The Effect of Flame Structure on Soot-Particle Inception in Diffusion Flames

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The effect of flame structure on soot particle inception is studied by varying the mixture fraction at stoichiometry $Z_{st}$ and, consequently, flame location. $Z_{st}$ is varied by reassigning the nitrogen from the oxidizer to the fuel such that the flame temperature is not changed. Strain rates in the coflow flame and flame heights in the coflow flame are measured in ethylene flames to identify the sooting limits based on the appearance of luminous soot. Numerical experiments with a coflow diffusion-flame code employing C2 kinetics are also performed to understand the effects of $Z_{st}$ on flame structure and to interpret the experimental results. The results show that as $Z_{st}$ is increased and the flame shifts towards the fuel, soot inception is suppressed. In the coflow flame, no luminous soot is detected at a strain rate greater than 60 s$^{-1}$ for $Z_{st} \geq 0.16$, as compared to 175 s$^{-1}$ for the ethylene/air flame, $Z_{st} = 0.064$. The laminar ethylene coflow flame is soot free at $Z_{st} \geq 0.72$, regardless of flame height. For small $Z_{st}$, where changes in $Z_{st}$ are primarily due to changes in fuel concentration, the effect on soot inception is primarily fuel dilution. For large $Z_{st}$, where the oxygen concentration is appreciably increased, the subsequent shift in the OH profile towards the fuel side of the flame can have a dramatic influence on inception. The shift in OH essentially narrows the soot zone suggesting that it may be possible to obtain soot-free conditions for many fuels if the structure of the flame can be adjusted to the extent that significant OH exists on the fuel side of the flame at a temperature of ca. 1300 K. The experimental and numerical results demonstrate that this requirement can be satisfied for ethylene flames.

INTRODUCTION

In contrast to premixed flames and shock tubes, diffusion flames experience dramatic gradients in concentration and temperature in the regions of soot formation and growth. This structure greatly complicates the process, yet the practical importance of diffusion flames requires that the implications of this structure on soot formation be understood. Furthermore, fundamental studies of soot formation in diffusion flames are often not as conclusive as those in premixed flames [1] and shock tubes [2], largely because the steep gradients can make data interpretation ambiguous. For example, although on kinetic grounds, it is apparent that the local fuel concentration and temperature must influence soot formation, it is less clear how perturbations in freestream conditions (e.g., oxygen concentration) affect soot formation in diffusion flames. The ambiguity arises from, among other things, the fact that any change in the freestream conditions of the fuel or oxidizer can change temperature, concentration and velocity [3]. Despite advances in understanding soot formation in diffusion flames, most studies have emphasized only the structure of the soot formation region [4–6], and not the structure of the reaction-zone itself. In this paper, we systematically vary the inner flame structure and determine how these variations influence soot inception.

Structural variations affect the temperature/species field relationships and these can affect soot inception. For example, if the region of large fuel concentration can be shifted to high temperatures then pyrolysis and soot-inception chemistry may be accelerated. Conversely, if the region of high oxygen concentration can be shifted towards the fuel-side of the flame, an inhibitory effect may be observed. These arguments are better appreciated by considering the simple diffusion flame described with the flame-sheet solution in mixture-fraction space. First, however, we will discuss how the flame location is shifted and then discuss the implications of the shift.

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The constraint that diffusion flames burn stoichiometrically in the limit of infinite Damköhler number implies that for an equi-diffusional system with a given stoichiometry, the adiabatic flame temperature $T_{ad}$ is uniquely defined. For example, the stoichiometry of an ethylene/air diffusion flame yields

$$C_2H_4 + 3O_2 + 11.28N_2 \rightarrow \text{products}$$

$$
(T_{ad} = 2371 \text{ K}). \quad (1)
$$

Provided that the stoichiometry in Eq. 1 is maintained, the nitrogen can be introduced with the fuel, oxygen or combinations of both, with no impact on the adiabatic flame temperature. In other words, the adiabatic flame temperature of a fuel mixture containing 1 mole of $C_2H_4$ and 11.28 moles of $N_2$ burning in pure oxygen is the same as that of pure $C_2H_4$ burning in air. Nonetheless, the flame location of the former will be much closer to the fuel. This location can be depicted by the stoichiometric mixture fraction $Z_{st}$ defined as

