A theoretical study on soot inception in spherical burner-stabilized diffusion flames

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Received 24 July 2003; received in revised form 23 August 2004; accepted 23 August 2004
Available online 23 November 2004

Abstract

The soot inception processes in nonpremixed flames have been studied in a spherical flame stabilized by a spherical porous burner to understand the effects of flame structure (stoichiometric mixture fraction) and hydrodynamics (flow direction). A simplified three-step model with high activation energies is employed to describe the fuel oxidation, soot/precursor formation, and soot/precursor consumption reactions, respectively. The fuel oxidation reaction also produces a radical necessary for the soot formation reaction. The scheme yields three distinct reaction zones, one for each of the three reactions. The spherical flame geometry is unique in being able to independently control the flow direction, either from the fuel to oxidizer or from the oxidizer to fuel, and the flame structure through inert distribution. Four limiting flames, namely the fuel/air flame, diluted-fuel/oxygen flame, air/fuel flame and oxygen/diluted-fuel flame with the first reactant being the one supplied from the burner, are studied to address the relative importance of hydrodynamics and flame structure on soot inception. The analysis yields a solution giving the flame response to the variations of the soot formation and consumption reaction rates, the mass-flow rate issued from the burner, the Lewis numbers, and the inert distribution. Results show that with negligible soot consumption reaction, soot/precursor production is greater with slower radical diffusion. In the presence of soot consumption reaction, the total amount of soot/precursor decreases with lower soot/precursor diffusion rate. The effect of burner flow rate is qualitatively similar to that of the soot/precursor formation reaction rate except that the flame temperature increases monotonically with the flow rate until the adiabatic limit is reached. The production of soot/precursor can be significantly suppressed or eliminated by redistribution of the inert gas from the oxidizer side to the fuel side regardless of the flow direction.

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Keywords: Soot inception; Diffusion flames; Spherical; Microgravity; Asymptotic analysis

1. Introduction

Recently, a number of studies have shown that the sooting characteristics of fuels in diffusion flames can be dramatically affected by changes in the stoichiometric mixture fraction [1–7]. The stoichiometric mixture fraction, $Z_{st}$, is the value of the mixture frac-
## Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a_{i,j})</td>
<td>integration constants</td>
</tr>
<tr>
<td>(B_j)</td>
<td>preexponential factor of reaction (R_j)</td>
</tr>
<tr>
<td>(c_p)</td>
<td>specific heat at constant pressure</td>
</tr>
<tr>
<td>(D_i)</td>
<td>mass diffusion coefficient of species (i)</td>
</tr>
<tr>
<td>(D_{a_j})</td>
<td>Damköhler number of reaction (R_j)</td>
</tr>
<tr>
<td>(E_j)</td>
<td>activation temperature of reaction (R_j)</td>
</tr>
<tr>
<td>(F)</td>
<td>fuel</td>
</tr>
<tr>
<td>(L_{e_i})</td>
<td>Lewis number of species (i)</td>
</tr>
<tr>
<td>(m)</td>
<td>mass-flow rate</td>
</tr>
<tr>
<td>(O)</td>
<td>oxidizer</td>
</tr>
<tr>
<td>(P)</td>
<td>combustion products</td>
</tr>
<tr>
<td>(q_{F,1})</td>
<td>heat of combustion per unit mass of fuel consumed in the primary oxidation reaction</td>
</tr>
<tr>
<td>(q_{S,3})</td>
<td>heat of combustion per unit mass of soot/precursor consumed in the soot consumption reaction (R3)</td>
</tr>
<tr>
<td>(\tilde{q}_S)</td>
<td>heat of combustion ratio, proportional to (q_{S,3}/q_{F,1}), defined after Eq. (7)</td>
</tr>
<tr>
<td>(R)</td>
<td>radical</td>
</tr>
<tr>
<td>(r)</td>
<td>radial spatial coordinate</td>
</tr>
<tr>
<td>(r_f)</td>
<td>flame sheet location</td>
</tr>
<tr>
<td>(S)</td>
<td>soot/precursor</td>
</tr>
<tr>
<td>(S_{1A,B})</td>
<td>soot index at the A side of the flame sheet with reactant B supplied from the burner</td>
</tr>
<tr>
<td>(T)</td>
<td>temperature</td>
</tr>
<tr>
<td>(T_f)</td>
<td>flame temperature</td>
</tr>
<tr>
<td>(\tilde{T}_{f,j})</td>
<td>(j)th order expansion of the nondimensional flame temperature (\tilde{T}_f)</td>
</tr>
<tr>
<td>(u)</td>
<td>radial flow velocity</td>
</tr>
<tr>
<td>(W_i)</td>
<td>molecular weight of species (i)</td>
</tr>
<tr>
<td>(Y_i)</td>
<td>mass fraction of species (i)</td>
</tr>
<tr>
<td>(Y_{i,b})</td>
<td>mass fraction of the reactant supplied from the burner</td>
</tr>
<tr>
<td>(Z)</td>
<td>mixture fraction</td>
</tr>
<tr>
<td>(Z_{st})</td>
<td>stoichiometric mixture fraction</td>
</tr>
<tr>
<td>(\zeta)</td>
<td>stretched coordinate in the soot/precursor formation and consumption regions, defined as ((\tilde{r} - \tilde{r}_f)/\delta)</td>
</tr>
<tr>
<td>(\theta_j)</td>
<td>(j)th order expansion of temperature in the oxidation region</td>
</tr>
<tr>
<td>(\Theta_j)</td>
<td>(j)th order expansion of temperature in the soot/precursor formation and consumption regions</td>
</tr>
<tr>
<td>(\lambda)</td>
<td>thermal conductivity</td>
</tr>
<tr>
<td>(\Lambda_i)</td>
<td>reduced Damköhler numbers defined in Eqs. (38) and (39)</td>
</tr>
<tr>
<td>(v)</td>
<td>stoichiometric coefficient</td>
</tr>
<tr>
<td>(\tilde{v}_F)</td>
<td>stoichiometric coefficient ratio defined as (v_{F,2}/v_{F,1})</td>
</tr>
<tr>
<td>(\tilde{v}_O)</td>
<td>stoichiometric coefficient ratio defined as (v_{O,3}/v_{O,1})</td>
</tr>
<tr>
<td>(\hat{v})</td>
<td>stoichiometric coefficient ratio defined as (\tilde{v}_F/\tilde{v}_O)</td>
</tr>
<tr>
<td>(v^*)</td>
<td>stoichiometric coefficient ratio defined as ((1 + \tilde{v}_O)/(1 + \tilde{v}_F))</td>
</tr>
<tr>
<td>(\xi)</td>
<td>stretched coordinate in the oxidation region, defined as ((\tilde{r} - \tilde{r}_f)/\varepsilon)</td>
</tr>
<tr>
<td>(\rho)</td>
<td>gas density</td>
</tr>
<tr>
<td>(\sigma)</td>
<td>stoichiometric oxidizer to fuel mass ratio</td>
</tr>
<tr>
<td>(\phi_{i,j})</td>
<td>(j)th order expansion of species (i) in the oxidation region</td>
</tr>
<tr>
<td>(\phi_{i,j})</td>
<td>(j)th order expansion of species (i) in the soot/precursor formation and consumption regions</td>
</tr>
</tbody>
</table>

### Subscripts

- \(b\) | conditions at the burner exit |
- \(F\) | fuel |
- \(O\) | oxidizer |
- \(R\) | radical |
- \(S\) | soot/precursor |
- \(T\) | temperature |
- \(\infty\) | conditions at \(r \to \infty\) |

### Superscripts

- \(-\) | solutions in the region between the burner and the flame sheet |
- \(+\) | solutions in the region outside of the flame sheet |
- \(\sim\) | nondimensional quantities |
tion $Z$ at the flame sheet, and can be written as

$$Z_{st} = Y_{O,b}/(Y_{O,b} + \sigma Y_{F,b}),$$

where

$$\sigma = (v_O W_O)/(v_F W_F)$$

is the stoichiometric oxidizer to fuel mass ratio, and the subscript “b” denotes conditions of the supply gases. All other notations are standard and are defined in Section 2.

Increasing $Z_{st}$ reduces soot formation and can eventually lead to complete elimination of soot formation and, thus, blue flames. These changes are observed even when $Z_{st}$ is varied in such a way that the flame temperature is invariant. Studies have been conducted in counterflow and coflow flames with qualitatively similar results. More recently experiments were performed with spherical flames produced in microgravity by Sunderland et al. [8]. The goal of those experiments was to evaluate the controlling mechanism for soot suppression with $Z_{st}$. Two possible mechanisms have been suggested to explain the observed results: one involving hydrodynamics and the other flame structure. Hydrodynamically, soot can be suppressed by increasing $Z_{st}$ because in the coflow and counterflow flames the direction of convection across the flame front is reversed [1,2]. For $Z_{st}$ less than 0.5 the streamlines are from oxidizer to fuel and for $Z_{st}$ greater than 0.5 they are from fuel to oxidizer. This reversal can lead to soot suppression because the heavier soot/precursors and particles will tend to follow the streamlines and be oxidized. The structural explanation is based on the variations of temperature and concentration fields that occur when $Z_{st}$ is varied. At low $Z_{st}$, the concentration of carbon in the high-temperature region is high while the concentration of oxidizer is low. On the other hand, at high $Z_{st}$ the oxidizer concentration is high and the carbon concentration is low. Consequently, low $Z_{st}$ conditions are conducive to soot formation and high $Z_{st}$ conditions are not.

While both mechanisms can explain the suppression of soot formation at high $Z_{st}$ in coflow or counterflow flames, it is not clear whether one mechanism is dominant. Recently microgravity experiments [8] were performed to attempt to evaluate this issue. The present work is intended to seek further insight into these observations using asymptotic methods.

The unique benefits that microgravity spherical flames afford in understanding the effects of $Z_{st}$ have been described in Sunderland et al. [8]. Briefly, the one-dimensional geometry allows for the independent variation of flame structure ($Z_{st}$) and convection direction (hydrodynamics). Flame structure is varied by removing inert from the oxidizer and adding it to the fuel in such a way that the adiabatic flame temperature $T_{ad}$ is not varied. This approach, called inert exchange, has been shown to be very effective in counterflow and coflow flames and $Z_{st}$ can be varied from about 0.05 to 0.8 without varying $T_{ad}$ [2,3]. The direction of convection can be varied by exploiting the unique aspects of spherical flames. This can be appreciated by referring to Fig. 1. By choosing either the oxidizer in the ambient or fuel in the ambient, the streamlines are forced to go from either the fuel to oxidizer or the oxidizer to fuel, respectively. In this way, one can study the effects of convection direction for a given $Z_{st}$ or the effects of $Z_{st}$ for a given direction of convection.

