Analysis of High Pressure H2/O2, H2/AIR AND KEROSENE/AIR Reacting Shear Flows

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ANALYSIS OF HIGH PRESSURE $H_2/O_2$, $H_2/Air$ AND KEROSENE/AIR REACTING SHEAR FLOWS

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Abstract

Direct Numerical Simulation (DNS) data for high pressure $H_2/O_2$ and $H_2/Air$ flames using the compressible flow formulation, detailed kinetics, a real fluid equation of state, and generalized diffusion are analyzed. The DNS is filtered over a range of filter widths to provide exact terms in the Large Eddy Simulation (LES) governing equations, including unclosed terms. The filtered heat flux vector is extensively compared with the heat flux vector calculated as a function of the filtered primitive variables (i.e. the exact LES term is compared with its form available within an actual LES). The difference between these forms defines the subgrid heat flux vector. The analyses are done both globally across the entire flame, as well as by conditionally averaging over specific regions of the flame; including regions of large subgrid kinetic energy, subgrid scalar dissipation, subgrid temperature variance, flame temperature, etc. In this work, both the subgrid heat flux vector and its divergence are found to be substantially larger in reacting flows in comparison with mixing due to the associated larger temperature gradients. However, the divergence of the subgrid heat flux vector tends to be significantly smaller than other unclosed terms in the energy equation with decreasing significance with increasing Reynolds number.

Then a reduced (29 step, 10 species) Kerosene/Air mechanism including a semi-global soot formation/oxidation model associated with an optically thin medium radiative heat flux model has been added to the same code to investigate soot formation/oxidation processes in a temporarily developing hydrocarbon flame operating at both atmospheric and
elevated pressures for both a real gas law (RGL) and the ideal gas law (IGL) equations of state (EOS). Both 3D the RGL and the IGL EOS predictions of the soot formation/oxidation processes good agreement with the limited literature of atmospheric pressure flames [45, 46, 96] has been achieved. High values of the soot volume fraction have been shown to be independent from high temperature flame regions by occupying the flame volumes whose temperature varies from 1300 K to 1800 K. Additionally, the soot number density has been shown to be highly dependent on the temperature, while the soot volume fraction is dominated by local flow characteristics which is also in good agreement with Ref. [96]. Lignell et al. [46, 45] have reported two distinct behaviors of soot mass fraction: I- the slow soot nucleation process has caused the soot mass fraction to be widely scattered in the flame, and II - turbulent transportation has carried the soot to the fuel rich region emphasizing the importance of the turbulence transportation in sooting flames. Similar behavior has also been observed in the current work. The soot generation rate has been shown to have a similar trend with soot mass fraction, while Lignell et al. [45, 46] have observed a high dependency on flame temperature for the soot generation rate.

Furthermore, a slight difference has been observed between the RGL and the IGL EOS model predictions of soot quantities in atmospheric pressure flames. This implies employing the IGL EOS might be reasonable to eliminate the complexity of mathematical models of real gas effects. In addition to the atmospheric pressure flames, inter-mediate (i.e. 5 and 10 atm) pressure flames are also investigated by artificially increasing the Planck mean absorption coefficient of the optically thin medium model (Lignell et al. [45] have reported due to the short simulation time, and small soot load in comparison to the domain size, radiative heat transfer is found to be insignificant). In adiabatic 5 and 10 atm flames slight differences for soot volume fraction, (fv), and the scalar dissipation rate are observed between the RGL and the IGL EOS models which are eliminated by the addition of the radiative heat loss. However, at 35 atm, the IGL EOS model has been shown to extremely
over-predict not only the flame temperature but also the soot quantities by 25% to 100% in comparison with the RGL EOS model predictions. In elevated pressure flames, a similar trend has been observed for the soot volume fractions and the soot number density (N), while high values of the soot mass fraction, ($Y_s$), exhibit a less scattered profile in the mixture fraction coordinate which is limited in the range of $\phi = 0.4$ to 0.9. The soot generation rate has been observed to have smaller standard deviation than its means in elevated pressure flames, while it has been observed to be larger than its means in atmospheric and intermediate pressure flames. As the time evolves in the RGL EOS elevated pressure flame by increasing the intensity of the flow more scattered trends have been detected in both flame characteristics and soot properties indicating soot properties are highly affected by the local flow characteristics.

After testing the validity of the current model with past literature, and revealing the importance of real gas effects on the soot formation/oxidation process, 2D DNS have been conducted for all cases to investigate pressure effects on the process in a much deeper manner. It has been known for decades that in hydrocarbon flames soot production has been increased by increased ambient pressure. Such a behavior has been noted in the current work by investigating flames of 1, 5, 10 and 35 atm with the RGL and the IGL EOS models. These predictions show the effects of pressure on the soot production/oxidation processes. For the first three pressures the IGL EOS model has not deviated from the RGL EOS model significantly. However, for the flames of 35 atm the differences becomes highly significant.

The unity Lewis (Le) number assumption on the soot formation/oxidation process has been studied in 3D DNS of atmospheric pressure flames for both the RGL and the IGL EOS models. The comparison of non-unity and unity Le number adiabatic atmospheric pressure flames has been done since it has a crucial importance to help to better understand the influence of the unity Le number assumption on these flames. In order to verify the acceptability of the unity Le assumption in hydrocarbon flames, similar trends in flame
structure and soot properties should be obtained. Testing the validity of the unity Le assumption for the adiabatic atmospheric pressure flame predictions of the RGL and the IGL EOS models has revealed that coupling the EOS models with the unity Le number assumption under-predicts the flame temperature and soot properties. The known effect of the unity Le number on the enthalpy has been observed in these atmospheric pressure flames. Ignoring non-unity Le number effects has been shown to under-predict the soot quantities by at least an order of magnitude.
I would like to dedicate my thesis to my parents, Kadir and Zeliha Korucu, and my brothers, Aydin, Mehmet and Metin Korucu, who have believed in me and supported me so that I could get my PhD degree abroad.
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Chapter 1

Introduction

High pressure combustion occurs in many devices, such as diesel, gas turbine, and rocket engines, where the reacting fluids are in supercritical pressure states. In this high pressure supercritical combustion regime, generalized diffusion phenomena, and real gas effects are often enhanced. Significant research has been conducted to show these often neglected diffusion phenomena and real gas effects can be necessary to accurately capture the physics of combustion processes; particularly at high pressure [26, 65, 67, 89, 66, 19, 37, 13, 46, 45, 53].

Because of ongoing advances in computational ability, numerical simulation has played an increasingly significant role in analyzing reacting flows; especially under supercritical conditions too costly and complex to be widely studied using experimental methods. The most accurate tool is Direct Numerical Simulation (DNS) which resolves all spatial and temporal scales of the flow field. However, limited literature exists for DNS of high pressure combustion involving real gas effects, detailed chemistry, and generalized diffusion phenomena. This lack of research stems from the complexities involved in accurately modeling the chemical and diffusion processes and large increase in computational resources needed to calculate all pertinent physics. This dissertation address several aspects of high pressure
combustion with detailed literature reviews provided in each individual chapter by topic.

The current work starts with an *a priori* analysis of subgrid heat flux in high pressure mixing and reacting $H_2/O_2$ and $H_2/\text{Air}$ flames. The subgrid analysis is performed in both globally and locally conditioned regions within two $H_2/O_2$, one $H_2/\text{Air}$ reacting and one $H_2/O_2$ purely mixing case with varying Reynolds (Re) numbers from 850 to 2500. The initial pressure is 100 atm for all $H_2/O_2$ flows, and 35 atm for the $H_2/\text{Air}$ flame. The results are reported in the Chapter 2.

In Chapter 3 soot formation/oxidations processes in high pressure *Kerosene*/Air flames with a semi-global soot model with an optically thin medium radiation heat transfer model are then analyzed. The simulations have been done with two different equations of state (EOS): I - the Peng Robinson [real gas law (RGL)] and II- t Ideal Gas Law (IGL) for four different initial pressures varying from 1 to 35 atm (i.e. two distinct simulations are obtained for each EOS model for 1, 5, 10 and 35 atm ambient pressures). For validation purposes the results of the atmospheric pressure flames have been compared to the limited literature [46, 45, 96] and a good agreement has been noted. However, the literature suffers from a lack of high pressure 3D DNS with real gas effects and generalized diffusion effects. The results of the current work are the first attempt to generate such data. Additionally, in Chapter 3, for 3D inter-mediate pressure flames, artificially increased radiation heat loss effects on the flame and soot properties have been investigated. Lastly, in this chapter we have investigated pressure effects for 2D data.

Finally, in Chapter 4, the study is extended to investigate the commonly used unity Le number assumption on the soot formation/oxidation processes associated with the RGL and the IGL models for 1, 5, 10 and 35 atm pressure flames. The chapter starts with reporting the Le numbers conditioned on mixture fraction of non-unity Le number cases for both EOS models for all ambient pressures considered in this work. Furthermore, the 3D adiabatic atmospheric pressure results of Chapter 2 are used to test the validity of the
unity Le number assumption associated with both EOS models. Later, the pressure and EOS model effects coupled with the unity Le number assumption on the soot and flame properties are studied.
Chapter 2

A *Priori* Analysis of Subgrid Scale Pressure and Heat Flux in High Pressure Mixing and Reacting Shear Layers

2.1 Introduction

To simulate practical reacting flows, which are often accompanied by high Reynolds numbers and complex geometry, Large Eddy Simulation (LES) is quickly becoming widely used [28, 30, 71, 35]. In LES, each primitive variable is filtered into a ‘resolved’ scale plus a subgrid scale fluctuating component; mathematically represented by $\Phi = \bar{\Phi} + \Phi'$. The LES governing equations yield several unclosed terms that may require modeling as they contain unresolved information about the subgrid scales. Of these unclosed terms, the filtered chemical reaction rate, $S_{Y_\alpha}$, and subgrid turbulent stresses, $\Pi_{ij} = \rho \left< u_i u_j - \rho \left< u_i \right> \left< u_j \right> \right>$, have received the most attention [43, 69, 77, 71, 72]. Here, $\left< \cdot \right>$ indicates density weighted Favre filtering that can be related to the standard filtering by the following expression, $\left< \Phi \right> = \frac{\rho \bar{\Phi}}{\bar{\rho}}$. Decomposition of a Favre filtered instantaneous variable includes the
Favre filtered component plus a fluctuating component, \( \Phi = \langle \langle \Phi \rangle \rangle + \Phi'' \).

In addition to these unclosed terms, the filtered heat flux vector, \( \overline{Q_j(\Psi)} \), may also be unclosed terms depending on the formulation, where \( \Psi \) is the set of composition variables (mass fraction, \( Y_a \) temperature, \( T \), density, \( \rho \), etc.). Few previous studies regarding \( \overline{Q_j(\Psi)} \) in the total energy equation exist. Subgrid mass flux vectors were recently studied by Foster and Miller [27, 28], who found that subgrid mass flux vectors can be important in reacting flows (see below).

The subgrid heat flux vector has been considered in: (1) natural convection dominated turbulent thermal flows [101, 92, 41], (2) forced convection dominated turbulent thermal flows [95, 64, 22] and (3) atmospheric turbulent boundary layers [75, 74, 12]. However, little literature addresses the subgrid heat flux vector in either supercritical reacting or non-reacting flows. Exceptions include Selle et al. [80] and Borghesi and Bellan [8] who provide \textit{a priori} and \textit{a posteriori} analyses of the subgrid heat flux vector for binary mixing. They pointed out in oxygen/hydrogen mixing that the gradient of the subgrid heat flux is of the same order of magnitude as the leading order terms in the filtered energy equation. The model introduced in Ref. [80] for the subgrid heat flux vector was further investigated in Refs. [87, 88] by Taskinoglu and Bellan. However, these studies have been limited to non-reacting mixing layers.

In a related matter, Foster and Miller [27, 28] have conducted \textit{a priori} analyses of DNS data of high pressure turbulent combustion to examine the importance of the subgrid mass flux vectors. They argue that for reacting flows a more detailed examination of unclosed subgrid terms is needed than purely global statistics. This is because turbulent flame dynamics can be strongly influenced by localized phenomena, including extinction and reignition. Therefore, in addition to a global analysis, they have also considered conditioning statistics on localized regions of the stoichiometric region, regions of large filtered subgrid scalar dissipation, large temperature subgrid variance, large reaction rate,
and large scalar subgrid variance. They show that even though the subgrid mass flux vector’s contribution is relatively small when analyzed globally, it is much more important for regions of large subgrid scalar dissipation and large subgrid temperature variance. Furthermore, the divergence of the subgrid mass flux vectors is of the same order, or larger, than the divergence of the filtered mass flux vectors in these regions. Foster and Miller [25] have also pointed out that with increasing Reynolds number, and filter widths, the contribution of the subgrid mass flux vector increases.

### 2.2 Objectives

Based on the above, the primary objectives of this chapter are to: I- conduct an *a priori* analysis of the subgrid heat flux vector, both in mixing and reacting flows, based on the DNS data generated in Refs. [27, 28, 25, 24], and II- test the hypothesis that the subgrid heat flux vector may require modeling in LES of realistic, high pressure, variable density, multicomponent, turbulent flames.

### 2.3 Governing Equations

The following sections in this chapter provide a post processing analysis of a previously generated DNS database of high pressure $H_2/O_2$ and $H_2/Air$ non-premixed shear flames. The DNS solves the fully compressible form of the continuity, momentum, total energy, and all species’ partial density equations in a temporarily developing reacting shear layer geometry (Fig. 2.1). The Peng-Robinson real gas state equation, real property models, and generalized heat and mass diffusion are also incorporated. The chemical reaction is calculated by a detailed pressure dependent 19-step and 8-species mechanism [84] for the $H_2/O_2$ flame. For the $H_2/Air$ simulations either a 4-step 3-species nitrogen mechanism is
added [84]. The details are provided in Refs. [27, 28, 25].

Equally spaced grid points from $0 \leq x_1 \leq L_1$ and $0 \leq x_3 \leq L_3$ in $x_1$ and $x_3$ directions are used for the computational mesh. In the $x_2$ direction an analytical mapping function proposed in [36] is employed to stretch the mesh in the cross-stream direction. In the vorticity thickness region a fine grid spacing ($\Delta x_2 \approx \Delta x_1$) is used, while the spacing stretches towards each of the free stream boundaries where $\Delta x_2 \approx 5\Delta x_1$ [24]. Assigning a coarser mesh in the free stream regions reduces the computational time significantly. Detail of the mapping the can be found in [24].

An eight order central explicit finite difference method and a forth order Runge-Kutta are employed to solve for the spatial and time derivatives, respectively. At each Runge-Kutta stage tenth order filtering is also applied to control spurious oscillations. The message passing interface (MPI) routines are used to parallize the code in all three directions. Further information about the numerical algorithms can be found in Ref. [38].

**Figure 2.1:** Direct Numerical Simulation domain schematic

The current work only provides the summarized forms of the governing equations. For a more detailed formulation see: Palle [65], Palle and Miller [66], Vasudevan [90] and
Foster [24]. The compressible form of the Navier-Stokes, energy, and species transport equations are:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_j} [\rho u_j] = 0, \tag{2.1}
\]

\[
\frac{\partial}{\partial t}(\rho u_i) + \frac{\partial}{\partial x_j} [\rho u_i u_j + P \delta_{ij} - \tau_{ij}] = 0, \tag{2.2}
\]

\[
\frac{\partial}{\partial t}(\rho e_t) + \frac{\partial}{\partial x_j} [(\rho e_t + P) u_j - u_i \tau_{ij} + Q_j + \sum_{\alpha=1}^{N} H'_{\alpha} J'_{j,\alpha}] = S_e, \tag{2.3}
\]

\[
\frac{\partial}{\partial t}(\rho Y_{\alpha}) + \frac{\partial}{\partial x_j} [\rho Y_{\alpha} u_j + J'_{j,\alpha}] = S_{Y_{\alpha}}, \tag{2.4}
\]

where \( t \) is time, \( x_j \) is the spatial coordinate vector, \( \rho \) is the mixture density, \( u_j \) is the mixture velocity, \( P \) is the pressure, \( \delta_{ij} \) is the Kronecker delta tensor, \( \tau_{ij} \) is the (Newtonian) viscous stress tensor, \( e_t \) the total specific energy (internal plus kinetic), \( Q_j \) is the Bearman-Kirkwood form of the heat flux vector, \( \sum_{\alpha=1}^{N} H'_{\alpha} J'_{j,\alpha} \) is the enthalpy flux (\( N \) is the total number of species) in which the partial molar enthalpy for species \( \alpha \) is \( H'_{\alpha} = \partial H'/\partial X_{\alpha} \) (\( X_{\alpha} \) is the mole fraction of species \( \alpha \)), and the molar mass flux vector for species \( \alpha \) is \( J_{j,\alpha} \).

The relation \( J'_{j,\alpha} = M_{\alpha} J'_{j,\alpha} \) (\( M_{\alpha} \) stands for the molecular weight of species \( \alpha \)) converts the molar flux vector to the mass flux vector, \( S_e \) is the chemical reaction source term for the energy equation \( (S_e = -\sum_{\alpha=1}^{N} \omega_{\alpha} \Delta H^0_{\alpha}) \), where \( \omega_{\alpha} \) is the reaction rate for species \( \alpha \) and \( \Delta H^0_{\alpha} \) is the enthalpy of formation. For Eq. (2.4), \( Y_{\alpha} \) represents the mass fraction of species \( \alpha \), and \( S_{Y_{\alpha}} \) is the chemical reaction source term for species \( \alpha \).

In order to capture real gas effects, the Peng-Robinson equation of state is used:

\[
P = \frac{RT}{V' - B_m} - \frac{A_m}{V'^2 + 2V'B_m - B_m^2}, \tag{2.5}
\]
where $V'$ is the specific molar volume, $R$ is the universal gas constant, and the temperature is $T$. The mixture parameters $A_m$ and $B_m$ are calculated by non-linear mixing rules found in Refs. [34, 57].

