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Effect of dimethyl ether (DME) addition on sooting limits in counterflow diffusion flames of ethylene at elevated pressures

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ABSTRACT

The effects of dimethyl ether (DME) addition to ethylene fuel on sooting tendencies with varying pressure were investigated in counterflow diffusion flames by using a laser scattering technique. Sooting limit maps were determined in the fuel (X_F) and oxygen (X_O) mole fraction plane, separating sooting and non-sooting regions. The results showed that when DME is mixed to ethylene, the sooting region was appreciably shrank, especially in the cases of soot formation/oxidation (SFO) flames as compared with the cases of soot formation (SF) flames. This indicated an inhibiting role of DME on sooting. An interesting observation was that the critical X_O required for sooting initially decreased and then increased with the DME mixing ratio to ethylene β for the cases of SF flames, exhibiting a non-monotonic behavior. This implied a promoting role of DME on sooting when small amount of DME is mixed to ethylene. As the pressure increased, the sooting region generally expanded. Specifically, the range of β in promoting soot formation extended with pressure. This implies that a strategy in reducing soot by adding DME to ethylene at high pressures required a large amount of DME addition. To interpret the observed phenomena, kinetic simulations including reaction pathway and sensitivity analyses were conducted with the opposed-flow flames model using the KAUST-Aramco PAH Mech. The results showed that the thermal effect of DME addition on sooting tendency monotonically decreases with β. The chemical effect was found to be the main contributor to the DME addition effect on sooting tendency, resulting in the non-monotonic sooting limt behavior. The pathway analysis showed the role of methyl radicals generated from DME promoted incipient benzene ring formtion when small amount of DME was added, which can be attributed to the soot promoting role of DME addition for small β.

1. Introduction

The combustion of fossil fuels provides energy, while accompanying hazardous emissions such as particulate matters and nitric oxides pose threats to the environment [1–4]. In particular, soot emission has been recognized to play detrimental roles on global warming and human health. To develop technologies for soot reduction, physical mechanisms of soot formation and oxidation at conditions relevant to practical combustion devices should be understood, which has motivated extensive studies on dominant factors controlling soot formation [5–9].

Fuel structure is one of the important factors that affect soot formation and thus the sooting propensity [10–16]. In this regard, several quantitative metrics have been proposed, including the threshold soot index (TSI) [10] and the yield soot index (YSI) [17], which are mainly based on coflow flames or the smoke points of pure fuels and air. Recently, a sooting limit map was investigated in counterflow diffusion flames (CDFs) by adopting light extinction/scattering techniques. Critical oxygen and fuel mole fractions [18,19] required for sooting were determined, from which the sooting temperature index (STI) and sooting sensitivity index (SSI) [19] were proposed. They revealed that the sooting tendency increased with the number of carbon atoms of fuels at a specified strain rate, and alkane fuels showed a lower sooting tendency than alkene fuels with the same number of carbon atoms.

Dimethyl ether (DME) is generally considered as a promising fuel additive or alternative fuel [4,20–26] because of low particulate emission, low auto ignition temperature, and rapid vaporization upon injection, which comes from its high cetane number (55 – 60) (suitable for compression ignition engines), high oxygen content (35 wt%), low boiling point (249 K), and relatively low vapor pressure (easy to handle and transport) characteristics. Additionally, DME can be produced from diverse sources such as natural gas, coal gasification, oil residues, and biomass [20]. The

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possibility of reduction of harmful emissions with DME addition was investigated in various laboratory scale flames and engine studies [27–30]. The flame structure and species composition with DME blending in iso-octane fuel was investigated experimentally and numerically in premixed low-pressure flames, and they revealed that the addition of DME decreased the formation of soot precursors while enhanced the production of formaldehyde [29]. However, in counterflow diffusion flames, a small amount of DME addition to ethylene fuel enhances the formation of small aromatic precursors and soot particles in the pyrolysis region based on laser-induced fluorescence and incandescence techniques [22,24]. These studies were mainly conducted under atmospheric or low-pressure conditions. Considering that pressure plays a crucial role in overall soot yield and practical combustion devices generally operated under high pressures [31,32], it is important to extend the DME mixing effect on sooting tendency to higher pressures.

In the present work, we systematically investigated the effect of DME addition to ethylene fuel in counterflow diffusion flames. Note that ethylene fuel has been extensively investigated for soot studies both experimentally and numerically including soot modeling [5,12]. Sooting limits were determined using a laser scattering technique and the effect of pressure was studied. Further, numerical simulation was conducted to provide chemical kinetic insights of the experimental observations.

2. Experiment and simulations

The apparatus consisted of counterflow burners, a flow control system and a laser light scattering setup. Two counterflow burner setups were employed, one dedicated at atmospheric pressure and the other for operation at elevated pressures. Atmospheric-pressure experiments were conducted on both burners to test the setup-dependency in sooting limit measurement.