Despite the apparent simplicity of these experiments there are complexities associated with the interpretation of results because residence times and radiative heat losses also vary. To gain insight into the experiments and to better understand the effects of flame structure and the effects of $Z_{st}$, we develop an analytical approach to model the flame. The approach employs a three-step chemical mechanism adopted in our earlier paper [7] to bring out the salient features of the effects of flame structure. Specifically, the mechanism includes a fuel oxidation step where the fuel is converted to products plus a radical. This step represents the fact that in the pyrolysis of the fuel, hydrogen is released, contributing to the H atoms that are needed for soot inception. The second step describes the process wherein this radical is employed in fuel pyrolysis, PAH growth, and particle inception. To simplify the system the second step assumes that the formation of soot, or more precisely soot/precursors, is from a reaction between the fuel and the radical. Effectively, what is being assumed is that the soot/precursor concentration is dependent on fuel concentration, e.g., the acetylene concentration is proportional to fuel concentration in the pyrolysis zone [2]. The third step is a soot/precursor oxidation step wherein the soot/precursor is oxidized by oxygen. Of course, in this analytical study this is analogous to considering that the concentration of the oxidizing species, e.g., OH, is proportional to the oxygen concentration.

With the analytical model we are able to evaluate whether the simplified mechanism captures the dominant features of the soot process and how structure influences the process. Furthermore, the relative importance of hydrodynamics and flame structure can be assessed.

2. Formulation

The problem of interest is a steady diffusion flame stabilized by a spherical porous burner maintained at a
constant temperature $T_b$ as shown in Fig. 1. A gas reactant, either the fuel or the oxidizer, is injected from the burner into an infinite quiescent environment of the other reactant at a temperature $T_{\infty}$. The burner is assumed to be ideal and the gravity is considered negligible so that the flow is spherically symmetric. The assumption of constant $T_b$ implies that the burner is cooled by a coolant and all the heat transferred to the burner is removed. A three-step representative reaction scheme following Chao et al. [7] is adopted for this analysis, and is given by

\begin{align}
\nu F + v_{O,1} O & \rightarrow \nu_{P,1} P + v_R R, \quad \text{(R1)} \\
\nu F + v_R R & \rightarrow \nu S, \quad \text{(R2)} \\
\nu S + v_{O,3} O & \rightarrow \nu_{P,3} P, \quad \text{(R3)}
\end{align}

where $\nu$ is the stoichiometric coefficient. These reaction steps respectively represent oxidation (R1), formation of soot/precursor (R2), and consumption of soot/precursor (R3). In the oxidation reaction, fuel ($F$) and oxidizer ($O$) are consumed in an oxidation region to produce final products $P$ and radical $R$ (e.g., H). Part of the radical is transported toward the fuel side and reacts with the fuel to produce the soot/precursor in a formation region adjacent to the oxidation region. Part of the soot/precursor subsequently penetrates through the oxidation region into the oxidizer side, and reacts with the oxidizer in a consumption region to produce more final products. Recognizing that the presence of soot is dependent on the presence of soot/precursors, the soot growth reaction will not be included and $S$ will herein be referred to as soot/precursor. For the case of fuel supplied from the burner, Region A in Fig. 1 is the soot formation region while Region B is the soot consumption region. For the case of oxidizer supplied from the burner, Region B is the soot formation region while Region A is the soot consumption region.

All three reactions are considered to follow second-order Arrhenius kinetics. Conventional assumptions are adopted to facilitate the analysis, including constant specific heat at constant pressure $c_p$, thermal conductivity $\lambda$, and the mass diffusivity of all species $\rho D_i$, where $\rho$ is the gas density and $D_i$ the mass diffusion coefficient of species $i$. Because the soot/precursor formation reaction (R2) is only slightly endothermic, this reaction will be assumed thermal neutral. Moreover, since the effects of fuel and oxidizer diffusivity are already well known [9], the Lewis numbers of the fuel and oxidizer are taken to be unity for simplicity. Radiative heat loss is not considered because the objective of this study is to isolate the fundamental effects of flame structure and flow direction, not to predict the effects of radiation on flame temperature, which can be significant in microgravity flames. With the above assumptions, the nondimensional conservation equations are given by

\begin{align}
\frac{1}{\tau^2} \left[ \frac{m}{d\tau} \frac{dF}{d\tau} - \frac{d}{d\tau} \left( \frac{r^2 dF}{d\tau} \right) \right] & = Da_1 \tilde{Y}_F \tilde{Y}_O \exp \left( -\frac{E_1}{T} \right) \\
& + \beta \tilde{q}_S Da_2 \tilde{Y}_S \tilde{Y}_O \exp \left( -\frac{\alpha E_2}{T} \right), \quad \text{(1)} \\
\frac{1}{\tau^2} \left[ \frac{m}{d\tau} \frac{dF}{d\tau} - \frac{d}{d\tau} \left( \frac{r^2 dF}{d\tau} \right) \right] & = -Da_1 \tilde{Y}_F \tilde{Y}_O \exp \left( -\frac{E_1}{T} \right) \\
& - Da_2 \tilde{Y}_F \tilde{Y}_R \exp \left( -\frac{E_2}{T} \right), \quad \text{(2)} \\
\frac{1}{\tau^2} \left[ \frac{m}{d\tau} \frac{dO}{d\tau} - \frac{d}{d\tau} \left( \frac{r^2 dO}{d\tau} \right) \right] & = -Da_1 \tilde{Y}_F \tilde{Y}_O \exp \left( -\frac{E_1}{T} \right) \\
& - \frac{\beta}{\nu} Da_2 \tilde{Y}_S \tilde{Y}_O \exp \left( -\frac{\alpha E_2}{T} \right), \quad \text{(3)} \\
\frac{1}{\tau^2} \left[ \frac{m}{d\tau} \frac{dR}{d\tau} - \frac{1}{Le_R} \frac{d}{d\tau} \left( \frac{r^2 dR}{d\tau} \right) \right] & = Da_1 \tilde{Y}_F \tilde{Y}_O \exp \left( -\frac{E_1}{T} \right) \\
& - \frac{1}{\nu} Da_2 \tilde{Y}_F \tilde{Y}_R \exp \left( -\frac{E_2}{T} \right), \quad \text{(4)}
\end{align}
\[
\frac{1}{r^2} \left[ m \frac{d \tilde{Y}_S}{d\tilde{r}} - \frac{1}{Le_i d\tilde{r}} \left( \frac{\partial^2 d \tilde{Y}_S}{\partial \tilde{r}^2} \right) \right] = Da_2 \tilde{Y}_F \tilde{Y}_R \exp \left( -\frac{\tilde{E}_2}{T} \right)
- \beta Da_2 \tilde{Y}_S \tilde{Y}_O \exp \left( -\alpha \frac{\tilde{E}_2}{T} \right),
\]

which are to be solved subject to the boundary conditions:

\[
\begin{align}
\tilde{r} &= 1: \quad \tilde{T} = \tilde{T}_b, \quad \tilde{Y}_i - Le_i^{-1} \left( d\tilde{Y}_i / d\tilde{r} \right) = 0; \quad i = R, S, \\
\tilde{m} \tilde{Y}_F - (d\tilde{Y}_F / d\tilde{r}) = \tilde{m} \tilde{Y}_{F,b}, \\
\tilde{m} \tilde{Y}_O - (d\tilde{Y}_O / d\tilde{r}) = 0,
\end{align}
\]

(fuel from the burner),

\[
\begin{align}
\tilde{m} \tilde{Y}_F - (d\tilde{Y}_F / d\tilde{r}) = 0, \\
\tilde{m} \tilde{Y}_O - (d\tilde{Y}_O / d\tilde{r}) = \tilde{m} \tilde{Y}_{O,b}
\end{align}
\]

(oxidizer from the burner),

\[
\tilde{r} \to \infty: \quad \tilde{T} = \tilde{T}_\infty, \quad \tilde{Y}_i \to 0; \quad i = R, S, \\
\tilde{Y}_F \to 0, \quad \tilde{Y}_O \to \tilde{Y}_{O,\infty}
\]

(fuel from the burner),

\[
\tilde{Y}_F \to \tilde{Y}_{F,\infty}, \quad \tilde{Y}_O \to 0
\]

(oxidizer from the burner).

In the above, the equations are nondimensionalized by

\[
\begin{align}
\tilde{Y}_F &= Y_F, \quad \tilde{Y}_O = \frac{v_{F,1} W_F}{v_{O,1} W_O} Y_O, \\
\tilde{Y}_R &= \frac{v_{F,1} W_F}{v_{R} W_R} Y_R, \quad \tilde{Y}_S = \frac{v_{F,2} W_F}{v_{S} W_S} Y_S, \quad \tilde{r} = \frac{r}{r_b},
\end{align}
\]

\[
\tilde{m} = \frac{c_p m}{4\pi r_b \lambda}, \quad \tilde{T} = \frac{c_p T}{q_{F,1}}, \quad Le_i = \frac{\lambda}{c_p \rho D_i},
\]

\[
\begin{align}
\alpha &= \frac{E_3}{E_2}, \quad \tilde{E}_j = \frac{c_p E_j}{q_{F,1}}, \quad Da_1 = \frac{r_b^2 c_p v_{O,1} W_O B_1}{\lambda}, \\
Da_2 &= \frac{r_b^2 c_p v_{F} v_{R} W_R B_2}{\lambda}, \quad \tilde{q}_S = \frac{v_S W_S q_{S,3}}{v_{F,2} W_F q_{F,1}},
\end{align}
\]

\[
\tilde{v}_F = \frac{v_{F,2}}{v_{F,1}}, \quad \tilde{v}_O = \frac{v_{O,3}}{v_{O,1}}, \quad \tilde{v}_S = \frac{v_S W_S q_{S,3}}{v_{F,2} W_F q_{F,1}}
\]

where \( T \) is the temperature, \( Y_i \) the mass fraction of species \( i \), \( r \) the radial spatial coordinate, \( r_b \) the radius of the burner, \( q_{F,1} \) the heat of combustion per unit mass of fuel consumed in the oxidation reaction, \( q_{S,3} \) the heat of combustion per unit mass of soot/precursor consumed in the soot consumption reaction, \( W_i \) the molecular weight of species \( i \), \( B_j \) and \( E_j \) the preexponential factor and activation temperature of reaction \( R_j \), \( Le_i \) the Lewis number of species \( i \), \( Da_j \) the Damköhler number of reaction \( R_j \), and \( \alpha \) and \( \beta \) the activation energy ratio and Damköhler number ratio between the soot/precursor consumption and formation reactions. The mass-flow rate, given by \( m = 4\pi r^2 \rho u \), where \( u \) is the radial flow velocity, is a constant from the continuity equation and controllable. Because the radical \( R \) (e.g., \( H \)) is generally lighter and the soot/precursor is heavier than the background gas, it is considered that \( Le_R < 1 \) and \( Le_S > 1 \). The Damköhler numbers are defined following that of the droplet burning because of the similarity in their geometry.