The effects of multicomponent, differential, and cross diffusion effects have been included into the equations employed for the DNS calculations. Under high pressures, these effects have the ability to be significant [65, 33, 32, 58, 26]. Harstad and Bellander derived the full form of the heat and mass flux vectors taking into account the aforementioned effects by using non-equilibrium thermodynamics and fluctuation theory (as kinetic theory is not applicable for high pressure dense fluids). The heat flux vector may be represented as a superposition of terms proportional to temperature, pressure, and mole fraction gradients:

$$Q_j = Q_j^T + Q_j^{X_1} + Q_j^{X_2} + \ldots + Q_j^{X_{N-1}}, \quad (2.6)$$

The superscripts indicate the variable gradient upon which the term is proportional to. The expanded heat flux form adopted in the DNS is derived from non-equilibrium thermodynamics and fluctuation theory:

$$Q_j = -\left\{ \kappa + \sum_{\beta=1}^{N-1} \sum_{\gamma=\beta+1}^{N} X_{\beta} X_{\gamma} \alpha_{BK}^{\beta\gamma} \frac{R \rho}{M_m} D_m^{\beta\gamma} \right\} \frac{\partial T}{\partial x_j}$$

$$- \sum_{\beta=1}^{N} \left\{ X_{\beta} \left[ \frac{M_{\gamma}}{M_m^2} X_{\gamma} \alpha_{BK}^{\beta\gamma} \rho D_m^{\beta\gamma} \right] V_{\beta} \right\} \frac{\partial P}{\partial x_j}$$

$$- \sum_{\eta=1}^{N-1} \sum_{\beta=1}^{\eta} \sum_{\gamma=\beta+1}^{N} \left\{ \frac{R T}{M_m^2} X_{\gamma} \alpha_{BK}^{\beta\gamma} \rho D_m^{\beta\gamma} \alpha_{D}^{\beta\eta} \right\} \frac{\partial X_{\eta}}{\partial x_j}, \quad (2.7)$$

where $N$ is the number of species, $M_m = \sum_{\alpha=1}^{N} X_{\alpha} M_{\alpha}$ is the mixture molecular weight and $X_{\alpha}$ and $M_{\alpha}$ are the mole fraction and molecular weight of species $\alpha$, respectively. In the above, $D_m^{\beta\gamma}$ represents the binary mass diffusivities, $\kappa$ is the mixture thermal conductivity,
and the thermal and mass diffusion factors are represented by $\alpha_{BK}^{\beta\gamma}$ and $\alpha_{D}^{\beta\gamma}$, respectively. The first term represents the expanded Fourier component. The remaining terms are referred to as Dufour diffusion and represent heat flux due to both mole fraction and pressure gradients. Similarly, the mass flux vector is:

$$
\overline{J}_j^\beta = -nD_m^{\beta\gamma} \sum_{\gamma \neq \beta}^N \left\{ X_\beta X_\gamma \frac{M_\beta}{M_m} \alpha_{BK}^{\beta\gamma} \right\} \frac{\partial \ln T}{\partial x_j} - \sum_{\gamma \neq \beta}^N nD_m^{\beta\gamma} \left\{ -\frac{M_\beta M_\gamma}{M_m M_m} X_\beta X_\gamma V'_\gamma + \frac{M_\gamma M_\gamma}{M_m M_m} X_\gamma X_\gamma V'_\beta \right\} \frac{\partial P}{\partial x_j} - \sum_{\eta=1}^{N-1} \left\{ \sum_{\gamma \neq \beta}^N \frac{M_\beta M_\gamma}{M_m M_m} X_\beta nD_m^{\beta\gamma} \alpha_D^{\beta\gamma} + \frac{M_\gamma M_\gamma}{M_m M_m} X_\gamma nD_m^{\gamma\beta} \alpha_D^{\gamma\beta} \right\} \frac{\partial X_\eta}{\partial x_j},
$$

where $n$ is the molar density ($n = \rho / M_m$), and $V'_\beta$ is the partial molar volume of species.

The simulations were conducted on the Palmetto Cluster at Clemson University with resolutions up to $560 \times 720 \times 336 \approx 135$ million grid points and using as many as 2016 processing cores ($Re_0 = 2500$). The initial vorticity thickness, $\delta_0$, is defined as the initial transition length from one free stream to the other. The “flame Reynolds number” is defined by $Re_F = U_0 \delta_0 \rho_0 / \mu_0$; where $U_0$ is the velocity difference between the two streams, $\rho_0$ is the averaged density, and $\mu_0$ is the averaged viscosity from each stream value. For the current work, 4 cases are considered as summarized in Table 2.1; including one $H_2/O_2$ purely mixing case, three $H_2/O_2$ reacting cases with different Reynolds numbers ($Re_0 = 850, 2500$) all at an ambient pressure $P_0 = 100$ atm, and one $H_2/Air$ reacting case with a different initial pressure ($P_0 = 35$ atm) and Reynolds number ($Re_0 = 2500$). Compared with the $H_2/O_2$ flame, the $H_2/Air$ flame is significantly diluted by $N_2$ which results in lower flame temperature and extensive local extinction; especially across the stoichiometric
mixture fraction \((\phi \approx 0.1)\) region. The mixture fraction, \(\phi\), is defined as:

\[
\phi = \frac{sY_{H_2} - Y_{O_2} + Y_{O_2}^0}{sY_{H_2}^0 + Y_{O_2}^0},
\]

where \(0 \leq \phi \leq 1\), \(s\) is the stoichiometric constant, \(Y_{H_2}\) and \(Y_{O_2}\) are the local mass fraction for \(H_2\) and \(O_2\), respectively, and \(Y_{H_2}^0\) and \(Y_{O_2}^0\) are the corresponding free stream values. In the pure fuel region \(\phi = 1\), and in the pure oxidizer region \(\phi = 0\). Intermediate values indicate varying degrees of mixedness. The mixture fraction and its dissipation are important quantities in many turbulent combustion models. The reason for considering each of these regions separately is that flame dynamics such as ignition, extinction and reignition are known to be highly localized. Therefore, if a model were deemed to perform well when only globally averaged it may not necessarily perform well locally in important flame regions.

\[
\begin{array}{cccccccc}
\text{Run} & \text{Oxidizer} & P_0 (atm) & \text{Type} & T_0 (K) & Re_0 & Re_0(t_f) & N_1 \times N_2 \times N_3 & t_f^* \\
1 & Oxygen & 100 & Mixing & 700 & 2000 & 15000 & 384 \times 384 \times 232 & 80 \\
2 & Oxygen & 100 & Reacting & 700 & 850 & 4500 & 384 \times 384 \times 232 & 80 \\
3 & Oxygen & 100 & Reacting & 700 & 2500 & 16000 & 560 \times 720 \times 336 & 115 \\
4 & Air & 35 & Reacting & 700 & 2500 & 18000 & 532 \times 630 \times 340 & 148 \\
\end{array}
\]

Table 2.1: Summary of simulation parameters from all cases considered. \(P_0\) is initial pressure, \(T_0\) is the initial temperature, \(Re_0\) the initial Reynolds number, \(Re_0(t_f)\) the Reynolds number calculated based on the vorticity thickness at the final simulation time, \(N_2\), \(N_2\), \(N_3\) are the number of grid points in the \(x_1\), \(x_2\), \(x_3\) direction respectively, and \(t_f^*\) is non-dimensionlized final simulation time.
2.4 Subgrid Analysis

A subgrid analysis relevant to LES is next performed. In LES a general filtered variable, $\Phi$, is defined by the convolution integral:

$$
\overline{\Phi}(x) = \int_{\Omega} G(x_j - x'_j)\Phi(x'_j)dx'_j,
$$

(2.10)

where $G(x_j)$ is the filter kernel defined over the domain $\Omega$. As compressible flow is considered for this work, employing the Favre filtering concept is necessary. The Favre filter is a density-weighted filtering operation, and is equivalent to a standard filter for constant density flows.

The subgrid analysis is done in both a 'global' flame manner (i.e. conditioned on $0.01 \leq \phi \leq 0.99$) as well as by conditioning on various specific regions within each flame. The reason for considering each of these localized regions listed in Table 2.2 separately is that flame dynamics such as ignition, extinction and re-ignition are known to be highly localized. Therefore, if a model were deemed to perform well when only globally averaged it may not necessarily perform well locally in important flame regions. Local regions are therefore analyzed by conditionally filtering on subsets of the global flame defined in Table 2.2, where $E(\Phi)$ indicates the expected value (average) over the global flame region $(0.01 \leq \phi \leq 0.99)$.

The current work uses a spherical top-hat filter with different diameters ($\Delta$) listed in Table 2.3. The filter diameter $3\delta_{u0}$ provides a realistic LES grid resolution as listed in Table 2.3. For example, with $\Delta = 3\delta_{u0}$, the $H_2/O_2$, $Re_0 = 2500$ reacting case will have grid resolution as $62 \times 80 \times 37$ in LES (with $\Delta x_{1,LES} = \Delta/2$) Therefore, most results herein
The global region

\[ 0.01 \leq \phi \leq 0.99 \]

The stoichiometric condition of \( H_2/O_2 \) flame

\[ 0.1 \leq \phi \leq 0.2 \]

The stoichiometric condition of \( H_2/Air \) flame

\[ 0.01 \leq \phi \leq 0.05 \]

Large filtered temperature variance

\[ \frac{\langle \langle T''^2 \rangle \rangle}{E(\langle \langle T''^2 \rangle \rangle)} \geq 2 \]

Large subgrid kinetic energy

\[ \frac{k_{sgs}}{E(k_{sgs})} \geq 2 \]

Large filtered mixture fraction variance

\[ \frac{\langle \langle \phi''^2 \rangle \rangle}{E(\langle \langle \phi''^2 \rangle \rangle)} \geq 2 \]

High OH reaction rate regions

\[ \frac{\omega}{E(\omega)} \geq 2 \]

High filtered scalar dissipation

\[ \frac{\chi^\phi}{E(\chi^\phi)} \geq 2 \]

Elevated temperature regions

\[ \frac{T}{T_0} \geq 2 \]

Table 2.2: The local regions within which statistical analyses have been done.

...are for a filter diameter of \( 3\delta_{\omega 0} \). However, the temporally developing shear layer can be seen as a small portion of a real combustion chamber. Larger filter diameters can be interpreted as relevant to simulations of larger domains. Figure 4 of Ref. [27] provides various spectra from the \( H_2/O_2 \) case at \( Re_0 = 2500 \) together with the relative filtering wavenumber (the middle of the three corresponding to \( \Delta = 3\delta_{\omega 0} \)). Note that the \( H_2/O_2 \) and \( H_2/Air \) flames behave quite differently. The former’s chemical reaction is near equilibrium whereas the latter displays large levels of local extinction. The diluted levels of oxygen in the \( H_2/Air \) flame lead to much smaller flame temperatures in comparison the \( H_2/O_2 \) flames. See Refs. [27, 28, 25] for details.

The exact (unclosed) filtered LES governing equations can be expressed as follows:

\[ \frac{\partial }{\partial t} \bar{\rho} + \frac{\partial }{\partial x_i} \left[ \bar{\rho} \langle \langle u_j \rangle \rangle \right] = 0, \]  

(2.11)

\[ \frac{\partial }{\partial t} (\bar{\rho} \langle \langle u_i \rangle \rangle) + \frac{\partial }{\partial x_j} \left[ \bar{\rho} \langle \langle u_i u_j \rangle \rangle + \bar{P} \delta_{ij} - \overline{\rho \delta_{ij}} \right] = 0, \]  

(2.12)
\[
\frac{\partial}{\partial t} (\bar{\rho} \langle \langle e_i \rangle \rangle) + \frac{\partial}{\partial x_j} \left[ (\bar{\rho} \langle \langle e_i \rangle \rangle)u_j + \bar{P}u_j - \bar{u}_i \bar{\tau}_{ij} + \bar{Q}_j + \sum_{a=1}^{N} H'_{a,j,a} \right] = \bar{S}_e, \tag{2.13}
\]

\[
\frac{\partial}{\partial t} (\bar{\rho} \langle \langle Y_\alpha \rangle \rangle) + \frac{\partial}{\partial x_j} \left[ \bar{P} \langle \langle Y_\alpha u_j \rangle \rangle + \langle \langle J'_{j,a} \rangle \rangle \right] = \bar{S}_{Y_\alpha}, \tag{2.14}
\]

\[
\bar{P} = \frac{RT}{V' - B_m} - \frac{A_m}{V'^2 + 2V'B_m - B_m^3}, \tag{2.15}
\]

The set of LES primitive variables is \( \Psi = [\bar{\rho}, \langle \langle u_j \rangle \rangle, \langle \langle e_i \rangle \rangle, \langle \langle Y_\alpha \rangle \rangle] \). Any term which cannot be directly obtained from this set of variables is unclosed and therefore requires modeling. For example, the non-linear convective term in Eq. 2.12 is typically decomposed as:

\[
\langle \langle u_i u_j \rangle \rangle = \langle \langle u_i \rangle \rangle \langle \langle u_j \rangle \rangle + \tau_{sgs,ij}, \tag{2.16}
\]

where \( \tau_{sgs,ij} = [\langle \langle u_i u_j \rangle \rangle - \langle \langle u_i \rangle \rangle \langle \langle u_j \rangle \rangle] \) and is referred to as the subgrid stress tensor. As DNS contains all the information about any instantaneous variable, the ‘exact’ subgrid information may be accessed by filtering the DNS data. This is referred to as the \textit{a priori} approach to LES analysis (in contrast to the \textit{a posteriori} approach in which an actual LES is performed).

Even though many unclosed terms exist in the LES filtered equations, this work focuses on the heat flux vector, Eq. (2.7). A detailed analysis and modeling for a canonical high pressure \( H_2/O_2 \) reacting shear layer is performed. The magnitude of the exact filtered heat flux, \( |\bar{Q}_j(\Psi)| \), is compared with the magnitude of the heat flux vector calculated using only filtered primitive variables, \( |\bar{Q}_j(\Psi)| \). Here, \( \Psi \) represents the set of primitive variables used in the DNS, \( \Psi = [\rho, u_j, e_i, Y_\alpha] \). Other variables such as temperature and pressure are obtained from this base set; eg. \( T(\Psi) \) and \( P(\Psi) \). Similarly, Eq. (2.7) provides the heat
flux vector, $Q_j(\Psi)$. For LES, variables other than the set $\Psi$ can only be approximated as functions of $\Psi$; eg. $T(\Psi)$, $P(\Psi)$, and $Q_j(\Psi)$. Therefore, terms calculated in such a manner are not necessarily either equal to, nor even approximately equal to, the terms in the exact LES equations. For example, $P(\Psi)$ appears in the exact LES momentum equation. However, only $P(\Psi)$ can be calculated. The difference, therefore, defines an unclosed ‘subgrid, or ‘subgrid scale (SGS) pressure; $P_{sgs} = P(\Psi) - P(\Psi)$. Subgird terms such as this may, or may not, require modeling depending in part on their magnitude with respect to both the resolved term [eg. $P(\Psi)$] as well as with respect to other terms in the LES equations.

<table>
<thead>
<tr>
<th>Run</th>
<th>$\Delta x_1$</th>
<th>$\Delta_1 = 3\delta_{\omega 0}$</th>
<th>$\Delta_2 = 4\delta_{\omega 0}$</th>
<th>$\Delta_2 = 5\delta_{\omega 0}$</th>
<th>$(N_1 \times N_2 \times N_3)_{LES}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.12$\delta_{\omega 0}$</td>
<td>25$\Delta x_1$</td>
<td>33$\Delta x_1$</td>
<td>42$\Delta x_1$</td>
<td>31$\times$31$\times$19</td>
</tr>
<tr>
<td>2</td>
<td>0.12$\delta_{\omega 0}$</td>
<td>25$\Delta x_1$</td>
<td>33$\Delta x_1$</td>
<td>42$\Delta x_1$</td>
<td>31$\times$31$\times$19</td>
</tr>
<tr>
<td>3</td>
<td>0.16$\delta_{\omega 0}$</td>
<td>19$\Delta x_1$</td>
<td>25$\Delta x_1$</td>
<td>32$\Delta x_1$</td>
<td>59$\times$76$\times$35</td>
</tr>
<tr>
<td>4</td>
<td>0.17$\delta_{\omega 0}$</td>
<td>18$\Delta x_1$</td>
<td>24$\Delta x_1$</td>
<td>30$\Delta x_1$</td>
<td>62$\times$80$\times$37</td>
</tr>
</tbody>
</table>

Table 2.3: Filtering diameter size used to postprocess the DNS data. 'Run' corresponds to the DNS cases presented in Table 2.1, $\Delta_1 = 3\delta_{\omega 0}$ is the spherical filter diameter this work focuses on, $\Delta x_1$ is the DNS grid spacing in the $x_1$ direction, $\delta_{\omega 0}$ is the DNS initial vorticity thickness, $(N_1 \times N_2 \times N_3)_{LES}$ are the equivalent LES grid resolutions in $x_1$, $x_2$, and $x_3$ directions, respectively, for $\Delta_1 = 3\delta_{\omega 0}$.

