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Strain rate effect on sooting characteristics in laminar counterflow diffusion flames

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ABSTRACT

The effects of strain rate, oxygen enrichment and fuel type on the sooting characteristics of counterflow diffusion flames were studied. The sooting structures and relative PAH concentrations were measured with laser diagnostics. Detailed soot modeling using recently developed PAH chemistry and surface reaction mechanism was performed and the results were compared with experimental data for ethylene flames, focusing on the effects of strain rates. The results showed that increase in strain rate reduced soot volume fraction, average size and peak number density. Increase in oxygen mole fraction increased soot loading and decreased its sensitivity on strain rate. The soot volume fractions of ethane, propene and propane flames were also measured as a function of global strain rate. The sensitivity of soot volume fraction to strain rate was observed to be fuel dependent at a fixed oxygen mole fraction, with the sensitivity being higher for more sooting fuels. However, when the soot loadings were matched at a reference strain rate for different fuels by adjusting oxygen mole fraction, the dependence of soot loading on strain rate became comparable among the tested fuels. PAH concentrations were shown to decrease with increase in strain rate and the dependence on strain rate is more pronounced for larger PAHs. Soot modeling was performed using detailed PAH growth chemistry with molecular growth up to coronene. A qualitative agreement was obtained between experimental and simulation results, which was then used to explain the experimentally observed strain rate effect on soot growth. However, quantitatively, the simulation result exhibits higher sensitivity to strain rate, especially for large PAHs and soot volume fractions.

1. Introduction

Soot emission from hydrocarbon fuels combustion is a significant environmental [1] and health concern [2] such that related regulations are becoming increasingly more stringent. A major approach of achieving soot reduction is through controlling its formation during the combustion processes. Consequently, a fundamental understanding of the detailed mechanisms leading to soot formation is required.

The chemistry for the formation of soot and its molecular precursors, i.e. polycyclic aromatic hydrocarbons (PAHs) can be orders of magnitude slower than that for main fuel oxidation [3]. Thus, PAH and soot formations are rate-limiting processes and could be affected appreciably by characteristic flow time scale. Therefore, the effect of flow time on soot and PAH formation is an important research topic.

Counterflow diffusion flame (CDF) is a canonical flame configuration to study flow time effect through variation in strain rate, which can be achieved by changing the nozzle exit velocity. CDF is also a convenient configuration for numerical modeling due to its quasi-one dimensional nature. In addition, it has important applications in laminar flamelet models for turbulent non-premixed flames [4]. With respect to sooting characteristics, the relative location of the flame surface and the stagnation plane differentiates CDF into soot formation (SF) and soot formation oxidation (SFO) flames [5]. Soot zone starts on the fuel side of the flame and are then transported toward the stagnation plane. In a SF flame, the stagnation plane is located on the fuel side of the flame, therefore the soot will be convected away from the high temperature flame zone without being oxidized. On the other hand, in a SFO flame, the stagnation plane is situated at the oxidizer side of the flame and the soot is expected to be convected toward the flame to be oxidized before leaking out.

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The effect of strain rate on soot and PAH formation in CDFs has been previously investigated both experimentally [6-12] and numerically [6,7,11,13]. The results generally showed that an increase in strain rate reduced both soot volume fraction (SVF) and PAH concentrations. Du et al. [14–16] measured the critical strain rate for soot onset based on the light scattering technique for various gaseous fuels, including the effect of dilution. Recognizing the importance of the effect of unsteady strain rate on flamelets [17–19], the response of SVF and PAH concentrations to unsteady strain rates in CDF were studied experimentally [8,12] and computationally [3,20,21]. These efforts have contributed significantly to our understanding of the interaction between characteristic flow time and soot/PAH formation. However, systematic data on detailed sooting structure in CDFs with varying strain rate remain limited, especially among different fuels. Notable sooting characteristics study in CDF involving various fuels includes Refs. [8,13,22].

Recently, sooting limit characteristic have been investigated in CDFs and sooting temperature index and sooting sensitivity index have been proposed by varying the nitrogen content in the fuel and oxidizer streams to achieve soot onset [23]. The sooting temperature index (STI) has been defined as either the minimum temperature that could generate soot for SF flames, corresponding to the case with the fuel mole fraction of unity, or the maximum temperature that could generate soot for SFO flames, corresponding to the case with the oxygen mole fraction of unity. These two STIs correlated well with the threshold soot index (TSI) for the tested fuels [23]. The sooting sensitivity index (SSI) was defined as the rate of dependence of STI on strain rate. It has been demonstrated that with varying strain rate in CDFs, STI correlated well with SSI for various fuels [23].

