



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, CO₂, 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 CO₂ 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 CO₂ dilution on soot. The findings of Schug et al. [6] implied a purely thermal effect of CO₂ dilution, whereas McLintock [7] concluded that the soot oxidation is enhanced chemically by CO₂. Chemical suppression effects of CO₂ 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 CO₂ 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 CO₂, Liu et al. [10] compared CO₂ dilution to the dilution by chemically inert CO₂. The reaction CO₂ + H → CO + OH was identified as the culprit for the chemical effect of CO₂ dilution [10]. Oxidation of soot precursors and nascent soot was found to be enhanced by the increased O and OH radicals upon CO₂ dilution. Further, chemical activity of CO₂ 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 CO₂ + H → 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 CO₂ addition would lessen the role of H-abstraction-C₂H₂-addition (HACA) mechanism in soot inception and soot particle size growth [12]. Measurements by laser-induced incandescence and transmission electron microscopy 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 CO₂ compared with those in N₂-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 CO₂ 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 CO₂ and N₂ 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 CO₂-diluted flames are compared with those for N₂-diluted flames. Different dilution rates with N₂ 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 CO₂ or N₂ 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 CO₂ and N₂ 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 lamp 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 N₂ and CO₂ 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 CO₂-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 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 buoyancy-controlled N₂- and CO₂-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 CO₂ and N₂ 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 ₂ H ₄ : N ₂	–	1:1	–	–
2	C ₂ H ₄ : N ₂	1:2	1:2	1:2	1:2
3	C ₂ H ₄ : N ₂	1:3	1:3	1:3	1:3
4	C ₂ H ₄ : N ₂	–	1:4	–	–
5	C ₂ H ₄ : CO ₂	1:2	1:2	1:2	1:2
6	C ₂ H ₄ : CO ₂	1:3	1:3	1:3	1:3

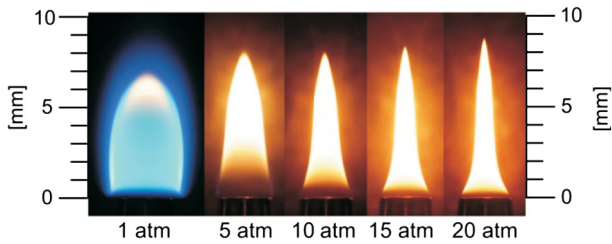


Fig. 1. Still photographs of N_2 -diluted-ethylene/air (1:3 by mass) laminar diffusion flames from 1 atm to 20 atm.

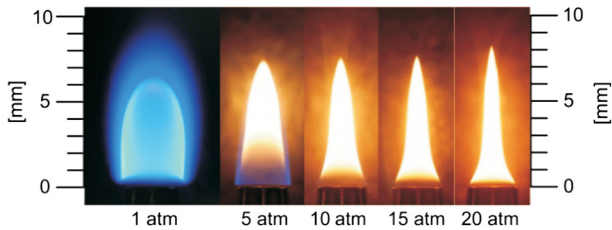


Fig. 2. Still photographs of CO_2 -diluted-ethylene/air (1:3 by mass) laminar diffusion flames from 1 atm to 20 atm.

Experimental results confirm that the flame cross-sectional area decreases with the inverse of pressure [14,22] in flames in which the fuel is not diluted. In diffusion flames in which the fuel is diluted, the flame height and diameter are only dependent on the fuel flow rate, and dilution with an inert does not significantly affect the flame shape at atmospheric conditions [23]. At elevated pressures, it was shown that the flame height in nitrogen diluted ethylene flames does not change with pressure as long as the fuel mass flow rate is kept constant [24]. As the mass flow rate of ethylene was maintained at a fixed value, it is possible to compare flame properties at equal heights in flames at different pressures since the residence times from the burner exit to the axial height of interest would be the same in all flames to a first approximation. The only exception is the zone close to the burner exit where velocities are expected to be higher with the diluent addition.

3.2. Soot yields

The sensitivity of the sooting propensity to pressure can be assessed by evaluating the variation of maximum soot yield with pressure. The soot yield can be defined as the percentage of the carbon in the fuel converted to soot. Soot yields were calculated using the methodology documented before [25–27].

Soot yields of N_2 -diluted flames at various dilution rates are shown in Fig. 3 as a function of the height above the burner rim. The first observation is that the suppression effect of N_2 dilution is not linear with the fuel/dilution mass ratio. Further, the maximum soot yield location moves downstream as the dilution rate is increased from 4 mm at 1:1 dilution to 6 mm at 1:4 dilution, Fig. 3. Comparison of soot yields of N_2 - and CO_2 diluted flames at 1:3 dilution ratio by mass clearly shows how the effectiveness of CO_2 diminishes with increasing pressure, Fig. 4. At 20 atm soot yields of both flames are almost identical, whereas at lower pressures suppression by CO_2 is higher than that of N_2 .

