Full Length Article

Effects of carbon dioxide and nitrogen addition on soot processes in laminar diffusion flames of ethylene-air at high pressures

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ABSTRACT

An experimental assessment of the influence of carbon dioxide and nitrogen dilution on sooting characteristics of laminar ethylene diffusion flames at pressures up to 20 atm is presented. Two dilution rates, defined as the ratio of mass flow of the fuel to that of the diluent gas, of 1:2 and 1:3 were used at all pressures with a fixed ethylene mass flow rate. A wider range of nitrogen dilution, from 1:1 to 1:4, was investigated at 10 atm. In the pressure range of interest and with the mass flow rates of fuel and diluents, resulting flames were stable and nonsmoking. Spectrally-resolved line-of-sight soot radiation measurements were obtained to infer the radial soot and temperature distributions within the flame envelope through an Abel inversion process. The sooting propensity, in terms of maximum soot yield, was found to be significantly lower with carbon-dioxide dilution in the pressure range of 1–15 atm but approached to comparable values to those with nitrogen-dilution at 20 atm. The implication of this finding is that the chemical suppression effect of carbon dioxide dilution, which was proven at atmospheric pressure previously, exists also at elevated pressures up to 15 atm and becomes relatively small at higher pressures. Variation of the maximum soot yields with pressure indicated that carbon dioxide-diluted flames show a relatively stronger dependence to pressure as compared to nitrogen-diluted flames. Temperatures decreased with increasing pressure as expected due to increasing radiative heat loss, and the peak temperatures were observed near the flame tips as a result of the heat release from soot oxidation.

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1. Introduction

Understanding the effects of carbon dioxide, CO_2, dilution is critical in pollutant control schemes such as multi-stage combustion and exhaust gas recirculation as well as in the oxyfuel combustion technology for CO_2 control, see, for example [1–4]. The influence of an additive on soot formation in diffusion flames could be any combination of the following effects [5]: (1) an effect due to dilution which changes the carbon composition of the fuel mixture, (2) a thermal effect resulting from the change in the physical properties of the fuel mixture, and (3) a chemical effect due to active chemical involvement of the additive. The results from earlier studies with carbon dioxide-diluted ethylene flames were contradictory regarding the nature of the effect CO_2 dilution on soot. The findings of Schug et al. [6] implied a purely thermal effect of CO_2 dilution, whereas McLintock [7] concluded that the soot oxidation is enhanced chemically by CO_2. Chemical suppression effects of CO_2 was confirmed by two experimental studies that were designed to separate the dilution, thermal, and chemical effects in laminar diffusion flames of ethylene [8,9]. It was argued that CO_2 dilution changes the concentrations of radicals such as O and OH, which are thought to be important in soot oxidation.

To isolate the chemical effect of CO_2, Liu et al. [10] compared CO_2 dilution to the dilution by chemically inert CO_2. The reaction $CO_2 + H \rightarrow CO + OH$ was identified as the culprit for the chemical effect of CO_2 dilution [10]. Oxidation of soot precursors and nascent soot was found to be enhanced by the increased O and OH radicals upon CO_2 dilution. Further, chemical activity of CO_2 leads to lower flame temperatures slowing the soot nucleation process. The study by Guo and Smallwood [11] argued that the reaction proposed by Liu et al. [10] affects mostly the inception and growth of soot rather than its oxidation. Consumption of H radical in $CO_2 + H \rightarrow CO + OH$ reaction slows down benzene formation and PAH growth by suppressing even-carbon-atom pathways. It is expected that reduction in H concentration upon CO_2 addition would lessen the role of H-abstraction-C_2H_2-addition (HACA) mechanism in soot inception and soot particle size growth [12]. Measurements by laser-induced incandescence and transmission electron microspcopy showed the effects of reduced soot inception

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and growth as soot particle size and number density are lowered in laminar propane diffusion flames diluted with CO_2 compared with those in N_2-diluted flames [13].

The current understanding that carbon dioxide dilution chemically suppresses soot formation is based on studies conducted mostly at atmospheric conditions. However, most combustion based energy conversion systems in transportation devices operate at pressures well above atmospheric due to size constraints and thermal efficiency considerations. Although the influence of pressure on soot process is significant [14], there is a lack of reliable combustion and soot models that are suitable for high pressure applications, and it is not certain whether the findings on the effect of CO_2 dilution at atmospheric pressure can be projected to elevated pressures. Our current understanding of pressure influence on soot inception, growth, and oxidation is very sketchy, and tractable measurements in flames at elevated pressures are desirable for a better insight.

The primary objective of this study is to assess the influence of CO_2 and N_2 dilution on soot formation processes at pressures above atmospheric. Soot volume fraction and temperature measurements were taken in laminar diffusion flames of diluted ethylene-air at pressures between 5 atm and 20 atm. Results for CO_2-diluted flames are compared with those for N_2-diluted flames. Different dilution rates with N_2 were also investigated at 10 atm to see the effects of inert dilution at a wider range.