To perform the analysis it must be recognized that all three reactions (\( R_1 \)–(\( R_3 \)) are confined to regions much thinner than the characteristic diffusion zone and that these regions are located near the flame sheet where stoichiometry between the fuel and the oxidizer is attained. The small parameter \( \varepsilon \) then is designated to be the ratio of the characteristic length scales for the oxidation and diffusion regions, and \( \delta \) is the ratio for the soot/precursor formation and diffusion regions. The solutions are expanded using \( \varepsilon \) and \( \delta \) where \( 1 \gg \delta \gg \varepsilon \). The logic of this ordering of length scales is based on the fact that experiments have shown that the minimum temperature required for soot formation in diffusion flames ranges from 1250 to 1650 K [10–14]. Thus, the region of soot/precursor formation is broad compared to the fuel oxidation zone. The soot/precursor consumption reaction, being driven by radicals, can occur at similar temperatures [15,16] such that its length scale is comparable to that of the soot/precursor formation reaction. Stated in asymptotic terms, the soot/precursor formation and consumption temperatures are sufficiently lower than the flame temperature of hydrocarbon–air flames that the effective activation energy of the soot/precursor formation reaction is much smaller than the activation energy of the fuel oxidation reaction (\( E_1 \gg E_2 \)), and the activation energies of the soot/precursor formation and consumption reactions are of the same order.

Equations (1)–(7) are then solved by activation energy asymptotics with the solutions between the burner and the flame sheet designated by superscript “–” while those outside of the flame sheet by superscript “+”. In the chemically inert outer regions, the process is controlled by the balance between diffusion and convection transport. Solving the source free form of Eqs. (1)–(5) subject to the boundary conditions in Eqs. (6) and (7), we obtain the outer solutions with those of the temperature, radical, and
Because the activation energy for the oxidation reaction is high, only an \( O(\epsilon) \) reactant leakage through the oxidation region is possible. Thus \( \bar{Y}_F \) and \( \bar{Y}_O \) are \( O(\epsilon) \) quantities and their detailed expressions are not important to this study. Similarly, for the case of oxidizer supplied from the burner, we have

\[
\bar{Y}_F^+ = \bar{Y}_{F,b} - \left[ \left[ a_{F,0}^+ + \Delta a_{F,1}^+ + O(\delta^2) \right] + O(\epsilon) \right] \times \exp(-\bar{m}/\bar{r}), \tag{12}
\]

\[
\bar{Y}_O^+ = \bar{Y}_{O,\infty} - \left[ a_{O,0} + \Delta a_{O,1}^+ + O(\delta^2) \right] + O(\epsilon) \times \left[ 1 - \exp(-\bar{m}/\bar{r}) \right]. \tag{13}
\]

Because the activation energy for the oxidation reaction is high, only an \( O(\epsilon) \) reactant leakage through the oxidation region is possible. Thus \( \bar{Y}_F^- \) and \( \bar{Y}_O^- \) are \( O(\epsilon) \) quantities and their detailed expressions are not important to this study. Similarly, for the case of oxidizer supplied from the burner, we have

\[
\bar{Y}_F^- = \bar{Y}_{F,b} - \left[ \left[ a_{F,0}^- + \Delta a_{F,1}^- + O(\delta^2) \right] + O(\epsilon) \right] \times \exp(-\bar{m}/\bar{r}), \tag{14}
\]

\[
\bar{Y}_O^- = \bar{Y}_{O,\infty} - \left[ a_{O,0}^- + \Delta a_{O,1}^- + O(\delta^2) \right] + O(\epsilon) \times \left[ 1 - \exp(-\bar{m}/\bar{r}) \right]. \tag{15}
\]

with \( \bar{Y}_F^- \) and \( \bar{Y}_O^- \) less important \( O(\epsilon) \) quantities. In the above, \( a_{i,j}^\pm \) are integration constants to be determined from the analysis.

The key steps for the rest of the analysis are presented in Appendix A and the detailed analysis can be found in Liu [17]. In the solution, the flame temperature is expressed by

\[
\tilde{T}_f = \tilde{T}_{f,0} - \delta \tilde{T}_{f,1} + O(\delta^2) \tag{16}
\]

and the location of the flame sheet is expressed by \( \tilde{r}_f \).

For the case of fuel supplied from the burner, the analysis yields nine algebraic equations,

\[
\frac{\tilde{T}_{f,0} - \tilde{T}_b}{1 - \exp(\bar{m}/\bar{r}_f)} + \frac{\tilde{T}_{f,0} - \tilde{T}_\infty}{\exp(\bar{m}/\bar{r}_f) - 1} = \frac{1 - \hat{q}_S\hat{v}}{1 + \hat{v}_F} \bar{Y}_{F,b} + \frac{\hat{q}_S\hat{v}\bar{Y}_{O,\infty}}{\exp(\bar{m}/\bar{r}_f) - 1} \tag{17}
\]

\[
a_{T,0} = \frac{(\tilde{T}_{f,0} - \tilde{T}_b)}{[\exp(-\bar{m}/\bar{r}_f) - \exp(-\bar{m})]}, \tag{18}
\]

\[
a_{F,0} = \frac{\bar{Y}_{F,b}\exp(\bar{m}/\bar{r}_f)}{\bar{m} - \tilde{T}_b}, \tag{19}
\]

\[
a_{T,0} = \frac{(\tilde{T}_{f,0} - \tilde{T}_\infty)}/[1 - \exp(-\bar{m}/\bar{r}_f)], \tag{20}
\]

\[
a_{O,0} = \frac{\bar{Y}_{O,\infty}}{1 - \exp(-\bar{m}/\bar{r}_f)}, \tag{21}
\]

\[
a_{R,0} = a_{F,0}^+ = 0, \tag{22}
\]

\[
a_{S,0} = \frac{a_{S,0}}{\exp(\bar{m}/\bar{r}_f) - 1} = \hat{v} \left[ \frac{\bar{Y}_{F,b}}{\exp(\bar{m}/\bar{r}_f) - 1} \right], \tag{23}
\]

to determine the 10 leading order constants, \( \tilde{T}_{f,0}, \tilde{r}_f, a_{T,0}, a_{F,0}, a_{O,0}, a_{R,0}, \) and \( a_{S,0} \), as well as six algebraic equations,

\[
\begin{align*}
(a_{F,1}^+ - a_{O,1}^-) & = \bar{Y}_{F,1} \left[ 1 - \exp(-\bar{m}/\bar{r}_f) \right] \\
-a_{F,1}^- & = \bar{Y}_{F,1}\exp(-\bar{m}/\bar{r}_f) \\
(a_{F,1}^- - a_{O,1}^+) & = \bar{Y}_{F,1}\exp(-\bar{m}/\bar{r}_f) + a_{O,1}^- \hat{v} \\
& = (a_{S,1}^- + a_{S,1}^-) \exp(-\bar{m}/\bar{r}_f) - a_{S,1}^+, \tag{26}
\end{align*}
\]

\[
\begin{align*}
& = \bar{v}_F (1 - \hat{q}_S\hat{v}) a_{R,1}^+, \tag{27}
& = \bar{v}_F (1 - \hat{q}_S\hat{v}) a_{R,1}^+ + (\hat{v} - 1) a_{S,1}^+, \tag{28}
& = 0, \tag{29}
& = \bar{Y}_{F,1} = -a_{T,1} \left[ \exp(-\bar{m}/\bar{r}_f) - \exp(-\bar{m}) \right]. \tag{29}
\end{align*}
\]

to determine the 9 \( O(\delta) \) constants, \( \tilde{T}_{f,1}, a_{T,1}^+, a_{F,1}, a_{O,1}^- + a_{S,1}^+, a_{R,1}^+, \) and \( a_{S,1}^- \). Four additional conditions are required to close the problem, which are provided by solving two nonlinear ordinary differential equations numerically with excess boundary conditions, given by

\[
\begin{align*}
d^2\Phi_{F,1}/d\xi^2 & = \Lambda_2 \Phi_{F,1} \Phi_{R,1} \exp(-\Theta_{F,1}^-), \tag{30}
\Phi_{F,1}(\xi \to -\infty) & = -a_{F,1}^- \exp(-\bar{m}/\bar{r}_f) \\
& = (-\bar{Y}_{F,b}\bar{m}/\bar{r}_f^2) \xi, \tag{31}
\Phi_{F,1}(\xi = 0) = 0, \tag{32}
\end{align*}
\]

\[
\begin{align*}
(d\Phi_{F,1}/d\xi)|_{\xi = 0} & = -\bar{m}\bar{Y}_{F,b}/\bar{r}_f^2 \left[ 1 + \bar{v}_F \right], \tag{33}
\end{align*}
\]

\[
\begin{align*}
da^2\Phi_{O,1}/d\xi^2 & = \Lambda_3 \Phi_{O,1} \Phi_{O,1} \exp(-\Theta_{O,1}^+), \tag{34}
\Phi_{O,1}(\xi \to \infty) = -a_{O,1}^+ \left[ 1 - \exp(-\bar{m}/\bar{r}_f) \right] + a_{O,0}(\bar{m}/\bar{r}_f) \exp(-\bar{m}/\bar{r}_f), \tag{35}
\end{align*}
\]
\[ \Phi_{O,1}^+(\zeta = 0) = 0, \quad (d \Phi_{O,1}/d \zeta)_{\zeta = 0} = \hat{m} \hat{Y}_{F,b}/[T_f^2(1 + \hat{v}_F)]. \quad (36) \]

In Eqs. (30) and (34),

\[ \Lambda_2 = 3^3 \text{Da}_2 \exp(-\hat{E}_2/\hat{T}_{f,0}) \quad (38) \]

is the reduced Damköhler number for the soot/precursor formation reaction,

\[ \Lambda_3 = 3^2(\beta/\hat{v})\text{Da}_2 \exp(-a \hat{E}_2/\hat{T}_{f,0}) \quad (39) \]

is the reduced Damköhler number of the soot/precursor consumption reaction, and

\[ \Theta_1^- = -a_{T,1}^- \left[ \exp\left( -\frac{\hat{m}}{R_f} \right) - \exp(-\hat{m}) \right] \]
\[ - \frac{\hat{m}}{R_f} \left[ \exp(\hat{m}/\hat{r}_f) - \exp(-\hat{m}) \right] \zeta, \quad (40) \]

\[ \Phi_{R,1}^- = a_{R,1}^- \exp(-L_e \hat{m}/\hat{r}_f) \]
\[ + (L_e \hat{v})[\Phi_{F,1} + a_{F,1}^- \exp(-\hat{m}/\hat{r}_f)] \]
\[ + \hat{Y}_{F,0} \left( \hat{m}/\hat{r}_f \right)^{\frac{1}{2}} \zeta, \quad (41) \]

\[ \Phi_{S,0}^+ = a_{S,0}^+ \left[ 1 - \exp(-L_e \hat{m}/\hat{r}_f) \right], \quad (42) \]

\[ \Theta_1^+ = \hat{g} \hat{S} \Phi_{O,1}^+ \]
\[ + (a_{O,1}^+ \hat{S} \hat{v} - a_{T,1}^+) \left[ 1 - \exp\left( -\frac{\hat{m}}{R_f} \right) \right] \]
\[ + \hat{m} \hat{g} \hat{v} \hat{Y}_{O,\infty} \left( \frac{\hat{r}_f}{\exp(\hat{m}/\hat{r}_f) - 1} - \hat{g} \hat{S} \hat{v} \hat{Y}_{O,\infty} \right) \zeta. \quad (43) \]

