ABSTRACT

Quantitative Characterizations of Coflow Laminar Diffusion Flames in a Normal Gravity and Microgravity Environment

Kevin T. Walsh

2000

Optical diagnostics are used to investigate a variety of laminar diffusion flames to assess the accuracy of existing flame models and to expand understanding of combustion fundamentals. This experimental flame study, which took place in both normal gravity and microgravity (µg) environments, proceeded in two phases. This first stage involved extending previous characterizations of a normal gravity flame to quantify trace species such as CH, CH*, and OH*, whose concentration and spatial distribution are sensitive tests of the detailed chemical kinetics needed to model pollutant formation. As a part of this study, reliable quantitative information was extracted from flame chemiluminescence measurements. Experimental capabilities were made compatible with existing µg facilities to extend this diagnostic technique to the microgravity environment of the KC-135 reduced-gravity aircraft, where a detailed study of flame structure was performed. After this work increased experimental expertise and understanding of microgravity diffusion flame behavior, an assortment of two-dimensional laser diagnostics were brought to the KC-135 to allow for the measurement of µg temperature, major species, and soot distributions to complement similar laboratory measurements. These experimental results are compared to the predictions of Professor Mitchell Smooke’s two-dimensional numerical model of this flame, in both 0 g and 1 g, that incorporates full transport and finite rate chemistry. The measurements performed in this work have afforded the most rigorous set of comparisons with flame computations to date, and have revealed discrepancies between measurement and computation not seen in previous laboratory studies of this flame.
Quantitative Characterizations of
Coflow Laminar Diffusion Flames in a
Normal Gravity and Microgravity Environment

A Dissertation
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of
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Doctor of Philosophy

by
Kevin T. Walsh

Dissertation Director: Professor Marshall B. Long

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TABLE OF NOMENCLATURE

\[ A_{21} \] - Einstein A coefficient for spontaneous emission
\[ A(\lambda) \] - absorption spectral profile
\[ [a] \] - particulate (or molecular) diameter
\[ [a(t)] \] - time-dependent particle size
\[ A_{\text{LIF}} \] - cross-sectional area of laser beam producing LIF signal
\[ A_{\text{Ray}} \] - cross-sectional area of laser beam producing the Rayleigh signal
\[ \alpha \] - molecular polarizability of individual molecules in a gaseous medium.
\[ \alpha_0 \] - static part of molecular polarizability
\[ b_{12} \] - rate constants for absorption
\[ b_{21} \] - are the rate constants stimulated emission
\[ c \] - speed of light in vacuum
\[ C \] - experimental calibration constant (Raman signal)
\[ d \] - apertured diameter of collection lens
\[ d_p \] - diameter of the primary particle in soot aggregate
\[ d_x \] - pixel size
\[ \Delta \lambda \] - differential wavelength element or band
\[ \Delta \nu \] - laser linewidth (FWHM)
\[ E \] - electric field of incident laser beam
\[ E(\lambda) \] - emission spectral profile
\[ E_{\text{LIF}} \] - laser energy per pulse producing LIF signal
\[ E_{\text{Ray}} \] - laser energy per pulse producing the Rayleigh signal
\[ \varepsilon \] - collection efficiency of the optics
\[ \varepsilon_0 \] - permittivity of free space
\[ f_B \] - fraction of ground state population in the state being pumped
\[ f_o \] - distance from collection lens to edge of imaged region
\[ F(r) \] - axisymmetric emissivity distribution
\[ \text{FWHM} \] - full width at half-maximum
\[ g \] - gravitational acceleration in local reference frame.
\[ g_{\text{EARTH}} \] - gravitational acceleration on Earth (9.8 m/s²).
\[ g(\nu) \] - molecular absorption lineshape
\[ \Gamma_A \] - dimensionless absorption overlap integral
\[ \Gamma_E \] - dimensionless emission overlap integral
\[ h \] - Planck’s constant
\[ \eta \] - detector efficiency
\[ I_0 \] - intensity of laser producing Raman signal
\[ I_{\nu}^0 \] - laser spectral irradiance in W/(cm²cm⁻¹)
\[ I_{\nu}(\nu) \] - spectral laser intensity profile in W/(cm²cm⁻¹)
\[ K \] - global strain rate at extinction
<table>
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<th>Definition</th>
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<td>$k_B$</td>
<td>Boltzmann’s constant</td>
</tr>
<tr>
<td>$k_i$</td>
<td>reaction rate $i$</td>
</tr>
<tr>
<td>$L$</td>
<td>burner separation in countercflow flame study</td>
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<tr>
<td>$L(\lambda)$</td>
<td>laser spectral profile</td>
</tr>
<tr>
<td>LIF</td>
<td>laser-induced fluorescence</td>
</tr>
<tr>
<td>LII</td>
<td>laser-induced incandescence</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>wavelength of incident laser light</td>
</tr>
<tr>
<td>$\lambda_{em}$</td>
<td>wavelength at the center of blackbody radiation</td>
</tr>
<tr>
<td>$M^*$</td>
<td>molecule in electronically excited state</td>
</tr>
<tr>
<td>M</td>
<td>molecule in electronic ground state</td>
</tr>
<tr>
<td>$\mu_g$</td>
<td>microgravity (reduced gravity)</td>
</tr>
<tr>
<td>$N$</td>
<td>number density of the gas flow used for calibration</td>
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<tr>
<td>$N(x,y)$</td>
<td>spatially-varying number density</td>
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<td>$N^*$</td>
<td>number density of excited-state molecules</td>
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<tr>
<td>$N_g$</td>
<td>number density of the ground state molecule of interest</td>
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<td>$N_i$</td>
<td>ground state in two level LIF model</td>
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<td>$N_e$</td>
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<td>$N_a$</td>
<td>number density of aggregates in probe volume</td>
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<td>$N_l$</td>
<td>number density of the Raman-active molecule</td>
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<td>$N_{STP}$</td>
<td>number density of gas at STP</td>
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<td>$n_i$</td>
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<td>$n_p$</td>
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<td>Nd:YAG</td>
<td>neodymium yttrium:aluminum garnet, a solid-state lasing medium</td>
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<td>$\Omega$</td>
<td>solid angle over which light is collected</td>
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<td>$\omega_0$</td>
<td>frequency of incident laser light</td>
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<tr>
<td>$\omega_v$</td>
<td>molecular vibrational frequency</td>
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<td>$p$</td>
<td>pressure (atmospheric in our work)</td>
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<td>$P$</td>
<td>dipole moment molecule</td>
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<tr>
<td>$P(a)$</td>
<td>distribution of particle sizes</td>
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<td>PAHs</td>
<td>polycyclic aromatic hydrocarbons</td>
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<td>$P_{21}$</td>
<td>rate constant for predissociation</td>
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<td>$\phi$</td>
<td>fluorescence yield [ = $A_{21}/(A_{21}+Q_{21})$ ]</td>
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<tr>
<td>$Q$</td>
<td>electronic coordinate</td>
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<td>$Q_{21}$</td>
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<td>$R(\lambda)$</td>
<td>Rayleigh spectral profile</td>
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<td>$R(x_0,y_0)$</td>
<td>triangular region over which light is collected in emission measurements</td>
</tr>
<tr>
<td>$\rho_r$</td>
<td>density of the fuel</td>
</tr>
<tr>
<td>$\rho_O$</td>
<td>density of the oxidizer</td>
</tr>
<tr>
<td>$r_i$</td>
<td>size of imaged region</td>
</tr>
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<td>$S_{LIF}$</td>
<td>measured laser-induced fluorescence signal</td>
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\[ S_{\text{LII}}(a,T) \] - the measured LII signal, which depends on temperature and particle size
\[ S_{\text{Ram},i}(x) \] - one-dimensional measured Raman signal of a species i
\[ S_{\text{Ray}} \] - measured Rayleigh signal
\[ S_{\text{Ray}}(x,y) \] - spatially-varying Rayleigh signal
\[ \sigma_i \] - collisional quenching cross section for species i with CH
\[ \sigma_{\text{Ram},i} \] - cross section of the Raman-active molecule
\[ \sigma_{\text{Ray}} \] - Rayleigh cross section
\[ T \] - temperature
\[ \tau \] - integration time, i.e. time over which signal is collected
\[ \tau_{\text{eff}} \] - effective lifetime of the excited state \[ \tau_{\text{eff}} = (A_{21} + Q_{21} + P_{21})^{-1} \]
\[ \tau_{\text{L}} \] - temporal length of the laser pulse
\[ [T(t)] \] - time-dependent temperature profile
\[ t_1 \] - time at which LII signal collection begins
\[ t_2 \] - time at which LII signal collection ends
\[ u_F \] - exit velocity of the fuel stream
\[ u_O \] - exit velocity of the oxidizer stream
\[ V \] - volume of laser probe producing Raman signal
\[ \langle v \rangle \] - thermal velocity of reduced-mass pair
\[ V_{\text{em}} \] - emission volume
\[ V_{\text{LIF}} \] - volume of laser probe producing Raman signal
\[ V_{\text{Ray}} \] - Rayleigh volume
\[ \nu \] - frequency of light, s^{-1}
\[ X_i \] - mole fraction of constituent i.
Chapter 1.
Introduction

Combustion is a central element in many critical technologies used by modern society in areas such as transportation, heating, and manufacturing. Most importantly, combustion accounts for approximately 85% of the world’s energy production, which makes combustion efficiency a vital economic interest. Despite their necessity, combustion processes produce troubling problems in the areas of pollution and human health, environmental effects, and climatic shifts. Although combustion has been used by mankind for centuries, there are still many areas of combustion science and technology that are not well understood. Basic combustion research seeks to produce fundamental knowledge of the interaction of fluids, chemistry, heat and mass transfer in combustion systems. This foundational knowledge can then be used to develop accurate simulations of complex combustion processes to be used one day in the design of practical combustion devices, such as aeroengines and power plants, with optimal efficiency and minimal pollutant production. As an integral part of combustion research, experiments are required to validate the predictions of these computational models and guide their development.

