NRC Publications Archive Archives des publications du CNRC

Experimental investigation of oxygen addition to fuel on soot formation in laminar coflow diffusion flames of ethylene and propane

Liu, Fengshan; Thomson, Kevin A.; Smallwood, Gregory J.

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.

Publisher's version / Version de l'éditeur:

Proceedings of the IMECE2006, IMECE2006-15988, pp. 119-125, 2006

NRC Publications Record / Notice d'Archives des publications de CNRC:

<https://nrc-publications.canada.ca/eng/view/object/?id=4c115426-db91-4bcf-bbbb-48b1fe627de3>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=4c115426-db91-4bcf-bbbb-48b1fe627de3>

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.

WR001844

CT-07781436-9

WR001844

CISTI ICIST

CT-07781436-9

Document Delivery Service
in partnership with the **Canadian Agriculture Library**

Service de fourniture de Documents
en collaboration avec la **Bibliothèque canadienne de l'agriculture**

THIS IS NOT AN INVOICE / CECI N'EST PAS UNE FACTURE

MARIA CLANCY
DGO
INST FOR CHEM PROCESS & ENVIR TECH
NATIONAL RESEARCH COUNCIL CANADA
M-12, ROOM 141, 1200 MONTREAL RD.
OTTAWA, ON K1A 0R6
CANADA

ORDER NUMBER: CT-07781436-9
Account Number: WR001844
Delivery Mode: XLB
Delivery Address:
Submitted: 2009/03/03 14:18:04
Received: 2009/03/03 14:18:04
Printed: 2009/07/30 15:04:12

Extended	Periodical	Fax - transcribed	CANADA

Client Number: MARIA CLANCY MAR 02 2009 # 11
Author: LIU F; RGOMSON K A; SMALLEWOOD G J
Date: 2006
Article Title: EXPERIMENTAL INVESTIGATION OF OXYGEN ADDITION TO FUEL ON SOOT....
Series Title: PROCEEDINGS OF 2006 ASME IMECE, NOV-5-10, CHICAGO, ILLINOIS, U S A, 2006

INSTRUCTIONS: ***ATTN: MARIA CLANCY*******

**Estimated cost for this 7 page document: \$0 document supply fee + \$0
copyright = \$0**

The attached document has been copied under license from Access Copyright/COPIBEC or other rights holders through direct agreements. Further reproduction, electronic storage or electronic transmission, even for internal purposes, is prohibited unless you are independently licensed to do so by the rights holder.

Phone/Téléphone: 1-800-668-1222 (Canada - U.S./E.-U.) (613) 998-8544 (International)
www.nrc.ca/cisti Fax/Télécopieur: (613) 993-7619 www.cnrc.ca/icist
info.cisti@nrc.ca info.icist@nrc.ca

National Research
Council Canada

Conseil national
de recherches Canada

Page

1 / 1

IMECE2006-15988

EXPERIMENTAL INVESTIGATION OF OXYGEN ADDITION TO FUEL ON SOOT FORMATION IN LAMINAR COFLOW DIFFUSION FLAMES OF ETHYLENE AND PROPANE

Fengshan Liu Kevin A. Thomson Gregory J. Smallwood
Institute for Chemical Process and Environmental Technology
National Research Council, Building M9, 1200 Montreal Road, Ottawa, Ontario, Canada

ABSTRACT

Investigation of the effect of oxygen addition to fuel on the visible flame appearance and soot formation characteristics of laminar diffusion flames is important to gain comprehensive understanding of gas-phase combustion chemistry and its interaction with soot chemistry. This paper reports experimental results of oxygen addition to fuel on the visible flame height and soot volume fraction distributions in axisymmetric coflow laminar ethylene and propane diffusion flames at atmospheric pressures. The carbon flow rate was maintained constant in all the experiments. Although many experimental studies have been conducted in the literature in this topic, the present investigation aimed at providing spatially resolved soot volume fraction distributions over the entire range of oxygen addition from no oxygen addition up to the point of flashback while keeping the carbon mass flow rate constant. The level of oxygen added to fuel right before flashback is about 45% (the percentage of oxygen addition is always by volume in this study) of the fuel flow rate in the ethylene flame and 300% of the fuel flow rate in the propane flame. As the added oxygen amount to ethylene increases, the visible flame height first increases. When the added oxygen flow rate is about 13% of the fuel flow rate, the flame becomes smoking, i.e., soot escapes from the flame tip. When the oxygen flow rate reaches about 42% of the fuel flow rate, the flame stops smoking. When oxygen was added to propane, the visible flame height linearly decreases with increasing the amount of oxygen. These very different effects of oxygen addition to ethylene and propane indicate that oxygen plays a drastically different role in the chemical pathways leading to soot formation in ethylene and propane flames. Distributions of soot volume fractions in these flames were measured using a 2D light attenuation technique coupled with the Abel inversion. The present study provides valuable experimental data for validating soot models.

