NRC Publications Archive
Archives des publications du CNRC

Influence of carbon dioxide dilution on soot formation in diffusive combustion

Gulder, O. L.; Baksh, M-Fazil

This publication could be one of several versions: author’s original, accepted manuscript or the publisher’s version. / La version de cette publication peut être l’une des suivantes : la version prépublication de l’auteur, la version acceptée du manuscrit ou la version de l’éditeur.

NRC Publications Record / Notice d'Archives des publications de CNRC:
https://nrc-publications.canada.ca/eng/view/object/?id=37e9451b-96a4-48b8-80cc-475e5013f3ab
https://publications-cnrc.canada.ca/fra/voir/objet/?id=37e9451b-96a4-48b8-80cc-475e5013f3ab

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at
https://nrc-publications.canada.ca/eng/copyright
READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L’accès à ce site Web et l’utilisation de son contenu sont assujettis aux conditions présentées dans le site
https://publications-cnrc.canada.ca/fra/droits
LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D’UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n’arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

National Research Council Canada
Conseil national de recherches Canada

INFLUENCE OF CARBON DIOXIDE DILUTION ON SOOT FORMATION IN DIFFUSIVE COMBUSTION*

Ö. L. Gülder and M. F. Baksh
National Research Council of Canada
IECE, Combustion & Fluids Engineering Lab., M-9
Ottawa, Ontario K1A 0R6, Canada

Introduction

Exhaust gas recirculation is one of the effective ways of controlling exhaust pollutants in compression ignition engines. Since the major species in the exhaust of a compression ignition engine are nitrogen, carbon dioxide, water vapor, oxygen and carbon monoxide, the influence of these gases on soot formation and oxidation is a current concern. The complex nature of the compression ignition engine combustion which makes it unsuitable for more fundamental studies also makes it difficult to interpret fundamental ideas regarding soot formation in the compression ignition engine context [1]. However, a basic understanding of soot formation and oxidation is central to the particulate emission control in combustion devices. One- and two-dimensional laminar diffusion flames are suitable devices to obtain this basic understanding.

The influence of carbon dioxide on soot formation in diffusion flames have been studied by several investigators [2-5]. Although the suppression of soot by carbon dioxide has been confirmed, a consensus has not been reached regarding the nature of this suppression. Arthur and Napier [2] observed that a concentration of 45% carbon dioxide completely suppresses luminosity in methane diffusion flames. They claimed that the suppression of soot by carbon dioxide due to purely thermal effects is unlikely, because (a) the critical carbon dioxide concentrations are not simply related to their thermal capacities, and (b) the reduction in luminosity is not linear with carbon dioxide concentration [2]. McClintok [3] suggested that the influence of carbon dioxide is exerted primarily in the soot oxidation zone where carbon dioxide presumably promotes soot burnout. Schug et al. [4] added up to 30% carbon dioxide to the fuel side (ethylene), and determined the influence of carbon dioxide as the increase in the fuel flow rate at the smoke point conditions. It was concluded [4] that the influence of carbon dioxide is predominantly thermal, with no measurable chemical influence, in laminar diffusion flames. More recently, Du et al. [5] demonstrated that carbon dioxide, whether added to the fuel side or oxidizer side can suppress soot formation chemically.

The objective of the present study is to quantify the influences of flame temperature, dilution, and direct chemical interaction when carbon dioxide is added to the fuel side in a coflow laminar diffusion flame. Experimental data for ethylene flames

---

* Combustion Institute/Canadian Section, Spring Technical Meeeting, May 10-12, 1993, Université Laval, Québec. Partial funding for the work presented here has been provided by EMR / PERD program.

diluted by carbon dioxide were compared to data from nitrogen diluted flames and to the flames in which the flame temperature is adjusted by varying the temperature of the reactants.

This work is a continuation of our previous efforts of deciphering the influences of fuel structure [6], flame temperature [7, 8], inert diluents [9], sulfur dioxide [10], and fuel-bound sulfur [11] on soot formation in diffusion flames.