Laser diagnostic techniques, such as laser-induced fluorescence, Rayleigh and Raman scattering, and laser-induced incandescence have been widely used to characterize combustion systems. These techniques can provide advantages such as species selectivity, high spatial and temporal resolution, and two-dimensional measurement which are not available with traditional, intrusive combustion measurement tools such as sampling probes and thermocouples. Laser diagnostics have an important role in verifying computational models [Xu 1993, Smooke 1996, Tanoff 1996] as well as in the
This thesis work is primarily experimental in nature, and all aspects of the flame computations performed within this study were done by Professor Mitchell Smooke at Yale University. In this dissertation, laminar methane/air diffusion flames are examined as the state-of-the-art combustion system which can currently be modeled with full transport and detailed finite rate chemistry. A variety of optical diagnostics are applied to these flames to assess the validity of this combustion model. This research pushes current knowledge of the fundamental chemical and fluid dynamic interactions of combustion to better understand flame stability, pollutant formation, and soot production.

As a part of this investigation, these flames are studied in a microgravity (μg) environment, in which gravitational forces are largely absent (g ≤ 1% g\textsubscript{EARTH}). Since combustion involves the production of high temperature gases whose low density results in buoyant motion, flame structure, temperature, and soot production are influenced by the presence of gravity. Accordingly, μg flame measurements and computations allow gravity to be considered as an independent variable to more fully characterize the performance of this computational model.

In Chapter 2, the optical diagnostics utilized in this work such as flame emission tomography, Rayleigh and Raman scattering, laser-induced fluorescence, and laser-induced incandescence are discussed. A simple two-level model of laser-induced fluorescence is described to provide the physics background necessary to interpret the CH concentration measurements made in Chapter 3. Flame emission and tomographic reconstruction, used extensively to determine radical concentrations as well as flame structure in Chapters 3 and 4, are illustrated also. Rayleigh scattering, used for
temperature measurements (Chapter 5) as well as calibration (Chapters 3 and 4), is reviewed along with Raman scattering, which is used for species concentration measurements. Finally, laser-induced incandescence is surveyed in terms of fundamental physics as well as practical measurement issues applicable to diffusion flame soot measurements, which are performed in Chapter 5.

Chapter 3 begins with an experimental and computational study of the CH radical in a normal gravity, laminar methane/air diffusion flame. The computational predictions of two chemical kinetic schemes are compared with the results of laser-induced fluorescence measurements to evaluate existing flame models. The second half of this chapter discusses a fundamental study of the excited-state CH (denoted CH*) and excited-state OH (denoted OH*) radicals in this flame. In this study, flame emission tomography is developed as a viable diagnostic technique through a thorough investigation of experimental issues, data analysis and interpretation, and the evaluation of chemical kinetic models.

The research of Chapter 3 indicated that quantitative number density measurements of CH* and OH* could be extended to a microgravity environment, where these radicals can serve as sensitive markers of flame structure. Accordingly, these normal gravity flame characterizations paved the way for a microgravity (µg) diffusion flame study, described in Chapter 4. This µg flame study required improvements in the computational model and modifications to the experimental approach to provide a comparison of measured and predicted flame properties for a variety of normal gravity and µg flames.

In Chapter 5, the µg flame structure information obtained in Chapter 4 is supplemented with a series of laser-based flame measurements to characterize these flames further and facilitate additional assessments of computational models.
Temperature measurements were performed to allow additional comparisons between measurement and computation. Fuel/air mixing in the flame anchoring region was studied with measurements of Raman scattering from methane and oxygen. Furthermore, two-dimensional laser-induced incandescence measurements were made to quantify changes in relative soot concentration and distribution when the influence of gravity is removed. The microgravity combustion studies performed in Chapters 4 and 5 have provided the most extensive tests of our combustion models to date.
Chapter 2.
Non-intrusive Combustion Diagnostic Techniques

2.1 Introduction

Combustion processes, such as those in the lifted, laminar flames investigated in this work, are sensitively stabilized and are therefore easily altered by the physical intrusion of measurement tools such as thermocouples and sampling probes [McEnaly 1999]. All measurements made in this work are multi-point and non-intrusive in nature, and require careful experimental design, image processing, and interpretation for meaningful results. Diagnostics divide between non-laser-based (flame chemiluminescence tomography) and laser-based techniques, which include Rayleigh scattering, Raman scattering, laser-induced fluorescence (LIF) and laser-induced incandescence (LII) [Eckbreth 1988].

Chemiluminescence (alternatively flame emission) tomography is used to measure the number density of light-emitting, chemically-excited flame radicals. The remaining diagnostics involve light scattering. The elastic scattering of light from molecules is termed Rayleigh scattering. Here the probed molecules do not change their energy state, the scattered light is not spectrally specific to any particular species, and the amount of light scattered is proportional to the total number density of scatterers. Another mechanism, called Raman scattering, is a weak inelastic process in which the scattered light is shifted from the laser wavelength by an amount determined by the vibrational or rotational modes of the probed molecule. Molecules with known energy states produce predictable wavelength shifts, which can allow for species-specific measurements. When spectrally-resolved Raman measurements are performed, it is often possible to monitor multiple species simultaneously using a single laser source. Laser-induced fluorescence (LIF) is a resonant process where the laser is tuned to an allowed electronic transition for
the molecule of interest. This resonance makes LIF species-specific, and highly sensitive since absorption cross sections increase drastically for resonant processes. In laser-induced incandescence (LII), the laser light heats up soot, which then black-body radiates. Provided certain conditions are met, the resultant signal is proportional to soot volume fraction. In the sections that follow, the basic theories for flame emission tomography, Rayleigh and Raman scattering, LIF, and LII are presented.

2.2 Chemiluminescence Tomography

Chemiluminescence is the emission of a photon by an atom or molecule which is chemically produced in an electronically-excited state. These reactions involve a molecule starting in this excited state $M^*$ giving up a photon and returning to ground state $M$, or

$$M^* \rightarrow M + h\nu.$$  \hfill (2.1)

The wavelength of the emitted photon is a signature of the molecule $M$, and the molecule-dependent rate at which this reaction occurs is called the spontaneous emission rate, described by the Einstein A coefficient. This rate constant is of order $10^6 \text{ s}^{-1}$ for CH* and OH*, the emitting flame species studied in this work.

The measured emission signal is given by [Hertz 1988]

$$S_{\text{em}} = \frac{1}{4\pi} A_{21} \tau V_{\text{em}} N^* \Omega \varepsilon \eta$$  \hfill (2.2)

where $A_{21}$ is the Einstein A coefficient (s$^{-1}$), $\tau$ is the integration time (s), $V_{\text{em}}$ is the emission volume (cm$^3$), $N^*$ is the number density of excited-state molecules (cm$^{-3}$), $\Omega$ is the solid angle, $\varepsilon$ is a calibration constant of the optics, and $\eta$ is the detector efficiency in
counts per photon. Note that since the chemically-excited radicals being measured are in steady-state, no correction for collisional quenching is required [Luque 2000].

The chemiluminescence image is acquired over the entire depth-of-field along a line-of-sight, so tomographic techniques are needed to reconstruct the three-dimensional quantity under investigation [Santoro 1981]. In particular, an Abel inversion can be used to reconstruct a cylindrically symmetric distribution from line-of-sight intensity measurements at a single angle [Hughey 1982]. Since the flame under investigation is axisymmetric, an Abel inversion seems appropriate. Strictly speaking, however, an Abel deconvolution is valid only when the measured signal is collected along infinitely thin, perfectly parallel rays [Dasch 1994]. This situation can only be approximated in a realizable chemiluminescence imaging system where non-parallel rays are collected in a cone, over a non-zero solid angle. These practical issues, which are shown to affect measurement accuracy but not the soundness of the theory, are examined in detail for the optical collection geometry utilized in an experiment described in Chapter 3.

A value must be determined for the calibration constants ($\Omega$, $\epsilon$, and $\eta$) to extract the excited-state species number density, $N^*$, from equation (2.2). Researchers have demonstrated that Rayleigh scattering on a gas of known temperature, pressure, and composition can be used to calibrate the chemiluminescence detection system [Luque 1996a]. After this calibration, a spectral throughput correction is performed and the number density of the chemically-excited flame radical is obtained, as described in detail in Ch. 3.
2.3 Rayleigh and Raman Scattering

Both Rayleigh and Raman scattering arise from the molecular polarizability, \( \alpha \), of individual molecules in a gaseous medium. This polarizability relates the induced dipole moment of the molecule (\( \vec{p} \)) with the incident electric field (\( \vec{E} \)) as

\[
\vec{p} = \varepsilon_0 \alpha \vec{E}
\]

where \( \varepsilon_0 \) is a proportionality constant known as the permittivity of free space. The polarizability is assumed to contain a static part (\( \alpha_0 \)) and a polarizability derivative with respect to some electronic coordinate \( Q \), which is to say

\[
\alpha = \alpha_0 + \left( \frac{\partial \alpha}{\partial Q} \right)_0 Q
\]

in which the zero subscript denotes the value of the derivative at the equilibrium position.

Assuming that the electric field of the incident light (at frequency \( \omega_0 \)) can be written as

\[
\vec{E} = \vec{E}_0 \cos(\omega_0 t)
\]

and that \( Q \) oscillates about an equilibrium position at the vibrational frequency of the molecule (at frequency \( \omega_v \)) we can further write

\[
Q = Q_0 \cos(\omega_v t).
\]

When equations (2.5) and (2.6) are substituted into (2.4), we have

\[
\vec{p} = \left[ \alpha_0 + \left( \frac{\partial \alpha}{\partial Q} \right)_0 Q_0 \cos(\omega_v t) \right] \varepsilon_0 \vec{E}_0 \cos(\omega_0 t).
\]

This expression is multiplied out and an identity involving products of cosines is used to obtain

\[
\vec{p} = \alpha_0 \varepsilon_0 \vec{E}_0 \cos(\omega_0 t) + \left( \frac{\partial \alpha}{\partial Q} \right)_0 \frac{\varepsilon_0 \vec{E}_0 Q_0}{2} \left[ \cos(\omega_0 - \omega_v) t + \cos(\omega_0 + \omega_v) t \right]
\]
The first term describes scattered radiation at the laser wavelength, which is the Rayleigh process, while the second and third terms show scattered light at a wavelength shifted from the incident by an internal frequency of the probed medium, namely Raman scattering [Eckbreth 1988]. Rayleigh scattering will be focused upon first.