INTRODUCTION

The effect of oxygen addition to fuel on soot formation in laminar hydrocarbon diffusion flames has attracted attention in the literature. Study of the effect of oxygen addition to fuel on soot formation in hydrocarbon diffusion flames is of interest both fundamentally and practically. From a fundamental point of view, addition of oxygen to fuel is one of many effective experimental tools for elucidating some specific aspects of the complex gaseous combustion chemistry and the mechanisms of soot formation and providing valuable experimental data for validating these models. For these purposes, addition of oxygen to fuel plays a similar role as addition of a small amount of hydrocarbon fuel to the fuel side of a laminar diffusion flame to gain further insights into soot chemistry (formation and oxidation) [1-3]. Practically, knowledge of the effect of oxygen addition to the fuel side of diffusion flames is clearly useful to improve our overall understanding of partially premixed combustion, a combustion mode found in many applications and has been the subject of intense research in recent years [4-7]. The early interest in the effect of oxygen addition to fuel, at least in part, was to investigate the effect of oxygen leaked through the diffusion flame front to the fuel side on fuel pyrolysis process and the subsequent soot formation in diffusion flames. Effect of oxygen addition to fuel on the flame structure and soot formation has received renewed interests in recent years due to the interest in partially premixed combustion.

The pioneering studies of the effect of oxygen addition to fuel on soot formation in hydrocarbon diffusion flames were conducted by Chakraborty and Long [8] and Dearden and Long [9] in 1968. Since our present interest lies in the effect of oxygen addition to ethylene and propane, the following discussion of the previous studies is focused on findings on these two fuels. In Ref.[8], Chakraborty and Long investigated the effect of oxygen addition to fuel on the amounts of chloroform-soluble material, polycyclic aromatic hydrocarbons (PAH), and carbonaceous residue in soot recovered from ethylene/air diffusion flames. They found that with increasing

the amount of oxygen to ethylene the amounts of all the materials investigated (notably soot and PAH) first increase to maxima and then decrease. Dearden and Long [9] investigated the effect of oxygen addition to the fuel side of ethylene and propane diffusion flames established on a Wolfhard-Parker slot burner. In their experiments, the carbon supply was maintained constant. The addition of oxygen to ethylene was found to significantly increase the overall sooting rate (the amount of soot mass collected over a period of time). However, the sooting rate of the propane flame was found to decrease linearly with the amount of oxygen added to fuel. Addition of oxygen to the fuel increases the flame temperature and the amount of C_2H_2 in the sooting region in both flames. They suggested that the enhanced sooting rate in the ethylene flames by oxygen addition is caused by increased temperatures in the fuel pyrolysis zone, leading to an increase in the amount of C_2H_2 in the luminous zone. Because of its different soot formation behavior from ethylene, Dearden and Long [9] suggested that the sooting behavior of propane is anomalous. Through a theoretical analysis and examination of available experimental data, Jones and Rosenfeld [10] later suggested that the sooting behavior of ethylene is anomalous, but not that of propane. The effect of oxygen addition to fuel on soot formation in ethylene and propane flames was later investigated by Wright [11], who employed a coflowing rectangular burner and analyzed the soot collected from the exhaust gases. The results of Wright showed that addition of oxygen to ethylene significantly increased the amount of soot emitted. Although the addition of oxygen to propane only slightly increased the amount of soot emitted, his results are qualitatively different from those of Dearden and Long [9], who found that addition of oxygen reduced the overall sooting rate of the propane flame. In addition, Wright [11] found that the amount of soot emitted from the flame drops rapidly when about 11% of the stoichiometric requirement in oxygen was added to both ethylene and propane. Wright suggested that the role of oxygen in promoting soot formation is catalytic rather than thermal, i.e., the added oxygen enhances the fuel pyrolysis rate through a catalytic effect, but not through the thermal decomposition of the fuel as a result of raised temperature. This suggestion is in disagreement with Dearden and Long [9], who speculated that the added oxygen enhances fuel pyrolysis through enhanced temperatures. No suggestion for the observed quantitative difference between ethylene and propane flames was offered by Wright [11].