Experimental Methodology

The fuel nozzle of the burner is a stainless-steel pipe of 12.7 mm inner diameter. Air is supplied from a concentric converging nozzle of 100 mm inner diameter. Both air and fuel streams are heated by regulated electric heaters. Temperatures of the reactants were monitored by thermocouples near the exit of the fuel and air nozzles, and kept within ±4 K of the desired reactant temperature. The measurement system and the details of the burner have been reported in detail elsewhere [7,8].

The line-of-sight average light extinction and the flame diameter were measured, as a function of the axial position, along the centerlines of ethylene diffusion flames. Experimental conditions are given in Table I. In the dilution experiments, the temperature of the reactants were kept at 623 K, and the amount of carbon dioxide in ethylene was varied from 0 to 0.55 mole fraction. For undiluted flame experiments, the temperature of the reactants varied from 623 K to 298 K to vary the flame temperature, Table I. Comparison of these two data sets to the measurements made in nitrogen diluted ethylene flames permits a quantitative assessment of the thermal, concentration, and chemical effects of carbon dioxide on soot formation.

TABLE I

Summary of experimental conditions and results. $X_{F,O}$ is the mole fraction of ethylene in the fuel gas mixture (the balance is carbon dioxide), T_r is the temperature of the reactants, T_{ad} is the adiabatic flame temperature corresponding to the initial temperature of reactants or the amount of carbon dioxide dilution, and F_{max} is the maximum soot volume fraction. Fuel flow rate is 3 mg/s.

DILUTION WITH CO_2 (T_r = 623 K)				HEATED REACTANTS ($X_{F,O}$ = 1)			
Key	$X_{F,O}$	T_{ad} (K)	F_{max} (ppm)	Key	T_r (K)	T_{ad} (K)	F_{max} (ppm)
C1	1.	2500	12.7	T1	623	2500	12.7
C2	0.9	2487	10.0	T2	573	2481	11.9
C3	0.8	2470	7.4	T3	523	2462	11.3
C4	0.7	2450	5.2	T4	473	2440	10.3
C5	0.6	2424	3.4	T5	373	2402	8.6
C6	0.5	2390	2.0	T6	323	2382	7.9
C7	0.45	2368	1.3	T7	298	2370	7.5

Results and Discussion

To make a quantitative comparison of the thermal, concentration, and chemical effects on soot formation, the following parameters in the diluted and the undiluted flames should be taken into consideration: the characteristic flame temperatures, residence times, the flame diameters, and fuel and oxidant mole fractions. We have taken the adiabatic flame temperature as the indicator of the temperature field of the lower regions of the flame where the soot inception and nucleation occur.

Using the data from nitrogen diluted and undiluted but temperature controlled ethylene flames, we have recently shown that [10] the maximum soot volume fraction in diffusion flames can be scaled as follows

$$F_{max} \sim \frac{X_{F,O}\sqrt{H}}{L_{max}} exp(-E_a/R^{\circ}T_{ad}) \tag{1}$$

where $X_{F,O}$ is the mole fraction of fuel in the fuel gas mixture, H is the visible flame height, L_{max} is the flame diameter at the axial location where the line-of-sight average soot volume fraction reaches the maximum, E_a is the activation energy and T_{ad} is the adiabatic flame temperature. The term $\sqrt{H}$ is proportional to the flame residence time, and L_{max} removes the effect of changing flame diameters with the amount of additive. The activation energy inferred from the experimental data was 200 kJ/mole [9, 10].