Rayleigh scattering is a result of a particle / light interaction where the particle diameter [a] is much smaller than $\lambda$, the wavelength of the incident light -- this implies that $\hat{E}$ is uniform across the particle. The scattered signal is proportional to the total number density in the probe volume and not spectrally specific to any given species. Rayleigh diagnostics are susceptible to interferences from other sources of elastic scattering, such as from particulates and spurious scattering off of surfaces [Gerry 1966, Bohren 1983]. If these interferences can be avoided, Rayleigh scattering has a high signal strength that can be used as a powerful combustion diagnostic [Muller-Dethlefs 1979, Dyer 1979, Dibble 1981, Zhao 1993].

Molecules of different gasses have different electronic structure, which determines molecular polarizability. This is indicated in the Rayleigh cross section ($\sigma_{Ray}$) of a given pure gas $i$, which at a 90° viewing angle can be written as

$$\sigma_{Ray} = \frac{4\pi^2(n_i - 1)^2}{N_{STP}^2 \lambda^4}$$  \hspace{1cm} (2.9)$$

where $n_i$ and $N_{STP}$ are the refractive index and number density of the gas $i$ at STP. Note that the index of refraction is related to molecular polarizability. The relative Rayleigh cross sections, written such that $\sigma_{Ray, N_2}=1$, of the major gasses found in our methane/air flames are shown in Table 2.1. The Rayleigh cross section of a gas mixture can be simply computed as

$$\sigma_{Ray, \text{mix}} = \sum_i X_i \sigma_{Ray, i}$$  \hspace{1cm} (2.10)$$
<table>
<thead>
<tr>
<th>Species</th>
<th>Relative Rayleigh Cross Section</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO₂</td>
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</tr>
<tr>
<td>O₂</td>
<td>0.86</td>
</tr>
<tr>
<td>CO</td>
<td>1.33</td>
</tr>
<tr>
<td>N₂</td>
<td>1.00</td>
</tr>
<tr>
<td>CH₄</td>
<td>2.27</td>
</tr>
<tr>
<td>H₂O</td>
<td>0.75</td>
</tr>
<tr>
<td>H₂</td>
<td>0.22</td>
</tr>
</tbody>
</table>

Table 2.1. Rayleigh cross sections of major flame species, given relative to nitrogen [Long, 1993].

where \( X_i \) is the mole fraction of constituent \( i \). Combining equation (2.10) and the information contained in Table 2.1, we see that the Rayleigh cross section can vary by more than a factor of two between the ambient air and unburned fuel regions in a flame.

In this work, Rayleigh scattering is used in two ways. When the elastically scattered signal is measured from a gas of known temperature, pressure, and composition, the Rayleigh signal can be used to calibrate the optical system, such as for emission and LIF measurements. For such calibration measurements, the Rayleigh signal \( S_{\text{Ray}} \) is most clearly written as

\[
S_{\text{Ray}} = \frac{N E_{\text{Ray}} V_{\text{Ray}} \sigma_{\text{Ray}}}{A_{\text{Ray}} h \nu \Omega \eta} \tag{2.11}
\]

where \( N \) is the number density of the gas flow used for calibration, \( E_{\text{Ray}} \) is the laser energy per pulse producing the Rayleigh signal, \( V_{\text{Ray}} \) is the Rayleigh volume, \( \sigma_{\text{Ray}} \) is the
Rayleigh cross section, \( h \nu \) the laser photon energy, and \( A_{Ray} \) the cross-sectional area of the laser beam. Since all of these factors are known, the calibration product \( (\Omega \varepsilon \eta) \) can be quantified, which can allow measured emission and LIF signals to be related to an absolute light level [Luque 1996a].

Rayleigh scattering can also be used to extract temperature information from measurements made in flames, even if the precise local gas mixture is unknown. In these flame measurements, the spatially-varying measured Rayleigh signal, \( S_{Ray}(x,y) \) is directly proportional to the number density, \( N(x,y) \), and Rayleigh cross section of the gas mixture \( \sigma_{Ray}(x,y) \), allowing us to write

\[
S_{Ray}(x,y) = c_0 N(x,y)\sigma_{Ray}(x,y)
\]  \hspace{1cm} (2.12)

where \( c_0 \) is a constant of proportionality that can be obtained explicitly by comparing equation (2.12) with equation (2.11). Since our measurements are made at atmospheric pressure \([p]\), we can use the ideal gas law to relate number density and temperature \((N=p/(k_B T))\) to obtain

\[
S_{Ray}(x,y) = k_0 \frac{\sigma_{Ray}(x,y)}{T(x,y)}
\]  \hspace{1cm} (2.13)

where \( k_0 \) is a constant determined from the Rayleigh signal in the ambient air region of the image, where \( T=300 \) K and \( \sigma_{Ray} \) is known. The acquired Rayleigh images are corrected for spurious laser scattering, detector response and vignetting, and laser beam non-uniformities. With the Rayleigh cross section initially assumed to be spatially constant, an initial temperature distribution is obtained by inverting this corrected Rayleigh image. An estimate of the variation of \( \sigma_{Ray} \) with temperature is then obtained from flame calculations, which is iteratively used to obtain a self-consistent temperature field. This process, described in detail in Chapter 5, results in a measured two-
dimensional temperature field that can be compared with computation. Note that the presence of sufficient quantities of soot may interfere with Rayleigh measurements and complicate the extraction of temperature information from the sooty portion of the flame [Smyth 1985].

The second and third terms of equation (2.8) describe spontaneous Raman scattering, which is spectrally shifted from the laser wavelength by an amount indicative of the energy levels of the probed molecule [Long 1977]. Radiation downshifted in frequency is termed Stokes, while the upshifted term is called anti-Stokes. While providing the advantage of species-specific measurements, Raman signals are extremely weak, with cross sections roughly three orders of magnitude lower than those for Rayleigh scattering. In this work, the probe laser was focused into a line to obtain maximum signal strength. The one-dimensional measured Raman signal of a species i, $S_{\text{Ram},i}(x)$, can be written simply as

$$S_{\text{Ram},i}(x) = CVI_0N_i(x)\sigma_{\text{Ram},i}$$

(2.14)

where C is an experimental calibration constant, V is the laser probe volume, $I_0$ the laser intensity, and $N_i$ and $\sigma_{\text{Ram},i}$ are the number density and cross section of the Raman-active molecule. Typically, the Raman signal imaged in a flame is divided by the Raman signal measured in the appropriate calibration gas. For the given species i, a full field of the gas at room temperature and atmospheric pressure would be used as a reference condition. This reference measurement allows us to write

$$\frac{[S_{\text{Ram},i}(x)]_{\text{Flame}}}{[S_{\text{Ram},i}]_{\text{Ref}}} = \frac{[N_i(x)]_{\text{Flame}}\sigma_{\text{Ram},i}[T_{\text{Flame}}(x)]}{[N_i(x)]_{\text{Ref}}\sigma_{\text{Ram},i}[T_{\text{Ref}}]} \frac{I_0,\text{Flame}}{I_0,\text{Ref}}$$

(2.15)

which can be simplified to
\[
\left[ N_i(x) \right]_{\text{Flame}} = \left[ N_i \right]_{0,\text{Ref}} \frac{I_{0,\text{Ref}}}{I_{0,\text{Flame}}} \frac{[S_{\text{Ram},i}(x)]_{\text{Flame}}}{[S_{\text{Ram},i}]} \frac{\sigma_{\text{Ram},i}[T_{\text{Ref}}]}{\sigma_{\text{Ram},i}[T_{\text{Flame}}(x)]}
\]

(2.16)

Note that the above equation indicates that the temperature variation of the Raman cross section must be taken into account. Raman spectra tend to shift as temperature increases and higher rotational levels become populated. The variation of \( \sigma_{\text{Ram}}(T_{\text{Ref}})/\sigma_{\text{Ram}}(T_{\text{Flame}}) \) in a given experiment depends on the spectral window of detection, the range of temperatures, and the energy level configuration of the particular species being measured.

Raman scattering has been widely utilized as a diagnostic in clean flames [Lapp 1974, 1976, Eckbreth 1981], in keeping with the assumption of the preceding discussion -- that no interferences exist within the spectral detection window. In practice, Raman measurements in many hydrocarbon flames result in fluorescence interferences from species such as \( \text{C}_2 \) and polycyclic aromatic hydrocarbons (PAHs), which are soot precursors. These interferences can present problems in interpreting measured signals [Aeschlimann 1975, Beretta 1985, Tanoff 1996]. This issue will be discussed in relation to the Raman measurements performed in Chapter 5.

2.4. Laser-Induced Fluorescence (LIF)

In LIF, the laser is tuned to a specific electronic transition of an atom or molecule, which results in a large scattering cross section and a high degree of species selectivity. For this reason, LIF is often used to detect combustion radicals which are important in pollutant formation but exist in small concentrations (< 0.1% mole fraction) [Crosley 1980]. Thirty or more combustion intermediates have been detected by LIF, many in flames [Crosley 1983]. A simplified, conceptual view of laser-induced fluorescence can
be seen in three steps. The first is an absorption of a laser photon, which excites an
electron from the ground state (state 1) to the excited state (state 2). There is then a finite
period of time before the molecule is de-excited and returns to the ground state. Some
fraction of these return transitions are the result of spontaneous emission, which provides
a detectable photon. This view is overly simplified -- states 1 and 2 are manifolds that
contain rotational and vibrational sub-states, so a spontaneously emitted photon may not
be at the laser wavelength.

