The first goal of this thesis is to validate a detailed co-flow flame soot formation model for high pressure applications. The second goal is to use this detailed model to understand the effect pressure has on soot formation. The third goal is to note any deficiencies in the model, and the fourth is to remedy these issues. The thesis is divided into two research studies. The first study validates the model for high pressure use against ethane-air co-flow diffusion flames from 2 to 15 atm. After validation, the results are used to determine the impact pressure has on the three main soot formation processes. It is determined that the original model could not account for the flame pre-heating effect. The second study addresses this issue by adapting the model to extend below the fuel tube exit plane, and includes conjugate heat transfer (CHT) between the fluid streams and solid fuel tube.
Acknowledgements

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Nomenclature

$A_s$  Soot surface area per unit volume of gaseous mixture

$A_v$  Avogadro’s Number

$C_{mass}$  The mass of a carbon atom

$C_{p,k}$  The $k$th species specific heat under constant pressure

$C_{p,s}$  The specific heat of soot

$C_p$  The specific heat of the mixture under constant pressure

$D_i^a$  The diffusion coefficient of the $i$th sectional aggregates

$K K$  The total gaseous species number

$N_a$  Aggregate number density

$N_i^a$  Number of $i$th sectional soot aggregates per unit mass of the gaseous mixture

$N_p$  Primary particle number density

$N_i^p$  Number of $i$th sectional primary soot particles per unit mass of the gaseous mixture

$Q_r$  The radiative heat transfer rate from soot and gaseous species CO, CO$_2$, and H$_2$O

$T$  Temperature

$V_{T_k}$  The $k$th species thermal diffusion velocity

$V_{T_s}$  Soot thermophoretic velocity

$V_k$  The $k$th species diffusion velocity

$W_k$  The $k$th species molecular weight
$W_s$ The molecular weight of soot

$Y_k$ The $k$th species mass fraction

$f_v$ Soot volume fraction

$g_z$ Axial gravitational constant

$h_k$ The $k$th species specific enthalpy

$h_s$ The specific enthalpy of soot

$p$ Pressure

$r$ Radial coordinate

$t$ Time

$u$ Axial velocity

$v$ Radial velocity

$z$ Axial coordinate

**Greek Symbols**

$\alpha$ The portion of active soot particle surface sites available for chemical reaction

$\dot{\omega}_k$ The $k$th species molar production rate per unit volume

$\dot{\omega}_s$ The molar production rate of soot

$\gamma$ Pyrene-soot surface condensation efficiency

$\mu$ Dynamic viscosity

$\rho$ Mixture density

**Subscripts**

$r$ The radial direction

$z$ The axial direction

**Acronyms**

CFD Computational Fluid Dynamics
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<tr>
<td>CHT</td>
<td>Conjugate Heat Transfer</td>
</tr>
<tr>
<td>CPU</td>
<td>Central Processing Unit</td>
</tr>
<tr>
<td>DOM</td>
<td>Discrete Ordinate Method (radiation model)</td>
</tr>
<tr>
<td>HACA</td>
<td>Hydrogen-Abstraction-Carbon-Addition</td>
</tr>
<tr>
<td>IARC</td>
<td>International Agency for Research on Cancer</td>
</tr>
<tr>
<td>MPI</td>
<td>Message Passing Interface</td>
</tr>
<tr>
<td>PAH</td>
<td>Polycyclic Aromatic Hydrocarbon</td>
</tr>
<tr>
<td>RTE</td>
<td>Radiative Transfer Equation</td>
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<tr>
<td>SIMPLE</td>
<td>Semi-Implicit Method for Pressure Linked Equations</td>
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<tr>
<td>TDMA</td>
<td>Tri-Diagonal Matrix Algorithm</td>
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Chapter 1

Introduction

1.1 Motivation

The use of controlled fires has played a major role in the day to day activities of human civilization. Combustion processes can be used for cooking food, light generation, home heating, tool manufacturing, power generation, and material synthesis. Fossil fuel combustion is the main source of today’s energy supplies (more than 90%) and it is likely to remain this way for many years to come [1].

Emissions from combustion processes have always been a topic of concern for combustion researchers. One such emission, soot, has garnered specific attention in recent years. This is due to atmospheric soot, often referred to as black carbon, or particulate, being known to have detrimental effects on both human health and the global environment. Soot particles, which are typically 10-300 nm in diameter, can be absorbed deeply into the lungs, causing respiratory diseases such as asthma and bronchitis. The particles can be small enough to pass into the bloodstream leading to possible heart disease [2]. Polycyclic Aromatic Hydrocarbons (PAHs), which are pre-cursors to soot formation, have been classified as known carcinogens by the International Agency for Research on Cancer (IARC) as well. Additionally, soot is an environmental concern as it is the second most important factor in global warming behind carbon dioxide, due to soot being a strong absorber of solar energy [3]. A portion of the sun’s energy is reflected when it hits the atmosphere or earth’s surface; however, an increase in soot deposits reduces the amount of energy that gets reflected, through thermal absorption and heating of the particles. Due to all these concerns, there is a need to reduce soot emissions from combustion devices. In order to do so, the factors that affect soot emission levels must be understood.

Due to the very complex nature of soot formation, it is still not fully comprehended; however, the
current theories are presented. Soot formation begins in the gaseous phase when aromatic rings are formed in the combustion process. These aromatic rings grow into PAHs through chemical reaction with other gas phase species. These PAHs will start to combine into solid particles, which is referred to as nucleation. Once these incipient solid particles have formed, several concurrent processes take place. The individual particles undergo growth via hydrogen-abstraction-carbon-addition (HACA) and PAH condensation. Additionally, particles may coalesce to form larger particles, or coagulate to form aggregates in a fractal, chain-like structure. Competing against these growth mechanisms is surface oxidation and aggregate fragmentation [4,5]. It should be noted that final soot emissions from a combustion process is determined by the difference of growth processes and oxidation; as long as all the formed soot is oxidized, there will be no emission.

Most experimental studies investigating soot formation have been performed at atmospheric pressure. This is due to the difficulty of achieving stable flames at elevated pressures. In order to gain an understanding of the underlying factors affecting soot formation mechanics, numerical simulations are required to look into the individual processes that occur. Since most experimental studies have been performed at atmospheric pressure, most numerical simulations have been performed at atmospheric pressure as well due to a lack of data for model validation. This is an issue, as most practical combustion devices operate at elevated pressures. In order to reduce emission levels from combustion devices, an understanding of the effect pressure has on soot formation mechanisms is required.

Recently, there has been an influx of quality high pressure data sets. Some researchers have utilized these data sets to validate soot formation models at elevated pressures. The results have been fairly positive; however, due to the simplified nature of the models that have been employed thus far, insights into the effect pressure has on the individual mechanisms of soot formation could not be gained. Zhang and co-workers [6–10] have developed a detailed numerical model that includes most of the processes believed to occur during soot formation. The model has gone through extensive validation at atmospheric pressure, although it has only been applied to high pressure flames in a preliminary investigation.

1.2 Objectives

The lack of numerical studies at elevated pressure utilizing a detailed soot formation model leaves a glaring gap in our knowledge of the factors that affect soot formation. To address this issue, the first objective is to validate the detailed numerical model developed by Zhang and co-workers [6–10] for high pressure use. Once validated, the second objective is to use the vast numerical data provided by the model to understand the effect that changing pressure has on soot formation. The third objective is to
1.3 Outline of the thesis

This thesis is composed of two journal articles that address the four objectives outlined above. Each publication contains its own literature review, methodologies, results and analyses, and conclusions, with additions to enhance the self-contained nature of a thesis. The first publication, which is presented in Chapter 2, is a study of elevated-pressure ethane-air co-flow diffusion flames utilizing the detailed numerical model and addresses the first three objectives. The numerical results are compared to the experimental data sets in [11,12] and subsequently used to determine the effect that pressure has on the various soot formation mechanisms. This article has been previously accepted for publication [13]. The fourth objective of addressing deficiencies is addressed in the second article presented in this thesis. The second publication looks at the effect of extending the computational domain below the fuel tube exit plane and including CHT between the gas phase and solid fuel tube on the same ethane-air flames studied previously, which is presented in Chapter 3. The effect that this modification has on the numerical results is investigated. This article has been submitted for publication at the time of writing this thesis. Finally, the overall conclusions and recommendations for the entire body of work are present in Chapter 4.
Chapter 2

Effect of Pressure on Ethane-air Coflow Diffusion Flames

2.1 Introduction

Soot formation in combustion devices is an important area of interest, as atmospheric soot is known to pose significant health risks and contribute to long term global climate change. Experimental and numerical studies can provide a fundamental understanding of the factors affecting soot formation, which would assist engineers in designing fuels, engines, and power generation systems that are cleaner and emit less soot into our atmosphere. Detailed numerical models must be employed to further our understanding due to the highly complex nature of soot formation. Any proposed model must be able to accurately predict soot formation at elevated pressures, as many combustion devices operate above atmospheric pressures.

2.1.1 Experimental studies at elevated pressure

There have been a number of experimental investigations of soot formation at elevated pressure, with most using a co-flow flame configuration. Measurements were performed by Flower and Bowman [14] of the line-of-sight integrated soot volume fraction in 1 to 10 atm ethylene flames. The maximum value of integrated soot in the flame was found to scale with $P^{1.2\pm0.1}$. This result was verified by Lee and Na [15], as they obtained a scaling of $P^{1.26}$ from 1 to 4 atm. McCrain and Roberts obtained measurements for path-integrated and local soot volume fractions for 1 to 16 atm for ethylene flames, and 1 to 25 atm for methane flames [16]. Finally, Geigle et al. measured centerline soot volume fraction in ethylene,
propane, and toluene flames from 1 to 5 atm \[17,18\].

Recently, an influx of investigations with radially-resolved soot volume fraction measurements at elevated pressures have occurred. Measurements of soot volume fraction and temperature for methane flames from 5 to 40 atm were performed by Thomson et al. \[19\]. Maximum soot volume fraction scaled with pressure according to $P^{2.0}$ for pressures from 5 to 20 atm, and $P^{1.2}$ for pressures from 20 to 40 atm. These measurements were extended to 60 atm, and then to 100 atm with methane-oxygen flames by Joo and Gülder \[20, 21\]. The latter investigation had the interesting result of soot volume fraction reaching a peak at 55 atm, then monotonically declining as pressure was raised further. Maximum soot volume fraction increased with a scaling of $P^{1.5}$ for pressures from 10 to 40 atm, $P^{-2.3}$ for pressures from 50 to 70 atm, and $P^{-7.6}$ for pressures from 70 to 90 atm. Additionally, Joo and Gülder \[22\] investigated soot formation in ethylene-nitrogen diluted flames from 10 to 35 atm. Another investigation by Bento et al. \[23\] utilized propane as the fuel, and covered a pressure range of 1 to 7.3 atm. In this investigation, maximum soot volume fraction scaled with pressure according to $P^{1.8}$ for pressures from 2 to 7.3 atm, and path-integrated soot was shown to scale as $P^{1.4}$. Finally, Mandatori and Gülder \[11, 12\] looked at soot formation in ethane flames from 2 to 33 atm. For all of these investigations, the flame cross-sectional area scaled with pressure according to $P^{-1.0}$.

### 2.1.2 Numerical studies at elevated pressure

While limited, efforts have been made to perform numerical studies of soot formation at elevated pressures as well. An initial investigation by Zhang and Ezekoye \[24\] utilized a highly-simplified 8-step chemical reaction scheme for a methane flame, along with a 4-step mechanism for soot formation developed by Fairweather et al. \[25\]. In this investigation, the increase of peak soot volume fraction with pressure was attributed to an increase in gas phase density. It was found that although particle inception and agglomeration rates increased with pressure, the overall rate for soot particle number density production decreased. This caused lower particle number densities as pressure was elevated. Liu et al. \[26\] applied an acetylene-based soot model proposed by Leung et al. \[27\] to methane flames from 5 to 40 atm, and validated against the data collected in \[19\]. Once again, the increase in soot volume fractions with pressure was determined to be primarily due to increases in gas phase density and subsequently species concentrations. Finally, simulations were performed for 0.5 to 5 and 10 to 35 atm ethylene flames and 1 to 60 atm methane flames at regular-gravity and micro-gravity using the Leung model by Charest et al. \[28–30\]. In the study of the 10 to 35 atm ethylene flames, the flow domain was extended to include part of the fuel flow tube to capture the effect of fuel pre-heating, which was shown to be significant.
by the study performed by Guo et al. [31]. Maximum soot volume fraction was found to increase with pressure, with the dependence of soot on pressure becoming weaker as pressure was increased. Prior to the investigation in this study, no attempts were made to perform detailed modeling of soot formation at elevated pressures.

