

Reduction of PAH and soot in premixed ethylene–air flames by addition of ethanol

Juntao Wu^a, Ki Hoon Song^a, Thomas Litzinger^{a,*}, Seong-Young Lee^a,
Robert Santoro^a, Milton Linevsky^a, Meredith Colket^b, David Liscinsky^b

^a Department of Mechanical and Nuclear Engineering, The Pennsylvania State University, University Park, PA 16802, USA

^b United Technologies Research Center, East Hartford, CT 06018, USA

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Abstract

Results are presented from a combined experimental and modeling study undertaken to understand the pathways by which the addition of ethanol to fuel-rich ethylene flames causes reductions in PAH and soot. The experimental work was conducted in a flat-flame burner at equivalence ratios of 2.34 and 2.64. Ethanol was added to the ethylene at two levels corresponding to 5 and 10% oxygen by weight in the fuel. Soot was measured by laser-induced incandescence calibrated with light extinction, and aromatic species were measured using laser-induced fluorescence. Modeling was based on a 1-D premixed flame model and kinetic mechanisms available in the literature. The modeling work captures the trends in aromatic species with changes in equivalence ratio and oxygen concentration in the fuel. However, the soot predictions do not match the increases observed at the higher equivalence ratio. Analysis of the modeling results for the lower equivalence ratio shows that the addition of ethanol to the ethylene reduces the aromatic species mainly by reducing the amount of carbon that is available to form precursor species.

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1. Introduction

The work presented here was conducted as part of a larger effort of a consortium of DoD laboratories, universities, and industry seeking ways to reduce particulate emissions from military gas turbine engines [1]. It has been estimated that U.S. military aircraft emit about 600,000 kg of particulate matter into the atmosphere each year [2]. All of this par-

ticulate matter (PM) is in the form of particles with diameters less than 2.5 μm . A growing body of evidence indicates that these small particles cause both health and environmental problems. To reduce the amount of particulate matter emitted from gas turbine engines, two approaches are possible: modification or redesign of the combustion system and modification of the fuel either by reformulation or additives. For existing engines, hardware retrofits are normally prohibitively expensive. Reformulation of the fuel is possible but certification of a new fuel could take years. Therefore fuel additives appear to be the preferred approach.

* Corresponding author. Fax: +1 (814) 865 4021.
E-mail address: tal2@psu.edu (T. Litzinger).

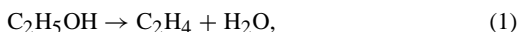
The objectives of the overall research program are to develop fundamental understanding of the complex interactions of additives with the processes that lead to PM emissions from military gas turbine engines and to use that fundamental understanding to select and investigate the most promising additives for reducing PM emissions. The overall research program involves testing in premixed and diffusion flames, a well-stirred reactor, a shock tube, an atmospheric spray burner, and a high-pressure turbulent combustion reactor. Understanding the effects of additives across this array of devices will permit a more complete understanding of the effects of additives in gas turbine combustors. The results presented here focus only on the premixed flame results.

Most additives investigated for reduction of PM emissions are metal-based [3]. However, metal additives may produce potentially harmful exhaust emissions and may adversely affect the engine itself, so organic compounds were selected for the initial phase of the study. An oxygen-containing additive was selected because of the large amount of research that has been performed to understand the potential of oxygen in the fuel to reduce emissions from diesel engines, see, e.g., Refs. [4–6]. Ethylene was selected as the fuel due to its use in many previous studies of soot formation in a variety of combustion devices. Another consideration in the selection of the fuel and oxygenated compound was the availability of chemical kinetic mechanisms for modeling the ethylene and ethanol.

No work was located in the literature that specifically investigated the effects of ethanol addition on soot from premixed ethylene–air flames. However, several studies of the effects of ethanol on PAH or soot formation in flames and shock tubes are available. The most closely related study is that of Inal and Senkan, who investigated the effect of three oxygenated additives, methanol, ethanol, and MTBE, on the formation of PAH and soot in laminar, premixed, atmospheric fuel-rich *n*-heptane (*n*-C₇H₁₆)/Ar/O₂ flames [7]. The equivalence ratio in their study was 2.10 and the oxygen weight percent in the fuel was kept at 2.7% for each *n*-heptane/oxygenate mixture. In flames containing oxygenated additives, they observed consistent reduction of low-molecular-weight hydrocarbons such as acetylene (C₂H₂), propyne (C₃H₄), diacetylene (C₄H₂), and vinylacetylene (C₄H₄). The addition of oxygenated additives also reduced aromatic species, including benzene (C₆H₆), phenylacetylene (C₈H₆), and larger aromatics such as naphthalene (C₁₀H₈) and pyrene (C₁₆H₁₀). Based on the results observed, they concluded that the reductions in PAH mole fraction in the oxygenated fuels were related to the concentrations of small unsaturated hydrocarbons and

these small hydrocarbon species must be the major precursors for the formation of PAH in flames.

Other investigators have studied the effects of ethanol addition on soot formation in shock tubes. Frenklach and Yuan investigated the effects of methanol and ethanol addition on soot formation from benzene during pyrolysis in shock tubes [8]. They studied temperatures from 1580 to 2395 K and alcohol/benzene molar ratios of 1 and 3.2, corresponding to oxygen weight percents of 14.5 and 28.3 for methanol. Their results demonstrated that while both alcohols reduced soot, ethanol was more effective. In interpreting their results, they concluded that the reduction of soot was due to the production of OH radicals that can oxidize soot precursors and soot particles, and the removal of hydrogen atoms. They emphasized that removal of H atoms plays an important role in the reduction of soot since H atoms reactivate relatively stable aromatic molecules to radicals and thus propagate the ring growth process. For ethanol the source of OH was the reaction sequence



that converts a reactive hydrogen atom to molecular hydrogen, leading to a slowdown in the formation of soot. For methanol, the OH radicals form directly from the decomposition of methanol.

On the other hand, Alexiou and Williams studied soot formation of binary mixture of toluene–methanol, toluene–ethanol, and toluene–oxygen in a shock tube and found more pronounced effects for methanol addition than for ethanol in reducing soot yield [9]. Alexiou and Williams claimed that because ethanol produces ethylene, which is a major source for acetylene formation in toluene pyrolysis, production of ethylene during ethanol decomposition offsets oxidation by OH radicals, leading to more soot formation than methanol. Their conclusion differs from that of Frenklach and Yuan, indicating that the effect of oxygenation may vary, depending on experimental conditions, flame types or parent fuel.

Finally, Ni et al. studied the effect of methanol, ethanol, 1-propanol, and 2-propanol addition to an ethylene, co-flow diffusion flame [10]. Only methanol caused substantial reductions in soot. Ethanol caused a small decrease, whereas the two propyl alcohols increased soot. Ni et al. noted that the pyrolysis of methanol would yield OH radicals that can oxidize soot precursors, whereas the pyrolysis of ethanol will yield primarily ethylene and water. Based on a comparison of the methanol results to results for methane addition to the ethylene flame, the authors noted that their results “strongly suggest that a chemical interaction is the dominant effect in soot suppression by methanol.”

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