Motivated by this and considering that practical fuels are mixtures of a large number of hydrocarbons, we extend our SSI study to include the effects of strain rate on soot formation for various fuels in actually sooting flames, in addition to the previous sooting limit study. We also note that studies with simultaneous PAH and soot measurements at various strain rates are limited, while such information could provide a useful database for soot model developments. In this regard, experimental studies were performed on the effect of strain rate on PAH and soot formation in counterflow diffusion flames. Four different fuels (C_2H_4, C_2H_6, C_3H_6 and C_3H_8) have been studied. Oxidizer having different oxygen mole fractions (X_O) was used to examine the effect of oxygen rich environment on soot formation at different strain rates. In addition, numerical modeling of soot formation using recently developed PAH and soot surface growth models were performed for ethylene flames.

The objectives of this study are to: (1) systematically measure and compare the effects of strain rate on soot and PAH formation in counterflow diffusion flames for various fuels with various level of oxygen enrichment; and (2) perform detailed modeling of the sooting structure of ethylene counterflow diffusion flame with varying strain rate and compare with experimental results, for a better understanding of the underlying mechanism regarding the strain rate effect.

It is worthwhile to note here that although some studies on the effects of strain rate on soot formation already existed in literature, we propose to enrich the database with new comprehensive dataset as well as detailed soot modeling with the following unique features: (1) we demonstrated that although at a fixed X_O, the sensitivity of soot loading to strain rates depends on fuel types, after matching peak soot volume fraction of different fuels at a reference strain rate (through adjusting X_O), the sensitivity of soot loading to strain rates becomes comparable among the tested fuels; (2) We measured PAH fluorescence at various different wavelengths for different size groups as a function of strain rate (and X_O) and compared the results among a variety of fuels. For different fuels with similar soot loading at a reference strain rate, their PAH fluorescence level can be significantly different and the sensitivity of PAH to strain rate is also different to that of soot. (3) For a better understanding, we conducted soot modeling for ethylene fuel at various strain rates with detailed gas-phase mechanisms including the formation of large PAHs up to coronene (A7) and compared with experimental data, which, to the authors' knowledge, has not been done before.

2. Experiment

The experimental apparatus consisted of a counterflow burner and mass flow controllers, a laser-induced incandescence (LII) setup for soot volume fraction (SVF) measurement, a laser-induced fluorescence (LIF) setup for PAH measurement and a light extinction/light scattering (LE/LS) setup for SVF, soot number density, and average size measurements. Details of the burner, LII, PAH LIF and LE/LS setups have been reported previously [24,25] and a brief description is given here. The counterflow burner has two opposing divergent-convergent nozzles with an i.d. of 10 mm and a separation distance of 8 mm. A concentric slit of 2 mm width provided a nitrogen shield. Commercially-pure grade fuel and O_2/N_2 mixtures were supplied through the lower and upper nozzles, respectively. The burner assembly was mounted on a two-dimensional translation stage with a movement increment of 100 µm. The nozzle exit velocities (V_0) tested were 10, 15, 20, 25 and 30 cm/s, these being controlled by mass flow controllers calibrated with precision positive displacement flow meters. It is worthwhile to point out here that, in the current study, we used global strain rate as represented by V_0. Although local strain rate obtained from measured velocity profiles [26,27] are a more accurate representation, global values as derived from asymptotic analysis [28] can also provide quantitative information sufficient for this study, considering that (1) in the current study we are more interested in the relative effect of strain rate increase rather than the absolute value of strain rates and that (2) global and local strain rate values were found to be correlated well linearly [7]. Thus, the usage of global strain rate is expected to be acceptable.

The LII technique was used to measure SVFs for ethane, ethylene, propane and propene flames at varying V_0, while the SVF for the ethylene flame was also measured using the LE/LS technique for cross-checking with the LII result and for modeling test. The flame length, required for the determination of local extinction coefficient in the light extinction technique, was determined from LII images.

In the LII setup, the second harmonic of a pulsed Nd:YAG laser beam (10 ns pulse duration at 10 Hz, 532 nm) was shaped to form a vertical sheet beam. The laser fluence was set to be within the saturation regime [25,29]. Signal detection was made by an intensified CCD (ICCD) camera with a band pass filter (center wavelength 400 nm, FWHM 40 nm [30,31]) to avoid noise from elastic scattering, C_2 swan band emission, as well as PAH fluorescence signal, considering that anti-Stokes component of the fluorescence signal is much lower than the stokes component [32,33]. The ICCD gate was opened 30 ns after the laser and the gate duration was set at 80 ns. This 30 ns delay also helped to reduce PAH interference since the lifetime of fluorescence is much shorter than the incandescence signal originated from black body radiation. The short gate width (80 ns) also enhanced the inhibition of the background incandescence signals from soot at flame temperature, which contributed to the background signals and was thus subtracted during post-processing.

For the PAH LIF measurements, a laser sheet was formed from a dye laser at around 283 nm with energy of about 1 mJ/pulse. It has been confirmed that there was no LII signal if such laser energy was used in the LII experiments. Therefore, LII interference with PAH fluorescence signal was avoided. Narrow band-pass

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