Following these early experimental investigations on the effect of oxygen addition to fuel on the sooting behavior of ethylene and propane diffusion flames in terms of global observations, several studies were carried out more recently [12-18]. Schug et al. [12] systematically investigated the effect of various diluents added to fuel on the sooting behavior of axisymmetric coflow laminar diffusion flames burning different hydrocarbons or their mixtures. The sooting behavior of the fuel mixtures was assessed by measuring two global parameters: the sooting fuel flow rate and the sooting flame height. Although their primary interest was not the effect of oxygen addition to fuel, their results indicate that the addition of oxygen to ethylene enhances its sooting tendency, in agreement with previous findings [8,9,10]. The experimental study of Wey et al. [13] is perhaps the first one to provide spatially resolved soot volume fraction distributions in laminar

coflow underventilated propane diffusion flames stabilized on a Wolfhard-Parker slot burner with and without oxygen addition. They measured soot volume fraction and temperature distributions using a light attenuation technique and thermocouples, respectively. It is worth pointing out that in the study of Wey et al. the flow rate of the fuel stream (propane + oxygen), rather than the carbon flow rate, was maintained constant. Their results showed that as more propane in the fuel stream is replaced by oxygen (up to 20%) the flame temperature increases slightly but soot volume fractions increase somewhat significantly. They also suggested that the enhanced soot formation by oxygen addition to propane is attributed to the catalytic effect of oxygen on fuel pyrolysis.

The first comprehensive experimental investigation on the effect of oxygen addition to fuel on soot formation in counterflow hydrocarbon diffusion flames was conducted by Hura and Glassman [14]. In this study they also provided a summary of previous studies on this subject prior to 1987. Hura and Glassman found that oxygen addition to fuel greatly enhances soot formation in ethene (ethylene) flames, but has negligibly small impact on soot formation in propane flames. They explained the different effects of oxygen addition to fuel in ethylene and propane flames by arguing that oxygen has a different catalytic effect on the pyrolysis process of ethylene and propane: the addition of small amount of oxygen leads to no overall increase in the extent of the radical pool, which governs the fuel pyrolysis rate, in propane flames but enhances the radical pool extensively in ethylene flames. Glassman and co-workers [12,14] believed that the initial pyrolysis of the fuel is the crucial step in soot formation in diffusion flames.

The different effects of oxygen addition to fuel in ethylene and propane diffusion flames were further studied by Hura and Glassman in both the counterflow and axisymmetric coflow configurations [15]. When oxygen was added to the fuel side, fuel flow rate was always kept constant. In counterflow flames, they showed that the addition of small amount of oxygen to propane actually reduces soot formation, in qualitative agreement with the results of Dearden and Long [9]. However, as more oxygen was added to propane the soot loading increased significantly to a maximum before decreasing when the equivalence ratio of the fuel/oxygen mixture approaches the flashback limit. The addition of oxygen to ethylene enhances soot formation until the equivalence ratio of the fuel/oxygen reaches about 6. Further increase in the amount of oxygen leads to a rapid decrease in soot loading. Their smoke height measurements in axisymmetric coflow flames are in agreement with the counterflow flame results. They also found that the added oxygen to ethylene prompts soot formation through enhancing soot inception rate, but not soot surface growth rate. Once again they explained the different effects of oxygen addition to ethylene and propane flames in terms of the catalytic effect of oxygen on the pyrolysis of ethylene and the dilution effect of oxygen in propane flames. Unfortunately, Hura and Glassman [15] did not measure soot volume fraction distributions in these coflow flames or visible flame height variation with the amount of added oxygen.

The effect of oxygen addition to fuel in ethylene and propane diffusion flames in both counterflow and axisymmetric coflow configurations was revisited by Du et al. [16]. The primary interests of Du et al. were to isolate the dilution, thermal, and chemical effects of CO_2 and O_2 addition through