The atmospheric-pressure counterflow burner (Setup 1) had a pair of contoured nozzles with an exit diameter of $d = 10\,\mathrm{mm}$ and a separation distance of $l = 8\,\mathrm{mm}$. Concentric slits with 2 mm width supplied shield nitrogen to prevent interference from ambient air. The high-pressure burner (Setup 2) was enclosed in a pressure vessel, and had $d = 8.5\,\mathrm{mm}$ and $l = 8\,\mathrm{mm}$. The width of the concentric slit for shield nitrogen was 9 mm. The pressure vessel had four optical windows providing 160° of optical access, whose details were reported previously [33]. An electronic back-pressure regulator controlled the vessel pressure within 2% fluctuations at a setpoint. Nitrogen flow across the optical windows prevented soot particle accumulation, however, routine cleaning of the windows was required to ensure data quality. For all flames in both setups, the global strain rate (a) was maintained at 50 s^{-1}, which was calculated based on the nozzle exit velocity ($U_0 = 20\,\mathrm{cm/s}$) and $l/2$ [34]. The flow rates of all gases were controlled by thermal-based mass flow controllers. Due to the saturated vapor pressure of DME (about 5 atm at room temperature), the maximum pressure investigated in this work was limited to 3.5 atm for a stable operation. At higher pressures, an appreciable flame fluctuation was observed due to the vapor pressure issue. A future study is required at higher pressure by resolving this issue.

For the laser light scattering setup, an Argon-ion laser (514.5 nm) was used as the light source, and lock-in amplifiers coupled with a mechanical chopper were utilized to improve the signal-to-noise ratio. The optical system was aligned to provide a uniform laser beam with a diameter of 100 µm. To monitor the laser intensity fluctuation, a reference signal was monitored by a photodiode. The scattering signal was measured by a photomultiplier tube (PMT) at 90° to the incident light. As spurious laser reflections from other material surfaces may induce errors in the scattering signals, signal testing using three gases (nitrogen, propane, and helium) was conducted following ref. [35]. Helium (99.9999 %) was regarded as the background case due to its extra-small (≈ 0) scattering cross-section relative to other gases. Since the actual scattering cross-section of helium was not exactly zero, the method could slightly overestimate the ratio of propane to nitrogen of 13.6, which was acceptable in determining sooting limits (this ratio was 12.7 in [35]).

The fuel stream was composed of ethylene (> 99.9 %), DME (> 99.9 %), and nitrogen (99.9999 %), while the oxidizer stream was oxygen (99.9995 %) and nitrogen. In the following, X_F (X_O) represent the fuel (oxygen) mole fractions in the fuel (oxidizer) stream with the rest of nitrogen in each stream. The DME mixing ratio (β) in the fuel stream was defined as the ratio of volumetric flow rates between DME and mixture fuel (ethylene + DME).

To investigate the potential reasons for the effect of DME addition on sooting limits especially at elevated pressures, kinetic simulations including reaction pathway analysis and sensitivity analysis were conducted using the opposed-flow module [36] in Chemkin-Pro package. The boundary conditions and other relevant input parameters used in the simulations were determined from the experimental conditions. Specifically, the simulated flame conditions were set to a global strain rate $a = 50\,\mathrm{s}^{-1}$, the inlet temperature was $T_\mathrm{in} = 298\,\mathrm{K}$, and the pressure ranged from $P = 1$ to 5 atm. The kinetic mechanism used here was KAUST-Aramco PAH Mech [37], where the base mechanism is AramcoMech 1.3 [38] validated against the DME/O_2/N_2 flame at high pressure (12.5 atm). Since the experiments were conducted at soot onset conditions, only gas-phase reactions were considered in the simulation.

3. Results and discussion

Scattering signal is sensitive to particle size, thus the scattering intensity from soot particles is significantly larger than that from gas molecules. Based on this, sooting limit can be determined by varying X_F and X_O following the method detailed in [18,19]. Figure 1 shows a typical procedure in determining sooting limits, where the scattering signal along the centerline of the burner was recorded at a spatial increment of 0.1 mm for the baseline ethylene flame ($\beta = 0$). The critical oxygen mole fraction ($X_\mathrm{O,cr}$) at sooting limit is determined at a specified X_F by varying the oxygen content at which the scattering signal varies drastically. In some cases, a critical fuel content ($X_\mathrm{F,cr}$) was determined at a specified X_O.

The scattering signals at the atmospheric pressure for $X_\mathrm{F} = 1.0$ at several X_O are shown in Fig. 1a, along with direct images of corresponding flames in the insets. At $X_\mathrm{O} = 0.16$, the scattering signal monotonically decreases from the fuel to the oxidizer side, as a result of the larger gas-phase scattering cross-section of the fuel molecule C_2H_4 than that of the oxidizer. At $X_\mathrm{O} = 0.17$, a local maximum appears due to the particle scattering from soot, which is also confirmed by the orange-color luminescence emitted by heated soot particles. This first appearance of the local maximum is judged as the critical sooting condition. A higher concentration of O_2 contributes to the increase in flame temperature and production of radical species in counterflow flames, which promotes incipient ring formation for soot inception and subsequent growth along with H abstraction from soot surface generating more active site for soot growth [39,40]. This may explain why the oxygen concentration has to exceed a threshold for soot formation to occur. As will be explained in detail later, these flame conditions correspond to the soot formation (SF) flame [41], where the flame is located on the oxidizer side from the stagnation plane, thus the convection is toward the fuel side in the region between the stagnation plane and flame. Since the concentrations of oxidizing species such as OH and O_2 are very low, oxidation effects on soot are minimal. A further increase in $X_\mathrm{O} = 0.18$ results in a significant increase in the magnitude of the local maximum due to appreciable soot formation. At 3.5 atm (Fig. 1b) with $X_\mathrm{F} = 0.75$, an increment of 0.01 of