The subject of the current work is in testing whether or not the subgrid heat flux vector defined by $Q_{sgs,j} = \overline{Q_j(\Psi)} - Q_j(\Psi)$ is significant for reacting flows. A priori testing using the DNS data is chosen as all forms of the heat flux and subgrid heat flux vectors can be calculated ‘exactly’. The ‘exact’ filtered heat flux vector, which cannot be calculated within an actual LES, is expressed as follows:
\[
Q_j(\Psi) = -\left\{ \kappa + \sum_{\beta=1}^{N-1} \sum_{\gamma>\beta} X_\beta X_\gamma (\psi) \alpha_\beta \alpha_\gamma \frac{R\rho}{M_m} D_m^{\beta\gamma} \right\} \frac{\partial T(\Psi)}{\partial x_j} (2.17)
\]

\[
- \sum_{\beta=1}^{N} \left\{ X_\beta \sum_{\gamma\neq\beta} \left[ \frac{M_\gamma}{M_m^2} X_\gamma (\psi) \alpha_\beta \alpha_\gamma \frac{\rho D_m^{\beta\gamma}}{\bar{\rho}} \right] V_{\beta}(\Psi) \right\} \frac{\partial P(\Psi)}{\partial x_j}
\]

\[
- \sum_{\eta=1}^{N-1} \sum_{\beta=1}^{N} \left\{ RT(\Psi) \sum_{\gamma\neq\beta} \left[ \frac{M_\gamma}{M_m^2} X_\gamma (\psi) \alpha_\beta \alpha_\gamma \frac{\rho D_m^{\beta\gamma}}{\bar{\rho}} \right] \alpha_\eta D(\Psi) \right\} \frac{\partial X_\eta(\Psi)}{\partial x_j},
\]

In contrast, the ‘resolved heat flux vector that can be calculated within an LES is:

\[
\bar{Q}_j(\bar{\Psi}) = -\left\{ \kappa(\bar{\Psi}) + \sum_{\beta=1}^{N-1} \sum_{\gamma>\beta} X_\beta(\bar{\Psi}) X_\gamma(\bar{\Psi}) \alpha_\beta \alpha_\gamma \frac{R\bar{\rho}}{\bar{M}_m} D_m^{\beta\gamma}(\bar{\Psi}) \right\} \frac{\partial T(\bar{\Psi})}{\partial x_j} (2.18)
\]

\[
- \sum_{\beta=1}^{N} \left\{ X_\beta(\bar{\Psi}) \sum_{\gamma\neq\beta} \left[ \frac{M_\gamma}{M_m^2} X_\gamma(\bar{\Psi}) \alpha_\beta \alpha_\gamma \frac{\bar{\rho} D_m^{\beta\gamma}(\bar{\Psi})}{\bar{\rho}} \right] V_{\beta}(\bar{\Psi}) \right\} \frac{\partial P(\bar{\Psi})}{\partial x_j}
\]

\[
- \sum_{\eta=1}^{N-1} \sum_{\beta=1}^{N} \left\{ RT(\bar{\Psi}) \sum_{\gamma\neq\beta} \left[ \frac{M_\gamma}{M_m^2} X_\gamma(\bar{\Psi}) \alpha_\beta \alpha_\gamma \frac{\bar{\rho} D_m^{\beta\gamma}(\bar{\Psi})}{\bar{\rho}} \right] \alpha_\eta \bar{D}(\bar{\Psi}) \right\} \frac{\partial X_\eta(\bar{\Psi})}{\partial x_j},
\]

where all the terms are dependent on the variable set \(\bar{\Psi}\) and can be calculated in an LES simulation. The difference between Eqs. (2.17) and (2.18) gives the subgrid heat flux vector which might require modeling and is the focus of the current chapter. The subgrid analysis aims to analyze the possible necessity of modeling the subgrid heat flux vector by calculating correlation coefficients and ratio distributions of \(|Q_j(\Psi)|\) to \(|\bar{Q}_j(\bar{\Psi})|\). Ratios relative to other subgrid terms in the filtered energy equation [Eq. (2.13)] are also considered. Note that there are a number of ways in which to interpret the proper calculation of \(Q_j(\Psi)\) within an actual LES. Equation (2.18) above can certainly be used directly. To be able to neglect the subgrid heat flux vector, the condition of \(\bar{Q}_j(\bar{\Psi}) \approx Q_j(\Psi)\) should at least be satisfied.
2.4.1 Subgrid Heat Flux Vector Analysis

The subgrid heat flux vector has been neglected in nearly all previous LES studies. No validation or testing of this assumption exists for reacting flows. The subgrid heat flux vector is defined by \( Q_{\text{sgs},j} = \overline{Q_j(\Psi)} - Q_j(\overline{\Psi}) \), where the filtered heat flux, \( \overline{Q_j(\Psi)} \), is calculated by directly filtering Eq. (2.7), and the resolved heat flux vector, \( Q_j(\overline{\Psi}) \), is calculated using the resolved primitive variables. The subject of this part of the current work is to analyze the subgrid heat flux vector. Four cases are focused on, including the \( H_2/\text{Air} \) reacting case with \( Re_0 = 2500 \), the \( H_2/O_2 \) purely mixing case with \( Re_0 = 2000 \), and the \( H_2/O_2 \) reacting cases with \( Re_0 = 850 \) and \( Re_0 = 2500 \) respectively. All results below use the filter diameter of \( \Delta/\delta_{\omega0} = 3 \).

To compare the filtered heat flux vector to the resolved heat flux vector, Fig. 2.2 presents PDFs of the ratio of \( |\overline{Q_j(\Psi)}| \) to \( |Q_j(\overline{\Psi})| \). The PDFs have a Gaussian like shape, with peaks located at \( |\overline{Q_j(\Psi)}|/|Q_j(\overline{\Psi})| \approx 1 \), indicating the resolved heat flux vector has similar magnitude as the filtered heat flux vector. Compared with the reacting cases, the resolved heat flux behaves differently in the purely mixing case, as these PDFs display a wider distribution of ratios. In addition, the resolved heat flux vector deviates more from the filtered heat flux vector in specific local regions; e.g., the resolved heat flux vector magnitude can be as small as half of the filtered heat flux vector magnitude, or as large as twice the filtered heat flux vector magnitude, in the stoichiometric mixture fraction region [Fig. 2.2(b)] and regions of large \( OH \) reaction rate [Fig. 2.2(c)]. Relatively little influence of the Reynolds number is observed although significantly different behavior is observed for the \( H_2/\text{Air} \) flame in comparison to the \( H_2/O_2 \) flames; presumably due to the high levels of extinction in the former. Note that only the reacting cases are shown in [Fig. 2.2(c)] since there is no \( OH \) reaction rate to condition on for the mixing case. Figure 2.3 further illustrates the difference between the filtered heat flux and the resolved heat flux by presenting PDFs.
of the ratio of $|\partial Q_j(\Psi)/\partial x_j|/|\partial Q_j(\overline{\Psi})/\partial x_j|$ (the divergence being the actual term in the energy equation). As differentiation enhances small scale features, these PDFs have a much wider distribution compared with those in Fig. 2.2.

Based on the above results, there is significant difference between the filtered heat flux and the resolved heat flux. The influence of the subgrid heat flux (i.e. $Q_{sgs,j} = Q_j(\Psi) - Q_j(\overline{\Psi})$) is next compared with that of other subgrid terms in the filtered energy equation. First, the subgrid heat flux is compared with the subgrid convective term, $(\rho e u_j)_{sgs}$. Figure 2.4 presents PDFs of the ratio of $|Q_{sgs,j}|/(\rho e u_j)_{sgs}$. In the global flame region [Fig. 2.4(a)] and regions of large OH reaction rate [Fig. 2.4(c)], the subgrid heat flux magnitude is typically 5% of the subgrid convective energy. However, this value can be up to as large as 25%. The subgrid heat flux vector is relatively larger in the stoichiometric surface [Fig. 2.4(b)] where it can exceed the magnitude of $(\rho e u_j)_{sgs}$ in the $H_2/O_2 Re_0 = 850$ flame. However, in general, the relative influence of the subgrid heat flux is significantly diminishing with respect to that of the subgrid convective term with increasing Reynolds number. A similar analysis and conclusions are drawn from Fig. 2.5 which shows PDFs of $|\partial Q_{sgs,j}/\partial x_j|/|\partial(\rho e u_j)_{sgs}/\partial x_j|$.

Finally, Figs. 2.6 and 2.7 provide a similar analysis of the relative effects of the subgrid heat flux in comparison to the subgrid pressure work term and its gradient, respectively. The subgrid heat flux is found to be more significant relative to these terms than to the subgrid convective terms. Again, for mixing cases the ratios are generally small. However, for reacting cases the subgrid heat flux is as important and generally larger than the corresponding pressure work terms. Nevertheless, as before, the trend is decreasing importance with increasing Reynolds number. This suggests that neglecting the subgrid heat flux in LES of high Reynolds number reacting flows is most likely reasonable.
Figure 2.2: PDFs of $|Q_j(\Psi)|/|Q_j(\Psi)|$, conditioned on: (a) $0.01 \leq \phi \leq 0.99$, (b) the stoichiometric surface, (c) $\omega^{OH}/E(\omega^{OH} \geq 2)$. 
Figure 2.3: PDFs of $|\frac{\partial Q_j(\Psi)}{\partial x_j}| / |\frac{\partial Q_j(\Psi)}{\partial x_j}|$, conditioned on: (a) $0.01 \leq \phi \leq 0.99$, (b) the stoichiometric surface, (c) $\omega_{OH}/E(\omega_{OH} \geq 2)$. 
Figure 2.4: PDFs of $|Q_{sgs,j}|/|(\rho e_1 u_j)_{sgs}|$, conditioned on: (a) $0.01 \leq \phi \leq 0.99$, (b) the stoichiometric surface, and (c) $\omega^{OH}/E(\omega^{OH}) \geq 2$.
Figure 2.5: PDFs of $|\partial Q_{sgs,j} / \partial x_j| / |\partial (\rho e_i u_j)_{sgs} / \partial x_j|$, conditioned on: (a) $0.01 \leq \phi \leq 0.99$, (b) the stoichiometric surface, and (c) $\omega^{\text{OH}} / E(\omega^{\text{OH}} \geq 2)$. 
Figure 2.6: PDFs of $|Q_{sgs,j}|/(P_{u_j})_{sgs}$, conditioned on: (a) $0.01 \leq \phi \leq 0.99$, (b) the stoichiometric surface, and (c) $\omega_{OH}/E(\omega_{OH} \geq 2)$.
Figure 2.7: PDFs of $|\partial Q_{sgs,j}/\partial x_j|/|\partial (Pu_j)_{sgs}/\partial x_j|$, in regions described by: (a) $0.01 \leq \phi \leq 0.99$, (b) the stoichiometric surface, and (c) $\omega^{\text{OH}} / E(\omega^{\text{OH}} \geq 2)$. 
Chapter 3

*Kerosene / Air Reacting Shear Flows*

Including Soot Formation/Oxidation

With Radiative Heat Transfer

3.1 Introduction

In combustion of hydrocarbons (i.e. fossil fuels) soot formation is an unavoidable phenomenon that has been studied by scholars for decades. Essentially, soot is formed in the fuel-rich side of the flame due to the lack of oxidizer (i.e. incomplete combustion) [59]. The gaseous hydrocarbons are transformed into soot which are basically solid state carbon [39]. In the processes of soot formation and oxidation there are 4 steps that occur: I - particle inception/nucleation which are considered as the origin of soot formation, II - a) surface growth which occurs when the gas phase species stick to the surface of soot particles [11], and b) coagulation where the small-scale soot particles unite to generate large-scale soot particles [11, 39], III - aging, and IV - surface oxidation where soot particles are oxidized by mainly \( O_2 \), O and OH molecules yielding gaseous CO and \( CO_2 \) etc. [11, 39]. Despite
extensive work, the interaction between flow characteristics and soot formation/oxidation are still relatively poorly understood. In order to gain more insight about this phenomenon researchers have used different soot models that can be sorted in a list of three: I - fully empirical correlations, II - semi-empirical schemes that involve using some experimental inputs to solve soot formation rate equations, and III - detailed models that attempt to solve the rate equations which are associated with soot formation [39]. Even though empirical models have been accepted and used widely, the dependency on experimental inputs reduces them to be defined and restricted by the experimental setups [39]. This has forced scholars to pursue more generalized alternative schemes (i.e. detailed chemistry models). The detailed chemistry models aim to define the chemical kinetics of polyaromatic hydrocarbons (PAH) and surface growth of soot particle globally [39].

However, when adopting such aforementioned soot models for sooting flames coupling with radiation heat transfer is typically required. The radiative heat transfer within both lightly and heavily sooting laminar/turbulent flames has been modeled in many ways. Due to the massive computational cost and work, relatively simpler radiation models have been adopted widely by the turbulent combustion community. These models primarily aim to couple thermal radiation and soot formation/oxidation processes under the following assumptions. Inherently, soot particles are much smaller than any flow or scalar length scales and possess a perfect spherical geometry [48, 59]. This assumption implicitly adopts Rayleigh Scattering, that was proposed by Lord Rayleigh [59]. Owing to having such small sizes, soot particles’ temperature is the same as the flame temperature, hence they emit radiative energy more actively than gaseous molecules (i.e. combustion products) do [59].

Assuming the participating medium is negligibly scattering and formed by a number of gray gases [59], leads to the concept of the weighted-sum-of-gray-gases (WSGG) approach. WSGG helps to calculate the absorption coefficient of the appropriate gases (i.e. in this case they are $CO_2$, $H_2O$ and $CO$) [59, 99]. The negligible scattering assumption with
Rayleigh scattering allowed for the calculation of the absorption coefficient of soot particles as well [59]. The optical thickness of the medium is also an important parameter for the radiation models. The medium can be optically thin or thick depending on how sooty the flame is [76]. Optically thin and gray medium assumption says that emitted radiation will leave the medium due to the lack of self-absorption or scattering [3, 86] (i.e. lightly sooting flames). While the Planck mean extinction coefficient is recommended for the optically thin problems, the Rosseland-mean extinction coefficient is recommended for optically thick mediums [3, 59]. Both optically thin and thick assumptions have been extensively used in order to analyze the interaction between soot formation/oxidation processes and radiative heat loss.

Inherently capturing soot formation/oxidation coupled with radiation heat loss is a highly challenging process. As a result of the complexity of this process researchers generally neglect real gas effects (i.e. ideal gas law is adopted), and use highly simplified forms of the mass and heat flux vectors (often with a unity Lewis number (Le) assumption). These simplifications may cause a failure while predicting the flames characteristics, since at high pressures many fuels exhibit real gas effects inconsistent with the ideal gas law. There is therefore a need for a better understanding of such phenomena.

The following sections of this chapter provide an analysis of a generated DNS database of both atmospheric and high pressures Kerosene/Air reacting shear flames. As before the DNS solves the fully compressible form of the continuity, momentum, total energy, and all species’ partial density equations in a temporarily developing reacting shear layer geometry. The Peng-Robinson real gas state equation, real property models, and generalized heat and mass diffusion are also incorporated. The chemical reaction is calculated by a reduced pressure dependent 29-step and 10-species (including soot) mechanism [93] for the Kerosene/O₂ flame. Nitrogen is assumed to be inert.
3.2 Objectives

Based on the above, the primary objectives of this chapter are to: (I) conduct a comparison between the results of the atmospheric pressure flames with the limited literature to validate the adopted soot model in this work, (II) investigate real gas effects on the soot formation/oxidation processes by conducting a comparison with the IGL EOS model results, (III) study effects of the pressure on the processes of soot formation/oxidation and effectivity of the IGL EOS model in flames at high ambient pressures.

3.3 Chemical Kinetics and Numerical Approach

The reduced chemistry which is the basis of the DNS calculation includes a pressure dependent 29-step 10-species and soot ($C_{12}H_{24}$, $O_2$, $H_2O$, OH, H, O, CO, $N_2$, $H_2$ and $CO_2$) mechanism developed by Wang [93]. Table 3.1 provides the details of the mechanism. In Table 3.1, Wang [93] has suggested $C_{12}H_{24}$ as a generic surrogate for kerosene or RP-1. $C_{12}H_{24}$ has been modeled kinetically having 41.7% paraffin and 58.7% naphthene to be matched to $\frac{41}{56}$ for paraffin/naphthene as reported in the literature. The balance between paraffin and naphthene is highly important in order to catch the correct soot prediction since naphthene’s breakdown process to soot is quicker than paraffin’s [93]. $C_{12}H_{24}$ has been modeled to be a mixture of many pure elements that have the same average thermophysical–chemical profiles of the reported kerosene/RP–1 [93]. Consequently, having $C_{12}H_{24}$ as one–formula parent fuel includes the advantages of multiple fuel models while increasing the computational efficiency [93]. A summary of the comparison of the surrogate fuel’s thermophysical –chemical characterization is given in Table 3.2 [93]. The detailed list of the data has been used for comparison, and can be found in Ref. [93].