2.1.3 Detailed modeling at atmospheric pressure

A highly detailed numerical model was developed by Zhang and coworkers [6–10]. Instead of utilizing acetylene as the main precursor to soot, it employed polycyclic-aromatic hydrocarbons (PAHs) as precursors. The model accounts for all processes that are believed to occur in soot formation, including PAH growth and particle inception, surface growth via hydrogen-abstraction-carbon-addition (HACA) surface chemistry and via PAH condensation, surface oxidation, particle coagulation and fragmentation, gas phase scrubbing, soot particle diffusion, and radiation [4,32]. Initially, it utilized the gas-phase and surface growth mechanisms of Appel et al. [5], which is primarily based on the HACA mechanism; however, in a recent work [10], the gas phase mechanism was replaced with that developed by Slavinskaya and Frank [33]. This mechanism includes numerous additional reaction pathways for PAH formation, updated reaction rates, and includes an expansion of the HACA mechanism to include abstraction and growth by additional species. Using this mechanism resulted in the correct order-of-magnitude prediction of soot volume fraction along the centerline of an atmospheric pressure ethylene flame, which had not been possible with the less comprehensive mechanism of Appel et al. [10]. A comprehensive validation against numerous sets of experimental data was performed at atmospheric pressure [10].

In the present study, this highly detailed model and gas phase mechanism used in [10] are validated for high pressure conditions without any tuning or modification of model parameters. The mechanisms by which pressure affects soot formation are investigated.

2.2 Numerical Model

2.2.1 Burner and flame description

The flames chosen for this investigation are the steady, non-smoking, coflow laminar ethane/air diffusion flames at pressures of 2 to 33 atm, studied by Mandatori and Gülder [11,12]. Fuel and air mass flow rates are kept constant as pressure is varied, with values of 0.0052 g/s and 0.12 g/s for the fuel and air streams, respectively. The flame heights varied from 0.7 cm at 2 atm to 1.0 cm at 15 atm, and then remained constant. These flames were chosen as there are many characteristics that make them suitable
for studying the effects of high pressure on soot formation with the current numerical model. There are radially resolved soot measurements, as opposed to only integrated soot volume fractions. The fuel is a \( C_2 \) fuel, which is beneficial as the model has been validated at atmospheric pressure for another \( C_2 \) fuel (ethylene) \[10\]. Finally, the 2 and 5 atm flames appear to be lifted flames, which will reduce the effect of pre-heating by conduction from the flame to the fuel and air streams. Flames above 15 atm appear to be significantly anchored to the fuel tube in the images in [11]. With anchored flames, there exists an additional complexity of defining temperature boundary conditions at the anchoring site, which is discussed in the present study. Attempts to model pressures above 15 atm proved to be unsuccessful due to convergence issues attributed to the increased anchoring of the flame.

A schematic of the burner, flame, and computational domain are depicted in Fig. 3.1. The fuel (ethane) enters the flame domain through the center tube, which has an inner diameter of 0.306 cm. Air enters the flame domain through a coflowing annular region, which has an inner diameter of 2.54 cm. In the experiments, glass beads were used to straighten the air flow to achieve close to a flat exit velocity profile, thus a flat profile is used in the model. The fuel nozzle did not have glass beads. In earlier investigations, the sensitivity of the solution to an assumed flat profile was negligible, and thus a flat profile for the fuel velocity is used as well [10]. The inlet temperature for both the air and fuel streams is set to 300 K. Due to conduction from the flame, it is probable that the fuel and air streams are not at 300 K when entering the computational domain [31]. The sensitivity to the inlet temperature boundary condition and an attempt to quantify the amount of pre-heating that occurs is presented later in this study.

### 2.2.2 Governing equations

For the gaseous phase, the fully coupled elliptical conservation equations for mass, momentum, energy, and species mass fraction are solved. The model utilizes the axi-symmetrical nature of the flame, and equations are solved in the two-dimensional (\( z \) and \( r \)) cylindrical co-ordinate system. A more comprehensive description of the governing equations, boundary conditions, and solution methodology can be found in [6–9]. Below are the governing gas phase equations:

**Conservation of mass:**

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \rho v \right) + \frac{\partial}{\partial z} \left( \rho u \right) = 0 \tag{2.1}
\]

**Conservation of axial and radial momentum:**
Figure 2.1: Schematic representation of the burner and flame, with coordinate axes and computational domain boundaries (image is not drawn to scale.)

\( \rho_v \frac{\partial u}{\partial r} + \rho_u \frac{\partial u}{\partial z} = -\frac{\partial p}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \mu \frac{\partial u}{\partial r} \right) + 2 \frac{\partial}{\partial z} \left( \frac{\partial u}{\partial z} \right) - \frac{2}{3} \frac{\partial}{\partial z} \left[ \frac{\mu}{r} \frac{\partial}{\partial r} \left( r \nu \right) \right] - \frac{2}{3} \frac{\partial}{\partial z} \left[ \frac{\mu}{r} \frac{\partial}{\partial r} \left( r \nu \right) \right] + \rho g z \) \hspace{1cm} (2.2)

\( \rho_v \frac{\partial v}{\partial r} + \rho_u \frac{\partial v}{\partial z} = -\frac{\partial p}{\partial r} + \frac{2}{r} \frac{\partial}{\partial r} \left( r \mu \frac{\partial v}{\partial r} \right) + \frac{\partial}{\partial z} \left( \frac{\partial v}{\partial z} \right) - \frac{2}{3} \frac{\partial}{\partial r} \left[ \frac{\mu}{r} \frac{\partial}{\partial r} \left( r \nu \right) \right] - \frac{2}{3} \frac{\partial}{\partial r} \left[ \frac{\mu}{r} \frac{\partial}{\partial r} \left( r \nu \right) \right] + \frac{2}{3} \frac{\partial}{\partial z} \left( \frac{\partial v}{\partial z} \right) - \frac{2 \mu}{r^2} \frac{\partial}{\partial z} \left( \frac{\partial u}{\partial z} \right) + \frac{2 \mu}{3 r} \frac{\partial}{\partial z} \left( \frac{\partial v}{\partial z} \right) \) \hspace{1cm} (2.3)

Conservation of species mass fractions:
Chapter 2. Effect of Pressure on Ethane-air Coflow Diffusion Flames

\[ \rho v \frac{\partial Y_k}{\partial r} + \rho u \frac{\partial Y_k}{\partial z} = -\frac{1}{r} \frac{\partial}{\partial r} \left( r \rho Y_k V_{k,r} \right) - \frac{\partial}{\partial z} \left( \rho Y_k V_{k,z} \right) + W_k \dot{\omega}_k \]  
\[(k = 1, 2, ..., KK)\]  

\( (2.4) \)

**Conservation of energy:**

\[ C_p \left( \rho v \frac{\partial T}{\partial r} + \rho u \frac{\partial T}{\partial z} \right) = \frac{1}{r} \frac{\partial}{\partial r} \left( r \lambda \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left( \lambda \frac{\partial T}{\partial z} \right) - \sum_{k=1}^{KK} h_k W_k \dot{\omega}_k \]

\[ - \sum_{k=1}^{KK} \left[ \rho C_{p,k} Y_k \left( V_{k,r} \frac{\partial T}{\partial r} + V_{k,z} \frac{\partial T}{\partial z} \right) \right] \]

\[ - \rho C_{p,s} Y_s \left( V_{Ts,r} \frac{\partial T}{\partial r} + V_{Ts,z} \frac{\partial T}{\partial z} \right) \]

\[ - h_s W_s \dot{\omega}_s + Q_r \]  
\[(2.5) \]

In the preceding equations, \( u \) is the axial velocity, \( v \) is the radial velocity, \( \rho \) is the mixture density, \( \mu \) is the viscosity, \( g_z \) is the acceleration due to gravity, \( Y_k \) is the \( kth \) species mass fraction, \( V_{k,r} \) and \( V_{k,z} \) are the \( kth \) species radial and axial diffusion velocities, \( W_k \) is the molecular weight of the \( kth \) species, \( \dot{\omega}_k \) is the production rate of the \( kth \) species due to chemical reaction, \( C_p \) is the mixture specific heat capacity at constant pressure, \( T \) is the temperature, \( \lambda \) is the mixture thermal conductivity, \( h_k \) is the specific enthalpy of the \( kth \) species, \( C_{p,k} \) is the specific heat capacity of the \( kth \) species at constant pressure, \( C_{p,s} \) is the specific heat capacity of soot at constant pressure, \( Y_s \) is the soot mass fraction, \( V_{Ts,r} \) and \( V_{Ts,z} \) are the soot radial and axial diffusion velocities, \( h_s \) is the specific enthalpy of soot, \( W_s \) is the molecular weight of soot (same as carbon), and \( Q_r \) is the radiative heat transfer by soot, H\(_2\)O, CO\(_2\), and CO, and is calculated using the discrete-ordinates method (DOM) and a statistical narrow-band correlated-\( k \)-based model developed in [34] and benchmarked in [35]. The DOM integrates the radiative transfer equation (RTE) along a prescribed number of discrete ordinates defined by a polar and azimuthal angle, which eases obtaining numerical solutions to the RTE. Below is the discrete ordinates form of the RTE in cylindrical co-ordinates:

\[ -\kappa I_{m,l} + \kappa I_b = \frac{\mu_{m,l}}{r} \frac{\partial I_{m,l}}{\partial r} + \xi_{m,l} \frac{\partial I_{m,l}}{\partial z} - \frac{1}{r} \left( \frac{\alpha_{m,l+1/2} I_{m,l+1/2} - \alpha_{m,l-1/2} I_{m,l-1/2}}{\omega_{m,l}} \right) \]  
\[(2.6) \]

where \( m \) is the polar angle index from 1 to \( M \), \( l \) is the azimuthal angle index from 1 to \( L(m) \), \( \mu \)
and ξ are directional cosines, κ is the spectral absorption coefficient, I is the spectral intensity, \( I_b \) is the spectral blackbody intensity, \( α \) is a geometric constant [36], \( ω \) is a weighting function associated with the direction defined by \( (m, l) \), \( r \) is the radial direction, and \( z \) is the axial direction. The set of discrete ordinates is defined using the \( T_3 \) quadrature [37].