The soot/precursor concentration at the boundary between the soot/precursor formation region and the fuel side inert region is defined as a soot index \( S_{I,F,F} \) whose value is obtained by evaluating \( \hat{Y}_S \) at the flame sheet as

\[ S_{I,F,F} = \hat{Y}_S(\hat{r}_f) = (a_{S,0}^- + \delta a_{S,1}^-) \]
\[ \times \exp(-L_e \hat{m}/\hat{r}_f) + \cdots. \quad (44) \]

The soot/precursor concentration at the boundary between the soot/precursor consumption region and the oxidizer side inert region is defined as another soot index \( S_{I,O,F} \) with its value given by

\[ S_{I,O,F} = \hat{Y}_S(\hat{r}_f) = (a_{S,0}^+ + \delta a_{S,1}^+) \]
\[ \times \left[ 1 - \exp(-L_e \hat{m}/\hat{r}_f) \right] + \cdots. \quad (45) \]

The subscript “F” after the “,” in Eqs. (44) and (45) represents the case of fuel supplied from the burner and the subscript “F” and “O” before the “,” represent the side of the flame sheet where the soot index is evaluated.

Similarly, for the case of oxidizer supplied from the burner, we have Eqs. (18), (20), (22), and

\[ \tilde{T}_{f,0} - \tilde{T}_b \]
\[ = 1 - \frac{\hat{q} \hat{S} \hat{v} \hat{Y}_{O,b}}{1 + \hat{v}_F \exp(\hat{m}/\hat{r}_f) - 1} \exp(\hat{m}/\hat{r}_f) - 1 \]
\[ = \frac{1 - \hat{q} \hat{S} \hat{v} \hat{Y}_{O,\infty}}{1 + \hat{v}_F \exp(\hat{m}/\hat{r}_f) - 1} + \hat{q} \hat{S} \hat{v} \hat{Y}_{O,b}, \quad (46) \]

\[ \tilde{a}_{O,0}^- = \hat{Y}_{O,b} \exp(\hat{m}/\hat{r}_f), \quad (47) \]

\[ \tilde{a}_{F,0}^+ = \hat{Y}_{F,\infty}/[1 - \exp(\hat{m}/\hat{r}_f)], \quad (48) \]

\[ \tilde{a}_{S,0}^- = \frac{a_{S,0}^-}{\exp(Le \hat{m}/\hat{r}_f) - 1} \]
\[ = \tilde{v} \left[ \frac{v^* \hat{Y}_{F,\infty}}{\exp(\hat{m}/\hat{r}_f) - 1} - \hat{Y}_{O,b} \right] \quad (49) \]

to determine \( \tilde{T}_{f,0}, \tilde{r}_f, a_{T,0}^+, a_{F,0}^+, a_{O,0}^-, a_{R,0}^-, \) and \( a_{S,0}^- \), as well as

\[ (a_{T,1}^- - a_{O,1}^- \hat{q} \hat{S} \hat{v}) \exp(-\hat{m}/\hat{r}_f) - a_{T,1}^- \exp(-\hat{m}) \]
\[ = a_{F,1}^- [1 - \exp(-\hat{m}/\hat{r}_f)], \quad (50) \]

\[ a_{F,1}^-(Le \hat{v}/\hat{F}) [1 - \exp(-\hat{m}/\hat{r}_f)] \]
\[ = (a_{R,1}^- + a_{O,1}^-) \exp(-Le \hat{R} \hat{m}/\hat{r}_f) - a_{R,1}^- \]
\[ = Le \hat{S} [a_{F,1}^- \exp(-\hat{m}/\hat{r}_f) - a_{F,1}^-] \]
\[ = (a_{S,1}^- + a_{S,1}^+) \exp(-Le \hat{S} \hat{m}/\hat{r}_f) - a_{S,1}^-, \quad (52) \]

\[ (1 + \hat{v}_F) [a_{T,1}^- \exp(-\hat{m}) + a_{T,1}^+] \]
\[ = (1 - \hat{q} \hat{S} \hat{v}) (a_{F,1}^+ + \hat{F} a_{R,1}^-), \quad (53) \]

\[ \tilde{v} [a_{F,1}^+ v^* - a_{R,1}^- (v^* - 1)] - a_{S,1}^+ = 0, \quad (54) \]

\[ \tilde{T}_{f,1} = -a_{T,1}^- [1 - \exp(-\hat{m}/\hat{r}_f)], \quad (55) \]

to determine \( \tilde{T}_{f,1}, a_{T,1}^+, a_{F,1}^+, a_{O,1}^-, a_{R,1}^-, \) and \( a_{S,1}^- \). Four additional conditions are provided by solving the following two nonlinear ordinary differential equations numerically with excess boundary conditions,

\[ d^2 \Phi_{F,1}^+/d \zeta^2 = A_2 \Phi_{F,1}^+ + \Phi_{R,1}^+ \exp(-\Theta_1^-), \quad (56) \]

\[ \Phi_{F,1}^+(\zeta \rightarrow \infty) = -a_{F,1}^- [1 - \exp(-\hat{m}/\hat{r}_f)], \]
\[ + a_{F,0}^+ \exp(-\hat{m}/\hat{r}_f) - \zeta, \quad (57) \]

\[ \Theta_1^-(\zeta = 0) = 0, \quad (58) \]

\[ \left( d \Phi_{F,1}/d \zeta \right)_{\zeta = 0} = \hat{Y}_{F,\infty} \left( \hat{m}/\hat{r}_f^2 \right) \]
\[ \left( \left( 1 + \hat{v}_F \right) \exp(\hat{m}/\hat{r}_f - 1) \right), \quad (59) \]

\[ d^2 \Phi_{O,1}^-/d \zeta^2 = A_3 \Phi_{S,0}^- \Phi_{O,1}^+ \exp(-\alpha \Theta_1^-), \quad (60) \]

\[ \Phi_{O,1}^- (\zeta \rightarrow -\infty) = -a_{O,1}^- \exp(-\hat{m}/\hat{r}_f) \]
\[ - \hat{Y}_{O,b} (\hat{m}/\hat{r}_f^2) \zeta, \quad (61) \]

\[ \Phi_{O,1}^-(\zeta = 0) = 0, \quad (62) \]
\[
\frac{d\Phi_{O,1}}{d\xi} = -\tilde{Y}_{F,\infty} (\tilde{m}/\tilde{r}_f) [1 + \tilde{v}_F] \exp(\tilde{m}/\tilde{r}_f) - 1]
\]

where

\[
\Theta_1^+ = -a_{T,1}^+ \left[ 1 - \exp \left(-\frac{\tilde{m}}{\tilde{r}_f} \right) \right]
\]

\[
+ \frac{\tilde{m}}{\tilde{r}_f} \exp(\tilde{m}/\tilde{r}_f) - 1 \right] \xi,
\]

\[
\Phi_{R,1}^+ = a_{R,1}^+ \left[ 1 - \exp \left(-\frac{LeR \tilde{m}}{\tilde{r}_f} \right) \right]
\]

\[
+ \frac{LeR}{\tilde{v}_F} \left[ \Phi_{F,1}^+ + a_{F,1}^+ \left[ 1 - \exp \left(-\frac{\tilde{m}}{\tilde{r}_f} \right) \right] \right]
\]

\[
- \tilde{Y}_{F,\infty} (\tilde{m}/\tilde{r}_f) \frac{\exp(\tilde{m}/\tilde{r}_f)}{\exp(\tilde{m}/\tilde{r}_f)} \xi,
\]

\[
\Phi_{S,0}^- = a_{S,0}^- \exp(-LeS \tilde{m}/\tilde{r}_f).
\]

\[
\Theta_1^- = \tilde{q}_S \tilde{v} \Phi_{O,1}^- + (a_{O,1}^- \tilde{q}_S \tilde{v} - a_{T,1}^-) \exp \left(-\frac{\tilde{m}}{\tilde{r}_f} \right)
\]

\[
+ a_{T,1}^- \exp(-\tilde{m}) \left[ \frac{\tilde{r}_f \tilde{v}_b - \tilde{r}_f}{\tilde{m}} \left[ 1 - \exp(\tilde{m}/\tilde{r}_f - 1) \right] \right] - \tilde{q}_S \tilde{v} \tilde{Y}_{O,b} \right] \xi.
\]

As in the case of fuel supplied from the burner, the soot/precursor concentration at the boundary between the soot/precursor formation region and the fuel side inert region is defined as a soot index \( S_{IF,O} \) with its value given by

\[
S_{IF,O} = \tilde{Y}_S (\tilde{r}_f) = (a_{S,0}^+ + ak_{S,1}^+)
\times \left[ 1 - \exp(-LeS \tilde{m}/\tilde{r}_f) \right] + \cdots
\]

The subscript “O” of \( S_{IF,O} \) represents the case of oxidizer supplied from the burner and the subscript “F” represents that the soot index is evaluated at the fuel side of the reaction region. For this case, the soot/precursor concentration in the soot consumption region is expected to be very small because the direction of convection is from the oxidizer side to the fuel side. The soot/precursor production is primarily dominated by the soot/precursor formation reaction. Therefore the soot index similar to Eq. (44) will not be discussed.

The formulation is closed by noting that although the present analysis can be completed without knowledge of flame extinction, it is based on the existence of a diffusion flame. This study is applicable only for a diffusion flame in Lii"'a's diffusion flame regime [18] and sufficiently far away from the extinction limit.