Paraffin (straight – chain hydrocarbon) and naphthene (cyclic – chain hydrocarbon)
irreversible breakdown processes have been depicted with two global steps, and wet–CO mechanism turns the intermediate species into the final products in Table 3.1 [93]. The reaction rates of global steps of paraffin and napthene have been adjusted in agreement with the ratio of two hydrocarbons, $C_{12}H_{24}$ (proposed surrogate by Ref. [93]). The forth–order polynomial temperature dependent heat capacity form is shown in Eq. (3.2) [93], and Eq. (3.1) [93] is integrated from Eq. (3.2) to calculate enthalpy [93]. The coefficients of Eq. (3.2) and (3.1) are listed in Table 3.3 [93].

\[
\frac{H}{RT} = a_1 + \frac{a_2}{2}T + \frac{a_3}{3}T^2 + \frac{a_4}{4}T^3 + \frac{a_5}{5}T^4 + \frac{a_6}{T} \quad \text{(3.1)}
\]

\[
\frac{C_p}{R} = a_1 + a_2T + a_3T^2 + a_4T^3 + a_5T^4 \quad \text{(3.2)}
\]

The DNS data is calculated by solving the governing equations for 3D, temporally developing, non-premixed reacting shear layers of $C_{12}H_{24}/Air$. Details of the computational domain, mesh and numerical scheme used can be found in Ref. [24]. The mixing occurs at the center of the domain. For the current work, 12 distinct reacting simulations are considered for $Re_F = 2500$ as shown in Table 3.4. Four of the simulations include artificially enhanced radiation heat flux due to the soot and CO$_2$, H$_2$O and CO gaseous species, in order to observe its effects on the soot production/oxidation processes and flame characteristics with generalized diffusion (i.e. non-unity Le number). The initial temperature is 700 K for all the cases and four different ambient pressure are employed for the current work to be 1, 5, 10 and 35 atm. The convective Mach number is set to be 0.35.
<table>
<thead>
<tr>
<th>Reaction</th>
<th>A</th>
<th>B</th>
<th>E/R</th>
<th>Form</th>
<th>Ref.</th>
</tr>
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<td><strong>Parafin Global Step</strong></td>
<td></td>
<td></td>
<td></td>
<td>$p^{0.3}[C_{12}H_{24}]^{0.5}[O_2]$</td>
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</tr>
<tr>
<td>$C_{12}E_1H_{24} + 6O_2 \rightarrow 12CO + 12H_2$</td>
<td>3.888E+04</td>
<td>1</td>
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<td></td>
<td></td>
<td></td>
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<td>[93]</td>
</tr>
<tr>
<td>$C_{12}H_{24} + 6O_2 \rightarrow 12CO + 12H_2$</td>
<td>2.132E+07</td>
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<td><strong>Wet CO Mechanism</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td>$H_2 + O_2 = OH + OH$</td>
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<tr>
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<td>2.190E+13</td>
<td>0</td>
<td>2.590E+03</td>
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<td>$OH + OH = O + H_2O$</td>
<td>6.023E+12</td>
<td>0</td>
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<td>[94, 21]</td>
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<td></td>
<td></td>
<td>[93]</td>
</tr>
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<td>$C_s + 0.5O_2 \rightarrow CO$</td>
<td>1</td>
<td>0</td>
<td>0</td>
<td>$72R_{ox}[C_s]/(\rho D_s)$</td>
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</tr>
<tr>
<td>$R_{ox} = K_ApO_2\chi/(1+K_2pO_2)+K_BpO_2(1-\chi)$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>[61]</td>
</tr>
<tr>
<td>$\chi = 1/(1+(K_T/K_B)pO_2$</td>
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</tr>
<tr>
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<td>1.5098E+04</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$K_B$</td>
<td>4.4600E-03</td>
<td>0</td>
<td>7.6497E+03</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$K_T$</td>
<td>1.5100E+05</td>
<td>0</td>
<td>4.8817E+04</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$K_Z$</td>
<td>2.1300E+01</td>
<td>0</td>
<td>-2.063E+03</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td><strong>Homogeneous Soot Oxidation</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[93]</td>
</tr>
<tr>
<td>$C_s + OH = CO + H$</td>
<td>1.2200E+09</td>
<td>0.5</td>
<td>0</td>
<td>Standard</td>
<td>[93]</td>
</tr>
</tbody>
</table>

2. Wet = CO mechanism flames initially contains small amount of hydrogen and water vapor.

Table 3.1: Kerosene quasi global combustion kinetics mechanism [93].
<table>
<thead>
<tr>
<th>Property</th>
<th>Kerosene</th>
<th>RP – 1</th>
<th>Surrogate Fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molecular Formula</td>
<td>-</td>
<td>-</td>
<td>(C_{12}H_{24})</td>
</tr>
<tr>
<td>Molecular Weight</td>
<td>175</td>
<td>172 – 175</td>
<td>168</td>
</tr>
<tr>
<td>Elemental Formula</td>
<td>-</td>
<td>(CH_{1.95} – CH_{2.0})</td>
<td>(CH_{2.0})</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(CH_{1.9423}, CH_{1.953})</td>
<td></td>
</tr>
<tr>
<td>Formula Weight</td>
<td>-</td>
<td>13.97 – 14.03, 13.97</td>
<td>14.03</td>
</tr>
<tr>
<td>(H_e) kcal/g</td>
<td>-10.278</td>
<td>-10.241</td>
<td>-10.278</td>
</tr>
<tr>
<td></td>
<td>-10.321</td>
<td>-10.356</td>
<td></td>
</tr>
<tr>
<td>(H_{f,298K}) kcal/mole</td>
<td>-</td>
<td>(-5.430/CH_{1.9423})</td>
<td>(-92.200/C_{12}H_{24})</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(-7.683/CH_{2.0})</td>
</tr>
<tr>
<td>(C_p) kcal/mol.K</td>
<td>-</td>
<td>101</td>
<td>103</td>
</tr>
<tr>
<td>Paraffins (n and iso), %</td>
<td>-</td>
<td>41</td>
<td>41.7</td>
</tr>
<tr>
<td>Naphthe, %</td>
<td>-</td>
<td>56</td>
<td>58.3</td>
</tr>
<tr>
<td>Aromatics, %</td>
<td>5</td>
<td>5</td>
<td>0</td>
</tr>
<tr>
<td>Olefins, %</td>
<td>1</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 3.2: Comparison of thermophysical–chemical characterization of model fuel with reported data [93].

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>1000 to 5000 K</th>
<th>300 to 1000 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a_1)</td>
<td>0.36440206E+02</td>
<td>0.39508691E+01</td>
</tr>
<tr>
<td>(a_2)</td>
<td>0.54614801E-01</td>
<td>0.10207987E+00</td>
</tr>
<tr>
<td>(a_3)</td>
<td>-0.16091121E-04</td>
<td>0.13124466E-04</td>
</tr>
<tr>
<td>(a_4)</td>
<td>0.21478497E-08</td>
<td>-0.76649284E-07</td>
</tr>
<tr>
<td>(a_5)</td>
<td>-0.10131180E-12</td>
<td>0.34503763E-10</td>
</tr>
<tr>
<td>(a_6)</td>
<td>-0.63890109E+05</td>
<td>-0.52093574E+05</td>
</tr>
<tr>
<td>(a_7)</td>
<td>-0.12798973E+03</td>
<td>0.21980951E+02</td>
</tr>
</tbody>
</table>

Table 3.3: Thermodynamic coefficients for \(C_p\) of \(C_{12}H_{24}\) [93].
Run Oxidizer $P_0(\text{atm})$ EOS Le $T_0(K)$ $Re_0$ $N_1 \times N_2 \times N_3$ $t_f^*$ Radiation
1 Air 1 RG ≠1 700 2500 320 × 516 × 180 65 No
2 Air 1 IG ≠1 700 2500 240 × 480 × 144 65 No
3 Air 5 RG ≠1 700 2500 304 × 564 × 180 80 No
4 Air 5 RG ≠1 700 2500 304 × 564 × 180 80 Yes
5 Air 5 IG ≠1 700 2500 304 × 564 × 180 80 No
6 Air 5 IG ≠1 700 2500 304 × 564 × 180 80 Yes
7 Air 10 RG ≠1 700 2500 304 × 480 × 180 80 No
8 Air 10 RG ≠1 700 2500 304 × 481 × 180 80 Yes
9 Air 10 IG ≠1 700 2500 304 × 564 × 180 80 No
10 Air 10 IG ≠1 700 2500 304 × 564 × 180 80 Yes
11 Air 35 RG ≠1 700 2500 304 × 630 × 210 80 No
12 Air 35 IG ≠1 700 2500 304 × 640 × 180 65 No

Table 3.4: Summary of simulation parameters from all cases considered. $P_0$ is initial pressure, $T_0$ is the initial temperature, $Re_0$ the initial Reynolds number, $N_1$, $N_2$ and $N_3$, are the number of grid points in the $x_1$, $x_2$, and $x_3$, direction respectively, and $t_f^*$ is non-dimensionalized final simulation time.

### 3.4 Soot Model

As discussed earlier, soot models consist of 4 steps: nucleation, growth, aging and oxidation. Due to the extremely complex nature of soot formation many researchers aim to reduce the complexity by only treating the most significant steps of the process [48]. Studies frequently investigate the soot load and its effect on flame characteristics and temperature due to radiative heat loss [48]. Researchers have reported that pressure and additive gaseous species have significant effects on the soot formation mechanism [14, 16, 17, 50]. With increasing ambient pressure soot formation (i.e soot volume fraction or soot mass fraction) increases dramatically due to the higher operating temperatures.

Due to the limitation of empirical and semi-empirical methods most studies incorporate detailed mechanism models. One model that has a 4-step soot model for non-premixed includes nucleation, growth, particle coagulation and oxidation has been proposed by Leung et al.[44]. Recently, Lignell et al. [46, 45] have conducted both 2-D and 3-D soot
analysis in an ethylene flame. Lignell [44] has used Leung et al.’s [44] model that assumes monodisperse size distribution defined by soot particle number density, n, and soot mass fraction [48]. The model has the following steps [48]:

I- Nucleation : \( C_2H_2 \rightarrow 2C_{(s)} + H_2 \)

II- Growth : \( nC_{(s)} + C_2H_2 \rightarrow (n + 2)C_{(s)} + H_2 \)

III- Oxidation : \( C_{(s)} + \frac{1}{2}O_2 \rightarrow CO \)

IV- Coagulation : \( (n) \rightarrow (n - 1) \)

where \( C_{(s)} \) corresponds to soot implying gas-state graphite carbon [44]. In the first step, Leung et al. [44] have assumed that \( C_2H_2 \), acetylene, begins the soot nucleation, but it is not necessarily the parent fuel of the mechanism. Acetylene is produced as a result of the fuel breakdown mechanism [44]. While naphthene and aromatic hydrocarbons are instantaneously condensed to form PAH which are known to be soot ancestors, paraffins have to be decomposed into napthenes and aromatics and eventually form PAHs [93]. This work adopts a soot formation global step that soot is formed directly from the parent fuel, \( C_{12}H_{24} \), instead of acetylene suggested by Ref. [93]. Even though in early stages soot contains large amounts of \( H_2 \) molecules, lack of generalized presentation of soot leads researchers to assume pure \( C_s \) [44]. The second step of the model presents the surface growth process assuming that \( C_2H_2 \) in the gas state sticks to the surface of soot particles [44]. Surface growth of soot usually occurs in the high-temperature flame regions [6]. Leung et al. [44] adopted the following reaction rate source term suggested by Harris et al.[31];

\[
R_2 = k_2(T)f(S)[C_2H_2],
\]

where \( [C_2H_2] \) is the molar concentration in \( \text{kmol} / \text{m}^3 - \text{mixture} \) and S is the soot surface
area in $m^2/m^3$ – mixture. Leung et al. [44] have defined the surface area as:

$$S = \pi (d_p^2)(\rho N) = \pi \left( \frac{6}{\pi \rho_s N} \frac{Y_s}{N} \right)^{\frac{3}{2}},$$  \hspace{1cm} (3.4)$$

where $d_p$ is the soot particle diameter ($250 \times 10^{-8}$ in this study [93]), $\rho$ is the gaseous species density, $\rho_s$ is the soot density taken here as 2000 kg/m$^3$ [48, 44, 93], $Y_s$ is the soot mass fraction, $N$ [particles/kg – mixture] [44]. The third step, the soot oxidation process includes $O_2$, OH and O molecules. Even though OH and O aggressively attack soot in order to oxidate it, in most cases they have been excluded from models since their mass fractions are insignificant in comparison to the $O_2$ mass fraction [76]. The current work adopts heterogeneous ($O_2$) and homogeneous (OH) soot oxidation steps [93]. The three steps given above lead to the number density equation [44]. The particle number density is reduced by agglomeration that can be summarized with the forth step listed above [44]. Since the current work is to study the soot effects on flame characteristics, only soot formation and soot oxidation mechanisms are adopted. For only comparison purposes with the literature, Eq. (3.5) is adopted from Ref. [102] to calculate the soot particle number density which is associated to the soot mass fraction:

$$N = Y_s 6 \left( \frac{\rho_t}{\rho_s} \frac{1}{\pi d_p^3} \right).$$  \hspace{1cm} (3.5)$$

In addition to the governing equations defined in the Chapter 2 Eq. (2.1 - 2.5) one additional governing equation is adopted to calculate the soot mass fraction of Ref. [48]:

$$\frac{\partial (\rho Y_s)}{\partial t} = - \frac{\partial (\rho Y_s v_i)}{\partial x_i} - \frac{\partial j_{s,i}}{\partial x_i} + S_{Y_s},$$  \hspace{1cm} (3.6)$$

$$\frac{1}{\rho_t} = \frac{Y_s}{\rho_s} + \frac{1 - Y_s}{\rho},$$  \hspace{1cm} (3.7)$$
where \( Y_s \) is the soot mass fraction, \( \rho \) is the gaseous species density calculated with Eq. (3.7) Ref. [102], \( j_{s,i} \) is the soot diffusion calculated with Eq. (3.8) [48], and \( S_{Y_s} \) is the reaction term for soot. The soot mass diffusion based on thermophoretic effects is calculated as:

\[
    j_{s,i} = -0.556 \rho Y_s \frac{\nu}{T} \nabla T. \tag{3.8}
\]

which is taken from Ref [48]. Since thermophoretic effects are considered insignificant in comparison to the turbulent advection, it is usually ignored [102]. However, it is included in the current study for the sake of completeness.

### 3.5 Radiation Model

A complete combustion model should include radiative heat transfer in order to predict the flame formation, production of solid particles (i.e. soot), localized radiative heat flux and flame temperature [91]. For decades, researchers have attempted to propose radiative heat transfer models which are not only low on computational cost, but also accomplish ‘accurate’ radiative heat loss predictions. Since chemical kinetics of soot processes are intensely temperature dependent, accurately predicting the flame temperature is critical in sooting flames [91].

The accuracy of combustion models can be affected by the accuracy of the radiative heat transfer model (RHTM) adopted to predict flame temperature [91]. Inaccurate prediction of flame temperature can cause combustion models to fail to estimate flame characteristics accurately. Choosing an appropriate RHTM is affected by the nature of the problem, computational costs, desired degree of accuracy, and medium of the system [91]. Optically thin and gray medium assumption has been adopted with a view of a cold
The coefficients used to calculate Planck mean absorption coefficient of \( H_2O \) and \( CO_2 \) [2].

<table>
<thead>
<tr>
<th></th>
<th>( H_2O )</th>
<th>( CO_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( C_0 )</td>
<td>-0.23093</td>
<td>18.741</td>
</tr>
<tr>
<td>( C_1 )</td>
<td>-1.12390</td>
<td>-121.310</td>
</tr>
<tr>
<td>( C_2 )</td>
<td>9.41230</td>
<td>273.500</td>
</tr>
<tr>
<td>( C_3 )</td>
<td>-2.99880</td>
<td>-194.050</td>
</tr>
<tr>
<td>( C_4 )</td>
<td>0.51382</td>
<td>56.310</td>
</tr>
<tr>
<td>( C_5 )</td>
<td>-1.8684 ( 10^{-5} )</td>
<td>-5.8169</td>
</tr>
</tbody>
</table>

Table 3.5: The coefficients used to calculate Planck mean absorption coefficient of \( H_2O \) and \( CO_2 \) [2].

surrounding leads to the following [2, 59, 91]:

\[
Q_{rad} = 4 \sigma \sum_{j=1}^{n} p_{\alpha_j} \times a_{p,\alpha_j} \times (T^4 - T_{\infty}^4)
\]

(3.9)

where \( \sigma \) is the Steffan–Boltzman constant (= \( 5.699 \times 10^{-8} \) W/m\(^2\)K\(^4\)), \( p_{\alpha_j} \) is the partial pressure of species \( \alpha_j \), \( a_{p,\alpha_j} \) is the Planck mean absorption coefficient of species \( \alpha_j \), \( T \) is the instantaneous flame temperature in Kelvins and \( T_{\infty} \) is the ambient temperature consisted with an assumption of emission-only medium [2].

The temperature dependent Planck mean absorption coefficient of \( H_2O \) and \( CO_2 \) are given by Eq. (3.10) [2], and the coefficients used to calculate the curve fit as listed in Table 3.5 [2].

\[
a_{p,\alpha} = C_0 + C_1 \times (\frac{1000}{T}) + C_2 \times (\frac{1000}{T})^2 + C_3 \times (\frac{1000}{T})^3 + C_4 \times (\frac{1000}{T})^4 + C_5 \times (\frac{1000}{T})^5
\]

(3.10)

Equation (Fig. 3.11) [2] gives the Planck mean absorption coefficient of CO and the coefficients for this curve fit as listed in Table 3.6 [2].

\[
a_{p,CO} = C_0 + T \times (C_1 + T \times (C_2 + T \times (C_3 + T \times C_4)))
\]

(3.11)

As mentioned earlier, soot particles are very small and Ragleigh scattering theory
Table 3.6: The coefficients used to calculate Planck mean absorption coefficient of CO [2].

<table>
<thead>
<tr>
<th></th>
<th>$T(K) \leq 750K$</th>
<th>$T(K) \geq 750K$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_0$</td>
<td>4.78690</td>
<td>10.0900</td>
</tr>
<tr>
<td>$C_1$</td>
<td>-0.09530</td>
<td>-0.01183</td>
</tr>
<tr>
<td>$C_2$</td>
<td>2.95775 $10^{-4}$</td>
<td>4.77530 $10^{-3}$</td>
</tr>
<tr>
<td>$C_3$</td>
<td>-4.25732 $10^{-7}$</td>
<td>-5.816909 $10^{-10}$</td>
</tr>
<tr>
<td>$C_4$</td>
<td>2.02894 $10^{-10}$</td>
<td>-2.5334 $10^{-14}$</td>
</tr>
</tbody>
</table>

can be adopted at relevant wavelengths [59]. The extinction coefficient can be estimated by [59]:

$$
\kappa_\lambda = \beta_\lambda = C_0 \frac{f_v}{\lambda}, 
C_0 = \frac{36\pi nk}{(n^2 - k^2 + 2^2) + 4n^2k^2} \tag{3.12}
$$

where $C_0$ is a soot index refraction dependent constant [59], and $f_v$ is the soot volume fraction ($f_v = \rho_t * Y_s / \rho_s$ [48]). Felske et al. [23] have proposed an average value for the Planck–mean and the Rosseland–mean extinction coefficient:

$$
\kappa_m = \beta_m = 3.72 f_v C_0 \frac{T}{C_2} \tag{3.13}
$$

where $C_2 = 1.4388 cmK$ is the second Planck function constant Eq. (3.13) can only be adopted while working with very small particles for both optically thin and thick mediums [59]. The radiative heat transfer can now be calculated by Eq. (3.9) coupled with the energy equation Eq. (2.3).
3.6 Real Gas and Pressure Effects on Soot Formation Including Radiative Heat Transfer

Practical devices such as gas turbines, internal combustion engines and diesel engines often operate at supercritical pressures [100]. In such systems adopting the IGL EOS has been known to be potentially invalid due to real gas effects [100]. Even though adopting more realistic mathematical models are required to eliminate the errors from the IGL EOS simplifications, researchers tend to employ the IGL EOS based models for atmospheric and elevated pressures [14] for both laminar [14, 15, 16, 17, 42, 49, 51, 50, 82, 83, 85] and turbulent [102, 6, 18, 76, 96, 7, 76, 99, 4, 98, 10, 29, 68, 86, 97, 79, 48] radiating non-premixed reacting flows. The literature suffers from nonavailability of turbulent non-premixed flame studies with high pressure real gas effects, and generalized diffusion phenomena.