This two-level model provides a mathematical description of LIF. In a system where
excited state 2 emits to ground state 1 and the emission is integrated over all time, the
fluorescence signal \( S_{\text{LIF}} \) is given by [Luque 1996a]

\[
S_{\text{LIF}} = \frac{1}{4\pi} N_2 A_{21} \tau_{\text{eff}} \mathcal{V}_{\text{LIF}} \Omega \epsilon \eta
\]

(2.17)

where \( N_2 \) is the number density of molecules excited to level 2 by the laser pulse
\((\text{cm}^{-3})\), \( A_{21} \) is the Einstein A coefficient \((\text{s}^{-1})\), and \( \tau_{\text{eff}} \) is the effective lifetime of the excited
state \((\text{s})\). This lifetime includes the effects of spontaneous emission \((A_{21})\), collisional
quenching \((Q_{21})\), and predissociation \((P_{21})\), i.e. \( \tau_{\text{eff}} = (A_{21} + Q_{21} + P_{21})^{-1} \) [Crosley 1981].
Note that in this work, knowledge of major species concentrations and temperatures is
required to compute the collisional quenching rate. \( \mathcal{V}_{\text{LIF}} \) is the volume probed \((\text{cm}^3)\) and
the remaining factors \((\Omega \epsilon \eta)\) are the calibration constants discussed earlier.

We now need a way to relate \( N_2 \) to the number density of the molecule of interest
before laser excitation occurred, \( N_0 \). The time dependence of the population in the
excited state can be written by the differential equation [Seitzman 1993]

\[
\frac{dN_2}{dt} = b_{12} N_1 - (A_{21} + Q_{21} + b_{21}) N_2.
\]

(2.18)
Here $b_{12}$ and $b_{21}$ are the rate constants for absorption and stimulated emission, respectively, which are related to the Einstein B coefficients for absorption or stimulated emission by

$$b = \frac{B}{c} \int I_{\nu}(\nu) g(\nu) d\nu$$  \hspace{1cm} (2.19)

where $I_{\nu}(\nu)$ is the spectral laser intensity profile and $g(\nu)$ is the molecular absorption lineshape. Accordingly, the laser lineshape can be written as [Partridge 1995]

$$I_{\nu}(\nu) = I_0^0 L(\nu)$$  \hspace{1cm} (2.20)

with the laser lineshape function $L(\nu)$ is normalized such that

$$\int_{-\infty}^{\infty} L(\nu) d\nu = \Delta \nu$$  \hspace{1cm} (2.21)

where $\Delta \nu$ is the laser linewidth (FWHM). Equation (2.19) can therefore be written as

$$b = \frac{B}{c} I_0^0 \int_{-\infty}^{\infty} J L(\nu) g(\nu) d\nu = \frac{B}{c} I_0^0 \Gamma_A$$  \hspace{1cm} (2.22)

where $\Gamma_A$ is a dimensionless absorption overlap integral whose magnitude varies between 0 and 1. If the spectral features of the absorption profile are of similar width to the laser beam profile, $\Gamma_A$ can be considerably different from 1, as is the case in our study of the CH radical in an atmospheric pressure flame. Equation (2.22) can be expressed in experimentally measurable parameters by writing out $I_0^0$ explicitly as

$$I_0^0 = \frac{E_{LIF}}{A_{LIF} \tau_L \Delta \nu}$$  \hspace{1cm} (2.23)

where $E_{LIF}$ is the laser energy per pulse, $A_{LIF}$ is the cross-sectional laser beam area, and $\tau_L$ is the temporal length of the laser pulse.

We now return to equation (2.18) and consider it in the limit of a low-intensity, short duration laser pulse of length $\tau_L$. In this limit, $N_1 > N_2$ and the depopulation term may
be neglected, and the population excited into level 2 during the laser pulse is given by direct integration to be

\[ N_2 = b_{12} N_1 \tau_L \]  

(2.24)

Equations (2.22), (2.23), and (2.24) can be utilized in equation (2.17) to obtain

\[ S_{\text{LIF}} = \frac{1}{4\pi} N_1 \frac{A_{21}}{A_{21} + Q_{21}} \frac{E_{\text{LIF}}}{A_{\text{LIF}} \Delta \nu} \frac{B_{12}}{c} \Gamma_A V_{\text{LIF}} \Omega \varepsilon \eta \]  

(2.25)

in a state where no predissociation occurs. \( N_1 \) is related to the total number density of the molecule of interest \( N_0 \) by the Boltzmann factor \( f_B \), which is the fraction of the total ground state population in the state being pumped. This fraction can be computed given knowledge of the temperature, the rotational and vibrational state being probed, and the spectroscopic constants of the molecule under investigation. With this we can develop a finalized formulation for the measured LIF signal as

\[ S_{\text{LIF}} = \frac{1}{4\pi} f_B N_0 \frac{A_{21}}{A_{21} + Q_{21}} \frac{E_{\text{LIF}}}{A_{\text{LIF}} \Delta \nu} \frac{B_{12}}{c} \Gamma_A V_{\text{LIF}} \Omega \varepsilon \eta \]  

(2.26)

The product \( (\Omega \varepsilon \eta) \) in this equation can be accounted for with a Rayleigh calibration shot, similar to that performed for chemiluminescence measurements. The remaining quantities in equation (3.11) can be measured or calculated to determine \( N_0 \), the number density of the trace species of interest. This process will be described in detail in Ch. 3.
2.5. Laser-Induced Incandescence (LII)

Combustion-generated carbonaceous particles, generally termed soot, have been studied in both simple flames and in practical combustion devices due to their role as a pollutant and as an agent of radiative heat transfer. The most robust non-intrusive soot measurement technique to date is known as laser-induced incandescence (LII). LII occurs when a particle absorbs energy from a laser beam, which rapidly raises the particle’s temperature to a level at which significant incandescence can take place [Mewes 1996]. The advent of LII has allowed for measurement of time-dependent soot fields, in flickering and turbulent flames as well as in practical combustion devices, that cannot be obtained with traditional probe measurements [Shaddix 1994, Case 1996, Geitlinger 1998, Wannier 1999, Zhao 1999].

The development of LII as a diagnostic technique took place in two steps. Initial LII analysis was theoretical, considering the basic physical processes involved in the heating of particles by a pulsed laser and the use of a time-dependent model for absorption, vaporization, and heat losses through conduction and radiation. This work resulted in an expression for the LII signal that was proportional to the soot volume fraction in the probed region, revealing potential applications as a diagnostic technique [Melton 1984]. The next phase of diagnostic development focussed on influence of soot properties (primary particle size, aggregate size, time-temperature histories) and the choice of measurement parameters (excitation/detection wavelength, pulse energy, gate timing) on the accuracy of soot volume fraction measurements using LII [Vander Wal 1994, 1996a, 1998a].

In considering the basic physics of soot heating and incandescence, a soot particle is idealized as an agglomeration of spherical primary particles in which the particles are not
too densely packed (i.e. fractal dimension less than two). A time-dependent model of particle heating and cooling processes was developed [Melton 1984]. The terms of this energy balance equation account for laser absorption, conductive heat transfer from particle to gaseous medium, heat expended in vaporization, thermal blackbody radiation, and heating of the soot particle. Soot volume fraction is described by three parameters: the diameter of the primary particle \( d_p \), the number of primary particles per aggregate \( n_p \), and the number density of aggregates in the volume \( N_a \). These quantities combine to describe the soot volume fraction as

\[
f_v = \frac{\pi}{6} N_a n_p d_p^3
\]  

(2.27)

The energy balance equation, containing the terms described above, is coupled with a continuity equation which describes the particle diameter decreasing with vaporization. These two equations are solved numerically to calculate the time-dependent particle size \( a(t) \) and temperature \( T(t) \) resultant from a given laser excitation (with a specified intensity/time profile and excitation wavelength). As a blackbody, the soot particle radiates isotropically into a wavelength band \( \Delta \lambda \) centered at \( \lambda_{em} \) according to the Planck blackbody formula multiplied by the available surface area. Accordingly, the time-varying emitted radiation can be written as

\[
I_{em}(t) \propto \frac{4\pi[a(t)]^2}{\exp[\hbar c / \lambda_{em} k_B T(t)] - 1} \Delta \lambda
\]  

(2.28)

The computed solutions for \( a(t) \) and \( T(t) \) can be substituted into this expression, which can then be integrated over a hypothetical detector gate time (between \( t_1 \) and \( t_2 \)) to obtain an expression for the measured LII signal \( S_{LII} \).
\[ S_{\text{LII}}(a, T) \propto \int_{t_1}^{t_2} \frac{4\pi[a(t)]^2}{\exp[\hbar c/\lambda_{em} k_B T(t)] - 1} \Delta \lambda dt. \quad (2.29) \]

When \( t_1 \) and \( t_2 \) are chosen to bracket the laser pulse, as suggested by [Melton 1984], the above integral works out to be proportional to soot volume fraction when evaluated numerically. When a distribution of particle sizes, \( P(a) \), exists, the measured signal is an integration of this distribution times the LII signal as a function of diameter \( [a] \) over the range of particle sizes in the distribution

\[ S_{\text{LII, total}} = \int_0^\infty P(a) S_{\text{LII}}(a, T) da \quad (2.30) \]

For a lognormal \( P(a) \), this more general situation does not change the result that the measured LII signal is proportional to soot volume fraction [Melton 1984].

While the theoretical calculations work out nicely, there are other issues involved in making LII measurements in diffusion flames, as was done in this work. Diffusion flames involve a wide range of soot volume fractions, but these volume fractions occur with very different number densities, aggregate sizes, particle morphologies, and temperature-time histories which may influence the LII signal in unknown ways [Vander Wal 1994]. These quantities can be affected by experimental parameters. For example, a high-power pulsed laser could change soot particle morphology, sinter particles together, or even vaporize them altogether [Vander Wal 1995, 1998b]. Furthermore, Melton indicated that longer detection wavelengths result in the most linear proportionality between the LII signal and soot volume fraction, while shorter detection wavelengths are typically used to avoid fluorescence interferences in flame measurements [Mewes 1997]. A set of experiments described in available literature identifies how to obtain the most accurate LII signal by appropriate choice of excitation and detection scheme, excitation intensity, and temporal collection.
The theoretical treatments of LII include detailed effects of soot particle size. Melton found that different sized particles reached different final temperatures before incandescence. Specifically, large particles are elevated to higher temperatures than smaller particles, which biases the LII signal toward larger particles. These models also predict a particle-size-dependent cooling rate. Since cooling rate depends on a particle’s surface-to-volume ratio, large particles will cool more slowly than smaller particles at the same temperature. This means that larger particles will contribute more to a time-integrated LII signal, further biasing the LII signal. Furthermore, shorter detection wavelengths bias the LII signal toward the particles which reach the highest temperatures, namely larger particles. Note that these particle size effects are used by some researchers to determine particle size and morphology information from time-resolved LII signals [Fillipov 1999, Konsur 1999].