$$Z_{st} = (1 + Y_{F,1}W_O Y_{O,0}W_F Y_F)^{-1}, \quad (2)$$

where $Y_i$, $W_i$, and $\nu_i$ are the mass fraction, molecular weight and stoichiometric coefficient, respectively, of the $i$th species [7]. The subscripts $F$ and $O$ refer to the fuel and oxidizer and 1 and 0 refer to the boundaries of the fuel and oxidizer, respectively. For the ethylene/air flame, $Z_{st} = 0.064$ and for the corresponding diluted-ethylene/oxygen flame, $Z_{st} = 0.78$.

The location of the flame can have a dramatic influence on overall flame structure. In Fig. 1 the flame-sheet solution is plotted for $Z_{st} = 0.064$ (Fig. 1a) and $Z_{st} = 0.78$ (Fig. 1b). Here the oxidizer boundary corresponds to $Z = 0$ and the fuel boundary to $Z = 1$. First consider the ethylene/air flame, which sits close to the oxidizer because of stoichiometric requirements. Letting the effective reaction boundaries for multistep chemistry be identified with temperature $T_{rb}$ then, as noted by Williams [8], the mean concentration of oxidizer within the reaction zone is less than that of fuel. This observation explains why one-step high-activation-energy asymptotic analysis predicts fuel, not oxidizer, leakage for small Damköhler number flames [8].

The premise of this argument can be exploited for soot-particle inception as well. From Fig. 1a it is evident that for the ethylene/air flame the fuel concentration is large in the high temperature regions of the flame, i.e., conditions are favorable for soot inception. However, changing the flame location will change this temperature/concentration relationship. This is evident in Fig. 1b for diluted-ethylene/oxygen, where the flame sits close to the fuel. In contradistinction to the $C_2H_4$/air...
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flame, the mean concentration of oxidizer in the reaction zone of this flame is much greater than that of the fuel. Also, the fuel concentrations and temperatures are less than in Fig. 1a; thus, fuel pyrolysis chemistry will be inhibited. This implies that the conditions in the ethylene/air flame are more conducive to soot formation than those in the diluted-ethylene/oxygen flame, even though they both have the same flame temperature.

In what follows, this reasoning is evaluated by conducting counterflow experiments where soot-inception limits and temperature are measured for \( \text{C}_2\text{H}_4/\text{O}_2/\text{N}_2 \) flames over a wide range of stoichiometric mixture fractions. Coflow measurements of flame height and temperature at the onset of luminous soot are also made. Numerical computations with C2 chemistry are performed for the counterflow geometry to interpret the influence of \( Z_d \) on fuel pyrolysis reactions and the basic structure of the reaction zone. From these data, the effects of flame structure on soot formation are assessed.

EXPERIMENTAL AND NUMERICAL

The counterflow and coflow burners are similar to those described in Du et al. [9]. In the counterflow geometry a flame is established between two 11-mm opposed jets spaced 8 mm apart. An annular coflow of nitrogen is added to both sides to eliminate oxidizer entrainment and to extinguish the flame that would exist outside the region of interest. One difference between this burner and that of Ref. 9 is that in this burner the jets emanate from uniform cross-section tubes instead of high contraction nozzles. To ensure uniform laminar flow, a honeycomb core is placed 70 mm upstream of the exit of each tube and another is placed 70 mm upstream of the first. The burner is simple to construct and the resulting flow out of the tube is expected to be somewhere between potential and plug flow [10]. Measured flame-extinction strain rates for the methane/air flame and soot-inception limits for ethylene/air flames are in excellent agreement with those measured with the nozzle-type burners [9], suggesting that the characteristics of the flow are similar.