Soot particle dynamics are described using a fixed sectional method, in which soot particle mass ranges are divided logarithmically into thirty-five discrete sections. Soot aggregates are assumed to be composed of spherical primary particles of equal size with a constant fractal dimension of 1.8 [38, 39]. Two transport equations are solved for each section, those being aggregate number density \( (N^a_i) \) and soot primary particle number density \( (N^p_i) \), where \( i = 1, 2, \ldots, 35 \). Details of the two-equation-per-section model can be found in [40, 41]. The soot sectional model includes several processes, those being nucleation \( (nu) \), surface growth \( (sg) \), PAH surface condensation \( (sc) \), surface oxidation \( (ox) \), coagulation \( (co) \), fragmentation \( (fr) \), particle diffusion \( (D^a_i) \), and thermophoresis \( (V_{Ts}) \). Below are the governing soot particle dynamics equations:

**Conservation of soot aggregate number density:**

\[
\rho v \frac{\partial N^a_i}{\partial r} + \rho u \frac{\partial N^a_i}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho D^a_i \frac{\partial N^a_i}{\partial r} \right) + \frac{\partial}{\partial z} \left( \rho D^a_i \frac{\partial N^a_i}{\partial z} \right) - \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho N^a_i V_{Ts,r} \right) - \frac{\partial}{\partial z} \left( \rho N^a_i V_{Ts,z} \right) + \rho \left( \frac{\partial N^a_i}{\partial t} \right)_{nu} + \frac{\partial N^a_i}{\partial t}_{co} + \frac{\partial N^a_i}{\partial t}_{sg} + \frac{\partial N^a_i}{\partial t}_{ox} + \frac{\partial N^a_i}{\partial t}_{sc} + \frac{\partial N^a_i}{\partial t}_{fr} \quad (i = 1, 2, \ldots, 35)
\]

**Conservation of soot primary particle number density:**

\[
\rho v \frac{\partial N^p_i}{\partial r} + \rho u \frac{\partial N^p_i}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho D^p_i \frac{\partial N^p_i}{\partial r} \right) + \frac{\partial}{\partial z} \left( \rho D^p_i \frac{\partial N^p_i}{\partial z} \right) - \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho N^p_i V_{Ts,r} \right) - \frac{\partial}{\partial z} \left( \rho N^p_i V_{Ts,z} \right) + \rho \left( \frac{\partial N^p_i}{\partial t} \right)_{nu} + \frac{\partial N^p_i}{\partial t}_{co} + \frac{\partial N^p_i}{\partial t}_{sg} + \frac{\partial N^p_i}{\partial t}_{ox} + \frac{\partial N^p_i}{\partial t}_{sc} + \frac{\partial N^p_i}{\partial t}_{fr} \quad (i = 1, 2, \ldots, 35)
\]
Nucleation is modelled based on the collision, and subsequent dimerization, of two pyrene molecules in the free-molecular regime, which serves as a connection between the gaseous phase chemical reaction mechanism and the first soot particle section. It was determined in recent work by Sabbah et al. [42] that this particular dimerization is not a complete depiction of physical reality but rather a modeling technique used to effectively link the gas and particle phases. The HACA mechanism developed in [5,43–45] is used to describe soot particle surface growth, with the implementation used in [5] and adapted in [10] being employed in this model. The empirical parameter $\alpha$, which relates to the portion of active soot particle surface sites available for chemical reaction, reconciles the inaccuracies of treating sites on the soot surface as corresponding sites on gaseous PAH molecules, and is set to a constant value for all simulations. The value of $\alpha$ directly influences calculated soot surface reaction rates. A sensitivity analysis is performed to assess the effect that $\alpha$ has on computed soot volume fractions for ranges of 0.0175 to 0.332. PAH condensation is modelled based on collision theory between pyrene molecules and aggregates, with a collision efficiency, $\gamma$, of 0.55 [9]. All other soot phenomena, including nucleation, surface growth, PAH surface condensation, surface oxidation, coagulation, fragmentation, particle diffusion, thermophoresis, and particle radiation are modeled in the same fashion as in [6,7,10,13,46–48]. It should be noted that sectional model does not include detailed modeling of the chemical structure of the soot surface.

2.2.3 Boundary conditions

The boundary conditions are as described in [6,7,10,13,46–48], with modifications made to accommodate the extension of the computational domain. Below are the boundary conditions utilized in this investigation:

\textit{Inlet:}

\begin{align}
  u &= u_{\text{fuel}}, Y_{\text{fuel}} = 1.0 & r \leq 0.1145 \\
  u &= 0, Y_k = 0 & 0.1145 \leq r \leq 0.238
\end{align}

(2.9)
\[ u = u_{\text{air}}, Y_{O_2} = 0.231, Y_{N_2} = 0.769 \quad r \geq 0.238 \]
\[ v = 0, N_i^a = 0, N_i^p = 0, T = 300K \]

Axis of symmetry

Zero-gradient conditions into the axis of symmetry are imposed as follows.

\[ \frac{\partial u}{\partial r} = 0, v = 0, \frac{\partial T}{\partial r} = 0, \frac{\partial Y_k}{\partial r} = 0, \frac{\partial N_i^p}{\partial r} = 0, \frac{\partial N_i^a}{\partial r} = 0 \quad (2.10) \]

Outer Radial Boundary

Zero-gradient conditions and species mass fractions consistent with ambient air are imposed at the outer radial boundary as follows.

\[ u = 0, v = 0, \frac{\partial T}{\partial r} = 0, Y_{O_2} = 0.231, Y_{N_2} = 0.769, \frac{\partial N_i^p}{\partial r} = 0, \frac{\partial N_i^a}{\partial r} = 0 \quad (2.11) \]

Outflow

Zero-gradient conditions for the outflow are imposed as follows.

\[ \dot{m}_{\text{outflow}} = \dot{m}_{\text{inlet}}, \frac{\partial v}{\partial z} = 0, \frac{\partial T}{\partial z} = 0, \frac{\partial Y_k}{\partial z} = 0, \frac{\partial N_i^p}{\partial z} = 0, \frac{\partial N_i^a}{\partial z} = 0 \quad (2.12) \]

2.2.4 Chemical kinetic mechanism

The mechanism that is used for this investigation is the one originally presented in [33], with modifications described in [10]. This mechanism is chosen due to its ability to accurately predict soot volume fraction levels in all regions of an atmospheric C\textsubscript{2} fuel (ethylene) flame [10]. This mechanism aims to accurately describe C\textsubscript{2}/methane oxidation, with PAH growth up to five aromatic rings, and has been validated for high pressures [33]. It is important to note that reaction rate constants are not modified as pressure is varied.
2.2.5 Numerical Method

The first computation domain used for this investigation extends 2.46 cm in the axial direction and 1.18 cm in the radial direction, and is divided into 192 \((z) \times 88 (r)\) control volumes. A non-uniform mesh is used to save computational cost while still resolving large spatial gradients. The grid is finest in the flame region with maximum resolutions of 0.005 cm between \(r = 0.0\ cm\) and \(r = 0.25\ cm\) in the radial direction, and 0.01 cm between \(z = 0.0\ cm\) and \(z = 1.34\ cm\) in the axial direction. Due to convergence issues with increasing pressure, a refined grid with maximum resolutions of 0.0032 cm between \(r = 0.0\ cm\) and \(r = 0.16\ cm\) in the radial direction, and 0.0068 cm between \(z = 0.0\ cm\) and \(z = 1.1\ cm\) in the axial direction is utilized. This grid is divided into 240 \((z) \times 130 (r)\) control volumes, and is employed for 10 atm simulations with \(\alpha\) greater than 0.039, and for all 15 atm simulations. Flat velocity profiles are assumed for the inlet fuel and oxidizer streams with values calculated from the pressure, inlet temperature, molecular weight, and mass flow rates, and summarized in Table 2.1. Symmetry, free-slip, and zero-gradient conditions are enforced at the centerline, the outer radial boundary, and the outflow boundary, respectively.

<table>
<thead>
<tr>
<th>Pressure (atm)</th>
<th>Fuel Velocity (cm/s)</th>
<th>Air Velocity (cm/s)</th>
<th>Grid ((z) \times (r))</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>2.89</td>
<td>10.38</td>
<td>192 \times 88</td>
</tr>
<tr>
<td>5</td>
<td>1.16</td>
<td>4.15</td>
<td>192 \times 88</td>
</tr>
<tr>
<td>10</td>
<td>0.58</td>
<td>2.08</td>
<td>192 \times 88 for (\alpha &lt; 0.039)</td>
</tr>
<tr>
<td>15</td>
<td>0.39</td>
<td>1.38</td>
<td>240 \times 130</td>
</tr>
</tbody>
</table>

As in previous works [6, 7, 10, 46], the finite volume method is used to discretize the governing equations. A staggered mesh is used with a semi-implicit scheme to handle the pressure and velocity coupling and to solve the discretized equations at each discrete time interval. Fig. 2.2 displays the structure of the staggered grid, with the \(u\) velocity control volumes offset vertically and the \(v\) velocity control volumes offset horizontally from the pressure control volumes. The species, energy, and soot conservation equations are solved using the pressure control volume layout.

The diffusive terms are discretized using a second-order central difference scheme while the convective terms are discretized using a power law scheme [49]. The thermal properties of the gaseous species and chemical reaction rates are obtained by using CHEMKIN subroutines [50, 51]. Transport properties which include mixture-averaged quantities for viscosities, conductivities, and diffusion coefficients, as well as thermal diffusion coefficients for H and H\(_2\), are evaluated using TPLIB [52, 53].
Pseudo-transient continuation is used to aid convergence from an arbitrary starting estimate [9]. At each pseudo-time step, after the momentum and pressure correction equations are solved globally [49], the gaseous species equations are solved in a coupled manner at each control volume to effectively deal with the stiffness of the system and speedup the convergence process (i.e., at a given timestep, a linear system coupling each of the species mass conservation equations is formed and solved at each control volume). After iteration of the species equations, the sectional transport equations are also solved simultaneously in a similar fashion. The energy equation is then solved to generate an updated global temperature field and a convergence check is performed. If the vector norms of the independent variables change by less than one one-thousandth of their value from one timestep to the next, then the algorithm is deemed to have converged. Otherwise time stepping proceeds, again starting with solution of the momentum and pressure correction equations. Care was taken to ensure that the computed solution was independent of the convergence tolerance used, by lowering the convergence tolerance by an order of magnitude and checking for changes in the solution.

Due to the computational intensity of the problem, solutions would be intractable with serial processing. Therefore, distributed-memory parallelization with strip-domain decomposition is employed. The computational domain is divided uniformly into \( NPROC \) subdomains with the boundaries of each subdomain perpendicular to the \( z \)-axis, where \( NPROC \) is equal to the number of grid lines in the axial direction. The algorithm uses the Message Passing Interface [54] library to distribute the workload. The computations are performed on the General Purpose Cluster (GPC) of SciNet, on twelve to fifteen
2.3 Results and discussion

2.3.1 Soot volume fraction as a function of $\alpha$

A sensitivity analysis is performed to determine the relationship between $\alpha$ and computed soot volume fractions in both the wing and centerline regions. A recent study [10] has reported the empirical parameter $\alpha$, which relates to the portion of active soot surface sites available for chemical reactions, to be 0.078 for an atmospheric pressure ethylene diffusion flame. The exact variation of $\alpha$ with flame parameters is unknown and remains an area of active investigation. In the present work, calculations are performed for $\alpha$ values of 0.0195, 0.039, 0.078, and 0.156 for each flame, and 0.332 for the 15 atm flame. The results, with lines of best fit for the data, are displayed in Fig. 2.3.

Figure 2.3: Maximum soot volume fraction on the wings and centerline of a coflow ethane-air diffusion flame as a function of $\alpha$ with lines of best fit, at pressures of a) 2 atm, b) 5 atm, c) 10 atm, d) 15 atm.
As pressure increases, centerline soot volume fraction displays a decreased sensitivity to $\alpha$. For 2 atm, centerline soot volume fraction varies by 210% for the range of $\alpha$ studied. As pressure increases to 5 atm, centerline soot volume fraction varies by 24%, and for 10 atm and 15 atm, centerline soot volume fractions varies by 7% and 43% respectively. For the 2 atm flame, centerline soot volume fraction varies linearly, and for the 5 atm it takes on a non-monotonic relationship wherein it increases from 4.0 ppm for $\alpha = 0.00195$ to 5.0 ppm for $\alpha = 0.078$, then decreases back to 4.948 ppm for $\alpha = 0.156$. For the 10, and 15 atm flames, there is a reduction in centerline soot volume fraction as $\alpha$ increases, although the sensitivity is rather low.

As pressure is increased, the character of the sensitivity of wing soot volume fraction to $\alpha$ changes. For the 2 atm flame, wing soot volume fraction is proportional to $\alpha^2$, while for the 5 atm and 10 atm flames, wing soot volume fraction varies linearly. For the 15 atm flame, wing soot volume fraction varies logarithmically with $\alpha$. In a similar manner to which $\alpha = 0.078$ was determined in [10], the dependence of $\alpha$ on pressure is studied in the following section.

### 2.3.2 Dependence of $\alpha$ on pressure

It was sought to determine if there was one value of $\alpha$ for which all computed peak wing soot volume fractions would match the experimental data. Additionally, the data set modelled in [10] is included in this analysis, to determine if the sought value of $\alpha$ can also accurately predict peak wing soot volume fraction for an atmospheric pressure ethylene flame. The experimental values were obtained using spectral soot emission (SSE) for the elevated pressure flames [11], and by line-of-site attenuation (LOSA) for the atmospheric flame [55]. The error in the SSE method is estimated to be 30 to 40% [56], with a relative uncertainty due to systematic and random errors of 30% at 2 atm, and 3% at all higher pressures [12]. The error for LOSA is estimated to be 20 to 30% [56]. Using these error values, and the variations of wing soot volume fraction with $\alpha$, it is found that for values of $\alpha$ between 0.055 and 0.067, the predicted peak (wing) soot volume fraction for all five flames are within the experimental error. As shown in Fig. 2.3, peak wing soot volume fraction is highly sensitive to $\alpha$, while peak centerline soot volume fraction is not sensitive to $\alpha$, which is the reason that $\alpha$ is selected based on experimental peak wing soot volume fraction.

