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Structure of soot field in laminar ethylene diffusion flames diluted with either carbon dioxide or nitrogen at elevated pressures

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Abstract

The effects of dilution on soot formation and flame structure in nitrogen- and carbon dioxide-diluted co-flow ethylene/air flames were studied experimentally at pressures up to 20 atm. A constant ethylene mass flow rate was maintained for all flames, and dilution rates were set at two ratios: 1:2 and 1:3 by mass (fuel to diluting gas ratio) at all pressures. For these dilution rates, flames were stable and nonsmoking within the pressure range. Radially resolved soot concentrations and temperatures were obtained by Abel inversion from the line-of-sight soot spectral emission measurements. Maximum soot volume fractions were consistently lower in carbon dioxide-diluted flames in the lower pressure range but approached similar values to those in nitrogen-diluted flames at 20 atm. This observation implies that the chemical soot suppression effect of carbon dioxide, previously demonstrated at atmospheric pressure, is also present at elevated pressures up to 15 atm, but fades off beyond 15 atm. 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.

Introduction

In aviation gas turbines and diesel engines, where the mode of operation is mainly non-premixed (diffusion), a reliable and consistent combustion process is possible with overall equivalence ratios much lower than the lean flammability limits. However, the formation of soot is an artifact of the diffusive combustion. Further, diffusive combustion is attended by relatively higher levels of nitric oxide formation. Exhaust gas recirculation and multi-stage combustion are some of the effective means of controlling pollutant formation in these combustion engines. In principle, some of the products of combustion are mixed with the reactants to control the combustion temperatures. Since major species in the combustion products of common fuels burning in air are carbon dioxide, water vapour, and nitrogen, the influence of these gases on soot formation and oxidation is a current concern. The subject matter of this study is an investigation of the effects of nitrogen and carbon dioxide dilution on soot formation in laminar diffusion flames of ethylene at elevated pressures.

An additive may affect soot formation in any combination of the following three ways [1]: (1) dilution effect due to the change in the carbon composition of the fuel mixture, (2) thermal effect due to the change in the physical properties of the fuel mixture, and (3) chemical

effect due to the chemical activity of the additive. Liu et al. [2] identified the reaction $CO_2 + H \rightarrow CO + OH$ as the culprit for the chemical effect of CO_2 dilution. Increased O and OH concentrations in the soot forming region were found responsible for the oxidation of the soot precursors and nascent soot. Chemical activity of CO_2 resulted in lower flame temperatures, which adversely affected soot nucleation. A subsequent analysis by Guo and Smallwood [3] suggested that the same reaction proposed by Liu et al. [2] mainly affects the inception and growth of soot rather than its oxidation. Reduced concentration of the H radical due to its consumption in $CO_2 + H \rightarrow CO + OH$ reaction mitigates benzene formation and its growth by suppressing even-carbon-atom pathways. Soot volume fraction and primary particle size measurements in a propane laminar diffusion flame by laser-induced incandescence and transmission electron microscopy techniques substantiate that soot inception and growth are suppressed as soot particle size and number density are lowered in flames diluted with CO_2 compared with those in N_2- diluted flames [4, 5].

In general, available experimental data in the literature indicates that carbon dioxide dilution chemically suppresses soot formation. However, the current understanding is based on studies conducted at atmospheric pressure. Although research has shown that soot formation is greatly influenced by the operating pressure [6], there is a lack of reliable combustion and soot models that are aimed for practical 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. It is therefore of interest to understand the effects of pressure on soot formation and destruction pathways in tractable flames with CO_2 dilution.