systematic variation and independent controls of flame temperature, mixture composition, and the chemical nature of the additives. Their results of soot particle inception limit of counterflow flames showed that addition of oxygen to ethylene significantly accelerates soot inception chemistry in ethylene flames. Oxygen addition to propane only slightly enhances soot inception chemistry when less than 40% of oxygen was added. However, the soot inception chemistry was found to be rapidly enhanced when more than 40% of oxygen was added to propane. When the effects of dilution, thermal, and chemical were isolated, they also found that oxygen chemically suppresses the soot inception chemistry in the propane counterflow flames when the added oxygen is less than 40%. As more oxygen was added to propane, the soot inception chemistry was found to be significantly accelerated. The line-of-sight integrated soot volume fraction measurements of Du et al. [16] in axisymmetric coflow ethylene and propane diffusion flames reveal a consistent trend on the effect of oxygen addition to fuel to that observed from the soot particle inception limit measurements in counterflow ethylene and propane flames. It is worth pointing out that in their coflow flame experiments, the fuel flow rate was adjusted in such a way that the peak soot concentrations occurred at roughly the same axial position. Although the results of Du et al. [16] on the effect of oxygen addition to ethylene on soot formation are in qualitative agreement with previous findings [8,9,11,12,14,15], their results on the effect of oxygen addition to propane on soot formation contradict those of Dearden and Long [9] and Wright [11], though they seem in qualitative agreement with those of Wey et al. [13] and Hura and Glassman [15].

Through a series of carefully controlled experiments in axisymmetric coflow diffusion flames aiming at separating the different effects of an additive (thermal, dilution, and chemical,) Gülder [17] found that oxygen addition to propane chemically enhances soot formation. His conclusion is in contrast with that of Du et al. [16] who claimed otherwise. The effects of oxygen and propane addition to ethylene on soot formation in counterflow diffusion flames were recently studied by Hwang et al. [18]. These authors suggested that the different effect of oxygen addition to ethylene and propane diffusion flames can be explained in terms of the importance of the recombination reaction of propargyl (C_3H_3) leading to polycyclic aromatic hydrocarbon (PAH).

The above literature review reveals that previous studies on the effect of oxygen addition to fuel conducted in 2D coflow diffusion flame configuration are in general qualitative in nature since they provided only some global soot related parameters, such as the smoke height [12], the visible flame appearance captured on photograph [15], line-of-sight average soot volume fraction along the flame height [16,17]. Although more detailed studies on the effect of air addition to fuel (partially premixed flames) exist, e.g. Mitrovic and Lee [5], the results from these studies do not represent the effect of oxygen addition due to the presence of a large amount of nitrogen, which significantly lowers the flame temperature and the soot yield. Therefore, there is lack of quantitative experimental data in soot volume fraction distribution and the visible flame height under different levels of oxygen addition to fuel, which are important for the validation of gaseous combustion chemistry and soot formation and oxidation models. The present study made an attempt to fill this gap by providing spatially resolved

soot volume fraction distribution and the visible flame height over a wide range of oxygen addition to the fuel side of laminar axisymmetric coflow ethylene and propane diffusion flames at atmospheric pressure. This study provides complementary information on the effect of oxygen addition to fuel in these flames to the previous study of Hura and Glassman [15]. It is also noted that there exists contradictory evidence on the qualitative effect of oxygen addition on sooting tendency in propane flames. In addition, the chemistry mechanism for the different effects of oxygen addition to ethylene and propane flames on soot formation has not been fully resolved. Although these topics deserve further experimental and numerical investigations, the present study made no attempt to address these two issues.

EXPERIMENT Burner

The co-annular laminar diffusion flame burner used in this study is almost identical to that used in previous studies [12,15] and is shown in Fig.1. The burner consists of a fuel tube with a 10.9 mm inner diameter centered in an air nozzle of 100 mm inner diameter. The wall thickness of the fuel tube is 0.9 mm. Before exiting the air nozzle the air stream passes through packed beds of glass beads and porous metal disks to ensure steady and uniform velocity distribution and to prevent flame instabilities due to disturbance in the air stream. Air was supplied from an in-house compressor and filtered. Both hydrocarbon fuels, ethylene and propane, and oxygen used in this study are research grade (more than 99.9% pure). The burner assembly was attached to a positioning platform with accurate vertical and horizontal movement capability. Both fuel and air were delivered to the burner at room temperature.

Fig.1 Schematic diagram of the co-annular laminar diffusion flame burner assembly.

Diagnostics

The visible flame height, defined as the tallest point along the flame centerline where the flame luminosity due to soot radiation is visible, and soot volume fraction distribution were acquired in the experiments. Both the visible flame height of a coflow laminar diffusion flame and the soot volume fraction distribution are important data for soot model validation. The former is an important global parameter and defines the point

where soot is completely oxidized and latter offers important information with regard to regions of soot inception and surface growth.

