Effect of mixing methane, ethane, propane and ethylene on the soot particle size distribution in a premixed propene flame

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ABSTRACT

The fuel mixing effect on soot formation was investigated based on a premixed propene flame, in which the C/O mole ratio was 0.6 and the maximum flame temperature was 1829 K. Different proportions of methane, ethane, propane and ethylene were mixed into the propene flame, with the constant C/O mole ratio and similar maximum flame temperatures and temperature–time histories. The particle size distribution function (PSDF) was measured in the burner stabilized stagnation (BSS) flame configuration, using the micro-orifice probe sampling technique and the scanning mobility particle sizer (SMPS). It was observed that the PSDF of the propene flame was not sensitive to methane, ethane or propane addition, while the number density of small nucleated particles was strengthened with ethylene addition. The particle growth in flame with a small amount of ethylene addition was stronger than that of either the pure propene or ethylene flame, indicating the synergistic effect between propene and ethylene on soot formation.

1. Introduction

It is well known that soot emissions from fossil fuel combustion jeopardize the environment and human health. In urban areas, particle emissions from gasoline and diesel vehicles are the primary mobile sources of particles pollution, so it is worthwhile to understand the soot formation processes in gasoline and diesel combustion. In the engine combustion processes, the large fuel molecules first decompose to such small pyrolysis fragments as ethylene, methane, propene, and it is therefore of practical significances to learn about the sooting characteristics of small-molecule hydrocarbons and their mixtures.

The synergistic effect of fuel mixtures on sooting propensity is an interesting research topic, which means that a fuel mixture produces more polycyclic aromatic hydrocarbons (PAH) or/and soot than the sum of those produced by individual components under similar temperature or/and equivalence ratio conditions. Frenklach et al. observed the fuel synergistic effects on soot formation in some binary hydrocarbon mixtures in a shock tube. Detailed kinetic modeling revealed that the synergistic effects were primarily caused by the acceleration of acetylene-addition reactions [1]. The synergistic effects were also observed in ethylene-based diffusion flames with ethane and propane addition [2, 3], methane addition [4], and dimethyl ether and ethanol addition [5]. All these results substantiated the importance of methyl radical for the formation of propargyl and the subsequent aromatics formation that relied on odd-carbon-numbered species. Trottier et al. observed a synergistic effect in some binary fuel mixtures in diffusion flames, but they emphasized the importance of even-carbon based reactions for the first aromatic ring formation [6]. Karatas et al. also found synergistic effects on soot formation in diffusion flames, however, they considered that their results were inconsistent with any existing mechanisms for the binary fuel synergistic effects explanation [7]. Similar to McEnally et al. [5], Liu et al. observed a synergistic effect when a small amount of dimethyl ether was added to ethylene in a coflow diffusion flame. Their numerical results found this was primarily due to the cyclization of l-C_6H_6 and n-C_6H_7 and to a much lesser degree due to the interaction between C_2 and C_4 species for benzene formation, rather than the propargyl self-combination reaction route [8]. In a previous study, the authors have investigated the methane-doped impacts on sooting behaviors in an ethylene-based burner stabilized stagnation (BSS) flame by evaluating the evolution of particle size distribution function (PSDF) of nascent soot [9], and found the synergistic effects with small methane-doped ratios (5% and 10%). The computational
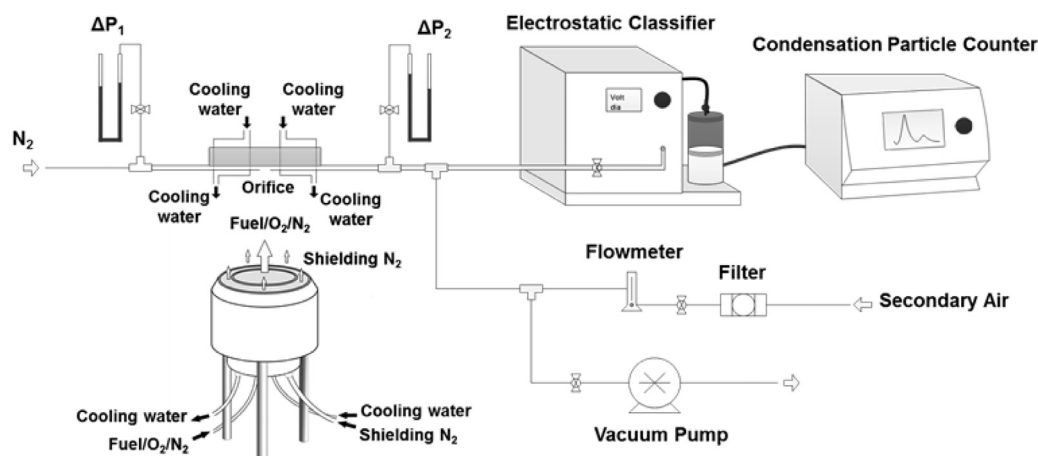
Fig. 1. Schematic of the experimental setup.

results indicated that small methane-doped amounts facilitated C_3H_3 and naphthalene formation, thus strengthening the sooting tendency.