As temperatures do not change much between both additives in this study, it is expected that most of the difference in soot volume fraction results is originating from chemical effects of CO_2 . The line of sight averaged temperatures along the flame axis are shown for CO_2 - and N_2 -diluted flames at 5 and 20 atm in Fig. 5. As expected, the changes in averaged temperatures between two diluents at a

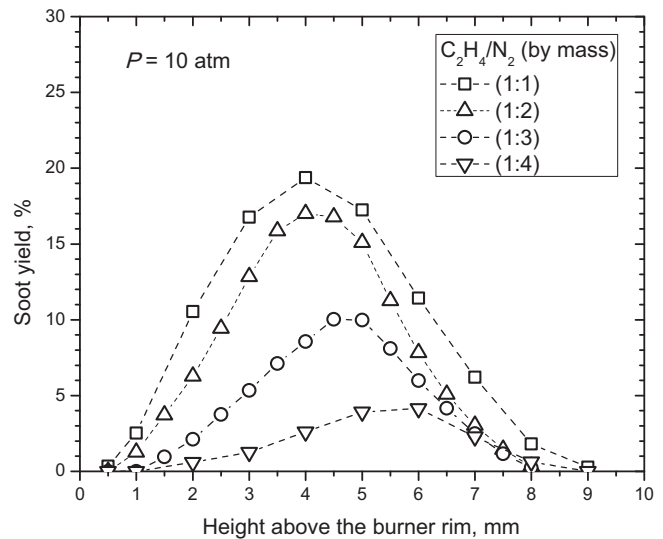


Fig. 3. Soot yields of ethylene flame diluted with nitrogen at various levels at 10 atm.

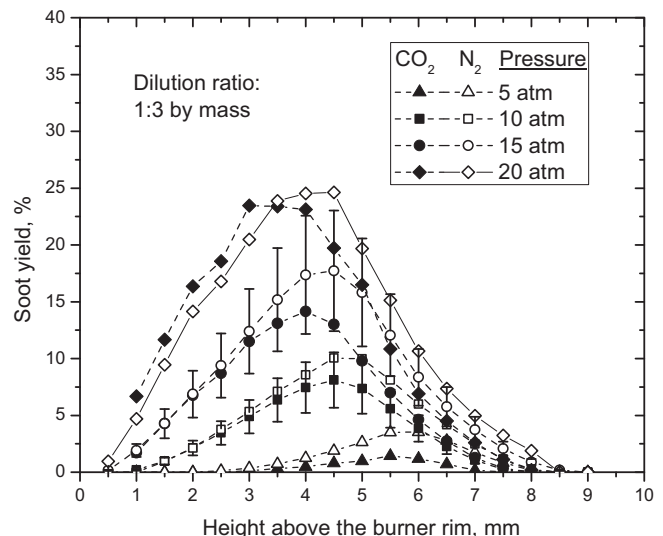


Fig. 4. Soot yields of ethylene/nitrogen and ethylene/carbon dioxide flames at various pressures; dilution ratio is 1:3 by mass (data sets 3 and 6 in Table 1).

given pressure are relatively small. Similar temperature profiles, but with smaller differences in corresponding temperatures, were observed at other pressures as well. Detailed radial temperature profiles corresponding to all data sets can be found in [28].

The discussion above comparing CO_2 dilution effects to those of N_2 is based on dilution ratios by mass which is required to have similar thermal effects so that the both diluents will have the same heat sink capacity. However, it is also necessary to consider equal dilution on a mole basis in comparing the chemical effects. In an experiment similar to the one reported here, an experimental design that would permit direct quantification of the chemical and thermal effects separately is difficult to achieve, if at all possible, unless the additives have the same molecular mass, similar molecular diffusivities and specific heats. In data set 2, the fuel mole fraction in the N_2 -diluted mixture is 0.5 whereas it is 0.52 in CO_2 -diluted data set 6, Table 1. These two data sets can be compared while keeping their flame temperature characteristics in mind. If the data sets 2 (1:2 dilution by mass) and 6 (1:3 dilution

