



# Soot particle size measurements in ethylene diffusion flames at elevated pressures



Scott A. Steinmetz<sup>a,\*</sup>, Tiegang Fang<sup>b</sup>, William L. Roberts<sup>a</sup>

<sup>a</sup> Clean Combustion Research Center (CCRC), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

<sup>b</sup> Department of Mechanical and Aerospace Engineering, North Carolina State University, Raleigh, NC 27606, USA

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## ABSTRACT

Soot particle size is investigated in laminar nitrogen-diluted ethylene coflow diffusion flames at 4, 8, 12 and 16 atm. Line of sight attenuation and scattering are used to measure two-dimensional soot volume fraction and particle size fields for the first time at elevated pressures. Soot volume fraction dependence on pressure is consistent with the observations of similar studies, scaling approximately with the square of pressure. Scattering intensity is analyzed through Rayleigh and Rayleigh–Debye–Gans polydisperse fractal aggregate theories to provide two estimates of particle size. An increase in overall particle sizes with pressure is found, consistent with similar one-dimensional studies. Particle diameters in the annulus of the flame increase faster with pressure than those on centerline. Contrary to previous studies, the dependence of particle size on pressure was found to taper off between 8 and 12 atm, with little observed growth beyond 12 atm. The measurements provide additional data for one of the International Sooting Flame (ISF) workshop's target pressurized flames.

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## 1. Introduction

One consequence of hydrocarbon based power production is the formation of soot. In addition to being detrimental to some combustion devices, soot is known to be harmful to the environment and to human health [1,2], and a better understanding of how it is formed and oxidized is key to reducing its net emission. Diesel and gas turbine engines are major contributors to global soot production, and while these devices operate at elevated pressures, soot studies are frequently carried out in atmospheric pressure conditions. However, pressure is an important parameter controlling soot formation and oxidation. There are a number of high pressure diffusion flame studies in the literature, and a comprehensive summary is given in [3]. As the turbulent jet flames of practical combustion devices are unsteady and difficult to probe, more tractable laminar coflow diffusion flames are typically studied, and the laminar flamelet concept allows our understanding of laminar flames to be applied to turbulent flames. These studies demonstrate a power-law dependence of soot yield on pressure, though with poorly understood scaling.

An important characteristic of soot that must be understood is its morphology, including distributions of primary particle size

( $d_p$ ), number of primary particles per aggregate ( $N$ ), and aggregate structure. New EURO 6 regulations specify both particulate mass and number densities, and therefore particle size. There are numerous challenges and uncertainties associated with determining morphology at elevated pressures [3–5]. Light scattering is frequently used for particle size measurement in atmospheric conditions [6–10], but optical access from several angles is usually necessary to determine aggregate properties. Time-resolved laser induced incandescence is able to resolve two-dimensional soot concentration and primary particle size, even in unsteady flows, but suffers from uncertainties in the effect of pressure on the rates of soot heating and cooling [5].

There is limited experimental information on particle sizes in high pressure diffusion flames, with this information frequently coming from engine studies [11,12]. However, Flower and Bowman [13] investigated soot size in a Wolfhard–Parker burner, though only to 2.5 atm, finding that particle sizes increase with pressure. Thomson et al. [4] determined effective soot particle size in methane–air flames from 5 to 40 bar using laser induced incandescence, at one flame height, finding a significant increase in particle size with pressure. Kim et al. [14] investigated diluted ethylene–O<sub>2</sub> flames up to 8 atm, finding an increase in centerline primary particle size with pressure through extractive sampling and transmission electron microscopy (TEM). These studies show the effect of pressure on particle sizes to be a function of axial and radial position in the flame, but spatially resolved high pressure

\* Corresponding author.

E-mail address: [scott.steinmetz@kaust.edu.sa](mailto:scott.steinmetz@kaust.edu.sa) (S.A. Steinmetz).

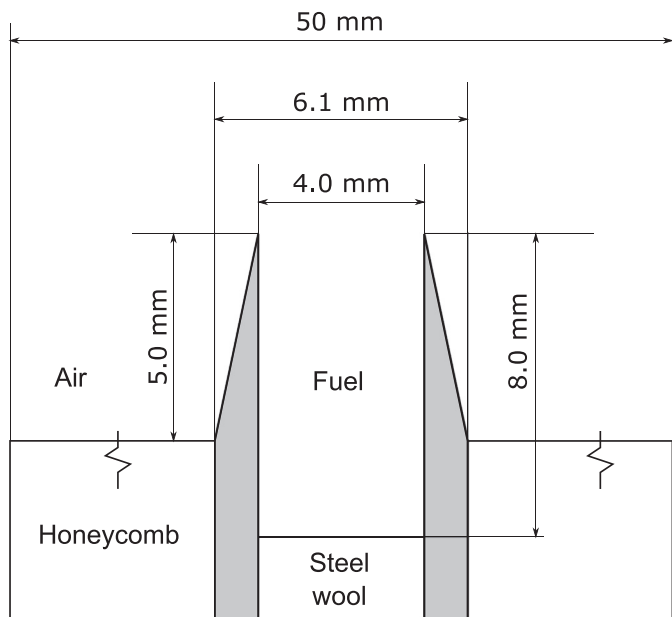


Fig. 1. Coflow burner exit geometry.

