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Effect of fuel composition on soot and aromatic species distributions in laminar, co-flow flames. Part 1. Non-premixed fuel

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ABSTRACT

The work described in this paper is part of Department of Defense-Industry-University collaboration to develop fundamental understanding of the effects of alternative fuels on emissions from military gas turbine engines. The study was conducted in an axisymmetric, co-flow, laminar flame configuration at atmospheric pressure to determine the effects of fuel structure on aromatic species and soot. Five fuels with varying molecular structure were investigated: pure *n*-dodecane and four binary mixtures: *m*-xylene/*n*-dodecane, *n*-heptane/*n*-dodecane, *iso*-octane/*n*-dodecane, and methylcyclohexane/*n*-dodecane. Flames with non-premixed and partially-mixed fuel jets were studied. This paper describes the results of flames with non-premixed fuel jets; a companion paper describes the results for the partially-premixed fuel jets. In all of the experiments, the total carbon flow rate and the fraction of carbon from the two components in the binary mixtures were kept constant to facilitate comparison among the fuels. A laser-induced fluorescence technique was used to obtain spatially-resolved information on the aromatic species. Signals from aromatic species were collected in two wavelength ranges corresponding to species with one or two rings and three–five rings. Laser-induced incandescence and laser extinction were applied to obtain two-dimensional soot volume fraction for all flames. The flames of the paraffin fuels are non-smoking and have similar spatial distributions of aromatic species and soot as well as maximum soot volume fractions. The flame from the *m*-xylene fuel is smoking and has spatial distributions of aromatic species and soot that are distinctly different than for the paraffin fuels. Maximum soot volume fraction produced by the *m*-xylene fuel is approximately three times that of the paraffin fuels.

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1. Introduction

The experimental investigation described in this paper was part of a collaborative Department of Defense-Industry-University research program to obtain data and develop fundamental models required for accurate simulation of emissions from military gas turbine engines burning alternative fuels [1]. The collaborative program was motivated by two major considerations. First, military gas turbine engines, which are the major source of emissions on military bases, consume approximately 70% of the fuel purchased by Department of Defense (DoD) [2]. Second, at the time the program began, very little information was available on the changes in emissions that could occur when alternative fuels are integrated into the DoD jet fuel supply. Subsequent to the start of this program, the Federal Alternative Jet Fuels Research and Development Strategy was written. This strategy includes a recommendation for

collecting fundamental data on the effects of alternative fuels on emissions [3].

The collaborative DoD-Industry-University program involved mutually supportive, tightly coupled experimental and computational efforts. A series of well-controlled laboratory experiments was designed and conducted that systematically progress in complexity in a way that permits collective analysis of the data to develop a fundamental understanding of the production of emissions from gas turbine engines burning alternative fuels under a wide range of conditions. Experimental facilities used in the program include: a well-stirred reactor in which chemical kinetics are the controlling process; shock tubes focusing on kinetics at realistic pressures; non-premixed and partially-premixed co-flow flame experiments with dependence on kinetics and molecular diffusion; and two combustor test rigs – a model combustor at Penn State and the Referee Combustor at Wright-Patterson Air Force Base, which include the composite effects of all the processes occurring in a gas turbine combustor operated at pressure including spray atomization, vaporization, diffusion, turbulent mixing, stretch, kinetics, and turbulent/chemistry interactions. Early in the program,

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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
LE	laser extinction
LIF	laser induced fluorescence
LII	laser induced incandescence
PAH	polycyclic aromatic hydrocarbons
TSI	threshold soot index
ϕ_{jet}	jet equivalence ratio
m-X	<i>m</i> -xylene/ <i>n</i> -dodecane mixture
C ₇	<i>n</i> -heptane/ <i>n</i> -dodecane mixture
i-C ₈	<i>iso</i> -octane/ <i>n</i> -dodecane mixture
MCH	methylcyclohexane/ <i>n</i> -dodecane mixture
C ₁₂	pure <i>n</i> -dodecane

the systematic increase in complexity was extended by the addition of experiments to study the effects of the test fuels on emissions from a turboshaft gas turbine engine (military designation T-63) using the test fuels designed for this program. Thus, the test devices in the overall program systematically cover key chemical and physical processes from devices in which chemical kinetics are the controlling process to a small gas turbine.

The studies reported in this paper were conducted in laminar co-flow flames to obtain data on the effects of fuel composition on aromatic species and soot under non-premixed and partially-premixed conditions, both of which occur within a gas turbine combustor. Laminar flames were chosen to minimize effects of turbulence–chemistry interactions, which could mask the direct chemical effects of changes in fuel composition. Experiments were conducted to obtain two-dimensional (2-D) images of laser-induced fluorescence (LIF) from aromatic species, collected over two wavelength ranges, and laser-induced incandescence (LII) from soot. The aromatic species are of interest in this study not only because they are key precursors to soot, but also because they are among the hazardous air pollutants emitted by military gas turbine engines [4,5]. In addition to the optical measurements, temperature measurements were made using thermocouples.