Liu et al. [49] have conducted an atmospheric (with the IGL EOS) laminar ethylene-air co-flow jet diffusion flame calculations employing the modified semi-empirical soot model of Ref. [44] and the optically thin (OTM) and the discrete ordinates radiation model (DOM) considering spectral absorption coefficients of H$_2$O, CO$_2$ and CO. Ref. [49] has employed a different soot oxidation model based on the results of Refs. [9, 62, 61, 60]. They have adopted the Nagle-Strickland-Constable O$_2$ soot oxidation model [61] with the rate constants of Ref. [60] and Bradley et al.’s [9] O radical soot oxidation rate constants. Neoh et al. [62] have shown the importance of O$_2$ and OH soot oxidations depending on these two species concentrations, and Bradley et al. [9] have stated that O radicals also oxidize the soot particles. Ref. [49] has reported that there is only a 5 K difference between the DOM and the OTM’s peak temperature predictions. The soot volume fraction, nucleation, growth and OH soot oxidation predictions have been noted to be better predicted with the OTM radiation model even though it slightly under-predicts the mentioned soot quantities [49].
Ref. [15] has employed an acetylene based semi-empirical soot model proposed by Ref. [44] and the DOM of Ref. [49] under atmospheric pressure for lightly sooting methane/air and heavily-sooting ethylene-air flames. The calculated temperature profile of the methane-air flame by Ref. [15] has been reported to be under-predicted by 100-200 K in the centerline and over-predicted at the outer surface of the flame in comparison to experimental data. The methane mole fraction is over-predicted in some regions of the flame by a factor of 4 relative to experimental results by Ref. [15]. While their model cannot exceed Ref. [83]’s predictions in accuracy for both temperature and the methane profiles, the peak soot volume fraction predictions have been reported to be better than Ref. [83]’s predictions. The peak soot volume fraction has been over-predicted in both Refs [15, 83]. The numerical results of the ethylene-air flame have been reported to be quite similar to those of Liu et al. [49]. Both Ref. [15] and [49] under-predict the peak temperature by ≈120 K. The peak soot volume fraction prediction is slightly over-predicted by ≈7 % in comparison to the experimental data by Ref. [15], while Ref. [49] has reported excellent agreement. Soot distribution predictions have been reported to be lower than the measured data in the centerline by Refs [15] and [49].

Charest [14] studied pressure and gravity effects on soot formation in two different laminar diffusion flames I - pure ethylene-air (0.5 to 5 atm) and II- pure methane-air (1 to 60 atm) by employing the IGL EOS, the semi-empirical soot model of Ref. [44] and the DOM for radiative heat flux calculations. Even though overall soot predictions are not in agreement with those from experimental data, the effects of pressure and prediction of the trends has been captured adequately in Ref. [14]. The predicted flame heights have been observed to be shorter, especially for the high pressures, due to the under-predicted near wall temperatures for the ethylene-air flames, whereas flame heights have been over-predicted in the methane-air flames (i.e. over-predicted near wall temperatures) for all pressures [14]. Charest Ref. [14] has addressed the differences between zero and normal-gravity flames at
the same operation pressures in the methane-air flames. In their work, zero-gravity flames tend to have higher soot, lower temperature and wider soot regions than normal-gravity flames when the pressure is higher than 10 atm [14].

Liu et al. [50] have studied axisymmetric co-flow laminar pure methane-air diffusion flames both numerically and experimentally to examine pressure effects on the flame structure and soot production at pressures varying from 5 to 40 atm by adopting the IGL EOS and a semi-empirical soot model from Ref. [44] including $O_2$, OH and O soot oxidation and the DOM [50]. The experimental results they have obtained (for pressures 1 to 80 atm) showed that flame height increases between 1 to 5 atm, and remains almost constant between 5 to 20 atm and then begins to lower with linear dependency on the pressure [50]. Their numerical model has slightly over-predicted the flame heights for 5 to 20 atm, and poorly catches the shrinkage of the flame for the higher pressures [50]. Liu et al. [50] have adopted two different surface growth rate models (linear and non-linear dependency on soot surface area) to examine the pressure dependency of soot load. They refer to the non-linear model as Model I while referring the linear model Model II [50]. Model I has been reported to anticipate the soot volume dispersions better than Model II, while it is unable to match the accuracy of Model II’s peak soot volume fractions for 5, 20 and 40 atm [50]. Furthermore, they have investigated the soot oxidation effect on soot volume fraction at the same pressures [50]. They have concluded that soot oxidation effect on soot volume fraction decreases with increasing pressure due to the reduction of $C_2H_2$ concentration while soot production increases which implies the importance of soot oxidation on soot volume fraction predictions [50].

Refs. [16] and [17] adopt the same numerical framework that has been employed by Ref. [14] for laminar flames at elevated pressures (1 to 20 atm) to compare the results with experimental data. Both works have adopted the non-linear soot surface growth rate model (Model I of Ref. [50]), OH and O soot oxidation steps are also added based on Ref.
Charest et al. [16] do not only inspect the pressure effect but also look for the effect of $CO_2$ addition to the fuel (methane) both experimentally and numerically. They have investigated one pure methane, one 80% methane-20% $CO_2$, and one 60% methane-40% $CO_2$ flame and they refer to them as F0, F20 and F40, respectively [16]. The experiments have been done at 5 atm and above for F20, and 10 atm and above for F40 flames [16]. While the soot volume fraction predictions have been reported to be significantly lower than the experimental results [16], the experimental trends have been well predicted by the model [16]. The ethylene-air flame in Ref. [17] has been diluted by $N_2$ to reduce the soot formation with ambient pressures varying from 10 to 35 atm. The model of Ref. [17] generally over-predicts the soot volume fractions while it captures the soot transportation in the flame that have been observed in their experiments.

Lautenberger et al. [42] have conducted a study for developing a new soot formation/oxidation model of non-premixed hydrocarbon flames and introduce a primal calibration to the new model. Soot formation and oxidation have been suggested to be temperature and mixture fraction dependent [42]. They have suggested an $O_2$ soot oxidation step that is independent of soot surface area, as the proposed $O_2$ oxidation is restrained by $O_2$ transport into the soot oxidation region (i.e. OH soot oxidation is a surface area dependent process) [42]. Later, by using the relationship between laminar smoke point height and the peak soot formation, the model has been universalized for multiple fuels (i.e. ethylene, propylene and propane) [42]. Their model differs in three ways from the general modeling approach by eliminating the soot number density conservation equation, soot formation is taken to be an explicit function of mixture fraction (i.e. independent of parent fuel and no precursor species, $C_2H_2$) and independent of fuel laminar smoke point height calculations [42]. The radiation heat flux has been calculated by employing the narrow band radiation model [42]. While their first attempt to globalize the model for the fuels mentioned above captures the trends of the experimental data, quantitatively it is in need to be improved [42]. The
simulations were conducted at atmospheric pressure with the IGL EOS [42].

Said et al. [76] have studied an atmospheric ethylene-air laminar flame a laminar version of GENMIX 1, and a turbulent flame (by employing an Eulerian-Lagrangian approach) with the OTM for radiative heat flux for both flames [76]. First, they have tested their semi-global soot model on an atmospheric laminar ethylene-air flame [76]. Soot number density has been ignored due to its insignificant effect on atmospheric diffusion flames. Instead of soot number density, a mean particle diameter of soot is adopted [76]. They have reported a fairly good agreement with experimental results of Ref. [78], and pointed out the radiation effect on sooting flames [76]. The existence of strong effect of temperature and oxidizer fluctuations’ effects on soot formation and destruction process have been emphasized (the observed deviation has been reported to be 200%) [76].

Bai et al. [4] have coupled a 2D Favre-averaged k-ε and the Boussinesq gradient-diffusion hypothesis for Reynolds stress and transport fluxes with a detailed laminar flamelet modeling for detailed soot model and the OTM has been used for radiative heat flux. Bai et al. [4] have not introduced a soot number density transport equation like Ref. [76]. The predicted mean mixture fraction, temperature and soot volume fraction has been reported to be in good agreement with measured data [4]. Radiation has been observed to be effective on soot formation/destruction at low scalar dissipation rates [4].

Yoo and Im [96] have modeled a steady 2D turbulent ethylene-air counterflow non-premixed flame using DNS with a semi-empirical soot model and the DOM for radiative heat flux using the S3D 2 code that solves the fully compressible, 3D reacting Navier-Stokes (N-S) equations with the IGL EOS [46, 45]. They have unveiled the interaction between soot formation/destruction and turbulence of the dependency of soot formation to local flow characteristics, temperature, and fuel load, and the soot number density dependency

---

1GENMIX is designed to solve 2D parabolic flows of high Peclet and Reynolds Numbers [1].
2S3D is a DNS code developed by Sandia National Laboratories that uses finite difference approximations to solve fully compressible N-S equations on Cartesian grids [46].
of the high temperature flame regions [96]. They have observed that turbulence causes an increase in soot load by creating additional flame volume, and a decrease soot load by depleting soot pockets out of the high-temperature flame regions (spending less time in the high-temperature flame regions) [96].

Lignell et al. [45, 46] have done two sets of DNS (2D and 3D) atmospheric ethylene-air flame to investigate soot-flame interactions and transport in an unsteady flame structure. The S3D code with the OTM has been used for radiative heat flux calculations. They have observed that flame curvature causes transportation of soot either into or away from the flame that leads to widely scattered soot in the mixture fraction coordinate [46, 45]. They have concluded the importance of considering unsteady and multidimensional effects on soot formation and transport in turbulent flames [46, 45]. Furthermore, they have emphasized the importance of radiative heat loss by running a 2D simulation for 50 ms [46], however, for 3D simulations radiative heat loss has been reported to be insignificant due to large domain size, short simulation run time and low soot concentration of the simulation [45].

Recently Bisetti et al. [6] have conducted a 2D DNS of soot formation in a detailed chemical mechanism of an atmospheric n-hephane/air non-premixed flame to examine the unsteady strain effects on soot growth and transport ignoring dissipation, body forces, and Soret and Dufor effects. They have reported that in high scalar dissipation regions soot formation and growth from PAH occurs and related it to the reduction of soot load at high mixing in turbulent sooting flames [6].

In the current work, a semi-global soot production/oxidation of Kerosene/Air turbulent flames has been investigated for atmospheric and elevated pressures using the OTM radiative heat flux assumption for non-unity Le numbers by employing a RGL and the IGL EOS. The results of atmospheric pressure flames have been compared with the limited literature [96, 46, 45]. However, since our research is more complex than the available literature, the results are the first attempt to study soot formation/destruction processes in
turbulent hydrocarbon flames with real gas effects at high pressure. The results are presented in Section 3.7. We start by comparing the RGL and the IGL EOS results for atmospheric flames and continue with the results of 5, 10 and 35 atm flames with \( \text{Re} = 2500 \) and \( \text{Ma} \) number \((= 0.35)\).

### 3.7 Results

The general tendency in the literature is using the IGL EOS for hydrocarbon flames due to the complexity of mathematical models of real gas effects. While Figs. 3.1 through 3.6 include the mean, rms and raw data (scatter) of normalized \( T(K) \), soot volume fraction \( (f_v) \), number density \( (N) \), soot mass fraction \( (Y_s) \), scalar dissipation \( (\chi_\phi) \) that are conditioned on mixture fraction at time of \( t_f = 65 \), Figs. 3.7 - 3.9 present contour plots of \( T(K) \), \( f_v \) and \( Y_s \) of the RGL and the IGL EOS model predictions of 1 atm flames at time of \( t_f = 65 \). Since the gaseous species obey the IGL EOS at low pressure and high temperatures, it is safe to say employing the IGL EOS for the flames operating at atmospheric pressure is probably a reasonable approach. The atmospheric turbulent flame investigation has significant importance to validate the current work since the literature only exist for atmospheric hydrocarbon flames of soot production/destruction processes. In this manner, the results of atmospheric pressure for different cases have been compared with the literature.

Yoo et al. [96] have stated that the high values of \( f_v \) do not necessarily exist in high temperature regions. Figures 3.2, and 3.8 support this by showing the high values of \( f_v \) exist in a wide portion of the flame volume whose temperature varies from \( \approx 1300 \) K to 1800 K. Furthermore, they have stated that \( N \) has been observed to be highly dependent on temperature, while \( f_v \) is dependent on local flow characteristics [96]. Figures 3.1 and 3.3 support their conclusion on \( N \). The high values of \( N \) occupies the high temperature flame region and is scattered widely where the temperature is scattered. Ref. [96] pointed out that
effects of transport and turbulence mixing must be taken into account in sooting turbulent flames. In addition, in Ref. [96], in the limited soot production regions, $f_v$ has relatively high values which has been explained by a large volume of soot transportation. Similar behavior has also been observed in the current work (Figs. 3.2 and 3.8). These results have been explained by the interaction between soot chemistry and transport [96]. Yoo et al. [96] have stated the peak of $f_v$ to be 2.4 times larger than the reported value in the literature of steady flames.

Refs [46, 45] have reported that the temperature is not scattered by implying the levels of mixing of their flame is low. In the current work, higher levels of mixing have been achieved (Figs. 3.1 and 3.7). This difference could be explained by comparing the scalar dissipation which have been calculated to be an order of magnitude larger than those in Refs [46, 45]. (since Ref. [96] and [6] have not provided any scatter plots of temperature, we are unable to compare the temperature scatter plots). Lignell et al. [46] have observed two distinct behaviors in $Y_s$ plots; I - slow soot nucleation rates force $Y_s$ to have a wide variation on the mixture fraction axes, and II - soot has been carried into the fuel rich side of the flow. As a result of the first behavior, $Y_s$ scatter plot present a dense band of $Y_s$ load below its higher range, and second behavior emphasizes the importance of the soot transportation in turbulent flames [46]. A similar trend has been shown in Fig. 3.4. The scatter plots of soot nucleation and growth rate have been highly affected by the temperature profile in Refs. [46, 45], while in the current work $w_{soot}$ has been shown to be similar to $Y_s$ scatter plots (Fig. 3.6). This could be explained with the different levels of mixing. In Ref. [45] the significant difference between the soot and gaseous species and how it affects the soot modeling in turbulent flames have been pointed out (i.e. difficulty of the soot modeling). They have stated that the conditional standard deviation (rms) of $Y_s$ are of the same order of magnitude with the conditional mean, whereas the gaseous species have an order of magnitude smaller rms values than their conditional means [45]. In contrast, the
soot nucleation has relatively small rms values since this process is only dependent on the gaseous species [45]. However, in the current work, while their first observation has been observed, the \( w_{soot} \) has a higher rms than its conditional means. Figures 3.6 and 3.4 present the \( w_{soot} \) and \( Ys \) conditional scatter, mean and rms of these soot quantities. Figure 3.4 is in good agreement with the first observation of Lignell et al. [46] while Fig. 3.6 shows a different trend for \( w_{soot} \) than the one in Ref. [46]. Since, in Ref. [46] the nucleation of soot (\( w_{soot} \)) has been shown to be extremely depended on the temperature profile, having a smaller rms of the \( w_{soot} \) is an expected behavior. However, in the current work, temperature is highly scattered (i.e. higher levels of mixing, Fig. 3.5) and the fuel breaks directly into soot; a conflicting trend has been observed for the \( w_{soot} \). In the current work, the rms of \( w_{soot} \) have been noted to be higher than the conditional means of the \( w_{soot} \).

Figure 3.1: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( < T/T_0 | \phi > \) for \( Re = 2500, Le \neq 1, \) and 1 atm ambient pressure at time of 65.

Figures 3.10 through 3.15 depict the soot and flame characteristics of adiabatic RGL and IGL model 5 atm flames. The high levels of mixing still present in both the RGL and the IGL EOS 5 atm flames indicate these two flames should possess similar flame characteristics as the atmospheric flames of the RGL and the IGL EOS shown above. As expected increasing pressure enhances the maximum flame temperature which has direct
Figure 3.2: On the left the RGL EOS model and on the right the IGL EOS model predictions of $\langle f|\phi \rangle$ for Re= 2500, Le ≠ 1, and 1 atm ambient pressure at time of 65.

Figure 3.3: On the left the RGL EOS model and on the right the IGL EOS model predictions of $\langle N|\phi \rangle$ for Re= 2500, Le ≠ 1, and 1 atm ambient pressure at time of 65.
Figure 3.4: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( < Y_s|\phi > \) for Re= 2500, Le ≠ 1, and 1 atm ambient pressure at time of 65.

Figure 3.5: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( < \chi|\phi > \) for Re= 2500, Le ≠ 1, and 1 atm ambient pressure at time of 65.
effects on the soot production/oxidation processes. Even though for the RGL and the IGL model flames’ temperatures we observe similar behavior in Fig 3.10, when the artificial radiation has taken into account IGL model under-predicts the flame temperature which is 55 K lower than the RGL flame temperature (i.e. 3% difference between the RGL and the IGL models). The artificially enhanced radiation heat loss results in the RGL flames to be 172 K, and IGL flames to be 224 K lower than the adiabatic flame temperature (Fig. 3.16). Figures 3.11 and 3.17 reveal the direct effect of temperature on soot production processes. For adiabatic flames of the RGL and the IGL models $f_v$ is $\approx 5 \times 10^{-5}$ ppm, while it is reduced by 10% for the RGL and 20% for the IGL models which emphasizes the significance of the temperature on soot characteristics. Furthermore, $f_v$ is still dominated by local flame characteristics (Figs. 3.10 3.16 3.11 3.17). However, the peak values of $N$ occupy the region where the high flame temperatures are observed once again, $N$ has started to immigrate to the fuel rich side of the flame. In contrast of the atmospheric flames (Fig. 3.3), the standard deviation of $N$ and $< N|\phi >$ conditioned one the mixture fraction are at the same order of magnitude. These two behaviors tell us that $N$ is no longer affected by the flame temperature and, like $f_v$ and $Y_s$, it will be dominated by local flame characteristics.
Figure 3.7: Contour plots of $T(\text{K})$ for pressure 1 atm, $\text{Le} \neq 1$ and $\text{Re}=2500$, top is the RGL model and bottom is the IGL EOS model predictions time of 65, 3D data.
Figure 3.8: Contour plots of $f v$ for pressure 1 atm, $Le=1$ and $Re=2500$, top is the RGL model and bottom is the IGL EOS model predictions time of 65, 3D data.
Figure 3.9: Contour plots of Ys for pressure 1 atm, Le≠1 and Re=2500, top is the RGL model and bottom is the IGL EOS model predictions time of 65, 3D data.
fv and $Y_s$ of the RGL and the IGL EOS flames at 5 atm (Figs. 3.11, 3.17, 3.12 and 3.18) have very similar behavior with fv and $Y_s$ of atmospheric pressure flames of the RGL and the IGL EOS (Figs. 3.2 and 3.4). Figure 3.13 and 3.19 indicate that slow soot formation rates still exist for both the RGL and the IGL EOS flames and the soot load is transferred to the fuel rich side. In addition to this, the peak value of $Y_s$ is shifted toward the fuel rich side of the flame (Fig. 3.13 and 3.19).