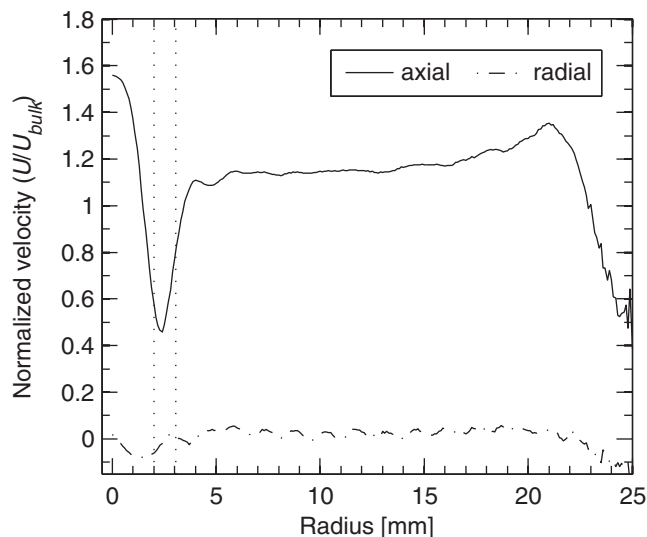


Fig. 2. Axial and radial mean velocity profiles of the cold flow, measured 1 mm above the nozzle outlet, with  $Re$  of the nozzle = 153, and  $Re$  of the coflow = 1980. Velocities are normalized by the corresponding bulk velocity. The dotted lines indicate the inner and outer radii of the nozzle.

information is still lacking. There have been several studies on the effect of pressure on particle sizes in premixed flames or shock tubes [15–19]. These flat-flame studies show an increase in particle size with pressure up to 10 bar, but a plateau or decrease in size at even higher pressures, while number density increases.

The objective of this work is to measure the effect of pressure on the two-dimensional fields of soot particle sizes in laminar coflow flames. This will add particle size information to the existing dataset [20–22] of one of the target pressurized flames for the International Sooting Flame (ISF) Workshop [23]. Line of sight attenuation (LOSA) is used to determine soot volume fraction, and laser scattering is measured to determine average soot particle size.

## 2. Experimental theory and method

### 2.1. Theory

Numerous sources in the literature can be referenced on the techniques used [6–10], but a brief description is given. Beer's law relates the transmissivity of light to the integral of the local extinction coefficient,  $\kappa_e$ , over the path length of an absorbing medium. Assuming extinction occurs only within the axisymmetric flame, local  $\kappa_e$  can be determined by tomographic inversion. The local extinction coefficient is related to soot volume fraction,  $f_v$ , using Rayleigh–Debye–Gans (RDG) theory

$$f_v = \frac{\kappa_e \lambda}{6\pi(1 + \rho_{sa})E(m)} \quad (1)$$

where  $m$  is the soot refractive index,  $E(m)$  is the imaginary soot refractive index function,  $\lambda$  is the wavelength of light, and  $\rho_{sa}$  is the

ratio of scattering ( $\kappa_s$ ) and absorption ( $\kappa_a$ ) coefficients. The optical properties of soot ( $m$ ,  $\rho_{sa}$ ) are not well known. In general, they are functions of morphology, particle diameter,  $d$ , and  $\lambda$ , and will vary throughout a flame due to differences in residence time, temperature, and chemistry [24]. As there has been very little investigation on soot morphology and optical properties at elevated pressures, values typical of atmospheric in-flame soot were used. Soot optical properties are assumed to be  $\rho_{sa} = 0.26 \pm 0.05$ ,  $E(m) = 0.37$ , and  $F(m) = 0.65 \pm 0.17$  based on dimensionless extinction coefficient measurements in ethylene coflow flames [24], with the uncertainties estimated based on the ranges of values measured in [24].

By measuring the intensity of vertically polarized light scattered from a vertically polarized incident beam,  $Q_{vv}$ , and assuming the particles are within the Rayleigh regime ( $d \ll \lambda$ ), an average weighted primary particle diameter can be found from the sixth to third moment ratio of particle probability functions [6,7] and is given by

$$D_{63} = \lambda \left( \frac{4 E(m) Q_{vv}}{\pi^2 F(m) \kappa_a} \right)^{\frac{1}{3}} \quad (2)$$

In addition to being biased to larger particles, a major limitation of this Rayleigh analysis is the neglecting of aggregation, thereby assuming that light is scattered by individual soot spherules. Research has shown that this limitation results in large overestimates of primary particle diameters [10]. For a more representative analysis, RDG polydisperse-fractal-aggregate (RDG-PFA) theory is used, assuming  $\pi d|m-1| \ll \lambda$ . The modulus of the scattering wave vector,  $q$ , can be found from  $q = 2k \sin \theta/2$ , where  $k = 2\pi/\lambda$ . Its inverse,  $q^{-1}$ , represents a length scale of the scattering

Table 1

Flow parameters.

| $P$<br>(atm) | $\dot{m}_{C_2H_4}$<br>(mg/s) | $\dot{m}_{N_2}$<br>(mg/s) | $\dot{m}_{air}$<br>(g/s) | $U_{noz}$<br>(cm/s) | $U_{cof}$<br>(cm/s) | Ratio | $Re_{noz}$ | $Re_{cof}$ | $Fr_{noz}$ | $T_{noz}$<br>(°C) | $T_{cof}$<br>(°C) |
|--------------|------------------------------|---------------------------|--------------------------|---------------------|---------------------|-------|------------|------------|------------|-------------------|-------------------|
| 4            | 1.37                         | 6.41                      | 1.25                     | 13.3                | 13.3                | 1.0   | 153        | 1980       | 0.143      | 62                | 21                |
| 8            | 1.37                         | 6.41                      | 2.51                     | 6.6                 | 13.3                | 2.0   | 153        | 3960       | 0.071      | 59                | 20                |
| 12           | 1.37                         | 6.41                      | 4.04                     | 4.4                 | 14.2                | 3.2   | 153        | 6350       | 0.048      | 56                | 20                |
| 16           | 1.37                         | 6.41                      | 4.42                     | 3.3                 | 11.7                | 3.5   | 153        | 7000       | 0.036      | 52                | 20                |

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