The test fuel set included five fuels. *n*-dodecane was the base fuel for all experiments; it was also used to test consistency of results throughout the study. In order to determine the effect of changes in chemical composition expected from the use of alternative fuels, four binary fuel mixtures were prepared containing *n*-dodecane and a compound representing one of the fuel major hydrocarbon classes found in “drop-in” alternative aviation fuels produced from non-petroleum sources – aromatic, normal paraffin, branched paraffin, and cyclo-paraffin. Colket et al. [6] define “drop in” aviation fuels as “composed solely of hydrocarbons (HCs), but produced from alternative sources such as bio-derived feedstocks.” These four hydrocarbon classes are also major constituents of conventional aviation fuels; however, in alternative aviation fuels, their concentrations may be substantially different than in petroleum-based fuels. The design of the study resulted in a very large amount of data, too much to be summarized effectively in a single publication. Therefore, the results were divided into two papers. This paper focuses on the non-premixed flames and includes detailed discussion of the experimental methods. The companion paper [7] focuses on the sets of data for the partially-premixed fuel jets, $\phi_{\text{jet}} = 24$ and 6, and includes an abbreviated discussion of the experimental methods.

2. Related work from previous studies

Previous studies in laminar diffusion flames have provided important insights into the fundamental processes related to soot formation occurring in practical combustion systems. These flames provide an environment that includes the key processes of soot formation, growth, and oxidation in a system with little fluid mechanic coupling. Furthermore, a number of researchers have simulated these flames in order to develop and validate detailed gas phase chemistry and soot models. In addition, the laminar flames permit easy access for use of diagnostics methods for species, temperature, and soot measurements. Research conducted in laminar diffusion and premixed flames has studied effect of pressure for e.g., [8–11], fuel molecular structure for e.g., [12], diluents for e.g., [13,14], and additives for e.g., [15–17] on the soot volume fraction. The present study investigates the effect of fuel molecular structure on aromatics and soot under non-premixed conditions, using high molecular weight fuels. This study is one of the first of its kind to study the effects of chemical structure using pre-vaporized high molecular weight fuels. In the remainder of this section, selected previous studies of the effects of fuel structure on soot and soot precursors are reviewed.

Calcote and Manos [18] studied the effect of molecular structure in C₂–C₁₆ fuels on soot formation in both premixed and diffusion flames. They developed the threshold soot index (TSI), ranking fuels from 0 (least sooting) to 100 (highest sooting), to provide a comparison of sooting tendency of different fuels. In diffusion flames, they found TSI increased with the increase in the number of the carbon atoms. The aromatics fuel had higher TSI as compared to the paraffin fuels for diffusion flames.

Olson et al. [12] measured TSI, soot concentration, and corresponding emission temperatures at half the total smoke point flame heights for 42 pure hydrocarbons in a laminar diffusion flame using a wick-fed burner at atmospheric pressure. They also compiled results from previous studies and reported data for 103 pure hydrocarbons. They found alkanes had lower sooting tendency as compared to the aromatics fuels. Furthermore, maximum soot volume fraction at flame half-heights for aromatic fuels was twice as large as compared to the alkanes. These studies began to quantify how fuel molecular structure affects sooting propensity of a fuel. McEnally and Pfeifferle [19] developed yield sooting indices (YSI) for 6 cycloaliphatics and 62 aromatics in a coflow methane/air non-premixed flame. The YSI work has been extended for oxygenated hydrocarbons [20], nonvolatile aromatic hydrocarbons [21], and unsaturated esters [22]. In these studies, they found YSI correlated well with TSI and they found YSI strongly dependent on the fuel molecular structure.

Similar to the investigation undertaken in the present study, Lemaire et al. [23] used two binary mixtures, Surrogate 1 with 70% *n*-decane/30% α -methyl-naphthalene by volume and Surrogate 2 with 80% *n*-decane/20% α -methyl-naphthalene by volume, to match a commercial low-sulfur Diesel fuel, which contained 23% *n*-paraffin, 25% branched paraffin, 7% cycloparaffin, 17% of aromatics, 8% of esters and 20% of unknown composition in volume %. Surrogate 2 was designed to match the TSI value of the commercial fuel. The fuels were atomized using a high-efficiency nebulizer and burned in a turbulent flame. Spatially resolved data on soot and soot precursors were obtained using LII and LIF, respectively. The results indicate that the Surrogate 2 produces very similar soot and soot precursors to the commercial diesel whereas Surrogate 1, which has more aromatics in the mixture, produces higher levels of soot.

In the surrogate fuel project led by Witkowski et al. [24], diesel fuel, gasoline, jet fuel, various synthetic fuels, and surrogate mixtures were studied in the laminar co-flow diffusion flame. Both LII and extinction measurements were taken to obtain the

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