While the addition of radiative heat loss causes a reduction on temperature, it has a very insignificant effect on the distribution of the flame temperature throughput the domain (Figs. 3.10 and 3.16). Figures 3.18 and 3.19 reveal the dramatic reduction on N and $Y_s$ for both the RGL and the IGL EOS flames by 10% and 25% for N, and 30% and 29% for $Y_s$ (i.e. radiative heat loss effect on soot properties). Figures 3.14 and 3.20 show the radiative heat loss on the scalar dissipation rate which turn out to be the most effected flow characteristics for both the RGL and the IGL EOS flames at 5 atm. While the scalar dissipation prediction of IGL is $\approx$ 16 times smaller than RGL model prediction for adiabatic flames of 5 atm (Fig. 3.14), with radiative heat loss the IGL model prediction of $\chi^{phi}$ becomes 15% larger than RGL model prediction (Fig. 3.20). Lastly, $w_{soot}$ predictions are about to same for the adiabatic flames of the RGL and the IGL models ($w_{soot}$ of IGL is 4% smaller than RGL) (Fig. 3.15), and it is reduced by $\approx$ 45% for both the RGL and the IGL EOS flames when radiation heat loss is added to the calculations (Fig. 3.21). Overall, the flame and soot characteristics of adiabatic flame at 5 atm is well predicted by the IGL EOS model.

Figures 3.22 through 3.27 depict the soot and flame characteristics of adiabatic 10 atm flames, while Figs. 3.28 to 3.33 present those of 10 atm with radiation heat loss. However, increasing ambient pressure effect on temperature increment appears in both the RGL and the IGL EOS adiabatic flames (Fig. 3.22), the IGL EOS model flame temperature displays a more scattered behavior than the RGL EOS model’s. The clear effect of temperature on soot properties is observed once again in the 10 atm flames. Figure 3.23 shows that fv is
Figure 3.10: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( < T / T_0 | \phi > \) for \( \text{Re} = 2500, \text{Le} \neq 1, \) and 5 atm ambient pressure at time of 80.

Figure 3.11: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( < f v | \phi > \) for \( \text{Re} = 2500, \text{Le} \neq 1, \) and 5 atm ambient pressure at time of 80.
Figure 3.12: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<N|\phi>$ for $Re=2500$, Le $\neq 1$, and 5 atm ambient pressure at time of 80.

Figure 3.13: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<Ys|\phi>$ for $Re=2500$, Le $\neq 1$, and 5 atm ambient pressure at time of 80.
Figure 3.14: On the left the RGL EOS model and on the right the IGL EOS model predictions of $\langle \chi | \phi \rangle$ for Re= 2500, Le ≠ 1, and 5 atm ambient pressure at time of 80.

Figure 3.15: On the left the RGL EOS model and on the right the IGL EOS model predictions of $\langle w_{soot} | \phi \rangle$ for Re= 2500, Le ≠ 1, and 5 atm ambient pressure at time of 80.
Figure 3.16: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< T/T_0|\phi >$ for $Re= 2500$, $Le \neq 1$, and 5 atm ambient pressure at time of 80 radiation included.

Figure 3.17: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< f|v|\phi >$ for $Re= 2500$, $Le \neq 1$, and 5 atm ambient pressure at time of 80 radiation included.
Figure 3.18: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<N|\phi>$ for $Re = 2500$, $Le \neq 1$, and 5 atm ambient pressure at time of 80 radiation included.

Figure 3.19: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<Y_s|\phi>$ for $Re = 2500$, $Le \neq 1$, and 5 atm ambient pressure at time of 80 radiation included.
Figure 3.20: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< \chi | \phi >$ for Re= 2500, Le ≠ 1, and 5 atm ambient pressure at time of 80 radiation included.

Figure 3.21: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< w_{\text{soot}} | \phi >$ for Re= 2500, Le ≠ 1, and 5 atm ambient pressure at time of 80 radiation included.
still under local flow characteristics in both the RGL and the IGL EOS adiabatic flames. Even though flame temperatures increase by increasing pressure a slight increment has been observed in soot load for 10 atm flames indicating the soot destruction process becomes more dominant for these flames. Figure 3.24 shows that for adiabatic flames the RGL EOS fails to predict the N profile, the addition of radiative heat loss to calculations prediction of N gets better and capture the expected behavior (i.e. temperature dominated behavior). The profiles of $Y_s$ of adiabatic flames are shown to be well predicted by both the RGL and the IGL EOS models (Fig. 3.25). The quantity of $Y_s$ are over-predicted by both models for adiabatic flames with the addition of radiative heat loss the peak value of $Y_s$ of the RGL model is reduced by $\approx 30\%$, while there is no significant change in the IGL EOS model flame (Fig. 3.31). In both the RGL and the IGL EOS the high levels of mixing still exist in 10 atm adiabatic flames, while the RGL EOS models fail to predict the correct trend of $\chi^\phi$ (i.e. $\chi^\phi$ is highly affected by the temperature profile). The IGL EOS model successfully captures the trend (Fig. 3.26). By the addition of radiation the prediction of $\chi^\phi$ of the RGL EOS model is reduced to be 22 times smaller than the adiabatic flame prediction and to adopt a gaussian-like distribution; while there is no significant change in the IGL model prediction (Fig. 3.32). Lastly, the soot production rate predictions slightly reduced by radiative heat loss in the RGL EOS model and has an insignificant effect in the IGL EOS model (Fig. 3.27 and 3.33).

Figures 3.36 through 3.42 examine the RGL and the IGL EOS effects on the soot formation/oxidation process at elevated pressure, 35 atm. As discussed earlier, with increasing pressure (i.e. increasing density) $f_v$ increases. The $f_v$ quantity differences between two pressures have been observe to be two orders of magnitude for both the RGL and the IGL EOS cases. Furthermore, at atmospheric pressure, $f_v$ scatter plots show a denser band under the higher range of $f_v$, while a conflicting trend has been observed for the high pressure flame (Figs. 3.2 and 3.36). As seen in Fig. 3.36 the conditioned rms and means of $f_v$ are of
Figure 3.22: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<T/T_0|\phi>$ for Re= 2500, Le ≠ 1, and 10 atm ambient pressure at time of 80.

Figure 3.23: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<f v|\phi>$ for Re= 2500, Le ≠ 1, and 10 atm ambient pressure at time of 80.
Figure 3.24: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( <N|\phi> \) for \( Re=2500, Le \neq 1 \), and 10 atm ambient pressure at time of 80.

Figure 3.25: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( <Ys|\phi> \) for \( Re=2500, Le \neq 1 \), and 10 atm ambient pressure at time of 80.
Figure 3.26: On the left the RGL EOS model and on the right the IGL EOS model predictions of $\langle \chi | \phi \rangle$ for $Re = 2500$, $Le \neq 1$, and 10 atm ambient pressure at time of 80.

Figure 3.27: On the left the RGL EOS model and on the right the IGL EOS model predictions of $\langle w_{soot} | \phi \rangle$ for $Re = 2500$, $Le \neq 1$, and 10 atm ambient pressure at time of 80.
Figure 3.28: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< T/T_0 | \phi >$ for $Re = 2500$, $Le \neq 1$, and 10 atm ambient pressure at time of 80 radiation included.

Figure 3.29: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< f v | \phi >$ for $Re = 2500$, $Le \neq 1$, and 10 atm ambient pressure at time of 80 radiation included.
Figure 3.30: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< N|\phi >$ for Re= 2500, Le $\neq$ 1, and 10 atm ambient pressure at time of 80 radiation included.

Figure 3.31: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< Ys|\phi >$ for Re= 2500, Le $\neq$ 1, and 10 atm ambient pressure at time of 80 radiation included.
Figure 3.32: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< \chi | \phi >$ for Re = 2500, Le ≠ 1, and 10 atm ambient pressure at time of 80 radiation included.

Figure 3.33: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< w_{soot} | \phi >$ for Re = 2500, Le ≠ 1, and 10 atm ambient pressure at time of 80 radiation included.
<table>
<thead>
<tr>
<th>P(atm)</th>
<th>EOS</th>
<th>$\rho (kg/m^3)$</th>
<th>Z (compressibility)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>RGL</td>
<td>$\approx2.97$</td>
<td>0.98</td>
</tr>
<tr>
<td>5</td>
<td>RGL</td>
<td>$\approx16.1$</td>
<td>0.91</td>
</tr>
<tr>
<td>10</td>
<td>RGL</td>
<td>$\approx35.7$</td>
<td>0.82</td>
</tr>
<tr>
<td>35</td>
<td>RGL</td>
<td>$\approx255$</td>
<td>0.4</td>
</tr>
<tr>
<td>1</td>
<td>IGL</td>
<td>$\approx2.92$</td>
<td>1</td>
</tr>
<tr>
<td>5</td>
<td>IGL</td>
<td>$\approx14.6$</td>
<td>1</td>
</tr>
<tr>
<td>10</td>
<td>IGL</td>
<td>$\approx29.3$</td>
<td>1</td>
</tr>
<tr>
<td>35</td>
<td>IGL</td>
<td>$\approx102$</td>
<td>1</td>
</tr>
</tbody>
</table>

Table 3.7: The density and compressibility of $C_{12}H_{24}$ at 1, 5, 10 and 35 atm flames.

the same order of magnitude, while the peak $f_v$ has been shifted toward to the leaner flame regions. The higher quantities of conditioned mean $f_v$ of the RGL EOS have been calculated to be in a wider part of the mixture fraction (i.e. between $\phi \approx 0.55$ to 0.78), while in the IGL EOS case it has a narrower conditioned mean $f_v$ profile (i.e. $\chi^\phi \approx 0.6$ to 0.8). Furthermore, the IGL EOS model over-predicts $f_v$ 70% larger than the RGL EOS model. This could be related to the higher temperature prediction of the IGL EOS model (Figs. 3.34 and 3.35).

The temperature difference between two cases is $\approx 530$ K, which means there is a 25% variation between the two peak mean temperatures. The IGL EOS model has predicted $Y_s$ to be 50% larger than the RGL EOS model. Similar trends of scatter plots of $f_v$ has been obtained for $Y_s$ scatter plots of elevated pressures. The IGL EOS model predicts the peak of conditioned $\chi^\phi$ to be 3 times larger than the RGL EOS model (Fig. 3.41). This difference can explain the large difference for larger $f_v$ and $Y_s$ quantities of the IGL EOS model. At elevated pressure, for both cases temperature has less scattered profile in comparison with atmospheric flames even though $\chi^\phi$ has been increased by the 2 and 3 orders of magnitude for both the RGL and the IGL EOS models, respectively. This difference may be due to the extreme density differences in the flame (the RGL EOS model has 86 times denser fuel, and, IGL EOS model 35 times denser fuel than atmospheric pressure flames Table 3.7).
The larger temperature, fv and Ys predictions lead to over-prediction of N (i.e. the IGL EOS model calculates N 20 times larger than the RGL EOS model). The low level of mixing in the IGL EOS case may help us to explain the higher temperature profile. The low mixing level implies a longer residence time for both the gaseous species and soot particles, and gives sufficient time to increase the chemical reaction and the heat release. The large differences in temperature, Ys and fv suggest an over-prediction of \( w_{soot} \) for the IGL EOS model (Fig. 3.42). However, the suggested lower rms value of the \( w_{soot} \) by Lignell et al. [45] may be seen for the RGL EOS model predictions of the \( w_{soot} \), while it is still extremely close to the mean of the \( w_{soot} \) for the IGL EOS model. Furthermore, the \( w_{soot} \) has a narrower profile than the atmospheric pressure flame for both elevated pressure flames. Such a behavior explains the trends of fv and Ys observed in the previous figures.

Figure 3.34: On the left the RGL EOS model and on the right the IGL EOS model predictions of \( \langle T/T_0 \rangle|\phi \rangle \) for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65.

Figures 3.43 - 3.48 show the evaluation of the RGL EOS flame at 35 atm. At time of 80, the flame temperature exhibit a slight reduction while becoming more scattered through mixture fraction axis (Fig. 3.43) (i.e. more turbulent flow). Figure 3.44 shows a 25% increase in fv, while it has been carried to the fuel rich region by the flow. N, at time of 80, now has two more distinct peaks located at \( \phi \approx 0.25-\approx 0.45 \) (Fig. 3.45). As the
Figure 3.35: Contour plots of T(K) for pressure 35 atm, Le=1 and Re=2500, top is the RGL model and bottom is the IGL EOS model predictions time of 65, 3D data.
Figure 3.36: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< f v | \phi >$ for Re = 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65.

Figure 3.37: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< N | \phi >$ for Re = 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65.
Figure 3.38: Contour plots of $fv$ for pressure 35 atm, $Le=1$ and $Re=2500$, top is the RGL model and bottom is the IGL EOS model predictions time of 65, 3D data.
Figure 3.39: Contour plots of Ys for pressure 35 atm, Le=1 and Re=2500, top is the RGL model and bottom is the IGL EOS model predictions time of 65, 3D data.
Figure 3.40: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< Y_s | \phi >$ for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65.

Figure 3.41: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< \chi | \phi >$ for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65.
time evolves, $Y_s$ becomes more scattered in agreement with the more scattered temperature profile (Fig. 3.46). Figures 3.47 and 3.48 also support more turbulent flame and has a larger standard deviation for $X^\phi$ and $Y_s$ at the region where we observe more scattered temperature profile.

Lastly, in this chapter we have investigated pressure effects on the soot formation/destruction processes in adiabatic flames in Figs. 3.50 - 3.54, only for 2D data, at the non-dimensional time of 80. Increasing ambient pressure results in increasing flame
Figure 3.44: The comparison the RGL EOS model predictions of $<f|\phi>$ for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65 and 80.

Figure 3.45: The comparison the RGL EOS model predictions of $<N|\phi>$ for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65 and 80.
Figure 3.46: The comparison the RGL EOS model predictions of $<Y_s|\phi>$ for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65 and 80.

Figure 3.47: The comparison the RGL EOS model predictions of $<\chi|\phi>$ for Re= 2500, Le ≠ 1, and 35 atm ambient pressure at time of 65 and 80.
temperature for both the RGL and the IGL EOS model flames. For the first three flames, the IGL EOS model well predicts the flame temperature while it highly over predicts the flame temperature. It deviates by $\approx 25\%$ from the peak flame temperature as stated before which will cause the over-prediction of the soot properties. Figure 3.50 presents the log-normal of $fv$ for four different pressures of the RGL and the IGL EOS models. This figure has two important trends to be considered. Firstly, it clearly shows that increasing the ambient pressure of the flame has a positive effect on $fv$ in all the flames. Secondly, the IGL EOS model under-predicts the $fv$ in the first 3 flames of 1, 5 and 10 atm, while it over-predicts $fv$ of 35 atm flame. The difference of atmospheric and 35 atm $fv$ predictions for the IGL EOS models is an order of magnitude larger than the RGL EOS model predictions’s difference. Figure 3.51 shows the pressure and the RGL and the IGL EOS assumption effects on $Ys$ predictions. This figure exhibits similar behavior with $fv$ and $N$ predictions, and it clearly states that the IGL EOS model over-predicts $Ys$ by $\approx 40\%$ in the flame at 35 atm. The destruction of the soot particles produces species of CO and H (i.e. from Table 3.1 $C_s + 0.5O_2 \rightarrow CO$ and $C_s + OH = CO + H$). In Figs. 3.52 and 3.53, CO and H mass fractions are over-predicted for the elevated pressure (i.e. 35 atm) flame. While the peak
mass fraction of CO is over-predicted by $\approx 28\%$ and for species H it is $\approx 35\%$. Finally, Fig. 3.54 presents the mass fractions of OH radical in the flames. This figure has importance to explain the flame characteristics since OH mass fraction is considered to highly affect flame temperatures. As mentioned earlier, the temperature at 35 atm for the IGL EOS model is much higher than the RGL EOS model’s flame temperature. The OH mass fraction is $\approx 45\%$ under-predicted in stoichiometric region where the heat release is expected to be very high (i.e. indication of the high levels of H consumption). Furthermore, for 5 and 10 atm flames the IGL EOS model predicts the OH mass fractions profiles to be very similar to each other, which can explain why the Ys, fv and N predictions have similar quantities in the IGL EOS model. Even though the IGL EOS model is capable of catching the profile of the soot properties for low operating pressures and high pressure flames, it fails when the pressure reaches the critical point. The employment of a RGL EOS model is crucial to predict the soot properties and transportation as well as the flame characteristics inadiabatic Kerosene/Air flame simulations appropriately.