The visible flame height was measured by a metered telescope. The soot volume fraction was measured using a 2D imaging technique based on light attenuation. This technique has been documented in detail in Ref.[19]. Only a brief description of the 2D imaging method is given here. The optical layout for the 2D transmission measurements is similar to that described in Ref.[19]. An arc of the Hg lamp was used as the light source. The detector was a Princeton Instruments CCD Model EEV-TE-1152x1242 with a Model ST138 controller. The detector array size is 1152x1242 pixels with a square pixel size of 22.4 μm . The detection wavelength is selected by the choice of the interference filter. A 20 nm bandwidth filter centered at 577 nm was used in our experiments, identical to the study of Snelling et al. [19]. Deconvolution of the transmission measurements from the 2D soot imaging using the Abel inversion algorithm of Dasch [20] yields the distribution of the transmission with respect to the radial position, which can be relate to the local soot volume fraction assuming the soot absorption function $E(m)$ is known [19,21]. Following the study of Snelling et al. [19], the soot absorption function $E(m)$ at $\lambda = 577 \text{ nm}$ was assumed to be 0.26 in the data analysis from transmission to soot volume fraction. Further details on the diagnostic method and data analysis can be found in Refs.[19,20,21]. In an experimental work such as this one, it is perhaps more important to consider the relative uncertainty in the soot volume fraction measurements rather than the absolute error, since we are most interested in the differences observed between different fuel conditions. The relative uncertainty error is dominated by uncertainty in the ratio of scatter to absorbed light, ρ_{sa} , which can be as high as 16% in the pure ethylene coflow diffusion flame [22]. The absolute uncertainty of the soot volume fraction measurements is dominated by uncertainty in the true value of the absorption refractive index function of soot, $E(m)$, which has an estimated uncertainty of 20% [23].

RESULTS AND DISCUSSION

The volume flow rates of hydrocarbon fuels (ethylene and propane) and oxygen were controlled using calibrated mass flow meters. The flow rate of the air stream was controlled with a rotameter. In all the experiments reported in this section the air stream flow rate was maintained constant at 4733.3 cm^3/s while the flow rate of ethylene and propane was kept constant at 3.23 cm^3/s and 2.16 cm^3/s , respectively. Therefore, addition of oxygen to either ethylene or propane led to a higher flow rate through the fuel nozzle than the pure fuel case. It is noted that the carbon flow rate was maintained constant throughout all the cases in both ethylene and propane flames. That is why the visible heights of the pure ethylene and pure propane flames are almost identical (see below).

Due to small oscillations in the visible flame tip, the visible flame height reported here is an average value over a period of a few minutes and has an estimated absolute error of about 1 mm.

Ethylene Flames

Representative photographs of the ethylene flames with different levels of oxygen addition acquired with a CCD camera of automatic mode (the exposure time may vary) are

shown in Fig.2. The equivalence ratio of these ethylene/oxygen mixtures is also indicated in the figure caption. The immediate impression from Fig.2 is that as more oxygen is added to ethylene the visible flame height first increases, then the flame emits soot, and finally becomes non-smoking again. This general trend of oxygen addition to ethylene agrees with the observations of Hura and Glassman [15], who reported that "under all conditions, initially non-smoking ethylene flames started smoking as oxygen was added." They also observed visually that the soot yield peaks in the equivalence ratio range of 8 to 9. We also observed, based on visual impression, that the ethylene flame emits most soot when the added oxygen is about 30% of the fuel, which corresponds to an equivalence ratio of 10, consistent with the findings of Hura and Glassman [15] and Wright [11].

Fig.2 Effect of oxygen addition to fuel on the visible appearance of the ethylene diffusion flame.

Fig.3 Variation of the visible flame height of the ethylene flame with oxygen addition.

Variation of the visible height of the ethylene flame with oxygen addition is shown in Fig.3. The circles and diamonds represent two trials. The close agreement between the two sets of data demonstrates that the repeatability of the visible flame height is very good. The visible height of the pure ethylene is about 64.5 mm. When the flow rate of oxygen added to ethylene is less than 11% of that of ethylene, the visible flame height increases almost linearly. Between 11% and 12% oxygen addition the visible flame height increases very rapidly. Beyond 12% oxygen addition the ethylene starts to emit soot, i.e., soot escapes the flame tip without being fully oxidized. When the oxygen addition reaches 41% of ethylene the flame becomes unstable in the sense that the visible flame tip oscillates vertically back and forth between smoking and non-smoking condition. At 42% oxygen addition the flame stops emitting soot. Within a narrow range of oxygen addition, 42 to 45%, the visible flame height decreases significantly from 69.5 mm to about 62 mm. When oxygen addition is more than 45%, the flame becomes unstable and starts to show tendency of flashback. The equivalence ratio of the ethylene/oxygen with an oxygen-to-ethylene ratio of 0.45 is $\phi = 6.67$. This flashback equivalence ratio for ethylene/oxygen mixture is excellent agreement with that reported by Hura and Glassman [15], who found that flashback occurs in the ethylene flame when the equivalence ratio falls in a narrow range of 6.5 to 7.3.