These effects will influence the choice of excitation wavelength, detection band, and timing in LII measurements. The most consistent LII signal is obtained with a prompt detection gate coincident with the laser pulse, with a premium placed on minimizing laser-induced photochemical interferences. UV photons have energies comparable with molecular bond energies and are therefore more likely to produce unwanted photochemical effects. A good LII excitation wavelength is considered to be that of the Nd:YAG fundamental, 1.06 μm, where no gas-phase molecular transitions have been seen. The choice of the spectral window of detection is guided by the desire to avoid fluorescence interferences and the large-particle detection bias inherent in LII. The presence of C₂ emission occurs in bands at 473, 516, and 573 nm combines with theoretical concerns to suggest long detection wavelengths (≥ 600 nm). [Vander Wal 1996a].
The influence of laser intensity on LII signal must be examined also. Theoretical calculations predict the LII signal to increase linearly with laser intensity until the laser-heated soot particles reach the vaporization temperature of carbon, at which point the signal flattens out. This behavior has been experimentally observed [Vander Wal 1994, 1998a]. At high enough laser fluences, significant amounts of soot can become vaporized, reducing the amount of LII signal observed. Operation in the plateau or “saturation” region balances the goal of high signal with the concern of intrusive soot vaporization by the laser probe. Note that the nature of the LII vs. laser intensity curve will likely change as a function of the flame studied, signal collection time, and excitation/detection scheme. The measurement of this curve is therefore necessary to experimentally characterize the flame under investigation.

Each of these concerns will be addressed explicitly in Chapter 5 when LII measurements are made in a methane-air diffusion flame, where theoretical accuracy is balanced against the need to make single-shot measurements in a microgravity environment.
3.1 Introduction

The CH radical has been studied extensively in fundamental flame research for a number of years [Bleekrode 1965, Smooke 1992, Cattolica 1984, Allen 1985, Joklik 1986, Garland 1986, Rensberger 1988, Berg 1997]. CH is examined here as a marker of the flame front as well as for its role as a key reactant in NO$_x$ formation through the prompt NO mechanism. This has been seen explicitly in previous computations in this flame, where a sharp increase in NO was observed at the flame front where CH concentration peaked [Marran 1996]. This NO formation is primarily due to a reaction between nitrogen and CH. Since the reaction rate of this CH-N$_2$ step is not precisely known, there is an uncertainty to calculated NO concentrations which could be clarified by quantitative CH measurements. Furthermore, since CH is a short-lived trace species that exists in a narrow spatial and temperature region within a flame, its concentration and spatial distribution are sensitive tests of detailed chemical kinetic models. In the first part of this study, quantitative measurements of CH are performed and comparisons are made with the computational predictions of different kinetic schemes.

Although flame emissions were examined spectrally decades ago and CH$^*$ has been recognized as the primary source of the ubiquitous “blue light” in low soot flames [Gaydon 1974], chemiluminescence is not well understood at a fundamental level. While chemically-excited CH and OH (denoted CH$^*$ and OH$^*$) have been investigated in previous research, quantitative measurements have yet to be compared to the predictions of detailed hydrocarbon flame computations [Porter 1966, Berman 1982, Hertz 1988, Dandy 1992, Devriendt 1996, Marchese 1996, Luque 1998]. The second part of this investigation
develops flame emission tomography as a viable combustion diagnostic in our flame and uses these measurements to assess the current state of CH* and OH* kinetics. Optical emission from flames is examined in anticipation of applying these measurement techniques to the study of microgravity flames.

3.2. Burner Configuration

The flame under investigation is a lifted axisymmetric laminar diffusion flame. The burner constructed for the experimental / computational study is shown in Fig. 3.1. The burner nozzle has an inner diameter of 4 mm and an outer diameter of 4.76 mm. The nozzle is surrounded by a 50 mm diameter coflow that helps isolate the flame from the ambient air as well as lift it off the burner surface. A 1/64” cell honeycomb flow straightener, designed to produce a radially-constant (plug flow) velocity profile and improve flame symmetry, is added to the top of the burner.

Methane, a simple fuel, is chosen to allow state-of-the-art computational modeling with detailed finite rate chemistry. To reduce sooting and soot precursors, the methane is diluted with nitrogen (65% CH₄ / 35% N₂ by volume). To allow continuity in the computational boundary conditions, the coflow and fuel stream are maintained at the same exit velocity (35 cm/s) with a plug flow profile. These flow velocities are large enough to lift the flame off of the burner face and eliminate heat loss due to thermal conduction while staying within the laminar flow regime. This fuel composition and flow field produces a lifted, blue flame 3 cm in height that stabilizes approximately 6 mm above the burner surface in normal gravity.
Figure 3.1 Burner design used for the axisymmetric lifted diffusion flame.
3.3 Previous Work

This flame, detailed above, has been characterized previously both experimentally and computationally [Smooke 1991, 1992, 1996]. Experimentally, temperature and major species (CH₄, N₂, O₂, H₂O, CO, CO₂, H₂) concentrations were measured simultaneously with Rayleigh and Raman scattering [Marran 1996]. Laser-induced fluorescence (LIF) measurements were performed to measure number densities of minor species. Quantitative, linear LIF measurements were made for OH and NO and qualitative measurements of CH have been made [Smooke 1992, 1996]. Modeling work has employed different kinetics schemes, including a 26 species C₂ hydrocarbon mechanism [Smooke 1992] and GRI Mech 2.11 [Bowman 1995]. Both produced excellent agreement for temperature and major species [Marran 1996]. Computed peak concentrations for NO and OH were within 30% and 15%, respectively, of their measured values.

In the following sections, the computational model and the various kinetics schemes used to predict CH in this flame are described. The experimental configuration is then specified and the details of CH concentration measurements are presented. Finally, the experimental results are compared with predictions of two kinetic schemes.

3.4 Computational Model

The computational model of Professor Mitchell Smooke solves the full set of elliptic two-dimensional governing equations for mass, momentum, species, and energy conservation on a two-dimensional mesh [Ern 1995]. The resulting nonlinear equations are then solved on an IBM RS/6000 Model 590 computer by a combination of time integration and Newton’s method. The chemical mechanisms employed were GRI Mech 2.11[Bowman 1995] and as well as a simpler mechanism mentioned earlier [Smooke
1992], referred to in this work as both the “alternate mechanism” and the “Yale mechanism.” These flame computations result in calculated temperature fields, velocity profiles, and species concentrations.

Accurate computations of the CH radical pose a difficult numerical problem. For example, the concentration of CH can change by an order of magnitude within 0.1 mm, which requires the adaptive grid to be extremely refined in the areas of high spatial activity.

3.5. Laser-Induced Fluorescence Measurement of CH

The laser beam used in the CH LIF experiment came from a dye laser containing Coumarin 440 pumped by the third harmonic of a Nd:YAG laser, operating at 10 Hz. The dye beam was split for power measurement, attenuated, shaped into a sheet, and passed across the jet centerline. This schematic is shown in Fig. 3.2. The R(7) line in the (0,0) band of the A-X system was selected for excitation after fluorescence intensity was measured as a function of wavelength. This measured excitation scan, performed in the saturation regime, is shown in Fig. 3.3. Although all three peaks are part of the R(7) line, the central peak seen at 426.777 nm represents an overlap between lambda-doublet components. This overlap enhances fluorescence intensity and was seen at low pressure by other researchers [Luque 1996b]. The laser sheet dimensions were measured to be 5.5 mm x 300 µm. For CH concentration measurements, the measured linewidth Δν of the beam was 0.16 cm⁻¹, with energies of 1 µJ/pulse and a pulse duration was 10 ns. These parameters correspond to a spectral density of order 10⁴ W/(cm² cm⁻¹), well below the saturation value of 10⁶ W/(cm² cm⁻¹) [Bonczyk 1979]. This operation in the linear regime was experimentally confirmed as well. Imaging was done with a cooled CCD camera and a lens-coupled image intensifier, which was gated coincident with the laser pulse to minimize
Figure 3.2. CH LIF experimental setup.
Figure 3.3 CH LIF excitation scan.
flame emission interferences. CH fluorescence was isolated with a short-pass interference filter that transmitted from 400 - 440 nm. A f/2 camera lens collected the CH fluorescence, and a pair of f/1.4 camera lenses focused the light from the back end of the intensifier onto the CCD chip. The imaged pixel volume was 30 x 30 x 300 µm³. The laser was set to 426.777 nm to record on-resonance images (I_{on}) and to 426.671 nm for off-resonance images (I_{off}). A flame luminosity image was also taken with the laser turned off (I_{lum}). Typical images were integrated over 6000 laser shots. The on-resonance image contained LIF and Rayleigh scattering on top of the flame luminosity background, while the off-resonance image had only Rayleigh scattering and flame luminosity. Since the laser energy differed by a small known amount between the on- and off-resonance measurements (E_{on} and E_{off}), the final LIF images were created as follows: \[ S_{LIF} = I_{on} - I_{lum} - (I_{off} - I_{lum}) \times \frac{E_{on}}{E_{off}} \]

