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## Partial premixing effects upon soot nanostructure

Chung-Hsuan Huang<sup>a,b</sup>, Randy L. Vander Wal<sup>a,b,\*</sup><sup>a</sup>The John and Willie Leone Family Department of Energy and Mineral Engineering, Penn State University, University Park, PA 16802, USA<sup>b</sup>The EMS Energy Institute, Penn State University, University Park, PA 16802, USA

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## ABSTRACT

Partial premixing within simple gas jet diffusion flames has a very long history – reaching back to the Bunsen flame. Yet HRTEM data of soot from such flames appears absent. Using benzene as the primary fuel with partial premixing tested the chemical path for C<sub>5</sub> production – proceeding through partial benzene oxidation yielding the phenoxy radical followed by CO loss to produce C<sub>5</sub>. A strong variation of lamellae curvature with oxygen content in the primary fuel stream was observed – reflecting the increasing C<sub>5</sub> production rate. Generality of the nanostructure dependence upon partial premixing and associated change in gas phase chemistry (compared to pure thermal pyrolysis) was demonstrated using an ordinary laboratory Bunsen burner with ethylene as fuel. Absent partial premixing, soot production is well described by the HACA mechanism, yielding benzenoid aromatics and soot nanostructure consisting of flat lamellae, without curvature, dissimilar to observations described here accompanying partial premixing.

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## 1. Introduction

Recent flame studies have shown that soot can possess profound differences in nanostructure depending upon its formation chemistry [1,2] and temperature history [3–5] with implications for oxidation [3,6–8]. By measurements and modeling, soot chemistry varies with fuel [9]. Russo et al. [1] recently found soot H/C to be highly dependent upon the parent fuel using ethylene versus methane in premixed sooting flames at similar equivalence ratio and temperature conditions. Additionally the aliphatic content was observed related to the growth history and chemical environment. In other highly detailed studies, applying HRTEM, elemental analysis, oxidative thermo-gravimetric analysis, and UV–vis spectroscopy, Alfe et al. [3] showed dependence of soot nanostructure upon fuel, and structure–property relationships between soot nanostructure and “bulk” properties. Apicella et al. [4] found that nanostructure evolved toward larger size aromatic systems throughout the soot formation region in premixed benzene and ethylene flames. Moreover the nanostructure order depended upon fuel, as did the soot H/C content. In prior studies Russo et al. [5] also found the H/C ratio dependent upon fuel, using premixed flames of methane, ethylene and benzene.

In engine studies, several different researchers have characterized nanostructure, identified its dependence upon fuel, engine or

combustion conditions, and its impact upon oxidation. For example, Boehman et al. [10] compared soot nanostructures of particulates produced from different fuels in a commercial direct injection (DI) diesel engine using HRTEM to show the potential impact of biodiesel blending on the low-temperature oxidation. Subsequent studies expanded upon the relationship between nanostructure and reactivity by comparison of biodiesel (Song, Lapuerta) and Fischer–Tropsch diesel fuel [11,12], impact of EGR [13], and engine load [14], speed and torque [15], Vander Wal first illustrated the impact of nanostructure upon reactivity [16], with illustration of its dependence upon fuel and formation conditions [17], extending this to oxygenated fuels [18] and also its impact upon reactivity [19]. Recently the impact of biodiesel fuel in a diesel engine [20] and of turbulence induced mixing (in a jet engine) upon nanostructure [21] was shown using HRTEM to identify fullerene nanostructure in the emitted PM.

Why is curvature, i.e. fullerene nanostructure of interest? As hypothesized it relates to the fuel air stoichiometry, namely the equivalence ratio  $\phi$  that is of great practical relevance. In diesel and gas turbine engines fuel atomization and vaporization produce fuel-rich regions that mix with air (concurrent with evaporation) leading to partial premixing. This study sought to test the path towards curvature and show the generality of partial premixing, using simple gas-jet flames to identify the origin of curvature and demonstrate the relevancy of partial premixing to nanostructure in soot.

In this study two flames were investigated to test the relation between C<sub>5</sub> chemistry and soot nanostructure. To identify limits,

\* Corresponding author at: The John and Willie Leone Family Department of Energy and Mineral Engineering, Penn State University, University Park, PA 16802, USA.  
E-mail address: [ruv12@psu.edu](mailto:ruv12@psu.edu) (R.L. Vander Wal).