In the counterflow flame we measure the sooting-limit strain rate \( K_L \), which is based on soot luminosity. This limit is defined as the condition where the yellow luminous zone in the central region of the flame is no longer visible [9]. To approach the limit, we gradually increase the flow rate of both streams until the central region is pure blue. The limit is also obtained by starting with a blue flame and reducing the strain until yellow luminosity is observed in the central region. A measurement is not accepted until both conditions agree. Nonetheless, the measurement is somewhat subjective and efforts have been made to ensure the results are reproducible. After a limit condition has been established, the velocity profile along the stagnation streamline is measured with laser-Doppler velocimetry. \( K_L \) is identified as the magnitude of the slope of the velocity profile upstream of the flame [9].

The coflow flame is run in the overventilated mode and the flow rate of the mixture is increased until visible soot first appears in the flame. The height of the flame is then measured with a cathetometer to give a qualitative indication of time for soot inception.

Temperatures are measured with BeO–YtO₃ coated Pt–Rh thermocouples. Corrections for radiative heat loss are not made. In the counterflow flame the thermocouple lead wires are oriented parallel to the flame to minimize conduction errors, and temperatures are measured at a reference strain rate of 100 s⁻¹. This strain rate is sufficiently below the extinction strain rate that the temperature does not increase with decreasing strain rate.

The numerical scheme employed was developed by Smooke [11]. The version of the code used here assumes potential flow outside the mixing layer. The reaction mechanism is a 134-step C2 mechanism with 29 species and is identical to that in Ref. 12.

RESULTS AND DISCUSSION

Experimental

In Fig. 2 the effect of \( Z_d \) on soot inception chemistry for \( \text{C}_2\text{H}_4/\text{O}_2/\text{N}_2 \) flames with molar stoichiometry given by \( 1/3/11.28 \) is demonstrated by plotting the sooting-limit strain rate.
flames, with C$_2$H$_4$/O$_2$/N$_2$ stoichiometries of 1/3/9 and 1/3/5.68, have adiabatic flame temperatures of 2519 and 2740 K, respectively. Due to thermocouple limitations the temperatures of these flames could not be measured. Figure 3 shows that these flames have similar trends to those in Fig. 2 and the trends persist over a wide range of $Z_{st}$. Table 1 shows that for these flames, both fuel and oxidizer mass fractions vary over a wide range.

Since in mixture fraction space the flame structure of the ethylene coflow flame is similar to that shown in Fig. 1, we expect that soot inception should be inhibited for large $Z_{st}$ in this flame as well. To characterize the influence of $Z_{st}$ on particle inception chemistry we measured the flame height for the onset of soot. This soot-limit flame height is not the smoke point, but rather the height at which yellow luminosity first appears in a blue flame. Since varying the flame height varies the residence time in the flame, the greater the height required to first see luminous emission from soot, the slower the soot chemistry. In Fig. 4 the soot-limit flame height is plotted as a function of $Z_{st}$ for the C$_2$H$_4$/O$_2$/N$_2$ system given in Eq. 1. For low $Z_{st}$ the flame has a high propensity to soot and consequently, the height is very small. Thus, during these measurements the bulk of the flame is affected by heat/radiative loss to the rim and, as shown in Fig. 4, flame temperatures are reduced. Consequently, the invariance in soot height with $Z_{st}$ for $Z_{st} < 0.6$ is not an intrinsic characteristic of the flame, but rather is an anomaly associated with the soot height measurement. Considering instead the behavior of flames with

![Diagram](image)

**Fig. 2** Experimental results of the limiting strain rate for soot suppression $K_L$ as a function of stoichiometric mixture fraction $Z_{st}$ for counterflow flames of C$_2$H$_4$/O$_2$/N$_2$ with molar stoichiometry given by 1/3/11.28. Also shown is maximum flame temperature at a common strain rate of $K = 100$ s$^{-1}$. The thermocouple wire diameter was 0.003 in. and radiation corrections were not made.

$K_L$ against $Z_{st}$. Table 1 lists freestream mass fractions for representative values of $Z_{st}$. $K_L$ represents a characteristic rate for soot-particle inception such that the observed decrease in $K_L$ with $Z_{st}$ implies that soot inception is inhibited [9]. (Data for $Z_{st} > 0.16$ could not be obtained with the existing burner because the flames are not stable for $K < 50$ s$^{-1}$). Also shown in Fig. 2 are maximum flame temperatures. As anticipated, the flame temperature is not dependent on whether the inert is added to the fuel or oxidizer. Thus, these near-equidiffusional flames can be assumed to have almost identical temperatures in the limit of large Damköhler number.