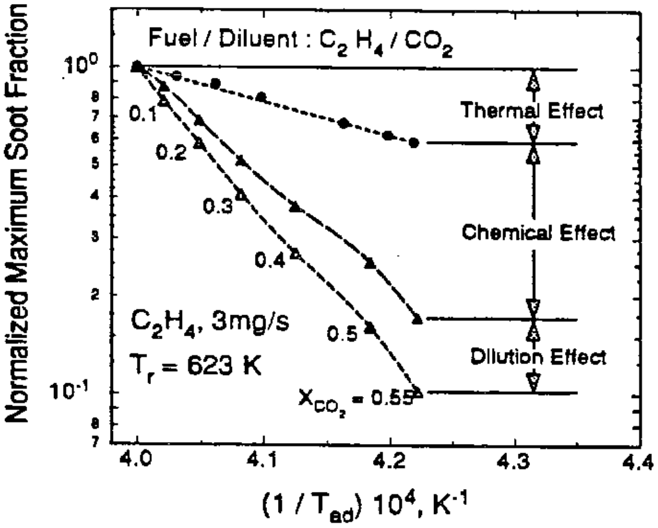

Fig. 1. The Arrhenius plot of the normalized maximum soot volume fractions versus the inverse of the adiabatic flame temperature showing the relative influence of temperature, concentration, and the direct chemical interaction. Round symbols are the data of the flames without CO_2 but with varying reactant temperatures. When the flame temperature varies while $X_{F,O}$=1, Eq.(1) gives the dotted straight line for E_a=200 kJ/mole. Short dash line with empty triangle symbols are those of CO_2 added flames (the lower curve). Full triangle symbols, connected with long dash curve, are the CO_2 added flame data that were evaluated using Eq.(1).

The normalized maximum soot volume fraction data are plotted as a function of the inverse of the adiabatic flame temperature in Fig.1. The dotted straight line shows the influence of the flame temperature imposed by the thermal inertia of added carbon dioxide, whereas the short-dash line (the lower curve in Fig.1) with empty triangle symbols shows the overall reduction in soot. When allowance is made for the fuel mole fraction, flame diameter, and the flame residence time (in accordance with Eq.(1)), we obtain the long-dash curve with full triangle symbols. If there were no direct chemical interaction effect of carbon dioxide this curve would coincide with the dotted straight line. Then, the difference between dotted straight line and the long-dash curve is due to the direct chemical interaction effect of carbon dioxide on soot formation rate, Fig.1.

In laminar coflow diffusion flames, maximum soot concentrations vary with the fuel flow rate. In this study, we investigated the influence of carbon dioxide for different fuel flow rates. Figure 2 shows that the effect of fuel flow rate on the effectiveness of carbon dioxide in reducing soot is negligible.

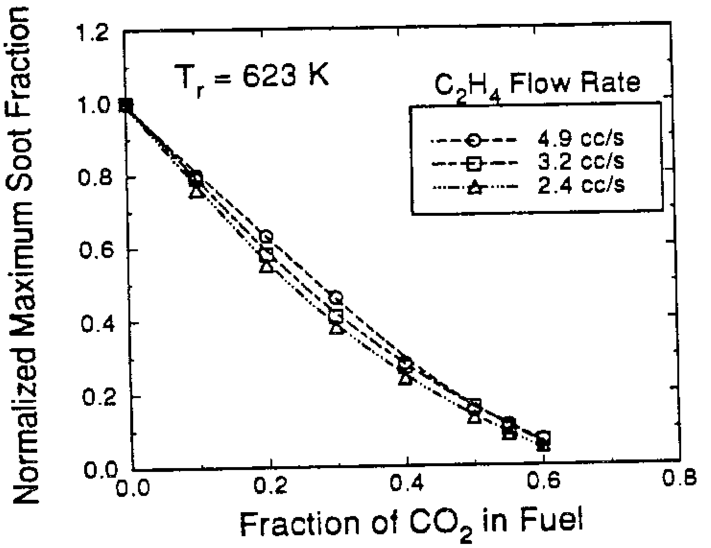

Fig.2. Influence of the ethylene fuel flow rate on the effectiveness of carbon dioxide in reducing the soot.

Although the mechanism responsible for the chemical suppression of soot in the presence of carbon dioxide is not known, Du et al. [5] suggested that carbon dioxide would elevate the concentrations of atomic oxygen and hydroxyl radical which could, in turn, increase the oxidative attack on soot precursors in the soot formation region of the flame. As a first approximation for the change in radical pool concentration, we plotted the change in the equilibrium hydrogen atom, oxygen atom, and hydroxyl radical concentrations with the amount of carbon dioxode in the fuel at a fixed temperature of 1800 K, Fig.3. It should be emphasized that this is a very crude approximation, because the radical concentrations approach toward equilibrium via slow three-body combination reactions with a typical characteristic time scale of ~ 3 ms in atmospheric flames, although radical production rates are relatively rapid (~ 20 μs) [12]. If we make the assumption that the equilibrium radical concentrations change in the same manner as the actual concentrations with the amount of carbon dioxide in the fuel gas stream, then it is possible to claim that increased concentrations of oxygen atom and hydroxyl radical oxidize the soot precursors in the soot formation zone leading to reduced soot concentrations. The change in hydrogen atom is relatively small, although it shows a slight decrease with increasing carbon dioxide fraction. A decrease in hydrogen atom concentration may contribute to the reduction in soot, since hydrogen atoms could speed up the soot formation rate by hydrogen atom abstraction from aromatic molecules [13].