2. Experimental method

The details of the experimental high pressure combustion chamber and the related measurement systems are described previously [15–18] and only essential features will be summarized. The cylindrical chamber is capable of operating up to 110 atm with an internal diameter and internal height of 0.24 m and 0.6 m, respectively. The laminar diffusion burner consists of a stainless steel fuel tube with a 3 mm inner diameter and an outer concentric air tube with a 25.4 mm inner diameter [15–18]. The outer surface of the fuel tube was chamfered to form a knife edge at the nozzle exit plane, which was helpful in improving the flame stability over a wide range of pressures. A flame enclosure with optical access shields the core flow from any disturbances that might be created inside the chamber.

Ethylene and either CO_2 or N_2 were mixed outside the pressure chamber in the desired ratio by two mass flow controllers and introduced into the burner from a single port. A wet-bubble-cell-based calibration system, traceable to NIST, was used to calibrate the mass flow controllers frequently. A constant mass flow rate of ethylene, 0.48 mg/s, was maintained at all pressures. This mass flow rate corresponds to a carbon flow rate of 0.41 mg/s, which matches the carbon flow rate of previous experiments performed by our research group [15–18]. The dilution rates of ethylene by CO_2 and N_2 by mass used in this work are listed in Table 1. Co-flow air was maintained at 0.34 g/s for all experimental conditions. The theory behind the spectral soot emission diagnostic (SSE) and its experimental layout, which was used in this study to measure

soot temperatures and concentrations, is described in detail previously [19]. In SSE, spectrally-resolved line-of-sight radiation emission from soot field within the flame envelope is measured along chords through the flame. A series of emission projections at a given height in the flame are inverted to obtain radially resolved emission rates from which radial temperature and soot volume fraction distributions can be inferred when soot optical properties are known [19,20].

The wavelength range over which the soot spectral emission was collected is 690–945 nm. The total array size of the CCD is 1340×400 pixels. However, due to the restricted size of the entrance slit, a region of interest of size 1340×80 pixels was selected. A horizontal spatial resolution of 70 µm over the depth of field defined by the burner nozzle exit diameter was estimated from knife-edge scans across a diffuse light source located at the object plane. The vertical spatial resolution was inferred to be approximately 290 µm. A pencil style neon calibration lamb was used for the calibration of the spectral axis of the CCD array. A filament lamp with calibration traceable to NIST, placed inside the chamber in place of the burner, is used for radiation intensity calibration.

3. Results and discussion

3.1. Visible flame shape

Photographs of ethylene diffusion flames diluted with of N_2 and CO_2 at various pressures, from atmospheric to 20 atm, are shown in Figs. 1 and 2 for 1:3 mass dilution (see Liu et al. [21] for 1:2 mass dilution pictures). Pressure changes the visible flame shape significantly. The luminous carbon zone is restricted to the tip of the flame at atmospheric pressure, and it expands as the pressure is increased. The size of the luminous zone in corresponding flames indicates qualitatively that CO_2-diluted flames produced less soot at lower pressures. All flames are attached to the fuel nozzle within the pressure range studied. The partially premixed blue zone at the periphery of the burner nozzle is more evident in the pictures for CO_2-diluted flames. As pressure is increased, the blue zone disappears, and the visible flame region expands towards the fuel nozzle. The flame height does not change considerably with pressure. Fully buoyancy-controlled N_2- and CO_2-diluted flame heights are around 9 mm and 8.5 mm, respectively. At 20 atm, soot wings are visible, although their height is not as high as the flame tip in 1:2 diluted flames for both CO_2 and N_2 dilution [21], indicating that flames at this dilution ratio and fuel mass flow rate are close to reaching their smoke points.

To compare laminar diffusion flame characteristics at various pressures for evaluating the pressure influence, the residence times, which scale with the square root of flame heights, should be controlled. In buoyancy-dominated co-flow diffusion flames, the fuel mass flow rate should be constant to keep flame heights independent of pressure. Independence of pressure of the flame height and flame residence time can be ensured if the flame cross-sectional area scales with the inverse of the pressure [5].

Table 1
Experimental conditions. Dilution ratios are by mass. The experimental results of the data sets 2 and 5 were originally used for verification of a numerical study and presented by Liu et al. [21].

Data set	Fuel:Diluent	5 atm	10 atm	15 atm	20 atm
1	$C_2H_4 : N_2$	–	1:1	–	–
2	$C_2H_4 : N_2$	1:2	1:2	1:2	1:2
3	$C_2H_4 : N_2$	1:3	1:3	1:3	1:3
4	$C_2H_4 : N_2$	–	1:4	–	–
5	$C_2H_4 : CO_2$	1:2	1:2	1:2	1:2
6	$C_2H_4 : CO_2$	1:3	1:3	1:3	1:3