### 3. Results and discussion

The sooting behavior is studied by adopting the thermo-chemical data closely approximating the ethylene/oxygen/nitrogen flames under atmospheric conditions, given by \( T_b = T_\infty = 300 \text{ K}, q_{F,1} = 10140 \text{ cal/g, } q_{S,3} = 13500 \text{ cal/g, } \lambda = 6.28 \times 10^{-5} \text{ cal/(cm K s), } c_p = 0.273 \text{ cal/(g K), } W_F = 28 \text{ g/mol, } W_O = 32 \text{ g/mol, } W_R = 1 \text{ g/mol, } W_S = 36 \text{ g/mol, } v_{F,1} = 1, v_{F,2} = 0.05, v_{O,1} = 2.9, v_{O,3} = 0.25, v_R = 0.4, v_S = 0.05, \text{ and } E_2 = 12590 \text{ K, corresponding to an activation energy of 25 kcal/mol, in the sample calculations. The selection of } v_{F,2} = 0.05 \text{ allows a maximum of 4.8% of ethylene to become soot/precursor and } v_R = 0.4 \text{ means 10% of the hydrogen in ethylene is converted to the radical in the oxidation reaction (R1). Other stoichiometric coefficients are adjusted to ensure mass conservation. Where applicable, the configuration and run conditions were those used in the experiments by Sunderland et al. [8]. The spherical porous burner is then 1/4 inch in diameter such that \( r_b = 3.175 \text{ mm, and the fuel flow rate is } 1.51 \text{ mg/s except when the effect of mass-flow rate is considered. The same four limiting flames as those studied by Sunderland et al., namely, the flames with (a) fuel issuing into air, (b) diluted fuel issuing into oxygen, (c) air issuing into fuel, and (d) oxygen issuing into diluted fuel, have been investigated. For Flames a and b, fuel is supplied from the burner such that the direction of convection is from fuel to oxidizer. Region A identified in Fig. 1 is the soot formation region and Region B is the soot consumption region. For Flames c and d, oxidizer is supplied from the burner such that the direction of convection is from oxidizer to fuel. Region B is the soot formation region and Region A is the soot consumption region. The flame structure (stoichiometric mixture fraction \( Z_{st} \)) is altered by exchanging the inert between the oxidizer and the fuel. Flames a and c are low \( Z_{st} \) flames \( (Z_{st} = 0.064) \) while Flames b and d are high \( Z_{st} \) flames \( (Z_{st} = 0.78) \). Except for the discussion on the Lewis number effects, the calculations were performed by taking \( \alpha = Le_R = Le_S = 1. \)

For Flame a, pure fuel is supplied from the burner and issued into an ambient such that \( Y_{F,b} = 1, Y_{O,\infty} = 0.233, m = 1.51 \text{ mg/s and the flow is from fuel to oxidizer. The soot/precursor formation region is between the burner and the flame sheet (Region A of Fig. 1) while the soot consumption region is located outside of the flame sheet (Region B of Fig. 1). Soot/precursors produced in the formation region are driven by the bulk flow to the soot consumption region before entering the ambient. Figs. 2 and 3 show, respectively, the soot index at the fuel side of the flame, \( S_{IF,F} \), and the soot index at the oxidizer side of the flame, \( S_{I O,F} \), as functions of the Damköh-
Fig. 2. Variation of the soot index at the fuel side, $S_{IF,F}$, with the Damköhler number of soot formation reaction, $Da_2$, for the fuel/air flame (Flame a). All the figures are for $\alpha = Le_R = Le_S = 1$ except for Fig. 21.

Fig. 3. Variation of the soot index at the oxidizer side, $S_{IO,F}$, corresponding to Fig. 2.

The Damköhler number for the soot/precursor formation reaction, $Da_2$, and three values of $\beta$. As shown in Fig. 2, by increasing $Da_2$ from an initial small value, $S_{IF,F}$ first increases, attains a maximum value, and then decreases, similar to that of the counterflow flame [7]. For the present study, the variation of $Da_2$ is caused by the change of reaction rate constant as shown in the nondimensionalization. When $Da_2$ is increased, the soot formation reaction is stronger so that more soot/precursor is produced. Simultaneously, a higher $Da_2$ yields a higher consumption of the radical $R$ such that the radical concentration at the boundary between the soot formation region and the fuel side transport region is reduced, and the production of the soot/precursor at this location is diminished. For values of $Da_2$ lower than that at the maximum $S_{IF,F}$, the radical concentration is sufficiently high so that the soot/precursor production is dominated by the reaction rate, and $S_{IF,F}$ increases with $Da_2$. After the maximum $S_{IF,F}$ is reached, the reaction rate is high so that the soot formation reaction is controlled by the availability of the radical, and $S_{IF,F}$ decreases with increasing $Da_2$. Such a decrease implies that the soot formation region is shifted toward the oxidation region and is narrower. This explanation also is applicable to the counterflow flame when the variation of $Da_2$ is caused by the change of reaction rate instead of the stretch rate. Fig. 2 also shows that with an increase in $\beta$, the soot consumption reaction becomes stronger such that $S_{IF,F}$ decreases, as it should.

Because the direction of convection is from the soot formation region to the soot consumption re-
Fig. 4. Variation of the flame temperature, $\tilde{T}_f$, corresponding to Fig. 2.

region, $S_{IF,F}$ only shows the soot/precursor concentration at the boundary between the soot/precursor formation region and the fuel side inert region. The net production of soot/precursor can better be exhibited by $S_{IO,F}$, the soot/precursor concentration at the boundary between the soot/precursor consumption region and the oxidizer side inert region, for all the soot/precursor produced in the formation region passes into the consumption region and, if not fully oxidized, it enters the inert region and appears in the combustion products. Fig. 3 shows that in the limit of negligible soot/precursor consumption reaction, $\beta = O(\delta)$, $S_{IO,F}$ increases with $Da^2$, and the rate of growth decreases with $Da^2$ because of the depletion of the radical. In the sample calculations the numerical value of $\beta$ is taken to be zero in the limit of $\beta = O(\delta)$ because only the leading order effects are considered. In the limit of infinitely large $Da_2$, all the radical is converted to soot/precursor and $S_{IO,F}$ reaches its maximum value. By increasing $\beta$, $S_{IO,F}$ decreases at any specified $Da_2$ as a result of enhanced soot consumption reaction. For $\beta = 0.05$, there exists a critical $Da_2$ at which $S_{IO,F}$ attains its maximum. For values of $Da_2$ larger than this critical value, $S_{IO,F}$ decreases with $Da_2$, indicating that the soot consumption reaction is faster than the transport of soot/precursor into the consumption region. A critical $Da_2$ also exists for $\beta = 0.025$, but at a larger value. When $\beta$ is sufficiently large, soot/precursor can be completely oxidized ($S_{IO,F}$ reduced to zero) as a consequence of the strong soot/precursor consumption reaction.

It is also observed by comparing Figs. 2 and 3 that the value of $S_{IO,F}$ is much lower than that of $S_{IF,F}$ under any given condition. Through the favorable convection, the soot/precursor produced in the soot formation region is convected into the soot consumption region in which it is oxidized. The flame is then soot free or nearly soot free outside of the oxidation region, and sooty in the soot formation region, in agreement with the experiments of Sunderland et al. [8]. This indicates that although favorable convection transports soot/precursor into the soot consumption region where it is oxidized, convection alone is not capable of suppressing soot formation to yield soot free (permanently blue) flames.

Recognizing that the production of soot/precursor reduces the conversion of chemical energy to thermal energy and, hence the flame temperature, the flame temperature is a parameter that quantifies the net soot/precursor production. By plotting the flame temperature $\tilde{T}_f$ versus $Da_2$ in Fig. 4, it is observed that in the limit of $\beta = O(\delta)$, the flame temperature decreases monotonically with increasing $Da_2$. For the other two values of $\beta$, there is a critical value of $Da_2$ at which $\tilde{T}_f$ attains its minimum, and the critical $Da_2$ decreases with increasing $\beta$. Moreover, for any given $Da_2$, $\tilde{T}_f$ is higher for larger $\beta$. These conclusions are consistent with our discussion of Fig. 3.

The discussion continues by studying the flame location, $\tilde{r}_f$, as a function of $Da_2$ in Fig. 5. This figure shows that in the limit of negligible soot consumption reaction, $\tilde{r}_f$ is independent of $Da_2$. For other values of $\beta$, the flame location increases with $\beta$ as well as $Da_2$ because more oxidizer is used to oxidize the soot/precursor. The reduction of the oxidizer supply into the oxidation region causes the flame to shift toward the oxidizer side. This behavior is qualitatively the same as that of the counterflow flame [7].

The effect of inert exchange, or flame structure, is examined by a limiting case of removing all inert gas from the oxidizer side and adding it to the
fuel stream to yield Flame b. For this flame, we have $Y_{O,\infty} = 1$ but $Y_{F,0}$ varies between 0.08143 and 0.08784 and $m$ varies between 18.53 and 17.19 mg/s ($18.73 \leq \bar{m} \leq 20.19$) depending upon the amount of soot production, to keep the fuel consumption rate of 1.51 mg/s. The results of $S_{IF,F}$, $S_{IO,F}$, $\tilde{T}_{f}$, and $\tilde{r}_{f}$ versus $Da_{2}$ for this flame are plotted in Figs. 6–9. Comparing Figs. 2–5 and 6–9, it is found that the behavior of these two limiting flames are qualitatively similar. Quantitatively, however, for Flame b the values of $S_{IF,F}$ and $S_{IO,F}$ in Figs. 6 and 7 are negligibly small in the same range of $Da_{2}$ as Figs. 2 and 3. Furthermore, much larger values of $Da_{2}$ are necessary to produce observable soot/precursor, $\tilde{T}_{f}$ is higher and $\tilde{r}_{f}$ is much smaller, and smaller values of $\beta$ are sufficient to activate significant soot consumption reaction.

By removing the inert from the oxidizer and supplying it with the fuel, as was done in Sunderland et al. [8], the initial fuel concentration is reduced while the initial oxidizer concentration is increased. Consequently, the flame moves closer to the burner and $\tilde{r}_{f}$ is decreased. The flame temperature, however, is increased because the heat transfer from the flame to the burner is reduced as a result of the high mass-flow rate out of the burner. The higher flow rate and smaller flame standoff distance then yield a much higher mass flux near the flame so that the soot formation region is compressed and soot consumption region is broadened. The thin soot formation region combined with a high velocity renders a
reduced residence time for the soot formation reaction so that a much higher $D_{a2}$ is required to produce soot/precursor and the amount of soot/precursor produced is reduced. Although the higher flame temperature favors soot/precursor production, the effect of residence time dominates the process because the increase in the flame temperature is insignificant as is shown from the comparison between Figs. 4 and 8, while the increase of the mass flux near the flame is relatively high. The approximately 12 times increase in the mass-flow rate and 50% decrease in the flame location yield an increase of the mass flux by approximately 40 times. The broadened soot consumption region then leads to more effective consumption of the soot/precursor so a smaller value of $\beta$ is sufficient to oxidize more soot/precursor. For the same range of $D_{a2}$, the diluted-fuel/oxygen flame (Flame b) is primarily soot free.