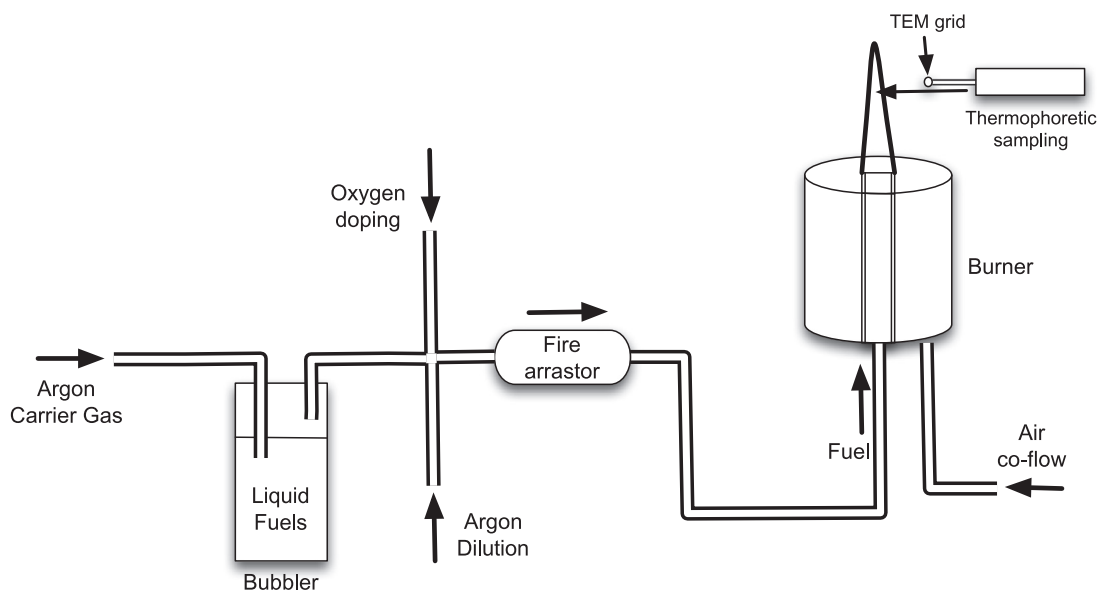


Fig. 1. The schematic of partially premixed diffusion flame system with Santoro-style burner.

soot from a partially premixed diffusion flame with benzene as fuel was studied for comparative nanostructure. Second, an ordinary Bunsen flame fueled with ethylene ( $C_2$ ) tested the generality of the  $C_5$  production by partial premixing, with evidence again being production of curvature in soot nanostructure.

## 2. Experimental

Bunsen flames are well known examples of partially premixed flames, typically stabilized on an open tube, as a gas jet. Variable premixing was introduced in a Bunsen flame configuration in order to stabilize the flame. The versatility of such flames is their stoichiometry. The relevancy of Bunsen flames to combustors and engines is that fuel sprays establish a diffusion flame that becomes partially mixed, the degree depending upon location and time [22]. Different fuels were used in this flame configuration with results presented below.

To test the oxidative pyrolysis chemistry leading to  $C_5$  formation, a gas jet diffusion flame was used with varied levels of partial premixing using benzene and oxygen. Given the high sooting propensity of benzene; nitrogen dilution was used to manage the soot load and maintain a close-tipped flame. Clearly the varied oxygen levels will inherently change the adiabatic flame temperature, but by nitrogen substitution, the same flame temperature may be maintained while keeping fuel concentration roughly constant [23].

Note that partial premixing is a term best applied for lower levels of oxygen, such as in diffusion flames with  $\phi > 3$ ; levels for which burner stabilized premixed flames are not operable. Streamlines in a buoyantly driven diffusion flame are convergent and non-monotonic in temperature and mixture fraction, with the exception of the center streamline. Guided by precedence of previous studies illustrating the sequence of soot inception and growth along this path, sampling of so-called mature soot was from this central axis with sampling approximately 5 mm below the visible flame tip.

### 2.1. Partially premixed diffusion flame

To investigate the effect of fuel/air partial premixing ratio on soot nanostructure, a co-flow burner with a central

Table 1

Tested fuels and the corresponding fuel/air equivalence ratios.

Fuels	Pure	10% $O_2$	20% $O_2$
$C_6H_6$	$\phi = \infty$	$\phi = 5.2$	$\phi = 10.4$

fuel jet (I.D. = 11.1 mm) surrounded by a co-flowing air stream (I.D. = 101.6 mm) Santoro-style burner was applied to produce the diffusion flame for soot particle formation and collection. In this study, the total flow rate of the central fuel jet was set at 300 sccm as the combination of methane base flame (to maintain main flame stability), dosed fuel and added oxygen. The co-flow was set at 43 slm air to stabilize the center diffusion flame. Benzene was fed by using argon as the carrier gas through a bubbler at ambient ( $20^\circ C$ ) temperature. The methane ( $CH_4$ ) and argon (dilution) mixture ratio was 1:1 to create non-sooting diffusion base flame (light blue flame). Oxygen at 0%, 10% and 20% (v/v) was added to the argon-fuel stream while maintaining the total flow rate at 300 sccm, thereby maintaining similarity of flame height. The tested fuel/air equivalence ratios are summarized in Table 1. Temperatures were not measured in the sooting region of these flames, but adiabatic estimates of peak flame temperature for these three cases were within 200 K of each other [24]. True values are considerably less, due to strong flame (soot) radiation [25,26].

Soot particles were directly collected within the close-tip diffusion flame by a thermophoretic sampling device with 300 mesh lacy carbon grid for further soot nanostructure HRTEM analysis. Thermophoretic sampling methodology has been well described and has a several-decade history. Briefly, the device was driven by a  $N_2$  cylinder, and to ensure one cycle sampling period can be done within  $50 \mu s$ , 25–30 PSI gas pressure was applied to the device, with further details and demonstrations presented elsewhere [27,28]. To avoid further oxidize collected-soot, only one sampling cycle was applied to sample collection. The soot samples were collected at the central axis of the flame at the location of peak soot. The spatial resolution is defined by the central 2-mm portion of the TEM grid, where variations in aggregate structure or primary particle size were not visibly evident during examination. The overall schematic of the flame system is shown in Fig. 1.

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