Approach

The experimental high pressure combustion chamber used in this study is described in detail in [7–10]. The design pressure of the chamber is about 110 atm, and its internal diameter and internal height are 0.24 m and 0.6 m, respectively. Optical access into the chamber is through three ports at 0°, 90°, and 180° locations allowing line-of-sight measurements as well as 90° scattering and imaging experiments. Constant ethylene mass flow rate of 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 [7—10]. The dilution rates (fuel:diluent) of 1:2 and 1:3 by mass were studied for each diluent. These dilution rates were chosen because (1) at higher dilution rates, soot concentrations were outside the dynamic range of our measurement system at the low end of the pressure range, and the flame attachment suffered from increased dilution, i.e., flames were lifted off, and (2) at lower dilution rates, smoke point of the flame was reached at pressures below 20 atm. Co-flow air flow rate was maintained at 0.34 g/s for all experiments. The theory and overall experimental layout of the spectral soot emission diagnostic (SSE) are described previously [11]. In SSE, line-of-sight radiation emission from

soot is measured along chords through the flame. A series of emission projections at a given height in the flame can be inverted to obtain radially resolved emission rates from which temperature and soot volume fraction can be determined when soot optical properties are known [12].

Results and Discussion

Still pictures of N2- and CO2-diluted ethylene/air diffusion flames at various pressures, up to 20 atm, are shown in Figs. 1–4. The flame shape is affected substantially by pressure. At atmospheric pressure, the luminous carbon zone is restricted to the tip of the flame, and it expands as the pressure was increased. From the size of the luminous carbon zone, it can be predicted that CO2-diluted flames produced less soot at lower pressures. All flames are attached to the fuel nozzle within the pressure range studied. At 5 atm, the more common triple flame attachment is apparent in the images. The partially premixed blue zone at the periphery of the burner nozzle is more evident in the pictures for CO2-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 developed N2- and CO2-diluted flame heights are around 9 mm and 8.5 mm, respectively. At 20 atm, soot wings are clearly visible, and their height is slightly shorter than the flame tip in 1:2 diluted flames for both CO2 and N2 dilution, implying that smoke point has nearly been reached in these flames.
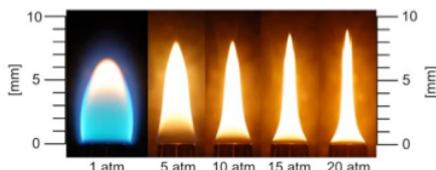

Fig. 1. Still pictures of N_2-diluted-ethylene/air (1:2 by mass) diffusion flames from 5 to 20 atm.
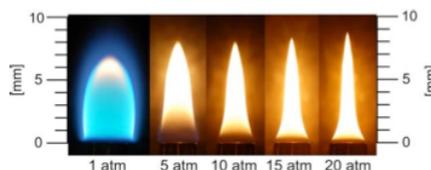

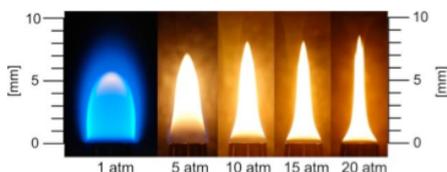Fig. 2. Still pictures of N_2-diluted-ethylene/air (1:3 by mass) diffusion flames from 5 to 20 atm.

Fig. 3. Still pictures of CO_2-diluted-ethylene/air (1:2 by mass) diffusion flames from 5 to 20 atm.
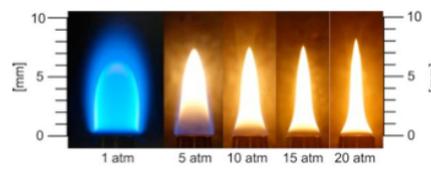

Fig. 4. Still pictures of CO_2-diluted-ethylene/air (1:3 by mass) diffusion flames from 5 to 20 atm.

Detailed radial distributions of soot volume fractions at various heights above the burner rim at 15 atm with nitrogen and carbon dioxide dilution are shown in Figs. 5 and 6, respectively. For all flames, soot first forms within an annular band near the burner rim at lower flame heights. In the region of one third to half of the soot region height from the base, soot particles begin to appear in the core of the flame in addition to the annular distribution of soot. Towards the tip of the flame, the annular distribution disappears and soot concentrations peak on the flame

centreline. As pressure is increased, soot volume fractions in all flames increase substantially.
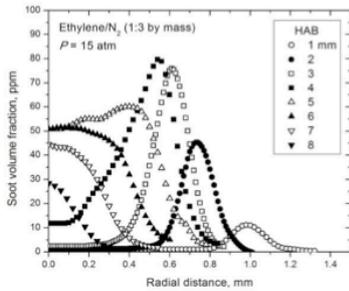

