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Effect of fuel composition on soot and aromatic species distributions in laminar, co-flow flames. Part 2. Partially-premixed fuel

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ABSTRACT

The goal of this work is to understand the effect of fuel molecular structure on soot precursors and soot in an axisymmetric, co-flow, laminar flame configuration at atmospheric pressure with partially-premixed fuel jets. Five fuels with varying molecular structure are investigated: methylcyclohexane/*n*-dodecane mixture, *n*-heptane/*n*-dodecane mixture, *iso*-octane/*n*-dodecane mixture, *m*-xylene/*n*-dodecane mixture and pure *n*-dodecane. The flames investigated have jet equivalence ratios of 24 and 6. The total carbon flow rate and carbon fraction of the two components are kept constant to facilitate comparison among fuels. A laser-induced fluorescence technique is used to obtain spatially-resolved polycyclic aromatic hydrocarbons, soot precursors, in the jet flames. The polycyclic aromatic hydrocarbons are identified into two classes: single/two ring aromatics (small) and aromatics having three to five rings (large). A laser-induced incandescence and laser extinction technique are applied to obtain two-dimensional soot volume fraction for all the flames. The experimental results indicate that the level of soot is highest for the *m*-xylene/*n*-dodecane fuel at approximately three times the peak soot levels in the paraffinic fuels. The data show the effects of premixing on the spatial distribution of aromatic species and soot, including the shift from a soot field that peaks near the flame front to one that has maximum soot volume fractions near the centerline in *m*-xylene/*n*-dodecane flame. The *iso*-octane/*n*-dodecane and methylcyclohexane/*n*-dodecane fuels show a similar transition of soot field that has an annular peak in the non-premixed flame to a more uniform soot field under premixed conditions. Normalizing the maximum soot volume fraction by the maximum for the *n*-dodecane base fuel, the data shows that, within measurement uncertainty, the effect of fuel structure on maximum soot volume fraction is independent of the equivalence ratio of the fuel jet.

1. Introduction

This paper is the second in a pair that present results from investigations of the effects of fuel composition on aromatic species and soot in laminar co-flow flames with non-premixed and partially-premixed fuel jets. The first paper, Wang et al. [1], focuses on flames with non-premixed fuel jets, whereas as this paper present results for flames with partially-premixed fuel jets. The research summarized in these papers was conducted as part of a Department of Defense-University-Industry collaboration. The major goals for the collaborative program were to obtain data and develop fundamental models to simulate emissions from military engines burning alternative fuels [2]. The collaborative program involved closely coupled experimental and modeling efforts. More background on the program are provided in [1,2].

The test fuel set included five fuels. *n*-Dodecane was used as the base fuel and was used to check consistency of flames and results for all the experiments. In addition to *n*-dodecane fuel, four binary fuel mixtures were prepared by mixing *n*-dodecane with *m*-xylene, *n*-heptane, *iso*-octane and methylcyclohexane. These four hydrocarbons represent the four major hydrocarbon classes found in alternative fuels – aromatic, normal paraffin, branched paraffin, and cyclo-paraffin. In order to effectively summarize all the important outcomes from this study, the results were divided into two papers. This paper focuses on the partially-premixed flames, $\phi_{jet} = 24$ and 6. It includes a brief summary of the experimental setup and measurement methods, which are discussed in detail in the companion paper of the flames with non-premixed fuel jets [1].

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Nomenclature

Symbol	name
2-D	two dimensional
DoD	Department of Defense
HAB	height above the burner
HACA	hydrogen-abstraction-acetylene-addition
Inf	non-premixed flame
LE	laser extinction
LIF	laser induced fluorescence
LII	laser induced incandescence
PAH	polycyclic aromatic hydrocarbons
TSI	threshold soot index
ϕ_{jet}/phi	jet equivalence ratio
m-X	*m*-xylene/*n*-dodecane mixture
C_7	*n*-heptane/*n*-dodecane mixture
i-C_8	*iso*-octane/*n*-dodecane mixture
MCH	methyl-cyclohexane/*n*-dodecane mixture
C_{12}	pure *n*-dodecane

2. Related work from previous studies

The work of Calcote and Manos [3] is one of the earliest systematic efforts to determine the effect of molecular structure on soot. They compiled results from other studies in both premixed [4–8] and non-premixed flames [9–14] for fuels from C2 to C16. In order to compare sooting tendency of different fuels, they developed the threshold soot index (TSI). In this method, the fuels were ranked from a value of 0 (least sooting) to 100 (highest sooting). In premixed flames and diffusion flames, they found that with the increase in the number of the carbon atoms TSI value increased. Additionally, in both non-premixed and premixed flames, the aromatic fuels had higher TSI as compared to the paraffin fuels. Furthermore, *iso*-alkane fuels had slightly higher TSI than *n*-alkane fuels.