In order to convert this measured LIF signal into a quantitative concentration measurement, a number of calibrations and corrections must be made. As shown in Ch. 2, the LIF signal per pulse in the linear regime is given by:

\[
S_{LIF} = \frac{1}{4\pi} f_B N_0 A_{21} A_{LIF} E_{LIF} B_{12} \Gamma_A V_{LIF} \Delta v \epsilon \eta \]

where \( B_{12}/c \) is the absorption rate, \( E_{LIF} \) is the laser energy per pulse, \( \Delta v \) is the laser linewidth, \( A_{LIF} \) is the cross-sectional area of the laser beam, \( N_0 \) is the number density of ground-state CH, \( f_B \) is the fraction of the ground state in the state being pumped, \( \Gamma_A \) is a dimensionless absorption overlap integral, \( A_{21} \) is the spontaneous emission rate, \( Q_{21} \) is the
**Figure 3.4** Measured CH LIF signal.
<table>
<thead>
<tr>
<th>Quantity</th>
<th>Symbol</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boltzmann fraction</td>
<td>$f_B$</td>
<td>0.07</td>
<td>(dimensionless)</td>
</tr>
<tr>
<td>Einstein A coefficient (CH)</td>
<td>$A_{21}$</td>
<td>$1.86 \times 10^6$</td>
<td>$s^{-1}$</td>
</tr>
<tr>
<td>Quenching rate constant</td>
<td>$Q_{21}$</td>
<td>$3.26 \times 10^8$</td>
<td>$s^{-1}$</td>
</tr>
<tr>
<td>Laser energy (per pulse)</td>
<td>$E_{LIF}$</td>
<td>1</td>
<td>$\mu J$</td>
</tr>
<tr>
<td>Laser Linewidth</td>
<td>$\Delta \nu$</td>
<td>0.16</td>
<td>$cm^{-1}$</td>
</tr>
<tr>
<td>Absorption rate (CH)</td>
<td>$B_{12}/c$</td>
<td>1243.3</td>
<td>$cm^{-1}cm^2/J$</td>
</tr>
<tr>
<td>Absorption overlap</td>
<td>$\Gamma_A$</td>
<td>0.17</td>
<td>(dimensionless)</td>
</tr>
</tbody>
</table>

**Table 3.1.** Values of selected quantities in equation (3.1).

collisional quenching rate, $V_{LIF}$ is the LIF pixel volume, $\Omega$ is the solid angle over which light is collected, $\epsilon$ is the efficiency of the collection optics, and $\eta$ is the detector efficiency in counts per photon [Luque 1996a]. The quantity that we are interested in determining is $N_0$, the number density of ground-state CH. The factors in equation (3.1) must be measured directly or determined from available literature; the value of many of these quantities is shown in Table 3.1.

The quantity $A_{21}/(A_{21}+Q_{21})$ represents the fraction of excited molecules that emit a photon and is called the fluorescence yield, $\phi$. The total collisional quenching rate for the excited state is $Q_{21} = \sum c_i k_i$, where $c_i$ is the concentration of species $i$ and $k_i$ is the collisional quenching rate constant for species $i$. This equation can also be written as
\[ Q_{21} = \sum \sigma_i N_i \langle v_{\text{CH}_2} \rangle, \] where \( \sigma \) is the temperature-dependent quenching cross section, \( N \) is the collider number density, and \( \langle v \rangle \) the thermal velocity of the reduced mass pair. An accurate calculation of the collisional quenching rate, therefore, requires knowledge of temperature and major species concentrations in the flame being measured. Computed temperature and concentration profiles of the major species (CH4, N2, O2, H2O, CO, CO2, H2), which were found to agree well with measurement, were used for these calculations. Computed CH images are clipped and thresholded at 10% of peak concentration to define spatial extent of the CH distribution in these calculations. This procedure is illustrated in the alternate mechanism in Fig. 3.5. The temperature in this CH-containing region can be seen to be nearly constant and is evaluated to be 1917 K ± 3%. This temperature value and its lack of spatial variation are confirmed by the computations of GRI Mech 2.11 as well. Values for \( \sigma_i \) (which are temperature dependent) were computed from the three parameter functional form given in the literature [Tamura 1997]. The quenching cross section of each major collider (at flame temperature) and the fitting function used are shown in Table 3.2. These cross sections facilitate the calculation of quenching images for both GRI Mech 2.11 and the alternate mechanism. These results are similar in nature to the temperature profile where CH is present in that the computed quenching rate is spatially constant and is independent of choice of kinetic mechanism. The resulting fluorescence yield \( \phi \) is 1/176. In this flame, N2 was responsible for over 60% of the quenching. Unfortunately, the CH/N2 quenching cross section has not been measured above 1300 K, making the extrapolation to flame temperatures a source of uncertainty. This uncertainty is discussed by [Tamura 1997], who estimate calculated CH quenching rates to be 20% accurate.
Figure 3.5 Calculation of flame-front temperature with alternate kinetic mechanism.
Table 3.2. Functional form of quenching cross sections given by [Tamura 1997] according to $\sigma(T) = AT^B \exp[-C/T]$. Cross sections are evaluated at 1917 K.

The Boltzmann factor, $f_B$, is calculated based on the molecule probed, temperature, and the excitation/detection scheme used. Fluorescence was detected over a spectrally broad region (40 nm) to avoid possible effects of rotational energy transfer [Garland 1985]. Spectroscopic rotational and vibrational constants were taken from available texts on molecular spectra [Huber 1979]. Given our choice of the R(7) line in the (0,0) band, $J=7$ and $v=0$, which at flame temperature of 1917 K results in $f_B=0.07$. This means that 7% of the ground state population is distributed among the three peaks of the R(7) line shown in Fig. 3.3. $\Gamma_A$ is a spectral integral of the overlap between the CH absorption profile $A(\lambda)$ and the laser beam profile $L(\lambda)$. This can be written explicitly as

<table>
<thead>
<tr>
<th>Collider</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>Cross Section $(\text{Å}^2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_4$</td>
<td>52.8</td>
<td>-0.5</td>
<td>84</td>
<td>1.15</td>
</tr>
<tr>
<td>O$_2$</td>
<td>8.61E-06</td>
<td>1.64</td>
<td>-867</td>
<td>3.27</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>166</td>
<td>-0.5</td>
<td>0</td>
<td>3.79</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>8.67E-13</td>
<td>3.8</td>
<td>-854</td>
<td>4.03</td>
</tr>
<tr>
<td>CO</td>
<td>8.31</td>
<td>0</td>
<td>0</td>
<td>8.31</td>
</tr>
<tr>
<td>N$_2$</td>
<td>1.03E-09</td>
<td>2.9</td>
<td>-192</td>
<td>3.77</td>
</tr>
<tr>
<td>H$_2$</td>
<td>221</td>
<td>-0.5</td>
<td>686</td>
<td>3.53</td>
</tr>
</tbody>
</table>
The laser profile was fit to a Gaussian distribution centered at 426.777 nm with a full width at half maximum (FWHM) of $\Delta \nu = 0.16$ cm$^{-1}$, which was measured using a grating spectrometer. The beam profile $L(\lambda)$ was normalized to $\Delta \nu$, expressed in nm. The absorption spectrum of CH (at atmospheric pressure with resolution $\Delta \nu$) was calculated with the LIFBASE program [Luque 1996c]. The absorption spectrum $A(\lambda)$ and the laser profile $L(\lambda)$ are shown Figure 3.6. The integrals described in equation (3.2) were performed numerically, which resulted in $\Gamma_A = 0.17$. The calculated absorption profile was needed because a CH excitation spectrum could not be measured practically in the linear regime given the long integration times needed to obtain reasonable signal levels. When evaluating the integral, both the computed absorption profile (with resolution $\Delta \nu = 0.16$ cm$^{-1}$) and fit beam profile had narrow features. Small variations in the peak of the Gaussian, hence small changes in the laser wavelength, affect $\Gamma_A$. Since the resettability of the dye laser is $\pm 0.02$ Å, the corresponding uncertainty in $\Gamma_A$ is 20%.

$$
\Gamma_A = \frac{\int A(\lambda)L(\lambda)d\lambda}{\int A(\lambda)d\lambda \int L(\lambda)d\lambda} \quad (3.2).
$$
Figure 3.6. Plot of spectral relationship between the CH absorption and laser line.
3.6. Rayleigh Calibration

Calibration of CH laser-induced fluorescence is somewhat problematic. CH is highly reactive, preventing reference to a known concentration, and is present in a very thin (~500 µm wide) region, making absorption calibration difficult. Rayleigh scattering can be used on the same optical setup to relate the measured signal to an absolute light level, and thus solve for the overall calibration product ($\Omega\varepsilon\eta$) in equation (3.1) [Luque 1996a].

For scattering from a homogeneous gas, the Rayleigh signal per pulse is given by

$$S_{Ray} = \frac{N E_{Ray} V_{Ray} \sigma_{Ray}}{A_{Ray} h\nu} \Omega\varepsilon\eta$$  \hspace{1cm} (3.3)

where $N$ is the number density of the gas flow used for calibration, $E_{Ray}$ is the laser energy per pulse producing the Rayleigh signal, $V_{Ray}$ is the Rayleigh volume, $\sigma_{Ray}$ is the Rayleigh cross section, $h\nu$ the laser photon energy, and $A_{Ray}$ the cross-sectional area of the laser beam. After Rayleigh measurements are made, the calibration product can be eliminated, resulting in a single expression for number density. Since the fluorescence beam occupies the same spatial region as the Rayleigh beam ($A_{Ray} = A_{LIF}$ and $V_{Ray} = V_{LIF}$), we now have

$$N_0 = \frac{4\pi S_{LIF} \Delta\nu N E_{Ray} \sigma_{Ray}}{S_{Ray} \left( \frac{B_{12}}{c} \right) E_{LIF} \Gamma A f_B \phi h\nu}$$  \hspace{1cm} (3.4)

as an expression for absolute number density of ground-state CH, with $\phi = A_{21}/(A_{21}+Q_{21})$.

Rayleigh calibration was done on a flow of clean air with the same optical setup and beam dimensions as used in LIF. Although calibration can be done with a single Rayleigh image, measurements were made over a range of energies to verify the linear relationship between the signal and laser energy and to confirm a zero intercept. The measured
calibration was linear and had an intercept within 1% of the origin. Energy per pulse was varied between 0.1 and 1.2 mJ, with an integration of 300 to 1500 shots.

