at $K = 30 \text{ s}^{-1}$. Recognizing that ethylene is one of the major olefins produced as an intermediate species in hydrocarbon combustion and that the study of sooting ethylene flames at elevated pressure conditions would enhance the fundamental understanding of soot formation processes in hydrocarbon flames [4], the current study aims to investigate the soot formation for counterflow non-premixed ethylene flames at elevated pressure conditions.

Regarding the essential influence of pressure on flame responses and dynamics, Law [17,18] stressed and demonstrated the importance of considering density (or pressure) weighting in the interpretation of various combustion phenomena. Namely, density-weighted diffusivities and mass fluxes, instead of the commonly used diffusivities and velocities, are the basic dynamic variables in the interpretation of the distinct roles of diffusive transport and chemical kinetics. Since vigorously-burning aerodynamically-strained non-premixed flames are controlled by convection and diffusion, and since density-weighted diffusivities are insensitive to pressure variations, their flame structures are correspondingly insensitive to pressure variations, provided that the strain rate is also density weighted [19]. Law and co-workers advocated through a series of studies that the density-weighted strain rate, instead of the strain rate alone, is the relevant parameter to represent the effect of stretch for the pressure effect on the extinction of counterflow non-premixed flames [20–22], on diffusive ignition limits [23–48], and on sooting limits of counterflow non-premixed flames [49,50]. Therefore, the influence of density (or pressure) variation on the extinction/ignition/soot inception chemistry can

be isolated from that on the fluid dynamic variation of the strain rate.

In view of the above, the current study of the effect of pressure on soot formation was investigated based on the following consideration. Since strain rate influences residence time and the flame structure of a counterflow non-premixed flame, and, thus, dictates the rate process of soot formation, it is necessary that the changes in the overall flame structure due to pressure variation are isolated in order to make meaningful comparisons. That is, a constant density-weighted strain rate was used to keep the flame thickness constant and to isolate the chemical effect on the flame, as conducted in the experiments of [19,24]. To our knowledge, this study is the first to carry out soot volume fraction measurements at elevated pressure conditions by keeping the density-weighted strain rate constant, with the objective of understanding the effect of pressure on soot formation in ethylene non-premixed combustion. In addition, we aim to provide experimental datasets of high quality that can be used for validation of numerical simulations and chemical kinetics of soot formation processes.

In the following section, experimental specifications, including details on the counterflow burner facility and the diagnostic techniques of LII and LE, will be discussed. The subsequent section will specify details on the counterflow non-premixed flame model employed and the gas-phase reaction mechanism taken from literature, which includes polycyclic aromatic hydrocarbon (PAH) chemistry up to pyrene (i.e., four-ringed aromatics). Then in later sections, the pressure effects on flame structure and soot volume fraction profile by keeping density-weighted strain rate or strain rate constant will be illustrated. Experimental results and their comparison to the simulated results will be also presented and discussed.

2. Experimental specifications

2.1. Counterflow burner facility

A counterflow burner facility was used in the current study to produce laminar counterflow non-premixed flames at varying pressures. This facility was designed to operate for pressures up to 20 atm and has proven capable of achieving stable counterflow premixed flames for liquid fuels at elevated pressures [51,52]. The high-pressure chamber body is made of 0.5 inch (12.7 mm) thick stainless steel with a 13 inch (330.2 mm) height and a 10 inch (254 mm) inside diameter. The pressure inside the chamber is controlled by regulating a back pressure valve at the top of the chamber. Two identical nozzles are placed opposed to each other inside the chamber, with the nozzle for the fuel stream located at the bottom facing upwards and the nozzle for the oxidizer stream located at the top facing downwards. Each convergent nozzle has an exit diameter of 10 mm. The separation distance between the two nozzles is 11 mm. Both fuel and oxidizer streams are diluted with nitrogen before flowing into the bottom and top nozzles, respectively. A shroud of nitrogen gas is also used to isolate the resulting flame from the ambience. Flow rates are regulated by sonic orifices of 1% accuracy. Four protruding flanges house cylindrical openings, which are positioned on the chamber such that each opening is perpendicular to its neighboring openings. Two of the openings, which provide the incident laser sheet and detection camera optical access to the flame, are sealed with quartz glass windows to minimize light attenuation. The other two openings are sealed with BK-7 glass to provide visual access for the operator. To prevent soot from accumulating on the inner surface of the windows, each window is purged by a nitrogen jet; these jets have a negligible effect on the flame. Another nitrogen flow is introduced from the bottom of the chamber to transport the soot particles out of the chamber without perturbing the flame.

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