Fig. 5. Radial soot volume fractions at various heights above the burner at 15 atm with nitrogen dilution (1:3 by mass).
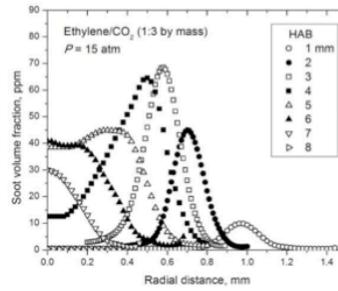

Fig. 6. Radial soot volume fractions at various heights above the burner at 15 atm with carbon dioxide dilution (1:3 by mass).

A plot of maximum soot yield as a function of pressure is shown in Fig. 7. Maximum soot yield increases to almost 40% for 1:2 diluted ethylene flames and 30% for 1:3 diluted ethylene flames at 20 atm. The percentage of fuel carbon conversion to soot can be represented in a power-law relationship of Y_s . Between 5 and 20 atm, the exponent n was estimated as 1.10 and 1.31 for 1:2 and 1:3 N_2-diluted flames, respectively, and as 1.34 and 2.01 for 1:2 and 1:3 CO_2-diluted flames, respectively. As expected, maximum soot yield decreases with increasing dilution rate. Maximum soot yield is lower for CO_2-diluted flames between 5 and 15 atm, suggesting that the reduction in soot concentration due to enhanced soot oxidation overpowers the increase in soot concentration due to enhanced inception between 10 and 15 atm. However, at 20 atm, both effects balanced each other so that the soot yields were similar for both diluents at the same dilution rate. Figure 7 also includes previously reported maximum soot yield data for n-heptane [10] and ethane flames [13] with a matching carbon mass flow rate. The n-heptane flame was diluted with N2 at approximately 1:2.1 by mass dilution rate. It can be seen that the change in soot yield with pressure was significantly higher for the n- heptane flame, implying that n-heptane flames are slightly more sensitive to pressure than ethylene and ethane flames. Undiluted ethane flames seem to produce similar soot yields to 1:2 diluted ethylene flames except that the sensitivity of soot yield to pressure of the ethane flame gets much lower with increasing pressure.
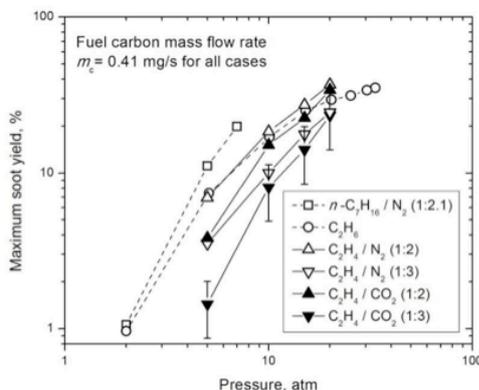

Fig. 7. Maximum soot yield of ethylene, n-heptane, and ethane flames for a fuel carbon mass flow rate of 0.41 mg/s. The n-heptane and ethane results were previously reported in [10] and [13] respectively. Fuel mass flow rates for n-heptane, ethane, and ethylene are 0.49, 0.52, and 0.48 mg/s, respectively. The ratios in parenthesis indicate fuel to diluent mass ratio.

Conclusions

Soot concentrations reduced everywhere in the flame with CO2 dilution at 5 atm as compared to N2 dilution, consistent with studies conducted at atmospheric pressure. As pressure was increased the effect of carbon dioxide dilution became more complex. Soot inception was enhanced, and the increase in soot oxidation with CO2 dilution became less effective on the overall soot yield with increasing pressure. These suggest that the chemical effect of CO2 dilution is more multi-faceted than suggested in studies conducted at atmospheric pressure. Increased soot inception with CO2 dilution at elevated pressures should be accounted for in soot models that are aimed for practical applications.

It was observed that the maximum soot yield of diluted n-heptane (1:2.1 by mass dilution), which was measured in a previous study, is higher than that of diluted ethylene (1:2 by mass dilution) at pressures above atmospheric.

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