These calibrations and calculations resulted in a two-dimensional number density profile of CH mole fraction. The peak measured ground-state CH number density was $1.53 \times 10^{13}$ cm$^{-3}$, which is combined with the computed CH temperature of 1917 K to yield a measured peak mole fraction of CH of $4 \times 10^{-6}$. The major sources of error in equation (3.4) come from several quantities. The developers of the Rayleigh calibration procedure used here assessed its relative error to be 10-15% [Luque 1996a]. The quenching correction and $\Gamma_A$ computation are each uncertain to 20%. Using error propagation techniques, these quantities combine to give us a CH concentration measurement accuracy of 30%.

### 3.7. Results and Discussion: CH

The measured CH profile can be compared with the predictions of each kinetic mechanism in Fig. 3.7. As with our previous investigations, GRI Mech 2.11 continues to produce good agreement with the overall flame length observed in the experiments, while significantly underpredicting the flame lift-off height. The alternate kinetic scheme is much more accurate in predicting lift-off height, but over predicts the overall flame length. Note that the extinction strain rate computed with GRI Mech 2.11 is nearly 20% higher than that obtained with the alternate mechanism, which may allow the flame to anchor in a region of higher strain; this question will be addressed in greater detail in Ch. 4.

The character of the CH distribution within the GRI-computed profile is in excellent agreement with the measurements, i.e., the highest CH concentration appears near the flame anchoring region, falling off to a nearly-constant level throughout the flame front, up
Figure 3.7. Comparison of CH measurements with the predictions of 2 kinetic mechanisms.
to and including the tip of the flame front. The measured peak mole fraction of CH was 4 x 10^{-6}, which is within 20% of the prediction of GRI Mech 2.11. Since the uncertainty in CH concentration measurement was 30%, this is considered to be excellent agreement.

Other than a better prediction of the lift-off height, the computed CH results are not as good with the alternate hydrocarbon kinetic mechanism. The location and distribution of CH is still within reasonable agreement, but the flame length appears to be over predicted. Additionally, the peak CH concentration is predicted to be 42% lower than the measured concentration. The sensitivity of the spatial characteristics and absolute concentrations on the choice of kinetic scheme reveals CH to be an important test of our rather well-validated flame model, as subtle differences are not seen between the two kinetic schemes for the prediction of major species and temperature profiles.

3.8. Chemiluminescence of CH* and OH*

Flame chemiluminescence, also known as optical emission, has long been a recognized combustion phenomenon [Gaydon 1974]. The oxidation of CH plays a central role in the production of chemically excited OH (A^2Σ, denoted OH*), which emits in the ultraviolet. This ultraviolet emission has been suggested as a measure of the final steps of the CHx reduction chain [Najm 1998]. Chemically-excited CH (A^2Δ, denoted CH*) is a source of blue light in low-soot flames, and may provide insight into the C_2 reaction chain [Najm 1998]. Despite the prevalence of CH* and OH* chemiluminescence, little quantitative work has been done either measuring or predicting the absolute concentrations of these species. Additionally, the space and power constraints imposed by microgravity experiments have generated a renewed interest in developing flame emission tomography as a simple diagnostic technique. As a consequence, a detailed study was performed to
understand how optical collection geometry affects measured intensity signals and resulting reconstructed emissivity profiles. Simulation results are compared with measurements to assess reconstruction errors resultant from these measurements. Measurements of chemically-excited flame radicals are relatively easy to make but have calibration difficulties similar to those of ground-state CH. Fortunately, the same Rayleigh calibrations can be applied to determine absolute intensity levels, which can be combined with a spectral throughput correction to quantify measured signals. These absolute concentration measurements are used to judge the predictive ability of currently available CH* and OH* kinetic mechanisms.

3.9. Flame Emission Measurements

OH* chemiluminescence occurs in a band centered at 305.4 nm. Excited-state OH measurements were made with a cooled CCD camera using a f/4.5 UV camera lens; the camera/lens system was placed 50 cm away to ensure a wide depth of field. A narrow bandpass UV filter was used (center 307 nm, 10 nm bandwidth). High emission signal levels were collected with 10 s integration times. Rayleigh calibration was done with a Nd:YAG-pumped dye laser utilizing sulfarhodamine 640 dye. The 612 nm output was frequency doubled to perform calibration measurements on clean air. Beam energy in the UV was varied from 0.3 - 2.0 mJ per pulse and integration times were 600 shots.

For CH* flame emission, the A^2Δ→X^2Π transition at 431.4 nm was imaged with the camera and lens system detailed above. An interference filter (center 431 nm, 10 nm bandwidth) was used to isolate the CH* emission. The calibration measurements were performed with a Nd:YAG-pumped dye laser. During Rayleigh calibration, the energy of the 431.5 nm beam was varied between 0.5 - 2.5 mJ, with an integration of 600 shots. A
schematic of the experimental setup can be seen in Fig. 3.8 - the laser-related parts of the diagram are shown in dashed lines to indicate that the laser is used for calibration only.

In brief, the emission signal is given by [Hertz 1988]

\[
S_{\text{em}} = \frac{1}{4\pi} A_{21} \tau V_{\text{em}} N^* \Omega \varepsilon \eta
\]  

(3.5)

where \( A_{21} \) is the Einstein A coefficient, \( \tau \) is the integration time, \( V_{\text{em}} \) is the emission pixel volume, \( N^* \) is the number density of excited-state molecules, \( \Omega \) is the solid angle, \( \varepsilon \) is a calibration constant of the optics, and \( \eta \) is the detector efficiency in counts per photon, as discussed for LIF in equation (3.1). The number density of electronically-excited molecules in the flame is a balance between production by chemiluminescent reactions and removal by electronic energy transfer collisions, reactive collisions of the excited molecules, and spontaneous emission. Thus, a steady-state concentration of excited radicals is observed in the flame and will be predicted by any model that includes both production and the removal pathways. As indicated in equation (3.5), this concentration produces optical emission at the radiative rate. Accordingly, no calculation of collisional quenching is required [Luque 2000].

As discussed in Ch. 2., flame emission measurements can be calibrated with Rayleigh scattering. Additionally, the spectral distributions of the Rayleigh signal and chemiluminescence signal differ, giving the two signals distinct spectral overlaps with the interference filter used in the detection system. Defining \( \Gamma_E \) to be the ratio of these two spectral overlaps, we can combine equations (3.3) and (3.5) to obtain

\[
N^* = \frac{4\pi S_{\text{em}} N E_{\text{Ray}} V_{\text{Ray}} \sigma_{\text{Ray}}}{S_{\text{Ray}} \Gamma_E A_{21} \tau V_{\text{em}} A_{\text{Ray}} h\nu}
\]  

(3.6)
Figure 3.8. Experimental setup of chemiluminescence experiment.
with $\Gamma_E$ explicitly defined as

$$
\Gamma_E = \frac{\int_0^\infty E(\lambda)F(\lambda)d\lambda}{\int_0^\infty R(\lambda)F(\lambda)d\lambda}
$$

(3.7)

where $F(\lambda)$ is the monochromatic filter throughput, $E(\lambda)$ the emission profile, and $R(\lambda)$ the Rayleigh profile, with the integrations taking place over all wavelengths $\lambda$. $E(\lambda)$ and $R(\lambda)$ are normalized such that

$$
\int_0^\infty E(\lambda)d\lambda = \int_0^\infty R(\lambda)d\lambda = 1
$$

(3.8).

3.10. The Abel Inversion

Since the emission images measured in this work are integrated through the collection optics along the line of sight, a tomographic inversion procedure is required to reconstruct the planar, spatially resolved emissivity distribution. The Abel inversion, used to recover axisymmetric radial profiles from line-of-sight intensity measurements, is an attractive candidate to reconstruct the measurements made in this flame [Hughey 1982]. However, an Abel deconvolution is valid only when incoming rays are parallel, while most practical optical setups used for emission imaging consist of single- and multi-lens systems that collect light in a cone, over a non-zero solid angle. To the best of our knowledge, no one has investigated how these effects influence the measured intensity profile and the resultant radial distribution computed with an Abel inversion. These effects could be important in interpreting the imaging measurements of both relative and absolute radical concentrations performed with emission tomography [Marchese 1996, Luque 1998]. For relative concentration measurements, the chosen collection optics may widen or distort the measured intensity profile relative to parallel ray collection. Determining absolute concentrations could potentially be problematic as well, since collection over a cone results
in higher peak signals than assumed by parallel ray collection. The relative scale factor is sensitive to the geometry of both the distribution being measured and the collection optics.

A ray tracing simulation was performed to quantify some of these issues to provide a meaningful interpretation of the measurements made in this work. Integrated CH* emission is imaged over the entire flame, and 1-D intensity profiles are extracted at various axial heights for comparison with the predictions of this simulation. Since these measurements had a high signal-to-noise ratio, no smoothing was required before the Abel inversion was performed.

An axisymmetric emissivity distribution $F(r)$ can be determined by measuring the line-of-sight integrated signal at a single angle and performing an Abel deconvolution. When infinitely thin, parallel rays are collected across the entirety of $F(r)$, the line-of-sight integrated intensity profile $I(y)$ is given by

$$I(y) = \int_{-\infty}^{\infty} F\left(\sqrt{x^2 + r^2}ight) \, dx$$

This parallel ray collection is illustrated in Fig. 3.9. The emissivity $F(r)$ can be recovered from projection data with the use of the Abel inversion, which is written analytically as:

$$F(r) = -\frac{1}{\pi} \int_{-\infty}^{\infty} \frac{I'(y)}{(y^2 - r^2)^{1/2}} \, dy$$  \hspace{1cm} (3.10)

where $I'(y) = dI/dy$. Several implementation procedures are available to perform this inversion. In this work, an algorithm equivalent to a two-point Abel deconvolution is utilized [Dasch 1994].

