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Numerical and experimental study of soot formation in laminar diffusion flames burning simulated biogas fuels at elevated pressures

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ABSTRACT

The effects of pressure and composition on the sooting characteristics and flame structure of laminar diffusion flames were investigated. Flames with pure methane and two different methane-based, biogas-like fuels were examined using both experimental and numerical techniques over pressures ranging from 1 to 20 atm. The two simulated biogases were mixtures of methane and carbon dioxide with either 20% or 40% carbon dioxide by volume. In all cases, the methane flow rate was held constant at 0.55 mg/s to enable a fair comparison of sooting characteristics. Measurements for the soot volume fraction and temperature within the flame envelope were obtained using the spectral soot emission technique. Computations were performed by solving the unmodified and fully-coupled equations governing reactive, compressible flows, which included complex chemistry, detailed radiation heat transfer and soot formation/oxidation. Overall, the numerical simulations correctly predicted many of the observed trends with pressure and fuel composition. For all of the fuels, increasing pressure caused the flames to narrow and soot concentrations to increase while flame height remained unaltered. All fuels exhibited a similar power-law dependence of the maximum carbon conversion on pressure that weakened as pressure was increased. Adding carbon dioxide to the methane fuel stream did not significantly effect the shape of the flame at any pressure; although, dilution decreased the diameter slightly at 1 atm. Dilution suppressed soot formation at all pressures considered, and this suppression effect varied linearly with CO_2 concentration. The suppression effect was also larger at lower pressures. This observed linear relationship between soot suppression and the amount of CO_2 dilution was largely attributed to the effects of dilution on chemical reaction rates, since the predicted maximum magnitudes of soot production and oxidation also varied linearly with dilution.

1. Introduction

Virtually all practical combustion devices burn high carbon-content fossil fuels such as coal, petroleum and natural gas. However, conventional sources of petroleum and natural gas are rapidly declining [1]. Additionally, fossil fuel combustion is responsible for nearly all of the anthropogenic emissions of nitrogen oxides (NO_x), carbon dioxide (CO_2), carbon monoxide (CO), soot, aerosols, and other chemical species that are harmful to human health and the environment. Gaseous biofuels, or biogas, are an attractive option to replace fossil fuels since they are environmentally friendly and can be produced locally [2]. They are also renewable, biodegradable, and generate exhaust gases of acceptable quality [3].

Biogases are produced in a variety of environments such as landfills, waste water treatment plants and biowaste digesters [4]. They typically consist of significant concentrations of methane (CH_4), carbon dioxide (CO_2) and nitrogen (N_2). Biogases are of particular interest because of their significant concentrations of CO_2 and/or N_2, both of which suppress soot formation in pure hydrocarbon flames [5–10]. The addition of inert gases such as CO_2 and N_2 to pure hydrocarbons reduces soot formation by reducing concentrations (dilution effect) and flame temperatures (thermal effect) [10–13]. Carbon dioxide also plays a chemical role by participating in reactions related to soot formation, providing an additional mechanism to suppress soot formation [11,12].

Most practical combustion devices, such gas turbine combustors and diesel engines, employ high-pressure turbulent flames. These types of flames are not easily characterized because of experimental limitations related to optical accessibility [14], complex flame geometries, and the vast range of time and length scales. As such, laminar flames with simple configurations are commonly studied. However, there are relatively few detailed fundamental studies on soot formation in laminar flames of biogases

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or fuels with similar compositions [5,11,12,15–19]. Furthermore, these studies were all carried out under atmospheric pressure, which does not accurately represent the conditions inside practical combustion equipment.

Pressure profoundly influences the structure and sooting characteristics of laminar diffusion flames through its effects on buoyant forces and chemical kinetics [9,20]. Since the buoyant acceleration scales with pressure-squared, increasing pressure drastically alters the shapes and sooting characteristics of flames [14,21–28]. As such, systematic fundamental studies of simple, small-scale premixed and non-premixed laminar flames are essential in order to develop the accurate physical models necessary to study high-pressure turbulent flames. The knowledge and detailed modeling of these laminar biogas flames, for which the full range of scales can be resolved, serves as a basis for the development of more practical turbulent combustion models.

Recently, several studies have focused on the effects of diluents on processes relevant to soot formation at elevated pressures. Yelverton and Roberts [29] investigated the effect of various diluents – N_2, Ar, He and CO_2 — on the smoke point heights of laminar methane- and ethylene–air flames between 1 and 8 atm. They found that smoke point heights increased with dilution at atmospheric pressure, but were insensitive to dilution at elevated pressures. The study also emphasized a diluent's effect on entrainment and mixing via changes in kinematic viscosity, which is more important in some cases than its effect on the heat capacity or chemical kinetics. Abhinavam Kailasanathan et al. [30] extended this study by measuring the effects of the same diluents on soot precursor formation and temperature in laminar ethylene–air diffusion flames at similar pressures (i.e., 1–8 atm). The study confirmed the superior soot suppression qualities of CO_2 as compared with the other diluents, even at elevated pressures. However, no measurements of soot concentrations were made in either of the two studies, and the maximum pressure considered was only 8 atm. Practical combustion devices such as gas turbine combustors or diesel engines operate at much higher pressures.