The range of stoichiometric mixture fractions considered in Fig. 2 is rather small. To obtain a wider range we have also considered higher temperature ethylene flames. These

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**TABLE 1**

Representative Flames for Various Molar Stoichiometries and Stoichiometric Mixture Fractions

<table>
<thead>
<tr>
<th>Molar Stoichiometry</th>
<th>$T_{ad}$ (K)</th>
<th>$Z_{st}$</th>
<th>$Y_{O,0}$</th>
<th>$Y_{F,1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C$_2$H$_4$/O$_2$/N$_2$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1/3/11.28</td>
<td>2371</td>
<td>0.064</td>
<td>0.233</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.23</td>
<td>0.284</td>
<td>0.276</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.78</td>
<td>1.00</td>
<td>0.081</td>
</tr>
<tr>
<td>1/3/9</td>
<td>2519</td>
<td>0.074</td>
<td>0.276</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.27</td>
<td>0.350</td>
<td>0.276</td>
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<tr>
<td></td>
<td></td>
<td>0.41</td>
<td>0.432</td>
<td>0.182</td>
</tr>
<tr>
<td>1/3/5.68</td>
<td>2740</td>
<td>0.099</td>
<td>0.376</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.33</td>
<td>0.506</td>
<td>0.30</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.66</td>
<td>1.00</td>
<td>0.15</td>
</tr>
</tbody>
</table>
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Fig. 3. Experimental results of the limiting strain rate for soot suppression \( K_L \) as a function of stoichiometric mixture fraction \( Z_{st} \) for counterflow flames of \( \text{C}_2\text{H}_4/\text{O}_2/\text{N}_2 \) with molar stoichiometry given by 1/3/9 and 1/3/5.68.

Fig. 4. Experimental results of the flame height at the onset of soot luminosity as a function of stoichiometric mixture fraction \( Z_{st} \) for coflow diffusion flames of \( \text{C}_2\text{H}_4/\text{O}_2/\text{N}_2 \) with molar stoichiometry given by 1/3/11.28. Also shown is the measured flame temperatures at three heights in each flame: \( \circ \), the flame tip \(( y = H/2 \) and \( y = R \), one burner radius \( R \) \( ( y = R ) \); the thermocouple wire diameter was 0.002 in. and radiation corrections were not made.

larger \( Z_{st} \), where the sooting-limit flame height is larger than the burner tube radius, the height is seen to approach infinity as \( Z_{st} \) approaches 0.72. Though the counterflow and coflow flames are not completely analogous, the results demonstrate that the suppression of soot chemistry with increasing \( Z_{st} \) is common to both flames.

Numerical

To understand how the change in flame structure that accompanies a change in \( Z_{st} \) can influence soot inception we have performed numerical calculations for the ethylene counterflow flame. Due to computational limitations no attempt was made to include soot chemistry in the model; instead the detailed structure for the C2 chemistry is predicted and its influence on soot chemistry is then inferred.

Numerical experiments were performed at a strain rate of 100 s\(^{-1}\) and will be presented for the (1) ethylene/air flame \(( Z_{st} = 0.064 )\), and (2) diluted-ethylene/oxygen flame \(( 8.1 \text{ mole\% C}_2\text{H}_4, Z_{st} = 0.78 )\). We have chosen to numerically study the widest range of \( Z_{st} \) possible so as to better delineate the effects of flame structure. A solution for \( Z_{st} = 0.23 \) was also obtained and the results from \( Z_{st} = 0.064 \) to 0.78 were found to follow a consistent trend. However, for \( 0.064 < Z_{st} < 0.23 \) the change in \( Z_{st} \) is primarily due to changes in fuel, not oxygen, concentration (see Table 1). The numerical results show that in this range the effect of \( Z_{st} \) is to dilute the fuel and intermediates. They also show, as anticipated in Ref. 3, that the local fuel mass fraction at a given temperature is nearly proportional to the initial (freestream) mass fraction, and the intermediates scale accordingly. For example, at 1600 K, \( Y_F(Z_{st} = 0.78)/Y_F(Z_{st} = 0.064) = 0.11 \), while the ratio of the freestream mass fractions \( Y_F,(Z_{st} = 0.78)/Y_F,(Z_{st} = 0.064) \) is 0.08.