Next, the impact of flow direction (convection) is investigated by interchanging the burner and ambient gases. Because the flow direction is from the oxidizer to the fuel-filled ambient for the inverted flame, the soot/precursor produced from the soot formation region is convected toward the fuel-rich region (away from the soot consumption region). The soot/precursor produced in the formation region (Region B of Fig. 1) can be transported to the soot consumption region (Region A of Fig. 1) only through diffusion that is against convection. It is then expected that the concentration of soot/precursor in the soot consumption region is low and the soot consumption reaction is difficult to occur. Therefore, the effect of convection (flow direction) is to favor soot/precursor formation and suppress soot/precursor consumption for the inverted flame. Indeed, all the calculations on Flames c and d show that the concentration of
soot/precursor in the soot consumption region is always negligibly small so $S_{I,O,O}$ will not be presented.

For Flame c, where air is issued from the burner into pure fuel, we have $Y_{F,\infty} = 1, Y_{O,b} = 0.233$, and $m$ varying between 20.46 and 22.22 mg/s ($22.33 \leq \dot{m} \leq 24.26$). The soot index $S_{I,F,O}$, flame temperature $\tilde{T}_f$, and flame location $\tilde{r}_f$ versus $Da_2$ are presented in Figs. 10–12. These figures show that the solutions are qualitatively similar to Figs. 2, 4, and 5, and as expected, extremely large values of $\beta$, 100 and 500 in the present calculations, are necessary to exhibit observable soot consumption reaction. Quantitatively, the flame is much closer to the burner as compared to Flame a (Fig. 12) because of the low supplied oxidizer concentration. Although the flame is close to the burner, the heat loss to the burner is actually low as a result of high burner flow rate, such that the flame temperature is high (Fig. 11). These behaviors are similar to those of Flame b. Among the four model flames studied, Flame c is the closest to the burner, having the highest mass-flow rate and the highest flame temperature. The high mass-flow rate and low flame standoff distance then yield the soot formation region (Region B in Fig. 1) broadened and the residence time for soot formation increased. The high flame temperature also enhances the rate of the soot formation reaction. For this flame, both the flame structure and the convection promote formation of soot/precursor so it is the sootiest among the four model flames. As is supported by Figs. 10–12, the values of $Da_2$ to yield substantial soot formation reaction are comparable to those of Flame a, and the
peak value of $S_{IF,O}$ is higher than that of $S_{IF,F}$ in Fig. 2.

The nonmonotonic behavior of $S_{IF,O}$ with $Da_2$ for $\beta = O(\delta)$ in Fig. 10 is not expected a priori. To better understand this behavior, the composite solution of the soot concentration in the fuel side of the flame, $\tilde{Y}_S^+$, is plotted versus the stretched spatial coordinate, $\zeta$, and presented in Fig. 13. Four values of $Da_2$ are selected to include both the ascending and the descending branches. For low values of $Da_2$, e.g., $Da_2 = 0.068$, the soot formation reaction is weak and the value of $\tilde{Y}_S^+$ are relatively small and the peak of $\tilde{Y}_S^+$ is located away from the oxidation region. With an increase in $Da_2$, soot/precursor is produced while the radical is consumed at a higher rate such that $\tilde{Y}_S^+$ is increased and the location of peak $\tilde{Y}_S^+$ shifts toward the oxidation region. For large values of $Da_2$, the soot formation reaction is strong, as illustrated by the $Da_2 = 2.49$ curve, and a large amount of soot is produced. Most of the radical is consumed in a narrow region near the oxidation region such that the peak of $\tilde{Y}_S^+$ is high and its location is at a small $\zeta$. The value of $\tilde{Y}_S^+$ then quickly drops for increasing $\zeta$ as a result of the low formation reaction rate. This behavior indicates that the soot formation region becomes thinner when $Da_2$ is increased because the radical is depleted near the oxidation region. Fig. 10 only represents a snapshot of the outer solution of $\tilde{Y}_S^+$ at a sufficiently large $\zeta$. Although $\tilde{Y}_S^+$ has a peak, the overall soot/precursor production increases monotonically with $Da_2$ as evidenced by the monotonic decrease in the flame temperature (Fig. 11).
The last of the four model flames, Flame d, is obtained by removing the inert from the air and introducing it into the fuel such that pure oxygen is injected into a diluted-fuel ambient, similar to the procedure to yield Flame b. For this flame, $Y_{O,b} = 1$, $Y_{F,\infty}$ varies between 0.08143 and 0.08784, and $m$ varies between 4.77 and 5.18 mg/s ($5.20 \leq \tilde{m} \leq 5.65$). Results of $S_{IF,O}$, $\tilde{T}_f$, and $\tilde{r}_f$ versus $Da_2$ are presented in Figs. 14–16 and as for Flames b and c, the behaviors are qualitatively similar to Figs. 2, 4, and 5. The solutions for $\tilde{T}_f$ and $\tilde{r}_f$ reveal that Flames a and d are similar and Flames b and c are similar. Flames a and d are relatively far from the burner because of the high concentration of ejecting reactant, while Flames b and c are much closer to the burner as a result of the low concentration of the ejecting reactant. However, the flame temperatures of Flames a and d are lower than those of Flames b and c owing to their low mass-flow rates and higher heat loss to the burner.

Regarding the soot/precursor formation, the low mass-flow rate and large flame standoff location for Flame d as compared to Flame c (about 23% of mass-flow rate and 2.5 times the flame standoff distance) render a much lower mass flux (about 3.6% of Flame c) near the reaction regions. The reduction in the mass flux yields a much narrower soot formation region and a much broader soot consumption region. It also promotes the diffusion of soot/precursor into the soot consumption region so that the soot consumption reaction is more effective. This is supported by the larger difference in all the solutions.
Fig. 15. Variation of the flame temperature, $\tilde{T}_f$, corresponding to Fig. 14.

Fig. 16. Variation of the flame sheet location, $\tilde{r}_f$, corresponding to Fig. 14.

responding to smaller values of $\beta$ as shown in Figs. 14–16. As a consequence to the modification in the reaction region thicknesses, the soot/precursor formation reaction is suppressed such that much less soot/precursor is formed, and more soot/precursor is depleted through the soot consumption reaction, so the total soot production is much lower. In addition, the reduced flame temperature further retards the soot formation reaction. Therefore, as for Flame b, the variation of flame structure through inert exchange from the oxidizer side to the fuel side significantly suppresses the soot/precursor production, as shown in Fig. 14 by the low $S_{IF,O}$, in agreement with experimental observations of Sunderland et al. [8].

Comparison of the four limiting flames discussed above delineates the relative importance between convection (flow direction) and flame structure (inert exchange) on soot inception. It is known from an earlier discussion that convection favors soot/precursor formation for Flames c and d but favors soot/precursor consumption for Flames a and b. In the meanwhile, flame structure favors soot/precursor production for Flames a and c but suppresses soot/precursor formation for Flames b and d. It is then clear that Flame c is sooty while Flame b is practically soot free. The low values of $S_{IF,F}$ and $S_{IO,F}$, as well as the high $Da_2$ shown in Figs. 6 and 7 for Flame b, and the high values of $S_{IF,O}$ shown in Fig. 10 support this argument. As to the other two flames, the high $S_{IF,F}$ for Flame a shows that although convection favors consumption of soot/precursor, it cannot prevent soot inception when the flame structure fa-
vors formation of soot/precursor. On the other hand, Flame d is soot free, despite the fact that convection favors soot/precursor production, because the flame structure suppresses soot/precursor formation. Thus, soot/precursor exists for the fuel/air flames (Flames a and c) while the diluted-fuel/oxygen flames (Flames b and d) are soot-free regardless of the convection direction. This suggests that although the effects of convection and flame structure are coupled and cannot be completely separated, the variation of flame structure (stoichiometric mixture fraction) that results from inert redistribution is primarily responsible for the suppression of soot inception and soot free flames. Although flow direction can lead to the consumption of soot/precursor, it is less important to soot inception in these low strained flames.

All the results discussed in the above were based on the variation of Da2, which is fixed when the system parameters are specified and the reactants are selected. Experimentally, it is more direct to vary the mass-flow rate while keeping Da2 unchanged. The flame response to the mass-flow rate is presented by taking Flame a as a representation and the sample calculations were performed using Da2 = 0.1 with the results of $S_{IF,F}$, $S_{IO,F}$, $\tilde{T}_f$, and $\tilde{r}_f$ versus $\tilde{m}$ plotted in Figs. 17–20. Figs. 17 and 18 show that the sooting behavior responding to the variation of $\tilde{m}$ is qualitatively the same as that of Da2 shown in Figs. 2 and 3. This is reasonable because Da2 is proportional to $m$ if it is defined following that of droplet burning. Moreover, Fig. 20 shows that the flame sheet and the reaction regions move outward almost linearly with increasing $\tilde{m}$, similar to that of quasi-steady droplet.
Fig. 19. Variation of the flame temperature, $\tilde{T}_f$, corresponding to Fig. 17.

Fig. 20. Variation of the flame sheet location, $\tilde{r}_f$, corresponding to Fig. 17.

burning, because both of these systems are controlled by a convection-diffusion balance in the gas phase. This outward flame motion, along with the stronger outward flow speed near the burner, results in a reduction of heat loss to the burner and a higher flame temperature as shown in Fig. 19. The effect of soot consumption reaction on $\tilde{T}_f$ and $\tilde{r}_f$, also consistent with the effect of $\beta$ in Figs. 4 and 5, is not observable in Figs. 19 and 20 because it is small compared to the variation caused by $\tilde{m}$. In the limit of sufficiently large $\tilde{m}$, $\tilde{T}_f$ approaches its adiabatic value. Only at this limit is the effect of $\beta$ on $\tilde{T}_f$ observable.

The effects of the activation energy ratio, $\alpha$, and the Lewis number of the radical, $Le_R$, have also been studied and found to be qualitatively the same as those in Chao et al. [7], i.e., the net soot/precursor production is higher and the flame temperature is lower for a larger $\alpha$ or $Le_R$. The Lewis number of the soot/precursor, $Le_S$, affects the sooting behavior through the soot consumption reaction and is significant only for Flame a. It is less important for Flames b and d because of the low soot/precursor formation, and Flame c because the convection suppresses the transport of the soot/precursor from the formation region to the consumption region. The sample calculations for Flame a were performed by taking $\beta = O(\delta)$ and 0.025, and the solution of the flame temperature $\tilde{T}_f$ is plotted versus $Da_2$ in Fig. 21. The dotted curve of Fig. 21 (also in Fig. 4) shows that in the limit of weak soot consumption reaction, $\beta = O(\delta)$, the solution is independent of $Le_S$. For nonzero $\beta$s, the flame temperature increases with $Le_S$, meaning
Fig. 21. Variation of the flame temperature, $\tilde{T}_f$, versus $D_{a2}$ for Flame a with $\beta = O(\delta)$ and 0.025. The figure shows the effect of the soot/precursor Lewis number, $L_{e_S}$, on the sooting behavior of Flame a.

that the amount of soot/precursor production reduces with higher $L_{e_S}$. Physically, when the diffusion rate of the soot/precursor is slower, more soot is accumulated in the soot consumption region such that more soot/precursor is oxidized and more heat is generated.