![Figure 3.49: The RGL and the IGL EOS pressure effects on mean variables Re=2500, left is RGL and right is IGL time of 80 2D data](image)

Figure 3.49: The RGL and the IGL EOS pressure effects on mean variables Re=2500, left is RGL and right is IGL time of 80 2D data
Figure 3.50: Pressure effects on means of $< f v | \phi >$ for Re=2500, Le≠1 and 1 atm on the left the RGL model predictions and on the right the IGL model predictions time of 80 2D data

Figure 3.51: Pressure effects on means of $< Y_s | \phi >$ for Re=2500, Le≠1 and 1 atm, on the left the RGL model predictions and on the right the IGL model predictions time of 80 2D data
Figure 3.52: The RGL and the IGL EOS pressure effects on mean variables Re=2500, left is RGL and right is IGL time of 80 2D data

Figure 3.53: The RGL and the IGL EOS pressure effects on mean variables Re=2500, left is RGL and right is IGL time of 80 2D data
Figure 3.54: Pressure effects on mean of $< Y_{OH} | \phi >$ for $Re=2500$, $Le\neq 1$ and 1 atm, on the left the RGL model predictions and on the right the IGL model predictions time of 80 2D data
Chapter 4

Differential Diffusion and Equation of State Effects on Soot Production/Oxidation Processes in Kerosene/Air Shear Flames

4.1 Introduction

Despite of intensive ongoing researches on developing new methods to predict soot formation, oxidation and transportation processes in turbulent flames, the literature suffers from lack of an adequate approach to this phenomenon. This usually emanates from assumptions of equal molecular diffusion of gaseous species (i.e. equal Schmidt numbers) and simply equating thermal diffusivity to mass diffusivity (i.e. unity Lewis number) [63]. The large and small scales of turbulence become detached with increasing Re number (i.e. turbulence intensity) reducing effects of differential diffusion which is associated with small scales (i.e. large Peclet number and heat release) [40, 63]. In turbulent flames with
The effects of differential diffusion in turbulent flames have been investigated in experiments by Refs. [5, 20, 54, 56, 81]. Bergmann et al. [5] have pointed out the significance of differential diffusion effects in turbulent $CH_4/H_2/N_2$ jet diffusion flames with $Re = 15,200$. Drake et al. [20] have shown that differential diffusion due to the preferential diffusion of $H_2$ becomes insignificant in $H_2/Air$ flames by increasing $Re$ number. Effects of differential diffusion on large scales have been observed by Smith et al. [81] in a $CO_2$ diluted $H_2$ flames at $Re = 30,000$. In a similar $CO_2$ diluted $H_2$ flame Masri et al. [54] have reported that differential diffusion becomes insignificant at $Re \approx 60,000$.

For different $H_2/N_2 - Air$ jet flames for the range $Re = 6,200$ to $8,800$ Meier et al. [56] have noted that differential diffusion is extremely important for temperature. The reader should note that based on aforementioned studies for the flames at low and intermediate $Re$ numbers differential diffusion should be taken into account for proper prediction of the flame.

Typically soot particles’ diffusion time scale is considerably larger than the gas species resulting in the soot particles to be governed by varied turbulent scales which causes soot particles to be transported throughout the flame, diverting the local flame characteristics [52]. It is known that lighter species (i.e. $H$ and $H_2$) are remarkably affected by differential diffusion [47, 52, 55, 70]. The diffusion of $H$ and $H_2$ reshape the soot particle bundles where the soot formation and soot surface growth occur in the presence of $H$ and soot precursors [52]. The differential diffusion has a role to draw $H$ toward to $C_2H_2$ and causes soot formation and growth processes to happen [52].

For the conditional moment closure (CMC) and Reynolds Average Navier-Stokes (RANS) models, the scalar dissipation rate has an important role to define the relation between scalar mixing and combustion modeling [52, 73]. It is also affected by Le number.
since it is the ratio of the thermal diffusivity to the mass diffusivity [52]. As stated in earlier paragraphs it is generally assumed that, turbulent mixing is the dominant power which governs species mass diffusivity, eliminating differential diffusion effects. This statement generally leads researchers to make the unity Le number assumption [52]. The transport of radicals, such as O, OH and H, from the flame kernel to the soot formation area is a very important phenomenon in the soot formation and growth processes [52].

The following section of this chapter presents an analysis of a generated DNS database of both atmospheric and high pressure Kerosene/Air reacting shear flames. The same reduced chemical mechanism including soot formation and oxidation processes has been coupled with the real and ideal gas laws and the unity Le number assumption to test the validity of these assumptions under realistic thermodynamic combustion regimes.

A semi-global soot production/oxidation of Kerosene/Air turbulent flames have been investigated for atmospheric and elevated pressures for the unity Le number assumption by employing a RGL and the IGL EOS model. The results of atmospheric pressure flames have been compared with the data obtained in the current work with non-unity Le number model of the RGL and the IGL EOS models. The results are presented in Section 4.3. We start by reporting the Le numbers of both EOS models associated with general diffusion of 1, 5, 10 and 35 atm flames. Later, we compare the unity Le number assumption of the RGL and the IGL EOS models’ results with the RGL and the IGL EOS results coupled with general diffusion at atmospheric pressure. Continued with comparing the unity Le number assumption results of the RGL and the IGL EOS models. Lastly, the effect of the widely used the unity Le number assumption and pressure effects on the soot production/oxidation processes are examined.
4.2 Objectives

Based on the above, the primary objective of this chapter is to examine the effects of the commonly used unity Le number assumption on the soot formation/oxidation processes in both atmospheric and elevated pressure flames for both a RGL and the IGL EOS models.

4.3 Results

Only 1 atm flames results of the RGL and the IGL EOS models of this chapter have been obtained with 3D DNS calculations; the rest of the results were obtained with 2D DNS calculations due to the computational limitations. Much deeper investigation is need to be done for future investigation of pressure and Le number effects associated with both a RGL and the IGL EOS for high pressure flames.

For the current work, ten distinct reacting simulations are considered for $Re_F = 2500$. The convective Mach number is set to be 0.35. Eight of the simulations have been conducted for 2D DNS (Table 4.1), and 2 of the 10 simulations have been conducted for 3D DNS (Table 4.2). The heat and mass flux equations with generalized diffusion (Eqs. (2.7) and (2.8) are reduced to the simplest forms to be used with the unity Le number assumption. While Eqs. (4.1) and (4.2) are the heat flux of Fourier’s law, and mass flux of Fickian’s law, respectively, Eq. (4.3) is the IGL EOS:

\begin{align*}
Q_j &= -\kappa \frac{\partial T}{\partial x_j}, \quad (4.1) \\
J_{j,k}' &= -D \frac{\partial X_a}{\partial x_j}, \quad (4.2) \\
P &= \rho R_m T, \quad (4.3)
\end{align*}

where $\kappa$ is the thermal conductivity, $D$ is the diffusion coefficient, $X_a$ is the mol fraction of
the species $\alpha$, $R_m$ is the gas constant of the mixture. For all cases the initial temperature is $T_0 = 700K$, and the ambient pressure varies between 1 and 35 atm (i.e. 1, 5, 10, and 35 atm).

<table>
<thead>
<tr>
<th>Run</th>
<th>Oxidizer</th>
<th>$P_0$(atm)</th>
<th>EOS</th>
<th>Le</th>
<th>$T_0(K)$</th>
<th>$Re_0$</th>
<th>$N_1 \times N_2$</th>
<th>$t_f^*$</th>
<th>Radiation</th>
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</thead>
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<td>400$\times$608</td>
<td>80</td>
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<td>IG</td>
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<td>2500</td>
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<td>No</td>
</tr>
<tr>
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<td>5</td>
<td>RG</td>
<td>1</td>
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<td>2500</td>
<td>304$\times$544</td>
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<td>1</td>
<td>700</td>
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<td>304$\times$544</td>
<td>80</td>
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</tr>
<tr>
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<td>Air</td>
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<td>RG</td>
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<td>700</td>
<td>2500</td>
<td>280$\times$480</td>
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<td>No</td>
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<td>10</td>
<td>IG</td>
<td>1</td>
<td>700</td>
<td>2500</td>
<td>280$\times$480</td>
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</tr>
<tr>
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<td>RG</td>
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</tr>
<tr>
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<td>Air</td>
<td>35</td>
<td>IG</td>
<td>1</td>
<td>700</td>
<td>2500</td>
<td>240$\times$640</td>
<td>80</td>
<td>No</td>
</tr>
</tbody>
</table>

Table 4.1: Summary of simulation parameters from all cases considered. $P_0$ is initial pressure, $T_0$ is the initial temperature, $Re_0$ the initial Reynolds number, $N_1$, $N_2$, are the number of grid points in the $x_1$, $x_2$ direction, respectively, and $t_f^*$ is non-dimensionalized final simulation time.

<table>
<thead>
<tr>
<th>Run</th>
<th>Oxidizer</th>
<th>$P_0$(atm)</th>
<th>EOS</th>
<th>Le</th>
<th>$T_0(K)$</th>
<th>$Re_0$</th>
<th>$N_1 \times N_2 \times N_3$</th>
<th>$t_f^*$</th>
<th>Radiation</th>
</tr>
</thead>
<tbody>
<tr>
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<td>IG</td>
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<td>700</td>
<td>2500</td>
<td>400$\times$672$\times$280</td>
<td>80</td>
<td>No</td>
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<td>IG</td>
<td>1</td>
<td>700</td>
<td>2500</td>
<td>400$\times$640$\times$300</td>
<td>80</td>
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</tbody>
</table>

Table 4.2: Summary of simulation parameters from all cases considered. $P_0$ is initial pressure, $T_0$ is the initial temperature, $Re_0$ the initial Reynolds number, $N_1$, $N_2$ and $N_3$, are the number of grid points in the $x_1$, $x_2$, and $x_3$, direction respectively, and $t_f^*$ is non-dimensionalized final simulation time.

We focus on the Le numbers of both EOS models adopted to create adiabatic flame DNS data. The list of the species pair numbers and species pair are listed in the Table 4.3. Figures 4.1 - 4.4 investigate the Le numbers calculated conditioned on the mixture fraction, $<Le|\phi>$. The red horizontal lines in each figure aim to indicate the $<Le|\phi>$ numbers for each species pair how different from unity Le number for each simulation considered in the current work. For all cases the species pairs 1, 10, 11, 12, 13, 14 and 17 possess higher
values than Le=1, while the rest stay below the red line for all the considered cases. Figure 4.1 shows the $<Le|\phi>$ numbers for the atmospheric pressure flames. As stated before, for the atmospheric pressure flames employing the IGL EOS might be safe with a non-unity Le number model. Figure 4.2 displays the $<Le|\phi>$ numbers for 5 atm flames for each EOS model employed in this work. The $<Le|\phi>$ numbers are to be indifferent from each other for each case. With increasing ambient pressure, an increasing trend for $<Le|\phi>$ is present in the 10 atm flames as well, while the $<Le|\phi>$ numbers of the RGL and the IGL EOS models still exhibit indifferent behavior from each other. Lastly, for 35 atm flames, the $<Le|\phi>$ numbers for the previously mentioned species pairs get $\approx 50\%$ larger than the atmospheric pressure flames, with the $<Le|\phi>$ numbers smaller than unity for the rest of the species pairs.

<table>
<thead>
<tr>
<th>Species Pair number</th>
<th>Species Pair</th>
<th>Species Pair number</th>
<th>Species Pair</th>
<th>Species Pair number</th>
<th>Species Pair</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$O_2$ − $C_2H_24$</td>
<td>10</td>
<td>$C_2H_24$ − OH</td>
<td>19</td>
<td>CO − CO$_2$</td>
</tr>
<tr>
<td>2</td>
<td>$O_2$ − CO</td>
<td>11</td>
<td>$C_2H_24$ − O</td>
<td>20</td>
<td>CO − H</td>
</tr>
<tr>
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<td>$O_2$ − OH</td>
<td>12</td>
<td>$C_2H_24$ − $H_2O$</td>
<td>21</td>
<td>CO − $N_2$</td>
</tr>
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<td>$O_2$ − O</td>
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<td>$C_2H_24$ − CO$_2$</td>
<td>22</td>
<td>$O_2$ − $H_2O$</td>
</tr>
<tr>
<td>5</td>
<td>$O_2$ − $H_2O$</td>
<td>14</td>
<td>$C_2H_24$ − $H$</td>
<td>23</td>
<td>$OH$ − $H_2O$</td>
</tr>
<tr>
<td>6</td>
<td>$O_2$ − CO$_2$</td>
<td>15</td>
<td>$C_2H_24$ − $N_2$</td>
<td>24</td>
<td>$OH$ − CO$_2$</td>
</tr>
<tr>
<td>7</td>
<td>$O_2$ − $H$</td>
<td>16</td>
<td>CO − OH</td>
<td>25</td>
<td>$OH$ − $H$</td>
</tr>
<tr>
<td>8</td>
<td>$O_2$ − $N_2$</td>
<td>17</td>
<td>CO − $O$</td>
<td>26</td>
<td>$OH$ − $N_2$</td>
</tr>
<tr>
<td>9</td>
<td>$C_2H_24$ − CO</td>
<td>18</td>
<td>CO − $H_2O$</td>
<td>27</td>
<td>$O$ − $H_2O$</td>
</tr>
<tr>
<td>28</td>
<td>$O_2$ − $CO_2$</td>
<td>22</td>
<td>$OH$ − $H_2O$</td>
<td>23</td>
<td>$CO_2$ − $H$</td>
</tr>
<tr>
<td>29</td>
<td>$O_2$ − $H_2O$</td>
<td>24</td>
<td>$OH$ − $N_2$</td>
<td>35</td>
<td>$CO_2$ − $N_2$</td>
</tr>
<tr>
<td>30</td>
<td>$O_2$ − CO$_2$</td>
<td>25</td>
<td>$OH$ − $N_2$</td>
<td>36</td>
<td>$H$ − $N_2$</td>
</tr>
</tbody>
</table>

Table 4.3: The list of Species Pair number and Species Pair.

We continue our analyses with comparing the non-unity Le number results of adiabatic 1 atm flames that have been analyzed in the previous chapter with the unity Le number assumption predictions of the RGL and the IGL EOS model flames. For both non-unity and unity Le number comparisons are made for the time of 65. Figures 4.5 - 4.13 compare the predictions of the RGL EOS model coupled with non-unity and unity Le number models. Figure 4.5 exposes how the temperature distribution is affected by the unity Le number assumption. The peak flame temperature is reduced by 25% by switching from the non-unity Le number to the unity Le model. Near the stoichiometric region of $\phi \approx 0.1$.
Figure 4.1: On the left the RGL EOS model and on the right the IGL EOS model Le number calculated for the adiabatic atmospheric pressure flame.

Figure 4.2: On the left the RGL EOS model and on the right the IGL EOS model Le number calculated for the adiabatic 5 atm pressure flame.
Figure 4.3: On the left the RGL EOS model and on the right the IGL EOS model Le number calculated for the adiabatic 10 atm pressure flame.

Figure 4.4: On the left the RGL EOS model and on the right the IGL EOS model Le number calculated for the adiabatic 35 atm pressure flame.
to $\phi \approx 0.3$ the unity Le number case has a substantially lower flame temperature. The top sub-figure of Fig. 4.6 presents the contour plot of the RGL EOS model with generalized diffusion and bottom one has been created with the same EOS model coupled with the unity Le number. It is clearly seen that a continuous hot flame packet exists in the top contour plot confirming the left sub-figure of Fig. 4.5. Another point is the how the flame structure has been modified by the unity Le number assumption. Now the flame possesses an intensified turbulent profile signifying that higher levels of mixing exists in this flame.

Figure 4.5: On the left the RGL EOS model $\text{Le} \neq 1$ and on the right the RGL EOS model $\text{Le}=1$ predictions of $\langle \frac{T}{T_0} | \phi \rangle$ for $\text{Re}=2500$, and 1 atm ambient pressure at time of 65.

As we move on investigating the soot properties expected significant reductions are exhibited. Figure 4.7 confirms the reduction in the $fv$ predictions (i.e. $fv$ is under-predicted by $\approx 90\%$ with the unity Le number assumption). Even though the local flow effects on $fv$ are still present, $fv$ presents more scattered behavior in contrast to the non-unity Le number predictions. Figure 4.8 reveals that an order of magnitude lower $fv$ predictions of the unity Le number assumption. Now, $fv$ is present in regions where it is not observed in the non-unity Le number predictions. Another soot property, $N$, is dominated by temperature while it is highly under-predicted by $\approx 54\%$ which confirms the effect of the under-predicted flame temperature on soot load. The under-prediction of the flame temperature leads to
Figure 4.6: The comparison of contour plots of T(K) top is Le≠1 and bottom is Le=1 both coupled with the RGL EOS model for pressure 1 atm and Re=2500 at time of 65 predictions of 3D data.
under-predictions of $Y_s$ by $\approx 90\%$ as well (Fig. 4.10). Comparison of Figs. 4.6, 4.8 and 4.11 shows how equating the thermal and mass diffusivity affects the soot load distribution in the flame kernel. The scalar dissipation rate is over-predicted by $\approx 400\%$ implying high levels of mixing which helps to explain why flame temperature, $f_v$, $N$ and $Y_s$ exhibit more scattered behavior than non-unity Le number predictions. Figure 4.12 displays how the scalar dissipation differs by adoption of the unity Le number assumption. Lastly, we focus on the soot generation rate which is under-predicted by $56\%$ (Fig. 4.13). This is one of the direct results of the under-predictions of the flame temperature and residence time (i.e. over prediction of scalar dissipation rate of the unity Le number) of the unity Le number model.

![Figure 4.7](image)

Figure 4.7: On the left the RGL EOS model $Le\neq 1$ and on the right the RGL EOS model $Le=1$ predictions of $<f_v|\phi>$ for $Re=2500$, and 1 atm ambient pressure at time of 65.

Figures 4.14 - 4.19 examine the effect of the unity Le number assumption effects coupled with the IGL EOS model. As it has been stated in the previous chapter, the IGL EOS model well predicts the flame and soot properties associated with generalized diffusion which suggests that the IGL EOS model coupled with the unity Le number assumption predictions might possess similar behavior observed in Figs. 4.5 - 4.13. While the flame temperature and soot property predictions are similar to those obtained with the RGL EOS and the unity Le number model, the scalar dissipation rate predictions displays a
Figure 4.8: The comparison of contour plots of $f_v$ for pressure 1 atm and Re=2500, top is $Le=1$ and bottom is $Le \neq 1$ predictions time of 65 3D data.
Figure 4.9: On the left the RGL EOS model $Le\neq 1$ and on the right the RGL EOS model $Le=1$ predictions of $<N|\phi>$ for $Re=2500$, and 1 atm ambient pressure at time of 65.

Figure 4.10: On the left the RGL EOS model $Le\neq 1$ and on the right the RGL EOS model $Le=1$ predictions of $<Y_s|\phi>$ for $Re=2500$, and 1 atm ambient pressure at time of 65.
Figure 4.11: The comparison of contour plots of Ys for pressure 1 atm and Re=2500, top is \( \text{Le}=1 \) and bottom is \( 1 \neq 1 \) predictions time of 65 3D data.
Figure 4.12: On the left the RGL EOS model $Le \neq 1$ and on the right the RGL EOS model $Le=1$ predictions of $< \chi | \phi >$ for $Re=2500$, and 1 atm ambient pressure at time of 65.