In Fig. 2.4, the peak experimental and computed wing soot volume fractions are compared, using an $\alpha$ of 0.061, which is in the middle of the acceptable values for $\alpha$, with error bars based on the experimental technique used. From Fig. 2.4, it can be clearly seen that for multiple pressures considering different experimental datasets, the predictions with $\alpha = 0.061$ fall within the experimental error bars.
These results show that when considering the error in the experimental data, \( \alpha \) is independent of pressure over the ranges of pressures simulated, 1 atm to 15 atm, which is the same finding as the experimental investigation of Kim et al. for ethylene-air diffusion and pre-mixed flames from 1 to 8 atm [57]. Additionally, this single value of \( \alpha \) can accurately predict the peak wing soot volume fraction for the atmospheric and elevated pressure flames considered in this investigation, which have different fuels, burners, and measurement techniques. This observation demonstrates that the model can be used for predicting peak soot formation at elevated pressures, as \( \alpha \) does not need to be altered to predict the trends in peak wing soot volume fraction as pressure is increased.

Fig. 2.5 shows the experimental and computed peak centerline soot volume fraction for \( \alpha = 0.061 \). The model consistently underpredicts the experimental data by a factor of 2 to 3, as frequently seen for laminar diffusion flames [58, 59]. Based on the low sensitivity of centerline soot volume fraction to \( \alpha \) shown in Fig. 2.3, especially in Fig. 2.3b,c,d at the higher pressures, this discrepancy would not be alleviated by varying \( \alpha \). This observation is consistent with the results in [10], wherein centerline soot volume fraction underprediction is attributed to possible errors in PAH chemistry (A4 formation) leading to errors in computed nucleation and PAH condensation rates. Another possible source of this underprediction may be uncertainty in the models employed for nucleation and PAH condensation.
2.3.3 Comparison of experimental and numerical soot volume fraction fields

Calculations with a value of $\alpha = 0.061$ are performed for 2, 5, 10, and 15 atm flames, and are compared with the experimental data sets [11,12].

The top left panel of Fig. 2.6 depicts a comparison of the computed soot volume fraction to the experimental contours for the 2 atm flame; top right for the 5 atm flame; bottom left for the 10 atm flame; and the bottom right for the 15 atm flame. The main trends observed in the experimental data set are well captured in the computed simulations, with soot volume fractions increasing with pressure on both the wings and the centerline. The model also predicts the distribution of soot within the flame with soot being formed in relatively high volume fractions on the centerline at 2 atm, and then migrating to the wings at higher pressures. Furthermore, the flame narrows as pressure is increased in the model and experiment, with the relationship $r_{flame} \propto P^{-0.5}$, which is attributed to the increased air entrainment caused by increasing bouyant forces [28]. $r_{flame}$ is taken as bounded by the maximum temperature contours [11,12]. An analytical analysis for the cause of the flame thinning is presented later in this study.

The location of the peak soot volume fraction on the wings and centerline is at a greater height above the burner, and at a greater radius in the computations than in the experiment, with the degree to which
this occurs increasing with increasing pressure. This tends to indicate a deficiency in the model that increases in severity with pressure. Additionally, the flame height is overpredicted in the computations, with increasing discrepancy as pressure increases. This is attributed to the assumed inlet temperature boundary condition of 300K, which becomes increasingly less accurate as pressure is increased. The associated effect on flame height is quantified later in section 3.5.

### 2.3.4 Effect of Pressure On Soot Formation

As pressure is increased, soot volume fraction increases along the wings and centerline. Calculated peak wing and centerline soot volume fractions are found to scale with $P^{2.49}$, and $P^{2.02}$, respectively. The scaling of peak wing soot volume fraction with pressure is similar to the values obtained experimentally for methane-air ($P^{2.0}$) [19] and propane-air flames ($P^{1.8}$) [23].

To understand the observed increase in soot volume fraction with pressure, the contributions to soot particle mass for a soot particle travelling along the pathline of maximum soot on the wings, from the three mechanisms of soot formation (inception, surface growth, and PAH condensation) are examined in Fig. 2.7. It can be seen that the mass added by all three mechanisms increases with increasing pressure,
with the exception of inception remaining constant as pressure increases from 10 atm to 15 atm. At all pressures, surface growth is the dominant mechanism, with condensation being more dominant over nucleation. Finally, the rate of increase of surface growth and nucleation with pressure reduces as pressure is increased.

Fig. 2.8 shows the contributions to soot particle mass for a soot particle travelling along the centerline from the three mechanisms of soot formation. As pressure increases, the mass contributed from all three mechanisms increases, which is similar to the trends on the wings. At 2 atm, the dominant mechanism is surface growth, with condensation slightly lower, and nucleation having a very small contribution. At pressures of 5 atm and greater, condensation overtakes surface growth and becomes the dominant mechanism. Although the model displays a consistent underprediction of centerline soot volume fraction by a factor of 2 to 3, the model very accurately predicts the trends in centerline soot volume as pressure increases. Therefore, investigating the contributions to centerline soot volume fraction by surface growth, PAH condensation, and nucleation is warranted.

To explain the trends of the magnitudes of three mechanisms of soot formation increasing with pressure, the effect that pressure has on these three mechanisms is investigated, beginning with surface growth.
Surface growth is modelled using the HACA surface growth mechanism [5]. To determine the amount of mass that is added by acetylene addition, the following equation is used for the surface growth rate, $R_{sg}$ [9]:

$$R_{sg} = \frac{2C_{mass}k_4[C_2H_2][\alpha A_sN]}{A_v} \chi_{c_{soot}} \tag{2.13}$$

where $k_4$ is the per-site rate coefficient for acetylene addition, $C_{mass}$ is the mass of a carbon atom, $[C_2H_2]$ is the molar concentration of acetylene, $A_s$ (cm$^2$/cm$^3$) is the average surface density of each soot particle, $N$ is the number density of particles (#/cm$^3$), $A_v$ is Avogadro’s number, and $\chi_{c_{soot}}$ is the number of dehydrogenate sites per cm$^2$ of soot surface area.

To understand why surface growth increases with pressure, Table 2.2 displays the values of parameters important to the surface growth model for the point of maximum mass addition by surface growth on the wings ($R_{sg} = R_{sg_{max}}$) within the 2 atm and 15 atm flames. The final column is the ratio of the given parameter at 15 atm to its value at 2 atm. Table 2.3 shows the same values along the centerline. Values for $C_2H_2$ are mole fractions, while $[C_2H_2]$ is the molar concentration of $C_2H_2$. Considering equation 2.13, surface growth varies linearly with $[C_2H_2]$, $\chi_{c_{soot}}$, $A_s$, and $N$.

From Tables 2.2 and 2.3, it can be seen the two most significant effects of pressure on the parameters
Table 2.2: Effect of Pressure on Surface Growth on the Wings

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_{sgmax} )</td>
<td>(g/cm³)</td>
<td>6.02e-4</td>
<td>2.55e-2</td>
<td>42.4</td>
</tr>
<tr>
<td>( \chi_{soot} )</td>
<td>( )</td>
<td>1.63e-2</td>
<td>1.13e-3</td>
<td>0.0696</td>
</tr>
<tr>
<td>C(_2)H(_2)</td>
<td>(mol/mol)</td>
<td>3.72e-3</td>
<td>2.23e-3</td>
<td>0.598</td>
</tr>
<tr>
<td>[C(_2)H(_2)]</td>
<td>(mol/cm³)</td>
<td>4.65e-8</td>
<td>2.20e-7</td>
<td>4.73</td>
</tr>
<tr>
<td>( A_s )</td>
<td>(cm²/cm³)</td>
<td>1.29</td>
<td>2.07e+2</td>
<td>161</td>
</tr>
<tr>
<td>( N )</td>
<td>(#/cm³)</td>
<td>5.46e+10</td>
<td>4.75e+10</td>
<td>0.870</td>
</tr>
</tbody>
</table>

Table 2.3: Effect of Pressure on Surface Growth on the Centerline

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_{sgmax} )</td>
<td>(g/cm³)</td>
<td>2.82e-4</td>
<td>3.98e-3</td>
<td>14.1</td>
</tr>
<tr>
<td>( \chi_{soot} )</td>
<td>( )</td>
<td>7.14e-3</td>
<td>2.99e-4</td>
<td>0.0419</td>
</tr>
<tr>
<td>C(_2)H(_2)</td>
<td>(mol/mol)</td>
<td>2.94e-3</td>
<td>2.64e-3</td>
<td>0.896</td>
</tr>
<tr>
<td>[C(_2)H(_2)]</td>
<td>(mol/cm³)</td>
<td>3.69e-8</td>
<td>2.48e-7</td>
<td>6.72</td>
</tr>
<tr>
<td>( A_s )</td>
<td>(cm²/cm³)</td>
<td>1.12</td>
<td>2.06e+1</td>
<td>18.4</td>
</tr>
<tr>
<td>( N )</td>
<td>(#/cm³)</td>
<td>8.53e+10</td>
<td>2.46e+11</td>
<td>2.88</td>
</tr>
</tbody>
</table>

For surface growth rates is \( A_s \) increasing, and \( \chi_{soot} \) decreasing, with secondary effects from increases in [C\(_2\)H\(_2\)]. Along the wings, the increases in \( A_s \) are more pronounced than decreases in \( \chi_{soot} \). Along the centerline, the increase in \( A_s \) and decrease in \( \chi_{soot} \) are fairly comparable, although the surface growth rates along the centerline still increase due to an increase in [C\(_2\)H\(_2\)]. The increases in [C\(_2\)H\(_2\)] are caused by increases in gas phase density, as mole fractions of C\(_2\)H\(_2\) do not vary significantly. It can therefore be concluded that the increases in surface growth rates seen as pressure increases relates to increases in soot area density and acetylene concentrations that more than offset decreases in \( \chi_{soot} \).

To explain the observed decreases in \( \chi_{soot} \), the equation used to calculate its value is examined. The following equation is used for \( \chi_{soot} \):

\[
\chi_{soot} = \frac{(k_1 \text{H} + k_2 \text{OH}) \chi_{soot-H}}{k_{-1} \text{H}_2 + k_{-2} \text{H}_2 \text{O} + k_4 \text{C}_2 \text{H}_2 + k_5 \text{O}_2}
\]  
(2.14)

where \( k_i \) is the per-site rate coefficient for reaction \( i \), \( \chi_{soot-H} \) is the number of reaction sites per cm\(^2\) of soot surface area, and is set to 2.3x10\(^{15} \) cm\(^{-2}\) [5], and H, OH, H\(_2\), H\(_2\)O, C\(_2\)H\(_2\), and O\(_2\) are mole fractions. Essentially, \( \chi_{soot} \) is calculated as the ratio of all reactions that act to abstract a hydrogen atom from the surface of a soot particle and create a de-hydrogenated site, to all those that would act to reduce the number of de-hydrogenated sites, which assuming a steady state condition, is the number of de-hydrogenated sites per unit surface area [9]. As pressure increases, the values for H\(_2\), H\(_2\)O, C\(_2\)H\(_2\), and O\(_2\) do not significantly vary; therefore, from equation 2.14 decreases in H and OH radical mole fractions will cause a decrease in \( \chi_{soot} \).
Tables 2.4 and 2.5 display the values of H and OH mole fractions and $\chi_{soot}$ at the point of maximum surface growth rate ($R_{sg} = R_{sg\text{max}}$) along the pathline of maximum soot along the wings, and along the centerline, respectively. Along the wings and centerline, the reduction in $\chi_{soot}$ is between the reductions observed for H and OH mole fractions, which is consistent with equation 2.14. The reduction in $\chi_{soot}$ is due to reduced radical mole fractions.