The results shown in Figs.2 and 3 indicate that addition of oxygen to ethylene first enhances soot formation when oxygen flow rate is less than about 30% of the fuel flow rate. At larger amount of oxygen addition, the role of the added oxygen to fuel is to suppress soot formation. While the chemical mechanism of soot formation enhancement by small amount of oxygen addition is not yet fully understood [14,17], the soot formation suppression by a large amount of oxygen addition is expected, as the fuel mixture becomes more premixed [15].

Soot volume fraction (SVF) distributions at several representative levels of oxygen addition, under non-smoking conditions, are displayed in Fig.4. Both the peak soot volume fraction and the visible flame height increase modestly with oxygen addition before the flame becomes smoking, Figs.4(a)-4(c). After the transition from smoking to non-smoking, both variables start to decrease, which is likely to be the consequence of enhanced soot oxidation under larger amount of oxygen addition. Although the overall shapes of SVF distribution at different amounts of oxygen addition are similar, i.e., the peak SVF always occurs in the annular region, the following two observations are made. First, the luminous region starts at a lower streamwise location closer to the burner exit as more oxygen is added to ethylene, which can also be visually observed from Fig.2. Secondly, SVF in the flame centerline region increases with increasing amount of oxygen addition, which is likely related to enhanced PAH production. The enhancement of PAH production by oxygen addition to ethylene was observed by Chakraborty and Long [8] and Hura and Glassman [14]. It is worth pointing out that the enhanced PAH formation has two implications to the increase in SVF in the flame centerline region shown in Fig.4 measured by the 2D imaging technique based on light attenuation. First, the enhanced light attenuation in the centerline region is indeed caused by increased concentration of carbonaceous material (soot). Secondly, it is also possible that the enhanced light attenuation is due to increased concentration of PAH or both

soot and PAH. Light attenuation techniques, such as the one used in this study, are unable to differentiate the contributions to light attenuation between soot and PAH.

Based on the measured SVF distributions shown in Fig.4, it is speculated that small amount of oxygen addition (less than 30% of fuel) to ethylene is to enhance both soot inception and surface growth processes. The net enhancement of soot oxidation by added oxygen is not materialized until the oxygen flow rate is more than 30% of that of ethylene. Nevertheless, our results demonstrate that oxygen addition to fuel has a strong effect on soot formation in axisymmetric coflow ethylene diffusion flames, which is consistent with previous findings [8,9,11,14-16].

Fig.4 Soot volume fraction distribution in ethylene flames at different levels of oxygen addition.

Propane Flames

Addition of oxygen to fuel results in less interesting variation in the visible flame appearance of propane flames in the sense that the flame never becomes smoking and the visible flame height decreases monotonically with the amount of oxygen addition. For this reason the flame photos at different levels of oxygen addition are not shown.

Fig.5 Variation of the visible flame height of the propane flame with oxygen addition.

Variation of the visible height of the propane flame with the amount of oxygen addition to propane is shown in Fig.5. As mentioned early, the carbon flow rate was maintained constant throughout this study. The visible height of the pure propane flame (about 66 mm) is almost identical to that of the pure ethylene flame (64.5 mm). As the amount of oxygen added to propane increases, the visible flame height decreases linearly up to 300% oxygen addition. At 300% oxygen addition, which corresponds to an equivalence ratio of $\phi = 1.67$, a stable flame can still be established. At a slightly higher oxygen addition (302%), the flame becomes unstable and flashback occurs. It is noticed that the flashback equivalence ratio for propane/oxygen mixtures, $\phi = 1.67$, is much lower than that reported by Hura and Glassman [15] who found that flashback occurred in the propane flame when the amount of oxygen addition leads to an equivalence ratio is in the range of 3.6 to 3.9. It is not clear about the cause of the different flashback equivalence ratios for propane/oxygen mixture between the present observation and the study of Hura and Glassman [15].

Fig.6 Soot volume fraction distribution in propane flames at different levels of oxygen addition.