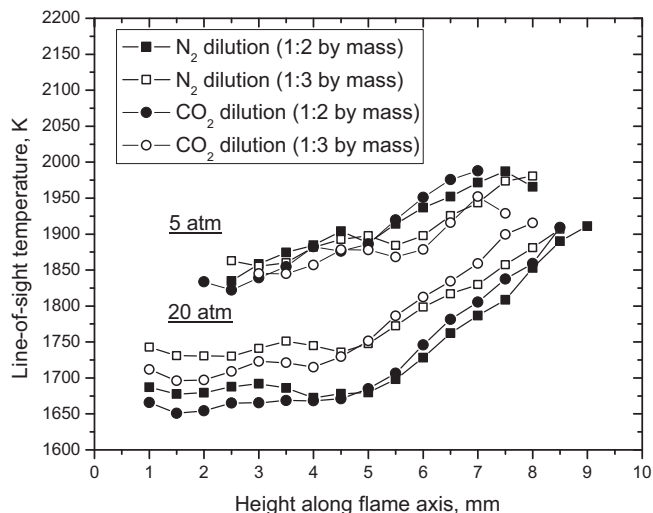


Fig. 5. Line-of-sight emission averaged soot temperature along the flame axis as a function of flame axial locations for various dilution rates at 5 and 20 atm.

by mass) in Table 1, which have similar dilution ratios by mole, are compared, it is seen that maximum soot yield ratios of nitrogen-diluted to carbon dioxide-diluted cases are 4.8 at 5 atm and reducing to 1.6 at 20 atm. This ratio is 2.3 at 10 atm and 1.9 at 15 atm, Fig. 6. In Fig. 5, line-of-sight temperature profiles at 5 atm, which could be considered as the representative overall temperature characteristics of the flames, for data set 2 and data set 6 display similar values but consistently lower about 25 K for CO₂-diluted case in the lower half of the flame where soot formation processes dominate. With these relatively small temperature differences, observed differences in soot yields could be attributed largely to the chemical suppression effects of CO₂. At 20 atm, temperatures are about 20–50 K higher for CO₂-diluted case in the lower half of the flame due to a lower radiative heat loss from the flame as a result of relatively lower soot concentrations. In view of these relatively low temperature differences, it is obvious that the chemical suppression effect of CO₂ decreases as pressure is increased from 5 to 20 atm.

Charest et al. [29] reported recently a numerical simulation of biogas surrogates, namely methane containing 20 and 40% CO₂,

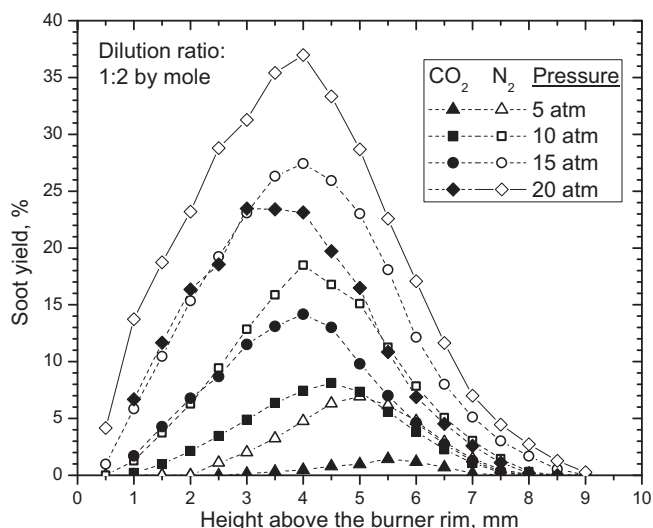


Fig. 6. Soot yields of ethylene/nitrogen and ethylene/carbon dioxide flames at various pressures; dilution ratio is about 1:2 by mole (data sets 2 and 6 in Table 1).

as fuels in laminar diffusion flames and assessed the influence of CO₂ content on flame structure and soot at pressures up to 20 atm. It was found that [29], similar to current results, CO₂ suppressed the soot formation chemically, however this suppression effect was much larger at lower pressures. Although the CO₂ dilution rates are much smaller in the simulation of Charest et al. [29] as compared to the dilution amounts in the current work, numerical findings support the experimental results reported here. As discussed by Liu et al. [10] for atmospheric pressure diffusion flames, the main chemical effect of CO₂ dilution is through the reaction $\text{CO}_2 + \text{H} \rightarrow \text{CO} + \text{OH}$ which lowers the rates of soot inception and C₂H₂ concentrations; this seems to be the case at elevated pressures as well [29]. The numerical simulation of N₂- and CO₂-diluted ethylene flames at 1:2 dilution ratio at elevated pressures by Liu et al. [21] concluded that CO₂ is still more effective than N₂ as a diluent to suppress soot formation at elevated pressures. The primary pathway for the chemical effect of CO₂ dilution was found to be through the reverse reaction of $\text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H}$ [21]. The chemical effect of CO₂ lowers the rates of soot inception, C₂H₂ addition, and PAH condensation. The effectiveness of the CO₂ chemical influence on soot formation suppression diminishes with increasing pressure. The diminishing effectiveness of the chemical influence of CO₂ dilution with increasing pressure is due to the significant decrease in the H radical mole fraction [21].