Table 2.4: Effect of Pressure on $\chi_{soot}$ on the Wings

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>(mol/mol)</td>
<td>5.43e-4</td>
<td>2.01e-5</td>
<td>0.04</td>
</tr>
<tr>
<td>OH</td>
<td>(mol/mol)</td>
<td>4.65e-4</td>
<td>3.11e-5</td>
<td>0.07</td>
</tr>
<tr>
<td>$\chi_{soot}$</td>
<td>( )</td>
<td>1.63e-2</td>
<td>1.13e-3</td>
<td>0.0696</td>
</tr>
</tbody>
</table>

Table 2.5: Effect of Pressure on $\chi_{soot}$ on the Centerline

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>(mol/mol)</td>
<td>2.21e-4</td>
<td>7.48e-6</td>
<td>0.03</td>
</tr>
<tr>
<td>OH</td>
<td>(mol/mol)</td>
<td>1.92e-4</td>
<td>9.43e-6</td>
<td>0.05</td>
</tr>
<tr>
<td>$\chi_{soot}$</td>
<td>( )</td>
<td>7.14e-3</td>
<td>2.99e-4</td>
<td>0.0419</td>
</tr>
</tbody>
</table>

To further investigate the effect pressure has on the species pertinent to surface growth over the flame region, Fig. 2.9 shows isopleths of soot volume fraction (top left), OH (top right), H (bottom left), and $\text{C}_2\text{H}_2$ (bottom right) mole fractions for the 2 atm flame, while Fig. 2.10 displays the same parameters for the 15 atm flame. While the mole fractions of $\text{C}_2\text{H}_2$ are similar in magnitude in the regions where soot is formed at low and high pressures, mole fractions of OH and H are lower in the 15 atm flame in the areas where soot is located, with the greatest reductions occurring along the centerline. This reduction in radicals is due to radical recombination reactions being favored as pressure increases.

As displayed in Fig. 2.9 and Fig. 2.10, and quantified in Tables 2.4 and 2.5 at the location of maximum surface growth rate, radical (H and OH) mole fractions are approximately an order of magnitude less at 15 atm than compared to 2 atm, and this causes the observed reduction in $\chi_{soot}$, with a greater reduction occurring along the centerline than on the wings. Additionally, the increased concentrations of $\text{C}_2\text{H}_2$ are due to increased gas phase density, and not increased mole fractions.

To explain the increase in $A_s$, it must be noted that $A_s$, or soot area density, is directly proportional to soot volume fraction. While soot volume fraction is a function of $A_s$ and N (or any other second parameter related to soot morphology such as soot particle size), it is shown that N does not change significantly with increasing pressure, thus $A_s$ is directly proportional to soot volume fraction. Therefore, Tables 2.2 and 2.3 show that a major contribution to the increase in surface growth rates is increases in
soot volume fraction. The result that increases in surface growth with pressure are heavily dominated by increases in soot volume fraction demonstrate that surface growth has a positive feedback quality, in that more soot allows for more soot to be created via surface growth. By examining equation 2.13,
this is due to soot volume fraction (or \( A_s \)) itself being part of the forward reaction rate calculation. In other words, soot can be thought of as both an input and a product of a global acetylene addition reaction. This positive feedback mechanism is initiated by the increases in gas phase density which cause surface growth rates to increase through increases in \([C_2H_2]\). It is concluded that surface growth rates increase as pressure increases due to increases in soot area density, caused by a positive feedback mechanism. Additionally, acetylene concentrations increase caused by increases in gas phase density, and the increases in \( A_s \) and \([C_2H_2]\) more than offset decreases in \( \chi_{soot} \), caused by reductions in H and OH mole fractions.

The effect that pressure has on PAH condensation will now be investigated. PAH condensation is modelled based on A4 (pyrene) concentrations and collisions with soot particles using collision theory. Therefore, PAH condensation is a function of A4 concentration and soot volume fraction (or soot surface area density \( A_s \), and soot particle density \( N \)), although the function is not linear. Tables 2.6 and 2.7 display the values for the parameters relevant to PAH condensation at the point of maximum mass addition by PAH condensation on the wings and on the centerline at 2 and 15 atm. A4 is the mole fraction of pyrene, and \([A4]\) is the molar concentration of pyrene.

### Table 2.6: Effect of Pressure on Condensation on the Wings

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_{max} )</td>
<td>(g/cm(^3))</td>
<td>8.98e-5</td>
<td>5.89e-3</td>
<td>65.6</td>
</tr>
<tr>
<td>A4</td>
<td>(mol/mol)</td>
<td>3.36e-7</td>
<td>1.20e-7</td>
<td>0.360</td>
</tr>
<tr>
<td>[A4]</td>
<td>(mol/cm(^3))</td>
<td>4.32e-12</td>
<td>1.19e-11</td>
<td>2.75</td>
</tr>
<tr>
<td>( A_s )</td>
<td>(cm(^2)/cm(^3))</td>
<td>3.67e-3</td>
<td>6.43e-2</td>
<td>17.50</td>
</tr>
<tr>
<td>( N )</td>
<td>(#/cm(^3))</td>
<td>1.30e+11</td>
<td>4.65e+10</td>
<td>0.360</td>
</tr>
</tbody>
</table>

### Table 2.7: Effect of Pressure on Condensation on the Centerline

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_{max} )</td>
<td>(g/cm(^3))</td>
<td>1.65e-4</td>
<td>8.45e-3</td>
<td>51.4</td>
</tr>
<tr>
<td>A4</td>
<td>(mol/mol)</td>
<td>6.98e-7</td>
<td>2.90e-7</td>
<td>0.420</td>
</tr>
<tr>
<td>[A4]</td>
<td>(mol/cm(^3))</td>
<td>9.31e-12</td>
<td>3.09e-11</td>
<td>3.32</td>
</tr>
<tr>
<td>( A_s )</td>
<td>(cm(^2)/cm(^3))</td>
<td>1.27e-4</td>
<td>1.14e-3</td>
<td>9.03</td>
</tr>
<tr>
<td>( N )</td>
<td>(#/cm(^3))</td>
<td>8.40e+11</td>
<td>4.94e+11</td>
<td>0.590</td>
</tr>
</tbody>
</table>

PAH condensation does not increase linearly with increasing \( A_s \), \( N \) and \([A4]\), thus the increase in \( R_{max} \) is not what would be expected by just multiplying the ratios of \([A4]\), \( A_s \), and \( N \). As with surface growth, the primary cause of the increase in condensation with increasing pressure is an increase in \( A_s \), with secondary effects from increases in \([A4]\) and decreases in \( N \). Once again, this increase in \( A_s \) is due to soot volume fraction itself being an input and product for PAH condensation reactions, thus being in
a positive feedback mechanism, which is initiated by increases in gas phase density. It is concluded that increases in PAH condensation are due to increases in $A_x$, caused by a positive feedback mechanism, and in $[A4]$, caused by increased gas phase density, which more than offset the decreases in $N$.

The effect pressure has on inception is now investigated. Inception is unique when compared to surface growth and condensation, in that it has no positive feedback mechanism. Based on collision theory, inception is modelled as solely a function of $[A4]^2$ and temperature. The following equation is used to model inception rates:

$$R_{inception} = \frac{\eta_{incep}[A4]^2\sqrt{T}}{A_v}$$

(2.15)

where $A_v$ is Avagadro’s number, $T$ is temperature (K), and $\eta_{incep}$ is a constant resulting from collision theory that includes the mass of carbon, density of carbon, Boltzmann constant, and disk diameter of an A4 molecule [4].

Given equation 2.15, inception rates vary with $[A4]^2$, and with the square root of $T$. Tables 2.8 and 2.9 display the values of A4 and $[A4]^2$ at the point of maximum mass addition by inception on the wings and on the centerline at 2 and 15 atm. Temperature is omitted as it is very similar for 2 atm and 15 atm along the wings and centerline.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_{max}$</td>
<td>(g/cm$^3$)</td>
<td>1.77e-5</td>
<td>4.81e-4</td>
<td>27.2</td>
</tr>
<tr>
<td>A4</td>
<td>(mol/mol)</td>
<td>3.68e-7</td>
<td>2.80e-7</td>
<td>0.760</td>
</tr>
<tr>
<td>$[A4]^2$</td>
<td>(mol/cm$^3$)</td>
<td>2.49e-23</td>
<td>6.86e-22</td>
<td>27.5</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit</th>
<th>2 atm</th>
<th>15 atm</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_{max}$</td>
<td>(g/cm$^3$)</td>
<td>5.68e-5</td>
<td>9.30e-4</td>
<td>16.4</td>
</tr>
<tr>
<td>A4</td>
<td>(mol/mol)</td>
<td>6.64e-7</td>
<td>3.29e-7</td>
<td>0.490</td>
</tr>
<tr>
<td>$[A4]^2$</td>
<td>(mol/cm$^3$)</td>
<td>8.61e-23</td>
<td>1.61e-21</td>
<td>18.7</td>
</tr>
</tbody>
</table>

Tables 2.8 and 2.9 show that the increases in maximum inception rates as pressure increases are solely due to increases in $[A4]^2$. This is a result of increased gas phase density with increasing pressure, which increases molar concentrations of A4, and subsequently inception rates. Given that inception rates are intrisically linked with A4 concentrations, the ability of the gas phase mechanism to predict A4 mole fractions, and subsequently concentrations, is highly important. In this study it is seen that A4 mole fractions do not significantly vary with increasing pressure. While Wartel et al. looked at PAH mole
fraction variation in low-pressure methane flames \cite{60, 61}, there is a need to experimentally investigate PAH mole fraction variation at elevated pressures.

To investigate the effect that pressure has on the relative contribution of the three mechanisms of soot formation, in Fig. 2.11a and b the contributions to soot particle mass for a soot particle travelling along the pathline of maximum soot on the wings, and along the centerline, from the three mechanisms of soot formation are examined. Here, the contributions are shown as percentages of the total mass added to soot. This is done to highlight the trends in the relative contributions of the mechanisms as pressure increases.

![Figure 2.11: Percentage of mass added to soot by inception, surface growth, and condensation, for a soot particle travelling a) along the pathline of maximum soot on the wings, and b) along the centerline, for 2 atm, 5 atm, 10 atm, and 15 atm ethane-air coflow diffusion flames.](image)

In Fig. 2.11a, surface growth is shown to be the dominant mechanism for soot mass addition on the wings, however, the general trend is an increasing role of condensation as pressure rises. In Fig. 2.11b, condensation is shown to increase in significance along the centerline as pressure rises, and actually is the dominant mechanism of soot mass addition along the centerline for pressures of 5 atm and greater. As pressure is increased, the role of condensation increases, while the role of surface growth decreases, in both the wing and centerline regions. To explain the trends of the relative contribution of surface growth decreasing and of condensation increasing, the dependance of surface growth and PAH condensation on $A_s$ and $\chi_{c,soot}$ is considered. The variation in PAH condensation with pressure is shown to primarily be a function of $A_s$, while the variation of surface growth with pressure is primarily a function of both $A_s$ and $\chi_{c,soot}$. $\chi_{c,soot}$ decreases with pressure, due to lower radical (OH and H) populations, while
Chapter 2. Effect of Pressure on Ethane-air Coflow Diffusion Flames

As increases with increasing pressure. The decreases in \( \chi_{\text{soot}} \) partially counteract the effect of \( \Delta s \) increasing, resulting in PAH condensation increasing more rapidly than surface growth with increasing pressure.

In summary, the reason pressure increases soot volume fraction is due to increases in gas phase density which increase reaction rates for inception, surface growth, and PAH condensation. Due to the positive feedback nature of surface growth and condensation, increases in surface growth and PAH condensation are beyond what would be projected from increases in gas phase density alone. The positive feedback effect is heavily dominant over the effect from increases in gas phase density. PAH condensation increases more rapidly than surface growth with increasing pressure due to reductions in \( \chi_{\text{soot}} \), which reduces the rate of increase of surface growth.