4. Concluding remarks

In this study, the sooting behavior of a spherical diffusion flame is analyzed by activation energy asymptotics with a simplified three-step kinetic scheme that describes the oxidation, soot/precursor formation, and soot/precursor consumption reactions. This geometry allows the independent variation of flame structure (stoichiometric mixture fraction) through inert redistribution and convection (flow direction) through exchange of burner and ambient reactants. Four limiting flames, namely, the flames with (a) ethylene issuing into air, (b) diluted ethylene issuing into oxygen, (c) air issuing into ethylene, and (d) oxygen issuing into diluted ethylene, are investigated and their soot production tendencies are compared.

The results showed that in the limit of negligible soot consumption reaction, the total amount of soot/precursor produced increases with the rate of the soot formation reaction until the diffusion controlled limit is reached. With the increase of the soot consumption reaction, there exists a critical soot formation reaction rate at which the net soot production is highest. The sooting behavior responding to the variation of the mass-flow rate and the rate of the soot formation reaction is qualitatively similar. Soot/precursor production decreases with a decrease in the radical Lewis number or an increase in the soot/precursor Lewis number.

Comparison among the four limiting flames reveals that the flames burning pure fuel with air (Flames a and c) produce soot and those burning diluted-fuel with oxygen (Flames b and d) are practically soot free regardless of the flow direction. Flow direction (convection) can reduce the total soot production by transferring the soot/precursor produced in the soot formation region to the soot consumption region such as in Flame a, but it is not capable of suppressing soot/precursor formation in these low strain flames. This suggests that flame structure (stoichiometric mixture fraction) has a profound impact on the soot inception of non-premixed flames and can result in permanently blue flames through inert redistribution.

Acknowledgments

This work was supported by NASA Grants NCC3-696 (B.H.C.) and NCC3-697 (R.L.A.). We gratefully appreciate many helpful discussions with Drs. David L. Urban, Peter B. Sunderland, and Dennis P. Stocker.

Appendix A. Asymptotic analysis

In the $O(\delta)$ soot/precursor formation region located in the fuel side of, and adjacent to, the oxidation region, only the soot/precursor formation reaction ($R_2$) is significant and only $O(\delta)$ variations on all quantities are possible. In this region, the stretched coordinate is defined as $\zeta = (\tilde{r} - \tilde{r}_f)/\delta$, where the small expansion parameter is given by $\delta = \tilde{T}_{2,0}/\tilde{E}_2$. 

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and the variables are expanded in terms of $\delta$ as

$$\tilde{Y}_\pm = [\tilde{T}_{f,0} - \delta \Theta_1^\pm - \delta^2 \Theta_2^\pm + O(\delta^3)] + O(\varepsilon), \tag{A.1}$$

$$\tilde{Y}_i^\pm = [\delta \Phi_{i,1}^\pm + \delta^2 \Phi_{i,2}^\pm + O(\delta^3)] + O(\varepsilon), \tag{A.2}$$

$$i = F, O, R, \tag{A.3}$$

In the above, the superscript “$-$” is for the case of fuel supplied from the burner, for which the soot formation is located between the burner and the reaction sheet such that $\zeta < 0$, and the superscript “$+$” is for the case of oxidizer supplied from the burner, for which the soot formation is located outside of the reaction sheet such that $\zeta > 0$. Because only an $O(\varepsilon)$ oxidizer leaks through the oxidation region into this region, it is required that $\Phi_{O,1}^\pm \equiv 0$. Substituting Eqs. (A.1)–(A.3) into Eqs. (1)–(5), expanding, then collecting terms of the same order in $\delta$, results in Eqs. (30), (56), and the following equations:

$$d^2 \Phi_{F,0}^\pm \over d\zeta^2 = d^2 \Theta_1^\pm \over d\zeta^2 = d^2 \Phi_{F,1}^\pm \over d\zeta^2 = \tilde{v}_F d^2 \Phi_{F,1}^\pm \over d\zeta^2,$$

$$= \left( \tilde{m} - 2\tilde{r}_f \right) \frac{d\phi_{F,0}^\pm \over Le_S \delta \zeta}{\delta \zeta^2} - \tilde{r}_f \frac{d^2 \Phi_{F,1}^\pm \over Le_S \delta \zeta}{\delta \zeta^2} \left( \tilde{m} - 2\tilde{r}_f \right) d\Theta_1^\pm \over d\zeta = 0, \tag{A.4}$$

$$- \tilde{r}_f^2 (d^2 \Theta_2^\pm \over d\zeta^2 - \tilde{q}_S \tilde{v} \Phi_{O,2}^\pm \over d\zeta^2) = 0,$$.

$$\tilde{m} \frac{(d\phi_{F,1}^\pm \over d\zeta) - \tilde{v}_F d\phi_{F,1}^\pm \over Le_S \delta \zeta}{d\zeta} \tag{A.5}$$

$$- 2\tilde{r}_f \left( \frac{d^2 \Phi_{F,1}^\pm \over Le_S \delta \zeta}{\delta \zeta^2} - \tilde{v}_F \frac{d^2 \Phi_{F,1}^\pm \over Le_S \delta \zeta}{\delta \zeta^2} \right) = 0.$$

$$\frac{d(\phi_{F,1}^\pm \over d\zeta) + d\phi_{S,1}^\pm \over d\zeta}{d\zeta} = \tilde{r}_f \frac{d^2 \Phi_{F,2}^\pm \over Le_S \delta \zeta}{\delta \zeta^2} + \frac{1}{Le_S} \frac{d^2 \phi_{S,1}^\pm \over d\zeta^2} - \tilde{v} \frac{d^2 \phi_{O,2}^\pm \over d\zeta^2} = 0. \tag{A.6}$$

The soot/precursor concentration is considered an $O(1)$ quantity since the soot/precursor formation and consumption reactions can have different rates.

For the case of fuel supplied from the burner, the required boundary conditions to solve the above equations can be obtained from matching the solutions in this region with the outer solutions in the fuel side. Matching is performed by first expressing the outer solutions in terms of $\zeta$, expanding in orders of $\delta$, and then equating the resulting expressions with Eqs. (A.1)–(A.3) as $\zeta \to -\infty$. Through the matching, we obtain Eqs. (18), (19), (22), (24, (31), and other conditions to solve Eqs. (A.4)–(A.7). Integrating the four expressions in Eq. (A.4) twice and Eqs. (A.5)–(A.7) once subject to the matching conditions, we obtain, for the case of fuel supplied from the burner, Eqs. (40), (41), and

$$\Phi_{S,0}^- = a_{S,0}^- \exp(-Le_S \tilde{m} / \tilde{r}_f), \tag{A.8}$$

$$\tilde{v}_F \Phi_{R,1}^- + \Phi_{S,1}^- \equiv \tilde{v}_F \Phi_{R,1}^- \exp(-Le_S \tilde{m} / \tilde{r}_f) + a_{S,1}^- \tilde{m} / \tilde{r}_f \times \exp(-Le_S \tilde{m} / \tilde{r}_f), \tag{A.9}$$

$$\Phi_{F,1}^- - \tilde{v}_F \Phi_{R,1}^- = \tilde{r}_f \left( \frac{d(\phi_{F,2}^\pm \over d\zeta) - \tilde{v}_F d(\phi_{F,2}^- \over Le_S \delta \zeta)}{d\zeta} \right)$$

$$= \frac{2\tilde{r}_f \tilde{r}_f \over m \left( d(\phi_{F,2}^\pm \over d\zeta) - \tilde{v}_F d(\phi_{F,2}^- \over Le_S \delta \zeta) \right)}{d\zeta} - \tilde{v}_F \Phi_{R,1}^- + \Phi_{S,1}^- \tag{A.11}$$

$$\tilde{v}_F \Phi_{F,1}^- \Phi_{R,1}^- \Phi_{S,1}^-$$

A similar approach for the case of oxidizer supplied from the burner can be performed with the matching at $\zeta \to \infty$. The matching yields Eqs. (18), (22), (48), and (57), and the integration of Eqs. (A.4)–(A.7) gives Eqs. (64) and (65) as well as

$$\Phi_{S,0}^+ = a_{S,0}^+ \left[ 1 - \exp(-Le_S \tilde{m} / \tilde{r}_f) \right]. \tag{A.13}$$
\[ \frac{\Phi^{+}_{R,1}}{L_R} + \frac{\Phi^{+}_{S,1}}{L_S} = a^{+}_{R,1} \hat{v}_F \left[ 1 - \exp \left( - \frac{\text{Le}_R \hat{m}}{\hat{r}_f} \right) \right] \]

\[ + \frac{a^{+}_{S,1}}{L_S} \left[ 1 - \exp \left( - \frac{\text{Le}_S \hat{m}}{\hat{r}_f} \right) \right] \]

\[ - \frac{a^{+}_{S,0} \hat{m}}{\hat{r}_f} \exp \left( - \frac{\text{Le}_S \hat{m}}{\hat{r}_f} \right) \xi, \quad (A.14) \]

\[ \frac{d(\hat{F}^+)}{d\xi} = \frac{d^2 \Phi^+_{F,2}}{d\xi^2} - \frac{\hat{v}_F}{\hat{r}_f} \frac{d^2 \Phi^+_{F,2}}{d\xi^2} \]

\[ + \hat{v}_F \left( \frac{\hat{m}}{\hat{r}_f} - \frac{2}{\hat{r}_f \text{Le}_R} \right) \frac{d\Phi^+_{R,1}}{d\xi} = 0, \quad (A.19) \]

\[ \frac{d^2 \Theta^+}{d\xi^2} - \frac{\hat{q}_S \hat{v} \Sigma^+}{d\xi} = 0, \quad (A.20) \]