Figures 5 and 6 show the species and temperature profiles for \( Z_{st} = 0.064 \) and 0.78, respectively at a strain rate of 100 s\(^{-1}\). As with the experimental temperature measurements in Fig. 2, the numerical results show that the flame temperatures are almost identical for the two flames (2156 K for \( Z_{st} = 0.064 \) and 2139 K for \( Z_{st} = 0.78 \)). On the other hand, the temperature and species profiles are quite different and the convective flow field has changed not only in magnitude but also sign. For \( Z_{st} = 0.064 \) the flame is on the oxidizer side of the stagnation plane and flow in the soot zone is from the oxidizer to the fuel side whereas for \( Z_{st} = 0.78 \) the flame is on the fuel side and flow is from the fuel to oxidizer. This change in the direction of velocity in the soot zone could influence soot growth since the particles will be carried with the flow. For \( Z_{st} = 0.064 \) they will be carried into a fuel rich region where
growth is possible, whereas for $Z_{st} = 0.78$ the young soot particles will be convected towards the oxidizer and rapidly be consumed. Therefore, the influences of surface growth on the final soot loading can be dramatically different for flames on opposite sides of the stagnation plane. In this study we are considering only the effect of structure on particle inception. Experimentally, the onset of luminous emission is nearly coincident with the onset of particles [9]. Thus, the measured limits indicate whether local conditions are conducive to particle formation, not surface growth.

Comparing the concentration and temperature fields in Figs. 5 and 6 we find, as anticipated by the flame sheet solution in Fig. 1, that as $Z_{st}$ increases, the oxygen shifts towards the fuel side of the flame and its concentrations are much higher in the high temperature zone. The fuel is also shifted towards the fuel side but its concentrations are lower. To quantify this displacement of oxidizer and fuel we have plotted the C/O ratio as a function of distance in Figs. 5 and 6. Unlike premixed flames the C/O ratio is a strong function of position (mixture fraction); it is low on the oxidizer side and increases rapidly to infinity as the pyrolysis zone is approached. Nonetheless, experiments by Santoro et al. [13] and Gomez et al. [14] have shown that soot will not form below 1300 K in diffusion flames. Considering this temperature as a lower limit we find that the ethylene/air flame has a C/O functional dependence that implies soot can be formed over a majority of the fuel-side of the flame. Conversely, the C/O ratio in the diluted-ethylene/oxygen flame is less than 0.85 except where the temperature is less than 1300 K. Noting that based on thermodynamics the critical C/O ratio for the formation of soot is nearly unity and for premixed ethylene/air flames it is 0.6–0.7 [15], then it is not surprising that the $Z_{st} = 0.78$ flame is soot free. $Z_{st}$ describes a level of mixedness of the fuel and

![Fig. 5. Numerical results for an ethylene/air counterflow diffusion flame ($Z_{st} = 0.064$) at $K = 100$ s$^{-1}$ with C$_2$ chemistry and in physical coordinates showing profiles of selected species and temperature.](image)

![Fig. 6. Numerical results for an 8.1% ethylene/oxygen counterflow diffusion flame ($Z_{st} = 0.78$) at $K = 100$ s$^{-1}$ with C$_3$ chemistry and in physical coordinates showing profiles of selected species and temperature.](image)
oxidizer in the soot forming region and thus offers an indication of whether it is possible to form soot.