2.3.5 Effect of Pressure on Flame Structure

The following analysis is an analytical solution to determine the variation of the radius of the flame with pressure. Beginning with a balance of the energy being conducted through the flame to the unburnt fuel, we have the following,

\[
\dot{m}c_p dT = kA_{\text{flame}} \frac{dT}{dz}
\] (2.16)

where \( \dot{m} \) is the mass flow rate of fuel \( (\dot{m} = \rho A_{\text{flame}} V) \), where \( V \) is velocity through the flame \([12]\), \( c_p \) is the specific heat capacity, \( \frac{dT}{dz} \) is the temperature gradient across the flame, \( k \) is the thermal conductivity, \( A_{\text{flame}} \) is the cross sectional area of the flame, defined as bounded by the maximum flame temperature \([11, 12]\). Substitution of \( \dot{m} = \rho A_{\text{flame}} V \), and setting \( dz \) equal to \( \delta \), the flame thickness, followed by re-arranging yields,

\[
V = \frac{k}{\delta \rho c_p}
\] (2.17)

Since \( k \propto P^0 \), \( c_p \propto P^0 \), \( \delta \propto P^{-1} \), and \( \rho \propto P^1 \), \( V \) must be \( \propto P^0 \). The numerical results are consistent with equation 2.17, as the velocity in the flame envelope does not vary substantially with pressure, and is consistent with the results found in \([26]\). Since \( \dot{m} \) is a constant in this investigation, at any height in the flame \( \rho A_{\text{flame}} V \) must be constant as well. Thus, \( A_{\text{flame}} \propto P^{-1} \), and \( r_{\text{flame}} \propto P^{-0.5} \), which is in agreement with the experimental and numerical results. In summary, since the mass flow rate is held constant, the amount of heat released or conducted by and from the flame must be constant. Since the flame thickness decreases with the pressure, \( A_{\text{flame}} \) must decrease as well so that the heat conducted from the flame remains a constant.
2.3.6 Inlet temperature boundary condition

Due to the lack of experimental temperature data in the plane of the computational domain inlet, there is uncertainty in the assumed inlet temperatures of 300 K for the air and fuel streams. To assess the error in this assumption, an analysis of the heat being conducted into the inlet of the computational domain is performed. An inlet temperature is estimated based on a balance of the heat being conducted into the inlet plane, with that being convected out of the inlet by the air and fuel streams, knowing that at a distance far beneath the burner exit, the air and fuel are at approximately 300 K. The following equation is used:

\[
T_{bc}(r) = \frac{T_2(r) + \frac{300 \Delta z m c_p}{k A(r)}}{1 + \frac{\Delta z m c_p}{k A(r)}}
\]  

(2.18)

where \(T_{bc}(r)\) is the corrected inlet temperature at radius \(r\) and \(z = 0\), \(k\) is the thermal conductivity of the gas, \(A(r)\) is the area of the first control volume face at radius \(r\), \(T_2(r)\) is the temperature at the first control volume above the inlet at radius \(r\), \(\Delta z\) is the control volume spacing, \(\dot{m}\) is the mass flow rate in the control volume, and \(c_p\) is the specific heat capacity of air or ethane. This method assumes that all energy conducted into the inlet will be added to the incoming streams, and does not dissipate radially. The method is not meant to be quantitatively precise, rather it is meant to assess the validity of the 300 K inlet condition, and the degree to which the flames are lifted. The results of this analysis for the 2, 5, 10, and 15 atm flame with an \(\alpha\) of 0.061 are shown in Fig. 2.12. The cause of the abnormality in the 10 atm profile near the peak value is due to the presence of the fuel tube in that region. It can be seen that the computed \(T_{bc}\) peaks at approximately 515 K at 2 atm, and 800 K at 15 atm. As pressure increases, the flame becomes increasingly anchored to the fuel tube (hotter at the inlet boundary), thus the inlet condition assumption worsens as pressure increases. Even at the lowest pressure, the flame is not truly lifted, and the assumption of the 300 K inlet is not ideal and may introduce some error into the calculations. Additionally, the results show that setting a temperature that does not vary with radius is also not fully accurate.

To investigate the effect that increasing the inlet temperature would have on predicted soot volume fractions, the 2 atm flame is modelled using boundary conditions of 400, 500, and 600 K. It is noted that testing variations of inlet temperature using constant values is not perfectly representative of the expected behavior, as the inlet temperature varies radially. However, given the lack of quantitative accuracy in the method used to calculate the inlet temperature only constant temperature values will be tested. Soot contours for the four inlet boundary conditions are compared in Fig. 2.13. As the inlet temperature is increased, peak soot volume fraction on the wings and centerline increase slightly. The
Figure 2.12: Corrected inlet temperature profile computed from an energy balance on the system for 2 atm, 5 atm, 10 atm, and 15 atm ethane-air coflow diffusion flames.

Figure 2.13: Comparison of soot volume fraction isopleths at 2 atm with inlet temperatures of (increasing from left to right) 300 K, 400 K, 500 K, and 600 K for an ethane-air coflow diffusion flame.
peak soot volume fraction on the wings increases by a factor of 1.83, while the centerline values increase by a factor of 1.97, by changing the temperature from 300 K to 600 K. Additionally, soot begins to form at a lower height above the burner and the overall flame height reduces, which is in better agreement with experimental data. These two analyses display the value that accurate inlet temperature profile boundary conditions could add to highly accurate soot formation modeling. However, the factor of approximately 2 change in soot with increasing inlet temperature is much less than the change in soot with pressure from 2 atm to 15 atm. Despite the error introduced by the inaccuracies in the temperature boundary conditions, the conclusions of how pressure effects soot formation reached by this investigation would not change if this source of error were removed.

2.4 Conclusions

Laminar, sooting coflow ethane-air diffusion flames at 2 atm, 5 atm, 10 atm, and 15 atm are numerically simulated using a detailed PAH-based sectional soot model. The trends observed in the experimental data are captured by the model, leading to the conclusion that $\alpha$, a parameter to describe soot surface reactivity, is independent of pressure and the model is able to predict soot formation at elevated pressures.

As in other investigations, centerline soot volume fraction is underpredicted by a factor of 2 to 3. However, use of a more accurate temperature boundary condition may partially alleviate this discrepancy. In addition, soot formation is delayed on the centerline when compared to the experimental data, and there is an outward radial shift of the location of peak wing soot volume fraction. Both of these discrepancies occur in greater magnitudes as pressure is increased. As pressure is increased, the flame cross-sectional area decreases according to $P^{-1.0}$ due to a constant mass flux and a thinning of the flame. Peak wing and centerline soot volume fractions are found to scale with $P^{2.49}$, and $P^{2.02}$ respectively. This scaling compares well to that observed experimentally for methane-air and ethylene-air flames.

Soot formation along the wings is seen to be surface growth dominated, while PAH condensation dominates centerline soot formation. Surface growth and PAH condensation increase along the wings and centerline of the flames with increasing pressure due to a positive feedback mechanism wherein growth and condensation increase soot surface area, in turn allowing for more growth and condensation. This positive feedback mechanism is initiated by increases in gas phase density. Conversely, inception is shown to increase solely due to increases in gas phase density. The decreasing significance of surface growth compared to PAH condensation with increasing pressure is shown to be due to reduced H and OH radical concentrations, with condensation proving to be an important mechanism of soot growth at elevated pressures. Finally, the assumption of the 300 K inlet temperature boundary condition is shown
to be inaccurate and there would be a benefit to better quantifying the conditions at the inlet. However, the validity of this assumption does not compromise the conclusions of this investigation. One way to obtain a more accurate boundary condition for the fuel tube exit plane would be to take radially resolved temperature measurements. Experimentally this may prove cumbersome or may have high uncertainty in the data, although feasibility was shown in [19]. Another solution would be to incorporate part of the fuel tube in the computational domain [31, 62], to allow the domain inlet to be at a location where pre-heating effects are negligible.
Chapter 3

The Effect of Conjugate Heat Transfer

3.1 Introduction

Atmospheric soot, often referred to as black carbon, or particulate, is known to have detrimental effects on both human health and the global environment. Soot particles, which are typically 10-300 nm in diameter, can be absorbed deeply into the lungs, causing respiratory diseases such as asthma and bronchitis. The particles can be small enough to pass into the bloodstream leading to possible heart disease [2]. Polycyclic Aromatic Hydrocarbons (PAHs), which are pre-cursors to soot formation, have been classified as known carcinogens by the International Agency for Research on Cancer (IARC) as well. Additionally, soot is an environmental concern as it is the second most important factor in global warming behind carbon dioxide, due to soot being a strong absorber of solar energy [3]. A portion of the sun’s energy is reflected when it hits the atmosphere or earth’s surface; however, an increase in soot deposits reduces the amount of energy that gets reflected, through thermal absorption and heating of the particles.

Due to these concerns, soot formation in combustion devices is an important area of interest. Experimental and numerical studies can provide a fundamental understanding of the factors affecting soot formation, which would assist engineers in designing fuels, engines, and power generation systems that are cleaner and emit less soot into our atmosphere. Experimental studies are best conducted utilizing co-flowing, laminar, diffusion flames, as their simple geometry and flow conditions allow for a focus on the mechanisms of soot formation. Even with the conditions in these experiments, detailed numerical
models must be employed to further our understanding due to the highly complex nature of soot formation. Any proposed model must be able to accurately predict soot formation at elevated pressures, as many practical combustion devices operate above atmospheric pressure.

3.1.1 Numerical studies at elevated pressure

While limited, efforts have been made to perform numerical studies of soot formation at elevated pressures. An initial investigation by Zhang and Ezekoye [24] utilized a highly simplified 8-step chemical reaction scheme for a methane flame, along with a 4-step mechanism for soot formation developed by Fairweather et al. [25]. Liu et al. [26] applied an acetylene-based soot model proposed by Leung et al. [27] to methane flames from 5 to 40 atm, and validated against the data collected in [19]. Finally, simulations were performed for 0.5 to 5 and 10 to 35 atm ethylene flames and 1 to 60 atm methane flames at regular-gravity and micro-gravity using the Leung model by Charest et al. [28–30]. The aforementioned investigations used some form of simplified soot formation model, which neglect the formation of PAH soot precursors. A second issue common to the majority of these previous works is that conduction from the flame to the fuel and air streams, which causes these streams to be pre-heated prior to the exit plane of the fuel tube, was neglected due to the confinement of the computational domains above the exit plane of the fuel tube. This pre-heating effect is of particular concern for elevated-pressure flames, which tend to become increasingly anchored to the fuel tube as pressure is increased. The effect was shown to have significant impact on soot formation in the study performed by Guo et al. [31], where the computational domain was extended below the fuel tube exit plane to capture the effect of fuel pre-heating, although the study was done for an atmospheric pressure flame. Charet et al. [28–30] employed this procedure of extending the domain in the 10 to 35 atm ethylene flames as well; however, in both of these studies with extended domains, conjugate heat transfer (CHT) between the fluid streams and the fuel tube was not modelled, rather an assumed fuel tube temperature or adiabatic conditions were used. While no investigations exist with CHT at elevated pressures, a study that did not include soot modelling was performed for an atmospheric methane-air pre-mixed co-flow flame [62].

3.1.2 Detailed modeling at elevated pressure

A highly detailed numerical model was developed by Zhang and coworkers [6–10]. Instead of utilizing a simplified acetylene-based soot formation model, it employs a polycyclic-aromatic hydrocarbons (PAHs) based sectional particle dynamics model, tracking shape and size distribution. The model accounts for all processes that are believed to occur in soot formation, including PAH growth and particle inception,
surface growth via hydrogen-abstraction-carbon-addition (HACA) surface chemistry and via PAH condensation, surface oxidation, particle coagulation and fragmentation, gas phase scrubbing, soot particle diffusion, and radiation [4,32]. Initially, it utilized the gas-phase and surface growth mechanisms of Appel et al. [5], which are primarily based on the HACA mechanism; however, in a previous work, the gas phase mechanism was replaced with that developed by Slavinskaya and coworkers [10,33,63]. This mechanism includes numerous additional reaction pathways for PAH formation, updated reaction rates, and includes an expansion of the HACA mechanism to include abstraction and growth by additional species.

As discussed in the previous chapter, in a recent work [13], the authors validated this detailed model for high pressure use against the elevated-pressure ethane-air flames from 2 to 15 atm experimentally studied in [11,12]. While the experimentally observed trends were reproduced accurately by the model, there were some areas of potential improvement. Peak centerline soot volume fraction values were within the correct order-of-magnitude; however, they were under-predicted by a factor of 2 to 3. Additionally, it was observed, both experimentally and numerically, that as pressure increased, the flame became increasingly anchored to the fuel tube, leading to excessive pre-heating of the air and fuel streams. The model as originally employed could not accurately account for this effect, as the computational domain began at the exit plane of the fuel tube.

3.1.3 Agenda

In the present study, the computational domain of the previously utilized highly detailed model is extended below the exit plane of the fuel tube for a set of elevated-pressure coflow laminar diffusion flames. Additionally, unlike the earlier investigations in [31] and [29], CHT between the fuel and air streams and the fuel tube is modelled to avoid having to prescribe an arbitrary temperature at the fluid/solid interface. CHT is implemented using the harmonic mean method, which was originally prescribed by Patankar in [49], and successfully used in previous studies involving CHT [64,65]. This method was chosen due to the relative ease with which it could be incorporated into the existing model, which was demonstrated in [31]. The effect that these two modifications have on the numerical predictions for soot volume fraction is investigated.