As discussed above, the previous explanations for the synergistic effects were not consistent with each other. In most of these studies, ethylene was selected as the base fuel. In addition to ethylene, propene is another important intermediate in liquid hydrocarbons combustion, and its fuel structure and sooting propensity are different from those of ethylene [10]. It is therefore necessary to investigate the sooting characteristics of propene and its mixture with other small hydrocarbons. The objective of this study is to investigate the sooting propensity of propene-based binary fuel mixture and the synergistic effect between the binary components by measuring the detailed PSDFs in a BSS flame configuration. Different proportions of methane, ethane, propane, and ethylene were mixed into the base propene flame (M1 flame studied in [10]), while keeping the same carbon to oxygen mole ratio (C/O) in the unburned gas. In addition, similar maximum flame temperatures and temperature–time histories were also held among the test conditions, and as such we could focus on the fuel composition effects on sooting propensity. The BSS flame configuration allowed for a well-defined boundary conditions for the probe-intruded flame and enabled less ambiguous comparison between experimental and modeling results [11].

2. Experimental and computational methods

The experimental and computational methods have been described in detail in previous work [10,12]. Figure 1 schematically shows the experimental setup. Briefly, the water-cooled flame burner was composed of a bronze porous plug of 5 cm in diameter and a concentric porous ring. Premixed atmospheric fuel/oxygen/nitrogen flowed into the plug, and a shroud nitrogen flowed through the ring to isolate the flame from the ambient air. A water-cooled aluminum alloy plate served as a surface for flow stagnation and sampling, so the BSS flame formed between the burner and the stagnation plate. The separation from the burner surface to the stagnation plate was the flame height, which could be adjusted by lifting the burner. More details about the BSS flame configuration could be found in [11,13].

The axial flame temperature field was measured by a S-type coated thermocouple. The thermocouple wire and bead diameter after coating was approximately 0.014 cm and 0.028 cm, respectively. The inlet/burner surface temperature was linearly extrapolated from the measured values in the pre-flame region closest to the burner surface [13,14]. A K-type thermocouple was embedded into the bottom of the stagnation plate to measure the downstream boundary temperature of the flame.

Radiation correction of the measured temperature was made using the Shaddix procedure [15]. In the radiation correction, the local transport and flow properties were calculated iteratively by a modified OPPDIF code [16,17]. The error of the measured temperatures mainly originated from the uncertainty of the thermocouple radiation coefficient, which led to the error in temperature radiation correction. The upper and lower limits of emissivity values in literature for the coating are 0.3 and 0.6, respectively [18]. The radiation corrected flame temperature was assumed to be the average of the temperatures corrected using the two limiting emissivity values [11,13].

To obtain reliable corrections for radiation heat loss, the gas physical properties were determined by a detailed simulation. The inlet and stagnation surface boundary conditions were used to solve the flame temperature and species concentration profile by the quasi one-dimensional OPPDIF code [11,13]. The burner-to-stagnation surface separation was smaller than the burner diameter thus the formulation introduced by Smooke could be employed [16]. The reaction kinetic model used was USC Mech II consisting of 111 species and 784 reactions [19].

A stainless-steel tube was embedded in the stagnation plate and the sampling orifice on the tube was 0.13 mm in diameter. The outer diameter and wall thickness of the sampling tube were 6.35 mm and 0.127 mm, respectively. The soot sample was drawn through the orifice into the tube and immediately diluted by high speed nitrogen flow (30 L/min at 298 K and 1 atm). A small fraction of the diluted sample flowed into the scanning mobility particle sizer (SMPS) system, consisting of a 3080 Electrostatic Classifier (EC) and a 3776 ultrafine Condensation Particle Counter (CPC). The EC was composed of a 3085 nano-Differential Mobility Analyzer (DMA) and a Kr-85 Neutralizer. With the scan time of 50 s up and 10 s retrace, particle size distribution was obtained for the diameter range from 2.5 nm to 66.1 nm.

The dilution ratio was determined as a function of pressure drop across the sampling orifice and calibrated by the bubble flowmeter method [11,13,20–22]. The pressure drop was controlled by regulating the secondary air flow and measured by two U-type water manometers at the upstream and downstream of the sampling orifice. For each flame studied, the optimum dilution ratio was where the dilution-corrected PSDF was insensitive to the dilution ratio, and the optimum dilution ratio ranged from about 600 to 1800 in this study. More information for the dilution ratio effects on the PSDFs could be found in the authors' previous work [12].