Distributions of the measured SVF at four levels of oxygen addition are shown in Fig.6. Some obvious effects of oxygen addition on SVF distribution are evident: the visible flame height decreases, the sooting zone moves closer to the burner surface, the peak SVF first decreases then increases, and the high soot concentration region moves from the annular region to the centerline with increasing amount of oxygen addition. These results qualitatively confirm previous findings that oxygen plays a very different role in soot formation in ethylene and propane flames. It is reasonable to assume that the monotonic decrease in visible height of the propane flames is due to somewhat insignificant effect of oxygen addition in enhancing, if not suppressing, soot inception and surface growth. The upstream move of the sooting zone to the burner exit surface is clearly related to the enhanced fuel pyrolysis process. It is worth pointing out that when a sufficiently amount of oxygen added to the fuel, either ethylene or propane, the overall flame is double structured: an inner fuel-rich premixed flame just above the burner exit and an outer diffusion flame. The unburned hydrocarbons and hydrogen from the inner fuel-rich premixed flame are consumed in the outer diffusion flame.

This overall flame structure has been shown numerically by Liu et al. [24] and briefly mentioned by Hura and Glassman [15].

Perhaps the most interesting observation from Fig.6 is the shift of the high soot concentration zone from the annular region to the flame centerline region, which has not been reported previously. While it is natural to link the increased soot concentration in the centerline region to PAH, such an explanation is not supported by available experimental evidence. To the contrary, Hura and Glassman [14] found from their experimental study in counterflow propane diffusion flames that oxygen addition leads to reduced PAH concentration. Since soot surface growth process is the dominant step for soot yield, the somewhat significantly increase in soot concentration in the centerline region at large amount of oxygen addition implies that there exists a large of amount chemical species responsible for soot surface growth other than PAH. The established species for soot surface growth are acetylene and PAH [25]. The study of Hura and Glassman [14] rules out the role of PAH and leaves acetylene the only possible species for soot surface growth in the centerline region. Further research is required to ascertain the mechanism for the significant increase in soot concentration in the centerline region.

CONCLUSION

A systematic experimental investigation on the effect of oxygen addition to fuel on soot formation in axisymmetric coflow laminar ethylene and propane diffusion flames was carried out. The present results once again show that oxygen has a very different effect on soot formation in these two hydrocarbon flames. Results of oxygen addition to ethylene qualitatively agree with previous studies. Although oxygen addition to fuel has less impact on soot formation in propane flames than in ethylene flames, this study observed a marked difference in the flashback equivalence ratio of propane/oxygen mixture than a previous study.

Besides some global observations the present study also obtained quantitative results on the visible flame height and spatially resolved soot volume fraction distributions. The complete shift of the high soot concentration region from the annular region to the centerline region in the propane flames under large amount of oxygen addition was observed for the first time. The experimental data obtained in this study are useful for validation of soot model.

ACKNOWLEDGEMENTS

The authors would like to thank Mr. Faz Baksh for his assistance in conducting the experimental work.

REFERENCES

1. Hamins, A., Anderson, D. T., and Miller, J. H., 1990, "Mechanistic Studies of Toluene Destruction in Diffusion Flames," *Combust. Sci. Tech.*, **71**, pp. 175-195.
2. Puri, R., Santoro, R. J., and Smyth, C. R., 1994, "The Oxidation of Soot and Carbon Monoxide in Hydrocarbon Diffusion Flames," *Combust. Flame*, **97**, pp. 125-144.
3. McEnally, C. S., and Pfefferle, L. D., 1998, "Soot Formation in Methane/Air Nonpremixed Flames Doped with Small Quantities of C3 Hydrocarbons," *Combust. Flame*, **112**, pp. 545-558.