To give an overall picture of the dilution effects, 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. To help with assessing the sensitivity of soot yield to pressure, soot yields could be represented by a scaling relationship as $Y_s \propto P^n$. Between 5 and 20 atm, the exponent *n* was estimated as 1.10 and 1.31 for 1:2 and 1:3 N₂-diluted flames, respectively, and as 1.34 and 2.01 for 1:2 and 1:3 CO₂-diluted flames, respectively.

3.3. Measurement uncertainties

Relatively higher soot concentrations at elevated pressures lead to non-negligible attenuation of emission by soot particles. This introduces some uncertainty into the measurements by the SSE technique. The maximum optical thickness, which is determined by the soot volume fraction in the measurement volume and the

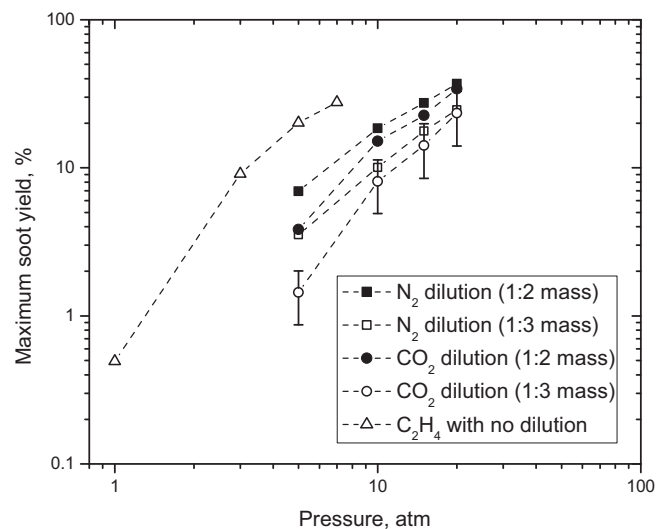


Fig. 7. Maximum soot yields of ethylene diluted either by nitrogen or carbon dioxide at two levels. The ratios in parenthesis indicates fuel to diluent mass ratio. Also plotted are the data for pure ethylene [30].

optical path length, encountered in these flames is about 2. The detailed modelling efforts reported in [19,31] indicate that when optical thickness approaches 2, uncertainty introduced by soot self-absorption to the value of soot volume fraction becomes significant but still is much smaller than the error contributed by the systematic uncertainty in soot absorption function $E(m)$ discussed below. However, the derived temperatures would be systematically lower by about 2% at the highest soot loading conditions [31].

One of the most significant source of systematic errors encountered in soot concentration and size measurements is the uncertainty in complex soot refractive index and its dependence on wavelength and temperature. A limited number of studies on the influence of temperature on soot refractive index imply that at typical flame temperatures representative of diffusion flames, the dependence may not be significant. To be consistent with our previous high-pressure soot measurements [14–18,22], we assumed that the soot refractive index does not have a significant dependence on temperature and wavelength and a value of 0.26 is assumed for the refractive index absorption function $E(m)$.

The maximum uncertainties reported in this work evaluated using the methodologies discussed in [15,19,31] are 40% in soot volume fraction and 5% in temperature measurements. It should be noted that a major part of the uncertainties are due to systematic errors. The error bars in Figs. 4 and 7 reflect the total uncertainty in soot measurements.

4. Conclusions

Soot spectral emission technique was used to measure the soot concentrations and temperature fields of co-flow laminar diffusion flames of N_2 - and CO_2 -diluted ethylene-air at pressures up to 20 atm to assess the relative suppression effects of the N_2 and CO_2 dilution of the flame. The dilution rates (fuel:diluent by mass) were chosen as 1:2 and 1:3 to achieve stable non-smoking flames within the pressure range. If the pressure dependence of the peak soot yield is approximated by a power law, then pressure exponent is about 1.1 and 1.31 for 1:2 and 1:3 N_2 -diluted flames, respectively, and 1.34 and 2.01 for 1:2 and 1:3 CO_2 -diluted flames, respectively, between 5 and 20 atm. Soot volume fractions were lower everywhere within the flame envelope with CO_2 dilution at 5 atm as compared to those with N_2 dilution. However, as pressure was increased further, the effect of carbon dioxide dilution became more complex. The sooting propensity, in terms of maximum soot yield, was found to be significantly lower with carbon-dioxide dilution in the pressure range of 5–15 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 but becomes relatively small at 20 atm.

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