Olson and Pickens [15] measured the TSI for 55 pure hydrocarbons from C3–C16 in a laminar premixed flame at atmospheric pressure. They found TSI increased with the increase in the number of carbon atoms for a given chemical class. The increase in TSI with carbon number was more rapid for molecules having less than six carbon atoms as compared to the molecules having more than six carbon atoms. Additionally, sooting tendency was dependent on the molecular structures, and the aromatic fuels had a higher TSI as compared to the alkanes.

Since the early work of Calcote and Manos, and Olson and Pickens, many studies have focused on understanding the pathways to soot precursors and soot in premixed flames. The early work on soot mechanisms focused on gaseous fuels, which includes flames of methane, ethylene, propane, and methane/ethane/propane fuels doped with higher hydrocarbons for e.g., [16–24]. These studies have aided in understanding the effect of premixing on soot through physical and chemical effects. In these studies, the soot volume fraction decreased in most of the flames with premixing. However, with ethylene and acetylene fuel, the soot volume fraction initially increased with slight premixing, and then decreased at higher level of premixing [19,25]. The study of partially premixed ethylene flames for e.g., [19,20] reported the peak soot volume fraction increases until an equivalence ratio of 24, and with further addition of air, the peak soot volume fraction decreases. The reason for this increase in soot with premixing has been attributed to the differences that occur in fuel breakdown due to oxidative pyrolysis, which provides favorable pathways to benzene formation [17]. Chernov et al. [26] numerically investigated the partial premixing effects on soot in ethylene flames. The numerical model found good qualitative and quantitative match to soot trends with premixing in ethylene flames studied by Arana et al. [19]. The authors' found that on the centerline, soot growth is due to both PAH and acetylene related processes; the higher inception rates led to the initial increase in soot yield. In annular region, soot growth is primarily due to HACA growth; higher acetylene yield resulted in the initial increase in soot yield for an equivalence ratio of 24.

In addition, soot has been studied in premixed flames of large *n*-alkane fuels [27–29]. Inal and Senkan [27] studied an *n*-heptane premixed flat flame at two different equivalence ratios, 1.91 and 2.1. They found that the flame with higher equivalence ratio produced higher concentrations of aromatics and soot. Abid et al. [28] studied the evolution of soot in a premixed flat flame using *n*-dodecane as the fuel at an equivalence ratio of 2 for two different maximum flame temperature of 1807 K and 1875 K. They found the soot size distribution was bi-modal, and the nucleation mode was stronger as compared to the ethylene flame. de Andrade Oliveira et al. [29] measured the PAH and soot in vaporized *n*-heptane and *n*-decane flames over an equivalence ratio range of 3.7 to 8.5. They found a linear correlation between peak PAH and peak soot in these two fuels.

Iso-octane flames have been studied previously by Paul and Datta [30]. This study investigated the structural change of soot precursors as a function of height above the burner (HAB) in a co-flow premixed flame. However, the soot volume fractions are not reported in this work. The study by Bakali et al. [31] on *iso*-octane and *n*-heptane premixed flat flames found differences in the C4–C7 intermediate decomposition products formed from these two fuels. These differences in the intermediate decomposition products from parent fuel underlines the importance of the chemical effect of fuel molecular structure on pathways to soot formation. The studies involving cycloalkane premixed flame include that of Alfe et al. [32], which investigated the soot nanostructure from a cyclohexane flame, and Li et al. [33] and Hansen et al. [34], which investigated the different pathways to benzene formation in cycloalkane fuels. However, these cycloalkane fuel studies do not report soot volume fraction measurements.

The aromatic fuel, *m*-xylene, has been studied previously in diffusion flames [35]; however, the effect of premixing on soot precursors and soot has not been investigated. The most studied aromatic fuel under premixed condition is benzene [18,36–38]. The most related study involving aromatic premixed flame is that of McEnally et al. [18]. They investigated effect of premixing in benzene-doped methane flames from non-premixed condition until equivalence ratio of three [18]. They found premixing resulted in an increase in the concentration of cyclopentadienyl, which can form naphthalene through the reaction of two cyclopentadienyl radicals. However, naphthalene concentration and soot reduced with increasing levels of premixing. Grieco et al. [38] investigated fullerenes formation pathways in a premixed benzene/oxygen flame.

3. Experimental setup and methods

Details of the experimental set up and the measurement methods are presented in Wang et al. [1]. Therefore, only a brief description is provided in this paper.

3.1. Burner and fuel vaporization system

The co-annular laminar flame burner used in this study is modified version of the burner used by Santoro et al. [39]. The original burner design is modified to prevent fuel vapor condensation in the fuel tubing with the use of heating tapes and insulation around the fuel tube. A part of the fuel tube is smaller and has 0.097 in.