3.2 Problem formulation

In this investigation, two models are compared; one without the inclusion of CHT and previously utilized in [13] and documented in the previous chapter, and one with the inclusion of CHT, which is presented in this study. The model without CHT shall herein be referred to as the Non-CHT Model, and the
model with CHT as the CHT Model.

3.2.1 Burner and flame description

The flames chosen for this investigation are the steady, non-smoking, coflow laminar ethane/air diffusion flames at pressures of 2 to 15 atm, studied experimentally by Mandatori and Gülder [11, 12]. These are the same flames that were studied previously in [13] with the Non-CHT Model, and presented in Chapter 2 of this thesis.

![Schematic representation of the burner and flame, with coordinate axes and computational domain boundaries for the Non-CHT and CHT Models (image is not drawn to scale.)](image)

A generic schematic of a co-flow burner, flame, and computational domain for the Non-CHT Model and CHT Model is depicted in Fig. 3.1. There are two regions of interest for soot formation in the flame, those being the wing region (depicted on the schematic) and the centerline region, which runs along the central axis of the flame. The computational domain for the CHT Model begins below the fuel tube exit plane at a location where temperature gradients are negligible, thus allowing for any pre-heating from the flame to the fluid streams to be captured. The fuel (ethane) enters the flame domain through the center tube, which has an inner diameter of 0.229 cm. The tube has a taper on its inner and outer surfaces. Air enters the flame domain through a co-flowing annular region, which has an inner diameter of 2.54
cm. The inlet temperature for both the air and fuel streams is set to 300 K for the Non-CHT Model and the CHT Model. During the previous study utilizing the Non-CHT Model, the assumed 300 K inlet temperature was shown to be erroneous due to the strong downward conduction of heat from the flame to the burner [13].
3.2.2 Governing Equations and Boundary Conditions

The governing equations and boundary conditions utilized are similar to those presented in the previous chapter. Due to the utilization of the harmonic mean method, no modifications to the governing equations are necessary; however, to ensure no convection of diffusion of species or soot particles across the fluid-solid interface, zero-gradient conditions are enforced for the fuel tube surface as follows.

\[
\frac{\partial Y_k}{\partial r} = 0, \frac{\partial N_i^p}{\partial r} = 0, \frac{\partial N_i^a}{\partial r} = 0
\]

3.3 Numerical method

3.3.1 Discretization techniques

As in previous works [6, 7, 10, 46], the finite volume method is used to discretize the governing equations. A staggered mesh is used with a semi-implicit scheme to handle the pressure and velocity coupling and to solve the discretized equations at each discrete time interval. The diffusive terms are discretized using a second-order central difference scheme while the convective terms are discretized using a power law scheme [49]. The thermal properties of the gaseous species and chemical reaction rates are obtained by using CHEMKIN subroutines [50, 51]. Based on sensitivity studies performed in [66], transport properties which include mixture-averaged quantities for viscosities, conductivities, and diffusion coefficients, as well as thermal diffusion coefficients for H and H\textsubscript{2}, are evaluated using TPLIB [52, 53].

The harmonic mean method works by setting a very large value for viscosity for the control volumes in the fuel tube region when solving the momentum equations, which allows a zero velocity inlet boundary condition to propagate and prevail in this region, consistent with it being a solid. To model the heat transfer in the fuel tube, values of specific heat capacity and conductivity for steel are assigned for the fuel tube region when solving the energy equation.

3.3.2 Computational domain

The computation domain used for the CHT Model in this investigation extends 3.45 cm in the axial direction and 1.18 cm in the radial direction, and is divided into 416 (z) \times 172 (r) control volumes. This grid has approximately twice the resolution in both the axial and radial directions when compared to the Non-CHT Model to allow for adequate resolution of the gradients near the fuel tube fluid-solid interface. A non-uniform mesh is used to save computational cost while still resolving large spatial gradients. The
first 0.5 cm in the $z$ direction are below the fuel tube exit plane. This distance is sufficient to ensure the temperature gradients at $z = 0.0$ (the bottom of the computational domain) are negligible. The mesh is initially fairly coarse in the axial direction in this region as gradients are low. The mesh progressively becomes finer, until at $z = 0.3$ it reaches its finest spacing. The grid is finest in the flame region with maximum resolutions of $4.3\times10^{-3}$ cm between $z = 0.3$ cm and $z = 1.6$ cm in the axial direction, and $2.5\times10^{-3}$ cm between $r = 0.0$ cm and $r = 0.30$ cm in the radial direction. Grid stretching is employed above $z = 1.6$ cm and beyond $r = 0.30$ cm utilizing a constant stretching factor. Fig. 3.2 displays the computational domain along with a representative temperature contour, with the black region of the contour being the fuel tube, and the utilized mesh.

![Figure 3.2: Computational domain with representative temperature contour and associated mesh utilized for the CHT Model. The black region of the contour represents the fuel tube.](image)

### 3.3.3 Solution method

Pseudo-transient continuation is used to aid convergence from an arbitrary starting estimate [9]. At each pseudo-time step, after the momentum and pressure correction equations are solved globally [49] by the tri-diagonal matrix algorithm (TDMA) for penta-diagonal matrices, the gaseous species equations are solved in a coupled manner at each control volume to effectively deal with the stiffness of the system and speedup the convergence process (i.e., at a given time step, a linear system coupling each of the species mass conservation equations is formed and solved using the Gauss elimination method at each control volume). After iteration of the species equations, the sectional transport equations are also solved simultaneously in a similar fashion. The energy equation is then solved using TDMA to generate an updated global temperature field and a convergence check is performed. As the solution increases
in its degree of convergence, the time-step used in the pseudo-time step process is increased. Initially, simulations are started at a time-step of $10^{-6}$, up to a maximal amount, of approximately $5\times10^{-5}$, that allows for the solution to proceed without divergence. Finally, if the vector norms of the independent variables change by less than one one-thousandth of their value from one time step to the next, then the algorithm is deemed to have converged. Otherwise time stepping proceeds, again starting with solution of the momentum and pressure correction equations. Care was taken to ensure that the computed solution was independent of the convergence tolerance used, by lowering the convergence tolerance by an order of magnitude and checking for changes in the solution.

Due to the computational intensity of the problem, solutions would be intractable with serial processing. Therefore, distributed-memory parallelization with strip-domain decomposition is employed as described in [67]. The computational domain is divided uniformly into $N_{PROC}$ subdomains with the boundaries of each subdomain perpendicular to the $z$-axis, where $N_{PROC}$ is equal to the number of grid lines in the axial direction. The algorithm uses the Message Passing Interface [54] library to distribute the workload. The computations are performed on the General Purpose Cluster (GPC) of SciNet, on twenty-six 16-core Intel Xeon E5540s with 2.53 GHz chip speeds and InfiniBand network interconnect.

3.4 Results and discussion

3.4.1 Computational Cost

Fig. 3.3 shows a comparison of the computational cost in CPU-hrs for the Non-CHT and CHT Model for the 2, 5, 10, and 15 atm flames. Please note the CPU-Hrs for the Non-CHT Model have been multiplied by a factor of 2 in Fig. 3.3. The CHT Model displays significant increases in computational cost over the Non-CHT Model for all simulated flames. The factor of increase in computational cost reduces with increasing pressure, with cost increasing by a factor of 6.5 for the 2 atm flame and 5.0 for the 15 atm flame. The mesh used for the CHT Model has approximately 4 times as many grid points as the Non-CHT Model. Additionally, the meshes for the two models have not been completely optimized as much as possible, thus it is difficult to compare the computational costs directly. However, the reduction in the factor of increase in computational cost with increasing pressure does display that the CHT Model actually has less convergence difficulties relative to the Non-CHT Model with increasing pressure. Additionally, it is observed that while there was a significant increase in the computational cost for the Non-CHT model when progressing from 10 to 15 atm, the CHT Model displays a very modest increase in computational cost. For the CHT Model at 15 atm, accounting for factors such as using a
shared computing system, 10 days are needed to obtain a solution using 416 processes.

Figure 3.3: Comparison of the computational cost of the Non-CHT Model and CHT Model for simulating ethane-air co-flow diffusion flames at 2, 5, 10, and 15 atm. The CPU-Hrs for the Non-CHT Model have been multiplied by a factor of 2.

3.4.2 Fuel Tube Exit Plane Temperature and Species

One of the conclusions mentioned from the previous chapter utilizing the Non-CHT Model was that the assumption of a 300 K inlet temperature was not accurate [13]. Fig. 3.4 displays radial profiles of temperature at the fuel tube exit plane computed by the CHT Model, compared to the 300 K assumption used with the Non-CHT Model [13]. This figure demonstrates that the 300 K inlet temperature assumption is erroneous, even at the lowest simulated pressure of 2 atm. Additionally, while the peak temperature at the fuel tube exit plane in the wing region is relatively insensitive to pressure, the peak temperature on the centerline increases appreciably with increasing pressure. At 2 atm, the peak centerline temperature is 531 K, while at 15 atm the peak centerline temperature is 773 K. This result demonstrates that as pressure increases, there would exist an increasingly hotter, very fuel rich region near the fuel tube exit plane on the centerline, which would enhance fuel pyrolysis.

Fig. 3.5 displays the acetylene (C$_2$H$_2$) mole fractions along the centerline at the fuel tube exit plane for the four simulated pressures. Acetylene is a product of ethane pyrolysis and an important species in soot formation chemistry. It can be seen that peak acetylene mole fractions on the centerline increase linearly with increasing pressure, confirming increases in fuel pyrolysis. This is a similar conclusion as the findings of the study in [29], regarding increasing fuel pyrolysis below the exit plane of the fuel tube with increasing pressure. This increase in fuel pyrolysis could not be captured with the Non-CHT Model, yet is important to flame and soot formation chemistry. Additionally, these results explain why
Chapter 3. The Effect of Conjugate Heat Transfer

Figure 3.4: Computed radial profiles of temperature for the CHT Model, compared with the Non-CHT Model, at the exit plane of the fuel tube for ethane-air diffusion flames at 2, 5, 10, and 15 atm.

Figure 3.5: Computed mole fraction of $\text{C}_2\text{H}_2$ in the centerline region at the exit plane of the fuel tube for the CHT Model for ethane-air diffusion flames at 2, 5, 10, and 15 atm.

the CHT Model has less convergence difficulty relative to the Non-CHT Model as pressure increases, since the 300 K boundary condition used in the Non-CHT Model becomes increasingly less physical as pressure increases.

Figure 3.6a displays the predicted peak centerline soot volume fraction for the four simulated pressures for the CHT Model and Non-CHT Model compared to the experimental data, with appropriate error bars based on the measurement techniques. Fig. 3.6b displays the same parameters for the wing region. While the Non-CHT Model accurately predicted the trends in peak centerline soot volume fraction as pressure increases, the values were under predicted by a factor of 2 to 3. With the CHT Model,

3.4.3 Soot Volume Fraction

Fig. 3.6a displays the predicted peak centerline soot volume fraction for the four simulated pressures for the CHT Model and Non-CHT Model compared to the experimental data, with appropriate error bars based on the measurement techniques. Fig. 3.6b displays the same parameters for the wing region. While the Non-CHT Model accurately predicted the trends in peak centerline soot volume fraction as pressure increases, the values were under predicted by a factor of 2 to 3. With the CHT Model,
peak centerline soot volume fraction values are within experimental error, a feat that was not previously possible for most soot formation models [10,26,46,59,68]. These noted improvements along the centerline are attributed to fuel pyrolysis that occurs within the fuel tube, which can only be captured by the CHT Model. Fig. 3.6b displays that the Non-CHT and CHT Model are both quantitatively accurate in the wing region, with minimal differences between the two models.

In addition to better agreement for the peak centerline soot volume fraction, the distribution of soot along the centerline has improved with the use of the CHT Model. Fig. 3.7a displays a comparison of the calculated centerline soot volume fraction at various heights above the burner, for the CHT and Non-CHT Models, to the experimental data for the 2 atm flame. Fig. 3.7b displays the same data for the 15 atm flame. The CHT Model predicts appreciable soot volume fractions at a lower axial height above the burner than the Non-CHT Model, which is in better agreement with the experimental data. This improvement at 2 atm is fairly minimal (Fig. 3.7a); however, it is quite substantial at 15 atm (Fig. 3.7b). This change in the predicted soot distribution is due to the increased pre-heating effect as pressure increases, which the Non-CHT Model cannot capture.