Figure 4.13: On the left the RGL EOS model $Le \neq 1$ and on the right the RGL EOS model $Le=1$ predictions of $< w_{soot} | \phi >$ for $Re=2500$, and 1 atm ambient pressure at time of 65.
significantly abnormal profile [by having a large standard deviation in the fuel rich region Fig.(4.18)].

Figure 4.14: On the left the IGL EOS model $L_e \neq 1$ and on the right the IGL EOS model $L_e = 1$ predictions of $< \frac{T}{T_0}\phi >$ for $Re=2500$, and 1 atm ambient pressure at time of 65.

Figure 4.15: On the left the IGL EOS model $L_e \neq 1$ and on the right the IGL EOS model $L_e = 1$ predictions of $< f_v|\phi >$ for $Re=2500$, and 1 atm ambient pressure at time of 65.

Figures 4.20 through 4.28 present a comparison of the unity $L_e$ number assumption coupled with the RGL and the IGL EOS model predictions of 3D atmospheric flames at time of 80. As the simulation time evolves from 65 to 80, the high flame temperature packets are transported from the stoichiometric region to the fuel rich region while insignificant decreases existed in both the RGL and the IGL EOS models as observed in Figs. 4.20
Figure 4.16: On the left the IGL EOS model Le≠1 and on the right the IGL EOS model Le=1 predictions of $<N|\phi>$ for Re=2500, and 1 atm ambient pressure at time of 65.

Figure 4.17: On the left the IGL EOS model Le≠1 and on the right the IGL EOS model Le=1 predictions of $<Y_s|\phi>$ for Re=2500, and 1 atm ambient pressure at time of 65.
Figure 4.18: On the left the IGL EOS model $L_e \neq 1$ and on the right the IGL EOS model $L_e = 1$ predictions of $< \chi | \phi >$ for $Re = 2500$, and 1 atm ambient pressure at time of 65.

Figure 4.19: On the left the IGL EOS model $L_e \neq 1$ and on the right the IGL EOS model $L_e = 1$ predictions of $< w_{soot} | \phi >$ for $Re = 2500$, and 1 atm ambient pressure at time of 65.
and 4.21. The slight decrease in the flame temperature in both flames lead to negligible reductions in $f_v$, $N$ and $Y_s$ (Figs. 4.22, 4.23, 4.24, 4.25, 4.26 and 4.28). The abnormal trend of the standard deviation of the scalar dissipation rate of the IGL EOS unity Le number model is increased by 68% at the final time of the simulation (Fig. 4.27).

Figure 4.20: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<T/T_0|\phi>$ for Re=2500, Le =1, and 1 atm ambient pressure at time of 80, 3D DNS data.

The examination of pressure effects on the flame structure and soot load with the unity Le assumption is also another focus of the current work. Figures 4.29 - 4.34 investigate how the flame temperature, soot properties (i.e., $f_v$ and $Y_s$) and species of CO, H and OH are influenced by increasing ambient pressure. Figure 4.29 displays the mean temperature profile of the RGL and the IGL EOS models coupled with the unity Le number assumption. For 1, 5 and 10 atm pressure flames, neither the RGL nor the IGL EOS models, has little impact on the mean temperature profiles; confirming a previously observed trend with the non-unity Le number predictions. However, by reaching the critical ambient pressure, the IGL EOS model over predicts the flame temperature. Predictions of $f_v$ are not differentiated by the EOS model adoption (Fig. 4.30). In contrast to $f_v$ predictions, the soot mass fraction predictions address the importance of the EOS model adoption (Fig. 4.31). While the $Y_s$ predictions of atmospheric and 5 atm flames for both models look nearly identical, the
Figure 4.21: The comparison of contour plots of T(K) for pressure 1 atm, Le=1 and Re=2500, top sub-figure is created the RGL EOS, and bottom one is created with the IGL EOS model at time of 80 for 3D data.
dissimilarity in $Y_s$ profiles starts with 10 atm flames. The RGL EOS model predicts $Y_s$ profile to ‘mimic’ $Y_s$ prediction of 5 atm while having slightly larger peak $Y_s$ values than the 5 atm prediction. The IGL EOS model predicts $Y_s$ to have a similar profile with the atmospheric pressure flame $Y_s$ prediction. The elevated pressure flame $Y_s$ predictions differs in two different manner. First, the peak value of $Y_s$ predictions of the IGL EOS model is larger than the RGL model prediction by 16%, and second $Y_s$ exhibits a more widened profile than the RGL EOS $Y_s$ profile does.

Figures 4.32, 4.33 and 4.34 have significance to demonstrate the soot oxidation process in the flame; since during direct oxidation CO and H is produced while OH is involved in the soot oxidation. The soot oxidation rate is seen to be almost identical for the atmospheric pressure flames for both EOS models, since $Y_{CO}$, $Y_H$ and $Y_{OH}$ have similar profiles in both flames. When pressure is increased to 5 atm, the oxidation of soot by $Y_{O2}$ is not affected by the EOS model adoption. However, $Y_{OH}$ shows a slight effect of the EOS model adoption on this soot oxidation step, since $Y_{OH}$ is slightly larger for the IGL EOS than the RGL EOS model prediction. When the pressure reaches 10 atm, $Y_s$, $Y_{CO}$, $Y_H$ and $Y_{OH}$ profiles support the idea that the soot oxidation rate is larger in the region $\phi \approx 0.4$ to 0.75.
Figure 4.23: The comparison of contour plots of $f_\nu$ for pressure 1 atm, $Le=1$ and $Re=2500$, top sub-figure. is created the RGL EOS, and bottom one is created with the IGL EOS model at time of 80 for 3D data.
Figure 4.24: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< N|\phi >$ for Re=2500, Le =1, and 1 atm ambient pressure at time of 80, 3D DNS data.

Figure 4.25: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< Y_s|\phi >$ for Re=2500, Le =1, and 1 atm ambient pressure at time of 80, 3D DNS data.
Figure 4.26: The comparison of contour plots of $Y_s$ for pressure 1 atm, $Le=1$ and $Re=2500$, top sub-figure. is created the RGL EOS, and bottom one is created with the IGL EOS model at time of 80 for 3D data.
Figure 4.27: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< \chi | \phi >$ for Re=2500, Le =1, and 1 atm ambient pressure at time of 80, 3D DNS data.

Figure 4.28: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< w_{soot} | \phi >$ for Re=2500, Le =1, and 1 atm ambient pressure at time of 80, 3D DNS data.
Figure 4.29: On the left the RGL EOS model and on the right the IGL EOS model predictions of \(< T/T_0|\phi >\) for Re=2500, Le =1, and 1 atm ambient pressure at time of 65, 2D DNS data.

Figure 4.30: On the left the RGL EOS model and on the right the IGL EOS model predictions of \(\log_{10} < f_v|\phi >\) for Re=2500, Le =1, and 1 atm ambient pressure at time of 80, 2D DNS data.
Figure 4.31: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<Y_s|\phi>$ for $Re=2500$, $Le=1$, and 1 atm ambient pressure at time of 80, 2D DNS data.

Figure 4.32: On the left the RGL EOS model and on the right the IGL EOS model predictions of $<Y_{CO}|\phi>$ for $Re=2500$, $Le=1$, and 1 atm ambient pressure at time of 80, 2D DNS data.
Figure 4.33: On the left the RGL EOS model and on the right the IGL EOS model predictions of $Y_H|\phi >$ for Re = 2500, Le =1, and 1 atm ambient pressure at time of 80, 2D DNS data.

Figure 4.34: On the left the RGL EOS model and on the right the IGL EOS model predictions of $< Y_{OH}|\phi >$ for Re = 2500, Le =1, and 1 atm ambient pressure at time of 80, 2D DNS data.
of the RGL EOS model predictions. In contrast to 10 atm flames, in 35 atm flames the soot oxidation rates become larger in the IGL EOS model predictions in the region $\phi \approx 0.25$ to 0.9.

So far we have compared the predictions of two different EOS models and two different diffusion models for four distinct ambient pressures separately. Since the current work is the first attempt to examine the effects of the EOS and diffusion models, now, we will examine the EOS, diffusion and pressure effects in the following from Figs. 4.35 through 4.40 for 2D data at time of 80. Figure 4.35 displays the mean temperature profiles for different ambient pressures while employing the RGL and the IGL EOS models coupled with the non-unity and unity Le number models. Figure 4.35 (a) examines atmospheric pressure flames by showing that employing either of the EOS models will have insignificant effects on the mean temperature; however, switching from the non-unity Le number to unity Le number will cause an $\approx 15\%$ temperature drop (i.e. soot load will be under-predicted). Figure 4.35 (b) displays the calculated mean temperature profiles for 5 atm flames. In the 5 atm flames we observe a slight temperature drop when the unity Le number assumption is employed (i.e. $\approx 10\%$ temperature drop has been observed between the non-unity Le number and the unity Le number cases coupled with the RGL EOS models). Even though both EOS models coupled with the unity Le number assumption slightly under-predicts the temperature profiles, it is capable of predicting the temperature behavior of the non-unity Le number cases for both EOS models for 5 and 10 atm flames [Figs. 4.35 (a) and (b)]. Lastly, we will check the mean temperature profile predictions for 35 atm flames calculated with two different EOS models coupled with either generalized diffusion or unity Le number model [Fig. 4.35 (d)]. Figure 4.35 (d) shows that both EOS and diffusion model adoptions have significant influence on the flame predictions. Alteration of the EOS model from the RGL to the IGL coupled with non-unity Le number causes an over-prediction of the mean flame temperature by $\approx 21\%$, while switching from the non-unity Le number to unity Le
number leads to under-predicting the mean peak flame temperature by \( \approx 18\% \) for the RGL and \( \approx 10\% \) for the IGL model. Overall the unity Le number assumption again fairly well captures the trends of the mean temperature profile of 35 atm flames.

Figure 4.35: The comparison of \( < T/T_0 | \phi > \) for different ambient pressures, the EOS models and diffusion models flames (a) 1 atm, (b) 5 atm, (c) 10 atm and (d) 35 atm ambient pressures.

Figure 4.36 (a) examines the impacts of the EOS and diffusion models made in the atmospheric pressure flames. The unity Le assumption over predicts \( \phi \) from \( \phi = 0 \) to \( \approx 0.7 \) in the mixture fraction region; while correct trends are caught by the unity Le number for both EOS models. Figure 4.36 (b) displays the \( \phi \) predictions of 5 atm flames,
and fv predictions are not differentiated from each other for both EOS and non-unity Le number case and unity Le. 10 atm flames predictions of the IGL EOS coupled with the unity Le number assumption are predicted to be one order of magnitude larger than the other three predictions in the region of $\phi \approx 0.45$ to 0.75 in Fig. 4.36 (c). When the unity Le number assumption is employed with the IGL EOS model, it fails to predict the fv profile except the regions of $\phi \approx 0.22$ to 0.45 and $\phi \approx 0.75$ to 1. Figure 4.36 (d) shows that either changing from the RGL to the IGL EOS or non-unity to unity Le number lead to mis-predictions of fv profiles in the region of $\phi \approx 0$ to 0.22 (i.e. fuel lean region).

Figure 4.37 (a) investigates $Y_s$ in the atmospheric pressure flames. As aforementioned, switching between the EOS models has insignificant impact on $Y_s$, however coupling the EOS models with the unity Le number assumption causes under-prediction of $Y_s$, while the trend of the $Y_s$ profiles are fairly well caught. Figure 4.37 (b) presents 5 atm flame prediction profiles of $Y_s$. By increasing pressure, soot load starts to be transported to the fuel rich side of the flame, while it is reduced by $\approx 35\%$ for the non-unity Le number predictions, whereas it is $\approx 12\%$ for the unity Le number predictions. In Fig. 4.37 (c), except the $Y_s$ prediction of the IGL EOS model coupled with the unity Le number assumption, soot load and the profiles follow a similar trend to each other. In the $\phi \approx 0.4$ to 0.75 $Y_s$ is over-predicted and from $\phi \approx 0.75$ to 1 under-predicted by the IGL EOS model employing the unity Le number assumption. From Figs. 4.35 4.36 and 4.37 support each other implying that the IGL EOS coupled with the unity Le assumption will fail to predict the flame and soot properties appropriately. Figure 4.37 (d) displays predictions of $Y_s$ profiles for the 35 atm flames. As it has been stated in the above, switching from the RGL to the IGL EOS results in over-predictions of flame temperature and mis-predictions of the soot properties. Coupling the unity Le number assumption with the IGL EOS leads to a similar observation for $Y_s$ throughout the entire mixture fraction region.

Figures 4.38 (a) and (b) show that the 5 atm predictions of $Y_H$ are very well predicted
Figure 4.36: The comparison of $<fv|\phi>$ for different ambient pressures, the EOS models and diffusion models flames (a) 1 atm, (b) 5 atm, (c) 10 atm and (d) 35 atm ambient pressures.
Figure 4.37: The comparison of $<Y_s|\phi>$ for different ambient pressures, the EOS models and diffusion models flames (a) 1 atm, (b) 5 atm, (c) 10 atm and (d) 35 atm ambient pressures.
for all the models considered in this work, while switching between non-unity and the unity Le number models causes significant failures in predictions of $Y_H$ in atmospheric pressure flames. Figure 4.38 (c) verifies the incapability of the IGL EOS model coupled with the unity Le number model to predict the flame characteristics by under-predicting $Y_H$ which is produced in the soot oxidation process. The under-prediction of $Y_H$ explains why we observe larger soot load in the 10 atm flame predictions of the IGL EOS model employing the unity Le number assumption. Figure 4.37 (d) shows the soot oxidation process in 35 atm flames. This sub-figure implies that the RGL and the IGL EOS models coupled with the non-unity Le number model predictions of $Y_{CO}$ should follow a similar trend with $Y_H$ since the heterogeneous and the homogeneous soot oxidation steps include both species to be produced. This sub-figure also reveals the fact that even though soot load is over-predicted by the IGL EOS coupling with the unity Le number, the soot oxidation process is under-predicted. In addition to that, although the RGL EOS coupling with the unity Le number assumption dramatically under-predicts the soot oxidation process.

Figure 4.39 displays $Y_{OH}$ predictions for the ambient pressures of 1, 5, 10 and 35 atm flames. While in Figs. 4.39 (b) and (c) $Y_H$ predictions are captured acceptably well for 5 and 10 atm flames, Figs. 4.39 (a) and (d) reveal the incapabilities of the IGL EOS model and the unity Le number assumption on the predictions of 1 and 35 atm flames.

Figure 4.40 shows the $Y_{CO}$ predictions of the RGL and the IGL EOS model employing either non-unity or the unity Le number models. Figure 4.40 (a) confirms the trends that have been observed previously on soot oxidation steps in atmospheric pressure flames. Again, $Y_{CO}$ predictions possess similar trends in all flames proving that soot properties are highly temperature dependent [see Fig. 4.35 (b)]. The predictions of $Y_{CO}$ in 10 atm flames help to better understand the soot load predictions of the IGL EOS model coupled with the unity Le number assumption confirming that the soot oxidation process is highly weakened by the choice of the EOS and the unity Le number assumption. Lastly, Fig. 4.40
Figure 4.38: The comparison of $< Y_H | \phi >$ for different ambient pressures, the EOS models and diffusion models flames (a) 1 atm, (b) 5 atm, (c) 10 atm and (d) 35 atm ambient pressures.
Figure 4.39: The comparison of $<Y_{OH}|\phi>$ for different ambient pressures, the EOS models and diffusion models flames (a) 1 atm, (b) 5 atm, (c) 10 atm and (d) 35 atm ambient pressures.
(d) displays the under-predictions of the $Y_{CO}$ supporting the idea of that the soot oxidation process weakens by increasing ambient pressure when the EOS model is coupled with the unity Le number assumption.

Figure 4.40: The comparison of $<Y_{CO}|\phi>$ for different ambient pressures, the EOS models and diffusion models flames (a) 1 atm, (b) 5 atm, (c) 10 atm and (d) 35 atm ambient pressures.
Chapter 5

Conclusions

DNS data for high pressure $H_2/O_2$ and $H_2/Air$ flames using the compressible flow formulation, detailed kinetics, a real fluid equation of state, and generalized diffusion are analyzed. For the $H_2/Air$ flame both the resolved heat flux vector and its gradient are as large as the filtered heat flux implying in this flame the subgrid heat flux vector may have an important effect. Similar behavior is noted in large reacting rate regions as well. While for low Re number reacting flow both the gradient of the heat flux vector and the subgrid heat flux vector itself are found to be important, these two terms become less important for higher Re number reacting flows in comparison with other unclosed terms in the energy equation for both mixing and reacting flows.

Soot production/oxidation investigations reveal the importance of real gas, generalized diffusion and pressure effects on a sooting hydrocarbon flames, ($Kerosene/Air$). The atmospheric pressure flame simulations of the RGL and the IGL models have good agreement with the limited literature. The results show that the IGL associated with generalized diffusion at atmospheric pressure flames may be a reasonable assumption, while it highly over-predicts flame temperature and soot quantities at 35 atm. Therefore, for high pressure flames the necessity of a real gas equation of state with generalized diffusion assumption
is required. The radiative heat flux is noted to be insignificant in the sooting flames due to small soot load in comparison to the domain size and the short physical time. In order to observe the effects of the radiative heat loss on intermediate pressure flames (i.e. 5 and 10 atm) an artificially increased Planck Mean Absorption coefficient has been employed. By the addition of the radiative heat loss slight differences have been reduced to be highly insignificant between the RGL and the IGL EOS models. Next, the unity Lewis number assumption associated with the RGL and the IGL models under-predicts soot quantities in 3D adiabatic atmospheric and 2D adiabatic intermediate and elevated pressure flames. The adiabatic 3D atmospheric pressure flames associated with the unity Le number assumption for the EOS models have revealed critical failures in flame structure predictions by possessing more turbulence and lower flame temperatures. Also, the pressure effect on the soot formation/oxidation processes is clearly observed. With increasing pressure, soot quantities and flame temperature are enhanced.
Bibliography