In the present study, the effects of composition and pressure on the structure and sooting propensity of methane-based, biogas-air laminar coflow diffusion flames were investigated. In particular, two different simulated biogas mixtures were examined through a combination of experimental and numerical means and compared with previous results obtained for pure methane–air flames [28,31]. Pressures ranging from 1 to 20 atm were considered.

2. Experimental methodology

The experimental apparatus, described in detail elsewhere [24,31–33], consists of a coflow burner installed inside a pressure vessel. It was designed to allow the burner operating pressure to be varied independently of the surrounding ambient conditions. The burner consists of an inner stainless steel fuel tube with a 3 mm inner diameter and an outer concentric air tube with a 25.4 mm inner diameter. The outer surface of the fuel tube was chamfered to form a knife edge at the nozzle exit plane, which was necessary to improve flame stability over a wide range of pressures. A chimney was also installed to improve flame stability by shielding the core flow from disturbances created inside the chamber.

The spectral soot emission (SSE) diagnostic technique was used to construct radial profiles of temperature and soot volume fraction at different axial heights along the burner axes [34]. In SSE, line-of-sight emission from soot is first measured along chords through the flame at various heights, and radially resolved emission rates are obtained using an Abel inversion procedure [35]. Temperature and soot volume fraction are then computed from these emission rates. Details of the inversion process and the theory applied to obtain temperature and soot volume fraction from the line-of-sight measurements are described by Snelling et al. [34].

In the current diagnostic setup, the flame was imaged using an achromatic doublet lens with a focal length of 300 mm and an *f*-number of *f*/48, positioned to provide a 1:1 magnification. It was imaged onto the entrance of a spectrometer and the output was focused onto a 16-bit charge-coupled device (CCD) detector (1340 by 400 pixels). The entrance of the spectrometer contains two slits: a vertical slit 25 μm in width, and a horizontal slit 290 μm in height. The apparatus has a horizontal and vertical spatial resolution of 70 and 290 μm, respectively. Soot emission was measured over the wavelength range from 690 to 945 nm. More details of the experimental setup are provided in [24,31–33].

A majority of the uncertainty in the experimental measurements for soot volume fraction and temperature result from assumptions that were made about the optical properties of soot, i.e., the dimensionless soot refractive index function, $E(m_\lambda)$, where m_λ is the complex refractive index of soot at the wavelength λ. The magnitude and variation of this function with λ must be known to estimate the soot volume fraction and temperature from the flame's emission [34]. Although there is a considerable amount of information about the optical properties of soot (see, for example, [36–38]), there is no real consensus on the topic [39]. Snelling et al. [34] compared SSE measurements for an ethylene diffusion flame with two-dimensional line-of-sight light attenuation (LOSA) measurements for soot concentration and coherent anti-Stokes Raman spectroscopy (CARS) measurements for temperature, and found that a constant refractive index function, $E(m_\lambda) = 0.26$, provided the best agreement. These authors demonstrated that changing $E(m_\lambda)$ from a constant function to a linear one that increased at a rate of 40%/μm resulted in a 3% increase in temperature and a 30% decrease in soot concentration. This represents an extreme case, since a linear regression of the experimental data for $E(m_\lambda)$ published by Krishnan et al. [38] yields a trend line with only 5%/μm variation in $E(m_\lambda)$. Here, a constant function with $E(m_\lambda) = 0.274$ was chosen based on the recommendations of Thomson et al. [24].

An uncertainty analysis was conducted by Thomson et al. [24] for a similar experimental setup. Based on this analysis, the uncertainty of the temperature and soot volume fraction measurements are 3.5% and 35–40%, respectively, both with a 95% confidence interval. This was confirmed for the current experimental apparatus by Karataş et al. [40]. More details of the uncertainty analysis for the SSE measurements are provided in [41].

Flames of two different methane/carbon dioxide biogas mixtures were investigated, hereafter referred to F20 and F40, and compared with pure methane flames, hereafter referred to as F0. The methane flames were previously studied by Joo and Gülder [31] and Charest et al. [28] over a range of pressures between 1 and 60 atm. Table 1 lists the compositions and total fuel mass flow rates for the three fuels.

For all the flames, constant mass flow rates for methane and air of 0.55 mg/s and 0.2 g/s were maintained, respectively. CO_2 was added to the methane fuel in the F20 and F40 flames, but the methane flow rates were not changed. Pressure varied between 1 and 20 atm; experiments were performed at 1, 5, 10, 15 and 20 atm. Experimental measurements for soot volume fraction and temperature were obtained in height increments of 0.5 mm and radial increments of 50 μm. However, because of low soot levels at lower pressures, reliable measurements could only be made by the SSE system at pressures of 5 atm and above in the F20 flames and 10 atm and above in the F40 flames. The SSE diagnostic technique relies on radiation emitted by soot only. Thus, measurements cannot be made in non-sooting flames. Measurements for the F20 and