### 3.4.4 Flow Field

For the 10 atm and 15 atm flames, a flow phenomenon is observed at the exit plane of the fuel tube near the centerline region. Fig. 3.8 displays streamlines for the 10 atm flame, zoomed in near the exit of the fuel tube (black region), which show the presence of a recirculation zone. This recirculation zone is not predicted at 2 atm or 5 atm. In order to gain an understanding as to why these recirculation zones were

Figure 3.6: CHT Model and Non-CHT Model computed peak a) centerline and b) wing soot volume fractions compared with experimental data for ethane-air diffusion flames at 2, 5, 10, and 15 atm. [11,12]
Figure 3.7: CHT Model and Non-CHT Model computed centerline soot volume fraction profile compared with experimental data for ethane-air diffusion flames at a) 2 and b) 15 atm. [11,12]

Figure 3.8: Computed flow streamlines near fuel tube tip for an ethane-air diffusion flame at 10 atm.

predicted, two cold flow cases (where no flame is present and the entire domain is at 300 K), utilizing the inlet flow conditions for the 10 atm and 15 atm flame respectively, are computed utilizing the CHT Model. For the two cold flow cases, in which no flame is present, the recirculation zones do not appear. These results indicate that the recirculation zone is being predicted due to the influence of the high temperature flame on the flow field. Fig. 3.9 displays the degree to which the air and fuel streams, in the vicinity of the fuel tube, are accelerated by the high temperature flame at the four simulated pressures. This is done by taking the ratio of the free-stream (outside the boundary layer) fluid speed in the vicinity of the fuel tube tip predicted by the CHT Model with a flame present, and dividing by the free-stream fluid speed that would be predicted based on a cold flow case, which is determined utilizing the mass flow rates used for the experiment and the geometry of the burner. This velocity ratio is determined for
both the air and fuel streams. It can be seen that as pressure increases, the velocity ratios for the air and fuel streams increase. This indicates increased flow acceleration for the fuel and air streams in the vicinity of the fuel tube tip as pressure increases.

![Figure 3.9: Ratios of the CHT Model computed free-stream fluid speed to the cold flow free-stream fluid speed in the vicinity of the fuel tube tip on the air and fuel sides of the fuel tube for ethane-air diffusion flames at 2, 5, 10, and 15 atm.](image)

Given these two analyses, the following conclusion with regards to the cause of the recirculation zones is made. The recirculation zones are caused by the increasing acceleration of the fluid stream on the air side of the fuel tube as pressure increases. This acceleration is caused by the increase in temperature on the air side of the fuel tube (Fig. 3.4 displayed temperatures of approximately 1700-1800 K). This rapidly accelerating air stream causes shear stress on the fuel stream in the vicinity of the fuel tube, thus causing the fuel stream to accelerate rapidly as well. Again, this acceleration of the fuel stream increases with increasing pressure. In order for continuity to be upheld, there must be a corresponding deceleration of the fuel stream in the centerline region. Eventually, this required deceleration is sufficiently large to cause the flow to reverse, and a recirculation zone forms. Experimentally, it would be very difficult to detect the presence of these recirculation zones, as they are located in the center of the flame, partially inside the fuel tube. Having a domain that extends below the fuel tube exit plane allows the CHT Model to capture the recirculation zones, which was not possible with the Non-CHT Model.

### 3.4.5 Importance of Gradients

It has been common practice in other simulations of co-flow flames to account for flame pre-heating by simply increasing the temperature at the boundary, either arbitrarily, or to match experimental temperature data [10, 35, 46, 59]. This brings forth the idea that if accurate temperature measurements could be made at the exit plane of the fuel tube, there would be no need for the computationally expensive CHT Model, as a well-defined temperature boundary condition could be prescribed in a Non-
CHT Model. In order to test this hypothesis, the Non-CHT Model is run for the 2 atm flame; however, the inlet boundary conditions are those determined by the CHT Model at the fuel tube exit plane. Additionally, the mesh of the Non-CHT Model is modified to be consistent with that of the CHT Model. If the Non-CHT Model is able to reproduce the results of the CHT Model, then the hypothesis will have been proven positive. It should be noted that this is a highly idealized case, as it is infeasible to take fully radially resolved measurements for temperature, velocities, density, and species mass fractions experimentally. Additionally, the 2 atm flame exhibits the least amount of flame pre-heating of all flames studied.

Figure 3.10: Comparison of computed temperature contours for the CHT Model and Non-CHT Model using boundary conditions predicted by the CHT Model for an ethane-air diffusion flame at 2 atm.

Fig. 3.10 compares temperature contours for the CHT Model and the Non-CHT Model with boundary conditions provided by the CHT Model. It is quite evident that the results for the two simulations are similar, but distinct, as the flame is shorter and has lower peak temperatures with the Non-CHT Model than with the CHT Model. The reason for this is the Non-CHT Model could not account for the gradients present at the fuel tube exit plane. Even though the values for temperature, velocity, and species mass fractions are the same between the two models at the fuel tube exit plane, a constant value assigned at a boundary condition (such as in the Non-CHT Model) implies a zero gradient condition below that boundary condition. This explains why the Non-CHT Model predicts a shorter flame, as numerically it is as though the fuel and air streams have been heated to, and held constant at, 600 K and 1800 K.
respectively prior to the fuel tube exit plane, which introduces an excess amount of heat. This analysis reinforces the notion that numerically accurate inlet boundary conditions are not sufficient to produce quantitatively accurate results, and the boundary must be located away from gradients. This result demonstrates that the previously used method of increasing the inlet temperature of the fuel stream to account for flame pre-heating is far from ideal and can introduce inaccuracies into the numerical results.

It should be noted that the degree of the inaccuracies will depend greatly on the degree to which the flame is anchored to the fuel tube. A lifted flame should not cause any significant pre-heating of the fuel and air streams, and thus in these cases the benefits of the CHT Model would be minimal. The CHT Model will have the greatest impact in cases were the flame is severely anchored to the fuel tube. As pressure is increased, it becomes increasingly difficult and infeasible to experimentally achieve stable lifted flames, thus the application of the CHT Model to model high pressure flames is recommended.

3.5 Conclusions

An improved co-flow flame and soot formation model is proposed, which extends the domain below the exit plane of the fuel tube and includes conjugate heat transfer (CHT) to model the heat transfer between the fluid streams and solid fuel tube. The CHT Model more accurately predicts centerline soot formation than the Non-CHT Model, which had been a well-known challenge in the literature. This is due to the CHT Model capturing fuel pyrolysis that occurs below the fuel tube exit plane. Additionally, the CHT Model uncovers the formation of a recirculation zone in the centerline region at the exit plane of the fuel tube at elevated pressures. The recirculation is caused by the increased flow acceleration of the fluid stream on the air side of the fuel tube with increasing pressure, causing acceleration of the fuel stream due to shear stresses.

It is determined that adjusting boundary conditions of the Non-CHT Model cannot reproduce the results of the CHT Model, due to the importance of boundary gradients. It is so demonstrated that obtaining temperature measurements at the exit plane of the fuel tube to be used as boundary conditions in a model without CHT cannot accurately account for the fuel pre-heating effect. When modelling flames that are not lifted off the burner, it is important to utilize a model that extends below the fuel tube exit plane if accurate predictions are to be made.
Chapter 4

Closure

4.1 Summary and conclusions

This project has been able to successfully utilize the detailed numerical soot formation code to model elevated-pressure flames. This is the first attempt at modelling elevated-pressure co-flow diffusion flames with a detailed soot formation model, including a detailed gas phase mechanism and soot aerosol dynamics model. The five major findings of the studies presented in this thesis are summarized below:

1. The models employed by the detailed code can accurately predict the experimentally observed trends in peak soot volume fraction with increasing pressure for ethane-air co-flow diffusion flames from 2 to 15 atm in the centerline and wing regions. The numerical results for peak soot volume fraction are within the experimental uncertainty in the wing region with the base model; however, capturing the flame pre-heating effect via extending the computation domain below the fuel tube exit plane is required to get quantitatively accurate results along the centerline.

2. Pressure does not have a uniform effect on the three main mechanisms of soot formation. Pressure dramatically increases HACA surface growth and PAH condensation rates due to the positive feedback nature of these two mechanisms. It is shown that while HACA surface growth dominates at lower pressures, lower abstraction rates caused by reduced radical populations lead to an increase in the relative importance of PAH condensation as pressure rises. This effect is most prominent on the centerline, where PAH condensation becomes the dominate mechanism for pressures of 5 atm and above.

3. The effect of flame pre-heating is not trivial for elevated-pressure flames, or more generally any flame that is not sufficiently lifted off the burner. Failing to capture this pre-heating effect in-
troduces errors into the numerical results, which increase with the degree to which the flame is anchored to the burner. Attempting to account for the pre-heating effect by altering boundary conditions does not lead to highly accurate numerical results, which are only obtained by extending the computational domain below the exit plane of the fuel tube to where gradients are negligible, such as in the case of the CHT Model.

4. The computational cost of utilizing detailed numerical codes at elevated-pressures is substantial and markedly higher than simulations at atmospheric pressure. Elevated pressure simulations cause the chemical and soot dynamics equations to become very stiff, and due to the flame thickness reducing at higher pressures, gradients increase as well. This necessitates the need for grid refinement and less aggressive time-stepping as pressure increases. Including CHT within the model further aggravates these issues due to the high gradients encountered at the fluid-solid interface.

5. Co-flow flames under elevated pressures can have a recirculation zone form near the exit plane of the fuel tube along the centerline. This is caused due to the increased shear stresses and subsequent acceleration of the fuel stream near the fuel tube tip as pressure is increased. The discovery of this phenomena is made possible by the CHT Model. A model that does not extend below the fuel tube exit plane cannot capture the occurrence of the recirculation zone. Experimentally these recirculation zones would be very difficult to detect due to line of sight issues caused by the close proximity to the fuel tube exit plane.

4.2 Recommendations for future work

Soot formation is a very complex and still ill-defined process. The addition of pressure as another factor that can influence soot formation complicates things further, both experimentally and numerically. There are still many studies that need to be performed to gain a full understanding of soot formation mechanics, particularly at elevated pressures. Based on the detailed modelling studies of soot formation at elevated-pressures performed in this thesis, the following future studies are recommended:

1. Application of the CHT Model to a wide range of data sets. The CHT Model has demonstrated its ability to accurately predict soot volume fraction at elevated pressures in both the wing and centerline regions. This model should be applied to as many high pressure data sets as possible, commencing with completion of the ethane-air data set up to 33 atm. Another data set of particular interest is the methane-oxygen co-flow flame data set which has experimental results up to 100 atm [21]. In addition to data at very high pressures, this work demonstrated an inverse
relationship between pressure and soot volume fraction for pressures greater than 50 atm. No other high pressure data set currently available displays a decrease in soot volume fraction as pressure increases. It is recommended that numerical modelling is utilized and leveraged to understand and explain this phenomenon.

2. Validation of soot particle and soot aggregate parameters at elevated pressure. While the CHT Model does give quantitatively accurate results for soot volume fraction at elevated pressures, no validation has been performed of soot particle or aggregate parameters. Primary particle number density, soot aggregate number density, and primary particle diameter all require validation at elevated pressures. Currently, this is hindered by the lack of available experimental data for these parameters.

3. PAH condensation study. As pressure increases, the role that PAH condensation plays in the total mass added to a soot particle increases, and can actually become the dominant mechanism. To date, there has been no experimental, or numerical, detailed study performed on PAH condensation, at either atmospheric or elevated pressures. Due to the prediction by the detailed code of the high importance of PAH condensation at elevated pressures, studies should be performed to fully characterize the nature of PAH condensation. In association with this study, predictions of PAH concentration levels at elevated pressures should be validated with experimental data.

4. Improvement of code efficiency and scalability. As pressure increases, the ability to perform parametric studies with the detailed code in a timely manner is diminished due to the increased computational cost. Increasing the code’s parallel efficiency and scalability will allow numerical results to be obtained in a shorter time frame. Additionally, this enhanced capability would increase the feasibility of modifying the detailed code for turbulent flame simulations, as in the code’s current state, such solutions would be nearly intractable.
Bibliography