Now considering the detailed flame structure in Figs. 5 and 6 we see that when the oxygen shifts towards the fuel pyrolysis zone there is a corresponding shift in the radical and intermediate profiles as well. These shifts are readily interpreted: The OH and O profiles are tightly coupled to the oxygen concentration and temperature through \( H + O_2 \rightarrow OH + O \). The H profile is coupled to the OH, O, and H\(_2\) profiles through \( OH + H_2 \rightarrow H_2O + H \) and \( O + H_2 \rightarrow OH + H \). Relative to the OH and O profiles, it is shifted somewhat towards the fuel because \( H_2 \) is produced during fuel pyrolysis and is a maximum in the fuel pyrolysis zone. Finally, the intermediates are coupled to the fuel and H profiles through the H-abstraction reactions. The effects of these structural changes with \( Z_{st} \) on soot inception chemistry can now be discussed.

Evident from Fig. 6 is that soot inception should be inhibited in large \( Z_{st} \) flames because \( O_2 \) and \( OH \) persist well into the fuel side, suggesting that if precursors and soot particles could form, they would also oxidize. Comparing mass fractions of \( O_2 \) and \( OH \) at 1500 K, we find that for \( Z_{st} = 0.78 \), \( Y_{O_2} = 7 \times 10^{-4} \) and \( Y_{OH} = 2 \times 10^{-8} \), while for \( Z_{st} = 0.064 \), \( Y_{O_2} = 3 \times 10^{-4} \) and \( Y_{OH} = 2 \times 10^{-6} \). Though these results are dependent on the kinetic mechanism, they demonstrate a substantial increase in the amount of oxidizer in the soot inception zone. The \( O_2 \) will attack precursors and soot at higher temperatures, but its mole fractions within the pyrolysis zone are not significantly greater than those of \( OH \) (and in some cases are less), suggesting that \( OH \) is the dominant oxidizer.

The compositional shifts can also affect fuel pyrolysis, soot precursor and inception reactions. With increasing \( Z_{st} \) the fuel concentration decreases, which lowers the pyrolysis rate. Yet the H radical is also displaced to the fuel side, which may have an accelerative effect on soot formation. The C\(_2\) chemistry used here, though inadequate to model ring formation, PAH growth and particle formation, can elucidate the effects of \( Z_{st} \) on fuel pyrolysis reac-
tions, which are known to play a key role in soot formation in diffusion flames [16].

The most notable difference in composition between these two flames is the smaller mass fractions for C\(_2\)H\(_2\), CH\(_4\), CH\(_3\), C\(_2\)H, and H\(_2\) in the \( Z_{st} = 0.78 \) flame. The maximum C\(_2\)H\(_2\) mass fraction in Fig. 6 is 22\% of that in Fig. 5. A reduction in C\(_2\)H\(_2\) is expected because the inlet fuel mass fraction \( Y_{F,1} \) of Fig. 6 is only 8\% of that in Fig. 5 and, as noted previously, \( Y_F \) scales reasonably well with \( Y_{F,1} \) at a given temperature, even though the flame location is shifted. At 1500 K, where the mass fraction of C\(_2\)H\(_2\) is a maximum in both flames, \( Y_F(Z_{st} = 0.78)/Y_F(Z_{st} = 0.064) = 0.14 \). Therefore, we might expect that the maximum C\(_2\)H\(_2\) mass fraction in Fig. 6 would be 8\%-14\% of that in Fig. 5. It is closer to 22\% because the shift in the H profile, discussed above, causes the H concentration to be higher for a given temperature so that pyrolysis reactions are enhanced.

A comparison of the net consumption rates of H radicals in the region of C\(_2\)H\(_4\) shows that the H-abstraction reactions are much more active for \( Z_{st} = 0.78 \).

This result implies that, although increases in \( Z_{st} \) do decrease the rate of fuel pyrolysis, the effect may, in general, not be large enough to completely suppress soot. Our findings suggest that the suppression of soot occurs because the soot zone finds itself between two boundaries. On the fuel side the soot-inception zone is confined to occur at temperatures greater than \( \sim 1300 \) K; on the oxidizer side the zone is confined by \( OH \) concentration. Therefore, as \( Z_{st} \) is increased, this zone becomes increasingly narrow until the flame can no longer support soot particle formation.