4. Gore, J. P., and Zhan, N. J., 1996, "NO_x Emission and Major Species Concentrations in Partially Premixed Laminar Methane/Air Co-Flow Jet Flames," *Combust. Flame*, **105**, pp. 414-427.
5. Mitrovic, A., and Lee, T.-W., 1998, "Soot Formation Characteristics of Laminar Partially Premixed Flames," *Combust. Flame*, **115**, pp. 437-442.
6. McEnally, C. S., and Pfefferle, L. D., 1999, "Experimental Study of Nonfuel Hydrocarbon Concentrations in Coflowing Partially Premixed Methane/Air Flames," *Combust. Flame*, **118**, pp. 619-632.
7. McEnally, C. S., and Pfefferle, L. D., 2000, "Experimental Study of Nonfuel Hydrocarbons and Soot in Coflowing Partially Premixed Ethylene/Air Flames," *Combust. Flame*, **121**, pp. 575-592.
8. Chakraborty, B. B., and Long, R., 1968, "The Formation of Soot and Polycyclic Aromatic Hydrocarbons in Diffusion Flames III—Effect of Additions of Oxygen to Ethylene and Ethane Respectively as Fuels," *Combust. Flame*, **12**, pp. 469-476.
9. Dearden, P., and Long, R., 1968, "Soot Formation in Ethylene and Propane Diffusion Flames," *J. Appl. Chem.*, **18**, pp. 243-251.
10. Jones, J. M., and Rosenfeld, J. L. J., 1972, "A Model for Sooting in Diffusion Flames," *Combust. Flame*, **19**, pp. 427-434.
11. Wright, F. J., 1974, "Effect of Oxygen on the Carbon-Forming Tendencies of Diffusion Flames," *Fuel*, **53**, pp. 232-235.
12. Schug, K. P., Manheimer-Timnat, Y., Yaccarino, P., and Glassman, I., 1980, "Sooting Behavior of Gaseous Hydrocarbon Diffusion Flames and the Influence of Additives," *Combust. Sci. Tech.*, **22**, pp. 235-250.
13. Wey, C., Powell, E. A., and Jagoda, J. L., 1984, "The Effect on Soot Formation of Oxygen in the Fuel of a Diffusion Flame," *Proceedings of the Combustion Institute*, **20**, pp. 1017-1024.
14. Hura, H. S., and Glassman, I., 1987, "Fuel Oxygen Effects on Soot Formation in Counterflow Diffusion Flames," *Combust. Sci. Tech.*, **53**, pp. 1-21.
15. Hura, H. S., and Glassman, I., 1988, "Soot Formation in Diffusion Flames of Fuel/Oxygen Mixtures," *Proceedings of the Combustion Institute*, **22**, pp. 371-378.
16. Du, D. X., and Axelbaum, R. L., and Law, C. K., 1990, "The Influence of Carbon Dioxide and Oxygen as Additives on Soot Formation in Diffusion Flames," *Proceedings of the Combustion Institute*, **23**, pp. 1501-1507.
17. Gülder, Ö. L., 1995, "Effects of Oxygen on Soot Formation in Methane, Propane, and n-Butane Diffusion Flames," *Combust. Flame*, **101**, pp. 302-310.
18. Hwang, J. Y., Chung, S. H., and Lee, W., 1998, "Effects of Oxygen and Propane Addition on Soot Formation in Counterflow Ethylene Flames and the Role of C₃ Chemistry," *Proceedings of the Combustion Institute*, **27**, pp. 1531-1538.
19. Snelling, D. R., Thomson, K. A., Smallwood, G. J., and Gülder, Ö. L., 1999, "Two-Dimensional Imaging of Soot Volume Fraction in Laminar Diffusion Flames," *Applied Optics*, **38**, pp. 2478-2485.
20. Dasch, C. J., 1992, "One-Dimensional Tomography: a Comparison of Abel, Onion-Peeling, and Filtered Backprojection Methods," *Applied Optics*, **31**, pp. 1146-1152.
21. Snelling, D. R., Thomson, K. A., Smallwood, G. J., and Gülder, Ö. L., 1999, "Two-Dimensional Imaging of Soot Volume Fraction in Laminar Diffusion Flames," *Applied Optics*, **38**, pp. 2478-2485.
22. Snelling, D. R., Thomson, K. A., Smallwood, G. J., Gülder, Ö. L., Weckman, E. J., and Fraser, R. A., 2002, "Spectrally Resolved Measurement of Flame Radiation to Determine Soot Temperature and Concentration," *AIAA Journal*, **40**(9), pp. 1789-1795.
23. Krishnan, S. S., Lin, K. C., Faeth, G. M., 2001, "Extinction and Scattering Properties of Soot Emitted from Buoyant Turbulent Diffusion Flames," *J. Heat Transfer*, **123**, pp. 331-339.
24. Liu, F., Guo, H., Smallwood, G. J., and Gülder, Ö. L., 2001, "Numerical Investigation of the Effect of O₂ Addition to Fuel on Soot Formation in Coflow Laminar C₂H₄/Air Diffusion Flames," *The Combustion Institute Canadian Section 2001 Spring Technical Meeting*, 13-15 May, 2001, McGill University, Quebec, Canada.
25. Frenklach, M., and Wang, H., 1990, "Detailed Modeling of Soot Particle Nucleation and Growth," *Proceedings of the Combustion Institute*, **23**, pp. 1559-1566.