Concluding Remarks

An overventilated coflowing axisymmetric laminar ethylene diffusion flame was used to study the influence of carbon dioxide addition to the fuel on soot formation. The experiments were designed such that the relative influences of temperature, fuel concentration, and direct chemical interaction can be isolated when carbon dioxide is added to the fuel side in varying amounts. A quantitative assessment of these three effects was made using an Arrhenius type expression which makes allowance for changes in flame profiles and flame residence times. It was shown that the direct chemical interaction of carbon dioxide plays a significant role in reducing the soot formation rate.

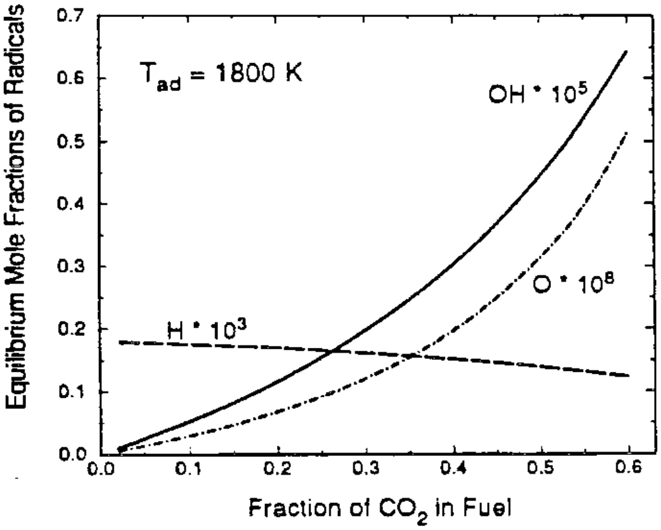

Fig.3. The change in the equilibrium concentrations of hydroxyl radical, oxygen atom, and hydrogen atom with the amount of carbon dioxide in the fuel gas stream.

References

[1] Heywood, J. B., Internal Combustion Engine Fundamentals,Ch.11, McGraw Hill, New York, 1988.
[2] Arthur, J. R., and Napier, D. H., *5th Symp. (Int'l) on Combustion*, p.303, Reinhold, New York, 1955.
[3] McClintock, I. S., *Combust. Flame*, **12**, 217 - 225 (1968).
[4] Schug, K. P., Manheimer-Timnat, Y., Yaccarino, P., and Glassman, I., *Combust. Sci. Technol.*, **22**, 235 - 250 (1980).
[5] Du, D. X., Axelbaum, R. L., and Law, C. K., *23rd Symp.(Int'l) on Combustion*, p.1501, Combustion Institute, Pittsburgh, 1991.
[6] Gülder, Ö. L., *Combust. Flame*, **78**, 179 - 194 (1989).
[7] Gülder, Ö. L., and Snelling, D. R., *23rd Symp.(Int'l) on Combustion*, p.1509, Combustion Institute, Pittsburgh, 1991.
[8] Gülder, Ö. L., *Combust. Flame*, **88**, 74 - 82 (1992).
[9] Gülder, Ö. L., and Snelling, D. R., *Combust. Flame*, **92**, 115 - 124 (1993).
[10] Gülder, Ö. L., *Combust. Flame*, **92**, 410 - 418 (1993).
[11] Gülder, Ö. L., and Glavinčevski, B., *Combust. Sci. Technol.*, **77**, 337 - 343 (1991).
[12] Barlow, R. S., Dibble, R. W., Chen, J. - Y., and Lucht, R. P., *Combust. Flame*, **82**, 235 - 251 (1990).
[13] Franklach, M., *22nd Symp. (Int'l) on Combustion*, p. 1075, The Combustion Institute, Pittsburgh, 1989.

Gulder

O.L. GULDER

The Combustion Institute
Canadian Section

1993 Spring Technical Meeting

09 to 12 May 1993
Université Laval
Québec QC

UNIVERSITÉ
LAVAL