This narrowing of the soot zone due to the shift in \( OH \) can be verified by considering what happens to the dark zone (the zone that is observed between the blue and yellow luminous zones in sooty flames) [17] if the oxygen index is increased in a neat fuel flame. The higher oxidizer concentration will shift the \( OH \) profile towards the fuel side of the flame and force the soot zone away from the location of maximum temperature \( X_f \). The amount of soot will increase because the temperature is increasing, but the edge of the soot zone on the
oxidizer-side will be further removed from $X_f$. This implies that the thickness of the dark zone should increase as the oxygen concentration increases even though the amount of soot may increase. This possibility has been confirmed in flames of ethylene burned in air, enriched oxygen and pure oxygen.

**CONCLUSIONS**

This study has evaluated how variations in $Z_{st}$ can affect flame structure and, consequently, soot-particle inception in diffusion flames. The results indicate that inception can be dramatically affected by flame structure through changes in fuel pyrolysis rates as well as oxidation rates adjacent to the pyrolysis zone. These findings, in addition to being of fundamental interest in understanding soot formation in diffusion flames, may be of practical interest as well, particularly when noting that it has been recently shown that increasing $Z_{st}$ strengthens the flame, making it more resistant to extinction by strain [18]. Therefore, if a portion of the nitrogen can be separated from the air feed, and redirected to the fuel, then the heat release and flame temperature can be maintained at that of the fuel burning in air, but soot formation will be suppressed and the flame will be strengthened. Recently, semipermeable membranes have been developed that satisfy these $N_2/O_2$ separation requirements quite well. The typical output of a commercial membrane system is a stream of 99.9% nitrogen by volume and another of 30% oxygen-enriched air [19]. If this $N_2$ stream is added to the fuel stream, the corresponding $Z_{st}$ is 0.4, which is sufficiently high to virtually eliminate soot in weakly strained ethylene flames.

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**REFERENCES**


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Comments

I. Kennedy, University of California, Davis, USA. Your figures show a linear relationship between the critical strain rate for suppression of inception and the stoichiometric mixture fraction. Is there a physical explanation for this relationship?

Authors’ Reply. To date we have not identified a simple phenomenological interpretation of the linear dependence of $K_L$ on $Z_a$. The consistent linear behavior in all three flames is intriguing but it is unknown whether this behavior can be extrapolated beyond the limited range of strain rates considered in these experiments. The process of soot inception in diffusion flames is complicated physico-chemical process involving fuel pyrolysis, precursor formation, nucleation as well oxidation. The change in structure with $Z_a$ will affect all of these processes and, while our results suggest that the rate of inception involves a balance between formation and oxidation, an interpretation of this linear relationship may not be possible without subsequent modeling, which we are presently engaged in.

M. B. Colket, United Technologies Research Center, USA. In an ethylene–air, opposed jet, diffusion flame, the stagnation plane is on the fuel side of the peak temperature. In such an environment, inception of soot particles and their growth occurs between the peak temperature regime and the stagnation plane. As they grow in size they are transported by convection to the lower temperature region of the flame (and grow by coagulation as they reach the stagnation plane). In flames in which the nitrogen is shifted to the fuel side, opposite physical conditions exist. You have interpreted your results principally, as being a chemical/dilution phenomena. To what extent do the above mentioned physical processes also contribute to your observed effects.

Authors’ Reply. The physical processes that you have described wherein the soot particles are convected by the streamlines, are relevant to the growth of soot particles after inception. However, our study has emphasized the inception process alone. The visual method that we use to identify $K_L$ has been shown to be highly effective at identifying the onset of soot inception [1]. Since these flames are virtually free of particles, particle transport does not influence our conclusions. This is an important point, particularly since it has been suggested in another paper in this Symposium [2] that particle transport is responsible for the elimination of soot when the flame is shifted to the fuel side. Our experimental observations and those of Atreya [3] (also see comment in Ref. 2) do not support this claim. Large soot concentrations have been measured in counterflow flames even when convection is driving the soot particles into the oxidizer. This is understandable because the inception region is finite and particle growth as well as inception can occur in this region.

REFERENCES