As in the soot/precursor formation region, for the case of fuel supplied from the burner matching the solutions in this region with the outer solutions in the oxidizer side yields Eqs. (20)–(22) and (35). Integrating the four expressions in Eq. (A.18) twice and Eqs. (A.19)–(A.21) once subject to the matching conditions, we obtain Eqs. (42), (43), and

\[ \Phi^{+}_{R,1} = a^{+}_{R,1} \left[ 1 - \exp \left( - \frac{\text{Le}_R \hat{m}}{\hat{r}_f} \right) \right], \quad (A.22) \]

\[ \hat{v}_F \frac{\Phi^+_{S,1}}{L_S} = -a^{+}_{S,1} \hat{v}_F \left[ 1 - \exp \left( - \frac{\hat{m}}{\hat{r}_f} \right) \right] \]

\[ - \frac{a^{+}_{S,1}}{L_S} \left[ 1 - \exp \left( - \frac{\text{Le}_S \hat{m}}{\hat{r}_f} \right) \right] \]

\[ + \frac{\hat{m}}{\hat{r}_f} \left[ \hat{v}_F \frac{\Phi^+_{S,1}}{L_S} \right] \exp \left( \frac{\text{Le}_S \hat{m}}{\hat{r}_f} \right) - 1 + a^{+}_{S,0} \exp \left( - \frac{\text{Le}_S \hat{m}}{\hat{r}_f} \right) \xi, \quad (A.23) \]

\[ \frac{d\Phi^+_{F,2}}{d\xi} = \frac{\hat{v}_F}{\hat{r}_f} \frac{d\Phi^+_{F,2}}{d\xi} \]

\[ = a^{+}_{R,1} \hat{v}_F \frac{\hat{m}}{\hat{r}_f} \exp \left( - \frac{\text{Le}_R \hat{m}}{\hat{r}_f} \right), \quad (A.24) \]

\[ \frac{\hat{r}_f^2}{\hat{m}} \left( \frac{d\Theta^+}{d\xi} - \frac{\hat{q}_S \hat{v} \Sigma^+}{d\xi} \right) \]

\[ - \Theta^+ + \frac{\hat{q}_S \hat{v} \Phi^+_{O,1}}{d\xi} \]

\[ = a^{+}_{R,1} - a^{+}_{O,1} \hat{q}_S \hat{v} - \frac{2}{\hat{r}_f} (a^{+}_{T,0} - a^{+}_{O,0} \hat{q}_S \hat{v}) \]

\[ \times \exp \left( - \frac{\hat{m}}{\hat{r}_f} \right) \xi, \quad (A.25) \]
\[
\frac{\tilde{r}^2}{\hat{m}} \left( \frac{d\Phi^+_{O,2}}{d\xi} - \frac{d\Phi^+_{F,2}}{d\xi} - \frac{1}{\text{Le}_S} \frac{d\Phi^+_{S,2}}{d\xi} \right) \\
- \hat{\nu}\Phi^+_{O,1} + \Phi^+_S, \\
= a^{+}_{S,1} + a^{+}_{O,1} \hat{v} - \frac{2}{\tilde{r}_f} \left[ a^+_{s,0} \exp \left( -\frac{\text{Le}_S \tilde{m}}{\tilde{r}_f} \right) \right] \\
+ a^+_{O,0} \hat{v} \exp \left( -\frac{\tilde{m}}{\tilde{r}_f} \right). \\
\]
\[\tag{A.26}\]

For the case of oxidizer supplied from the burner, the matching yields Eqs. (18), (22), (47), (61), and the integration of Eqs. (A.18)–(A.21) gives Eqs. (66) and (67) as well as
\[
\Phi^+_{R,1} = a^{+}_{R,1} \exp(-\text{Le}_R \tilde{m}/\tilde{r}_f), \\
\hat{\nu}\Phi^+_{O,1} = \frac{\Phi^+_S}{\text{Le}_S} = -a_{O,1} \hat{v} \exp \left( \frac{\tilde{m}}{\tilde{r}_f} \right) \\
- a^+_{S,1} \exp \left( \frac{\text{Le}_S \tilde{m}}{\tilde{r}_f} \right) \\
- \frac{\tilde{m}}{\tilde{r}_f^2} \left[ \hat{v} \hat{Y}_{O,b} + a^+_{S,0} \exp \left( -\frac{\text{Le}_S \tilde{m}}{\tilde{r}_f} \right) \right] \xi, \tag{A.27}\]

\[
\frac{d\Phi^+_{F,2}}{d\xi} = \frac{\hat{v}_F \Phi^+_{O,2}}{\text{Le}_R} - \frac{d\Phi^+_{S,2}}{d\xi} \\
- \frac{\tilde{m}}{\tilde{r}_f^2} \left[ \hat{v} \hat{Y}_{O,b} + a^+_{S,0} \exp \left( -\frac{\text{Le}_S \tilde{m}}{\tilde{r}_f} \right) \right] \xi. \tag{A.28}\]

\[
\frac{\tilde{r}^2}{\hat{m}} \frac{d\Phi^+_{O,2}}{d\xi} - \tilde{q} S \frac{d\Phi^+_{O,2}}{d\xi} - \Theta^+_1 + \tilde{q} S \hat{v} \Phi^+_{O,1} \\
= -a^{+}_{T,1} \exp(-\tilde{m}) \\
+ \frac{2}{\tilde{r}_f} \left[ a^+_{T,0} \exp \left( \frac{\tilde{m}}{\tilde{r}_f} \right) - \tilde{q} S \hat{v} \hat{Y}_{O,b} \right] \xi. \tag{A.30}\]

\[
\frac{\tilde{r}^2}{\hat{m}} \left( \frac{d\Phi^+_{O,2}}{d\xi} - \frac{d\Phi^+_{F,2}}{d\xi} - \frac{1}{\text{Le}_S} \frac{d\Phi^+_{S,2}}{d\xi} \right) \\
- \hat{\nu}\Phi^+_{O,1} + \Phi^+_S \\
= \frac{2}{\tilde{r}_f} \left[ a^+_{S,0} \exp \left( -\frac{\text{Le}_S \tilde{m}}{\tilde{r}_f} \right) \right] + \hat{v} \hat{Y}_{O,b} \xi. \tag{A.31}\]

In the \(O(\varepsilon)\) oxidation region, only the oxidation reaction (R1) is significant. The coordinate in this region is stretched as \(\tilde{r} = \tilde{r}_f + \varepsilon \xi\), where \(\varepsilon = \tilde{T}_f / \tilde{E}_1\) and the variables are expanded as
\[
\tilde{T} = \tilde{T}_f - \varepsilon \left[ \ Theta_1 + \delta \Theta_2 + O(\varepsilon^2) \right] + O(\varepsilon^2), \tag{A.32}\]
\[
\tilde{Y}_j = \varepsilon \left[ \phi_{i,1} + \delta \phi_{i,2} + O(\varepsilon^2) \right] + O(\varepsilon^2), \quad i = F, O, \tag{A.33}\]

\[
\tilde{Y}_R = \left[ \delta \phi_{R,0} + O(\delta^2) + O(\varepsilon/\delta) \right] \\
+ \varepsilon \left[ \phi_{R,1} + \delta \phi_{R,2} + O(\delta^2) + O(\varepsilon/\delta) \right] + O(\varepsilon^2), \tag{A.34}\]

\[
\tilde{Y}_S = \left[ \tilde{Y}_{S,0} + \delta \phi_{S,0} + O(\delta^2) + O(\varepsilon/\delta) \right] \\
+ \varepsilon \left[ \phi_{S,1} + \delta \phi_{S,2} + O(\delta^2) + O(\varepsilon/\delta) \right] + O(\varepsilon^2). \tag{A.35}\]

Substituting Eqs. (A.32)–(A.35) into Eqs. (1)–(5), expanding and collecting terms of the same order in \(\varepsilon\), we obtain
\[
\frac{d^2 \tilde{Y}_{S,0}}{d\xi^2} = \frac{\tilde{d}^2 \phi_{S,0}}{d\xi^2} = \frac{d^2 \phi_{R,0}}{d\xi^2} = 0, \tag{A.36}\]
\[
\frac{d^2 \Theta_1}{d\xi^2} - \frac{d^2 \phi_{F,1}}{d\xi^2} = \frac{d^2 \phi_{F,1}}{d\xi^2} - \frac{d^2 \phi_{O,1}}{d\xi^2} \\
= \frac{\tilde{r}_f^2}{\text{Le}_R} - \frac{d^2 \phi_{F,1}}{d\xi^2} = \frac{1}{\text{Le}_R} \frac{\tilde{m}}{\tilde{r}_f} \frac{d\tilde{Y}_{S,0}}{d\xi} = 0, \tag{A.37}\]
\[
\frac{d^2 \phi_{F,2}}{d\xi^2} - \frac{d^2 \phi_{O,2}}{d\xi^2} = \frac{d^2 \phi_{F,2}}{d\xi^2} + \frac{1}{\text{Le}_R} \frac{\tilde{m}}{\tilde{r}_f} \frac{d\phi_{R,0}}{d\xi} \\
= \frac{d^2 \phi_{F,2}}{d\xi^2} - \frac{d^2 \phi_{O,2}}{d\xi^2} = \frac{1}{\text{Le}_R} \frac{\tilde{m}}{\tilde{r}_f} \frac{d\phi_{S,0}}{d\xi} = 0. \tag{A.38}\]

An additional equation with the reaction term can also be derived, but will not be presented because it is not of interest to this study.

The required boundary conditions to solve Eqs. (A.36)–(A.38) are derived from matching the solutions in the oxidation region with those in the soot/precursor formation and consumption regions as \(\xi \to \pm \infty\) and \(\xi \to 0\). Matching is performed by first letting \(\xi = (\varepsilon/\delta) \xi\), substituting into the solutions in the soot/precursor formation and consumption regions, expanding the expressions for proper ordering, and equating with Eqs. (A.32)–(A.35). For the case of fuel supplied from the burner, matching yields Eqs. (24), (29), (32), (36), and the conditions to solve Eqs. (A.36)–(A.38). Integrating the three expressions in Eq. (A.36) twice and the four expressions in each of Eqs. (A.37) and (A.38) once subject to the matching conditions and then applying the solutions in the soot/precursor formation and consumption regions yield Eqs. (17), (23), (25)–(28), (33), and (37). In the limit of extremely slow soot/precursor
consumption reaction, $\beta$ is taken as an $O(\delta)$ quantity such that Eq. (34) can be integrated subject to Eqs. (35)–(37) to yield $a_{O,1}^+ = 0$ and the flame location is determined by

$$\tilde{r}_f = \tilde{m}/\ln\left[1 + \left(\tilde{Y}_{O,\infty} (1 + \tilde{v}_F)/\tilde{Y}_{F,b}\right)\right].$$

(A.39)

For the case of oxidizer supplied from the burner, matching yields Eqs. (50), (55), (58), (62), and the conditions to solve Eqs. (A.36)–(A.38). Integrating the three expressions in Eq. (A.36) twice and the four expressions in each of Eqs. (A.37) and (A.38) once subject to the matching conditions and then applying the solutions in the soot/precursor formation and consumption regions yield Eqs. (46), (49), (51), (52–(54), (59), and (63). In the limit of slow soot/precursor consumption reaction, the flame location is at

$$\tilde{r}_f = \tilde{m}/\ln\left[1 + \left(\tilde{Y}_{F,\infty}/(1 + \tilde{v}_F)\tilde{Y}_{O,b}\right)\right].$$

(A.40)

References