A ray tracing algorithm was developed to simulate measured integrated intensity profiles. The coordinate system used is diagrammed in Fig. 3.10. The function $F(r)$,
Figure 3.9. Intensity profile resultant from parallel ray collection.
Figure 3.10. Coordinate system used in ray tracing program to simulate light collection from a cone.
centered at \((x=r_i, y=0)\), is represented on a pixelated grid. The pixel width is \(dx\) and the imaged region is a square of side length \(2r_i\). A lens of diameter \(d\) is placed a distance \(f_o\) from the near edge of the imaged region. In two dimensions, the integrated intensity from the point \((x_0,y_0)\) is collected from the two triangular regions, labeled \(R(x_0,y_0)\), bounded by the rays shown in the diagram. The total integrated intensity at \(y_0\) is the sum of the signals collected as \(x_0\) is varied. This is expressed mathematically as

\[
2D: \quad I(y_0) = \sum_{x_0=0}^{2r_i} \sum_{x=0}^{2r_i} \sum_{y \in R(x_0,y_0)} F\left(\left(\frac{(x-r_i)^2}{\xi^2} + \frac{y^2}{\eta^2}\right)^{1/2}\right)
\]  

(3.11)

The equations of the two lines that define \(R(x_0,y_0)\) are shown in Fig. 3.10. Note that in the limit of \(f_o \to \infty\), this region collapses to the line \(y=y_0\) and parallel ray collection is recovered. This was confirmed in the simulation.

Extending the simulation to three dimensions requires the inclusion of all signals collected in a three-dimensional cone. This cone is formed by rotating the triangular regions \(R(x_0,y_0)\) about the chief ray, which goes between \((-f_o,0)\) and \((x_0,y_0)\). To perform this three-dimensional intensity collection, the measured distribution is assumed to be a stack of planes with thickness \(dx\) and emissivity \(F(r)\). Contributions from adjacent planes are added by performing additional two-dimensional sums, over regions defined by the intersection of the collection cone with the appropriate plane.

The magnitude of the simulated intensity distributions from the cone-collected signals and the parallel ray signals is different due to geometric distortions as well as changes in the solid angle of collection. To account for this, the laser calibration method described earlier was simulated using the same ray tracing program. Signals collected from a 300 \(\mu\)m thick laser beam were simulated, and a scale factor was found between the parallel ray and cone-collected signals, which varied as the simulation parameters \((f_o, d, dx \text{ and } r_i)\) were
changed. This scale factor was computed in each case to allow direct comparison of simulated parallel ray-collected and cone-collected signals.

For CH* chemiluminescence imaging measurements reported earlier, light was collected with a single lens of diameter \( d = 22 \) mm a distance \( f_o = 500 \) mm away from the edge of the imaged region. The depth of field was determined to be greater than the flame width and the measured pixel size (\( dx \)) was 68 \( \mu m \). To match measured intensity profiles, \( F(r) \) was modeled as a Lorentzian on one side and a step function on the other. The geometrical parameters used in the flame measurements were used for our simulation, with the peak location and width of \( F(r) \) varied as free parameters to best agree with measured data.

The simulated and measured signal intensities are shown along with the parallel ray signal on the left side of Fig. 3.11. The simulated distribution has a peak at \( r = 3.9 \) mm; \( r_i \) was 12 mm. The measured and simulated signals agree quite well. Both are shorter and wider than the parallel ray profile, with higher magnitudes along the centerline.

The simulated signals can now be Abel inverted, allowing the calculated \( F(r) \) to be compared to the emissivity reconstructed from parallel rays, as seen on the right side of Fig. 3.11. The actual \( F(r) \) used in the simulation, not shown, is nearly indistinguishable from the reconstructed parallel ray profile. The integrated signal from cones with a finite solid angle gives a computed emissivity profile that is shorter and wider than the parallel ray profile. There is also a one pixel difference in the location of the peak. After the Abel inversion is performed, there exists low-magnitude residual noise from the centerline to where the distribution peaks, which is an artifact of all Abel inversion algorithms.

For these chemiluminescence measurements, the spatial resolution is estimated to be 2 pixels, or \( \sim 140 \) \( \mu m \). Therefore, the one-pixel difference in peak location and the two-pixel
Figure 3.11. Comparison of simulated and measured intensity profiles (left) and the corresponding reconstructed emissivity profiles (right).
difference in the full-width at half maximum between the reconstructed and actual F(r) fall within the experimental uncertainty. These CH* emissivity profiles can therefore be used to determine flame shape and lift-off. The 30% inaccuracy in peak signal, however, is significant, and needs to be combined with other experimental uncertainties to assess the accuracy in the measured peak CH* concentration.

As a general trend, errors in the reconstructed profiles tend to diminish as \( f_o/d \) approaches infinity and collected rays become more parallel. However, the magnitude of these errors depends on the distribution being measured, F(r), as well as the optical collection geometry [Walsh 1999]. This is shown in Fig. 3.12 (b), where the peak percent error is plotted as a function of \( f_o/d \) for three different simulated distributions, shown in Fig. 3.12 (a), that were similar in spatial extent. The value of \( f_o/d \) used in the flame emission measurements described in this work, which was constrained by signal-to-noise considerations, is also indicated in Fig. 3.12 (b). The simulation calculations were performed with \( f_o = 1000 \) mm, \( dx = 0.068 \) mm, and \( r_i = 12 \) mm. For a Gaussian profile, reconstruction errors are small over a wide range of collection geometries. However, for a top-hat profile of radius 5 mm, the reconstructed peak values are significantly overpredicted. As noted previously, the peak of the narrow CH* distribution is underpredicted. This differing behavior in identical collection geometries suggests that detailed, distribution-specific calculations are required to quantify measurement errors and interpret measured signals in emission tomography applications where parallel ray collection is required.
Figure 3.12. (a) Plot of the F(r) used in (b) to show reconstruction errors for different F(r).
3.11. Image Corrections

Appropriate background images, taken for both CH* and OH* with the flame extinguished, are subtracted from the raw emission signal. Normally, quantitative imaging techniques also require correction for variations in the throughput of the imaging optics. However, due to the large object distance and depth-of-field, the variation in throughput was less than 2% across the image (determined from the laser calibration images) and no response correction was included. An Abel inversion is then performed to recover a two-dimensional, in-plane intensity distribution proportional to number density.

A value for $\Gamma_E$ must be computed to correct for the throughput differences of the emission and calibration signals. The Rayleigh signal has a spectral width equal to the laser linewidth of 0.03 Å, and therefore passes through the interference filter at the laser wavelength only. In contrast, the chemiluminescence signals are spectrally broad, making the amount of light collected dependent on the entire spectral overlap with interference filter in the optical setup. LIFBASE [Luque 1996c] was used to calculate the CH* and OH* spectral emission profiles $E(\lambda)$ at flame temperature and a spectrophotometer was used to measure interference filter transmission, $F(\lambda)$. These spectral relationships are shown in CH* in Fig. 3.13. The resulting correction factor, $\Gamma_E$, is 0.78 for CH* and 0.99 for OH*.

The computed temperature in the spatial location of the computed CH* and OH* profiles was determined to be spatially-constant at 1917 K, similar to ground-state CH. The absolute number densities that result from corrected measurements are combined with these computed temperatures to provide CH* and OH* mole fractions. The peak mole fraction of CH* was $1.2 \times 10^{-11}$, while the corresponding peak of OH* was $4 \times 10^{-11}$.

Uncertainty in these measurements comes from the Rayleigh calibration and the flame emission signal. For a perfectly axisymmetric flame, the integrated flame emission signal
would be symmetric about the jet centerline. In our measurements, slight differences existed between the two halves of the measured signal. For certain fuel mixtures, the peak concentration can change by as much as 15% when the other half of the flame is inverted. The developers of the Rayleigh calibration procedure used here assessed its relative error to be 10-15% [Luque 1996a]. These factors combine to produce a measured mole fraction accuracy of 25% for CH* and OH*, which is then combined with the 30% systematic error uncovered in the ray tracing simulation. These errors are comparable to those of other trace species measurements in this flame.
3.12. Kinetics Modeling

To model chemiluminescence, the species CH* and OH* were added to the kinetic mechanisms of GRI Mech 2.11 and the alternate hydrocarbon mechanism described earlier. Reactions describing the formation and destruction of CH* and OH* were then formulated with appropriate rate constants and added to the mechanisms. All CH*/OH*-related rate constants used in this study are detailed in Table 3.3.

CH* is produced chemically via the reaction of the ethynyl radical with monatomic and diatomic oxygen:

I)  \( \text{C}_2\text{H} + \text{O}_2 \leftrightarrow \text{CH*} + \text{CO}_2 \)

II) \( \text{C}_2\text{H} + \text{O} \leftrightarrow \text{CH*} + \text{CO} \).

The rate constants have been reported as \( k_I = 3.6 \times 10^{-14} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1} \) and \( k_{II} = 1.8 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1} \) [Devriendt 1996]. The uncertainties associated with these reaction rates are about 40%. Destruction reactions occur by spontaneous emission (CH* → CH + hv) and collisional quenching. Quenching was modeled with seven different reactions, each involving CH* and a major chemical species in the flame (CH₄, N₂, O₂, H₂O, CO, CO₂, H₂). Species-specific, temperature-dependent quenching rates were taken from [Tamura 1997]. The heat of formation of CH* was set at 66.3 kcal/mole above that of ground-state CH, based on the energy of the spontaneously emitted photon.

OH* formation was modeled with a single reaction

III) \( \text{CH} + \text{O}_2 \leftrightarrow \text{OH*} + \text{CO} \).

The rate constant \( k_{III} = 1 \times 10^{-13} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1} \) deduced from the flame measurements of [Porter 1966], is utilized. The heat of formation of OH* was set at 93 kcal/mole above that of ground-state OH. From the rate constant for all products of CH + O₂ measured by
<table>
<thead>
<tr>
<th>Reaction</th>
<th>A</th>
<th>B</th>
<th>Ea</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{C}_2\text{H}+\text{O} \leftrightarrow \text{CH}^*+\text{CO} )</td>
<td>1.08E+13</td>
<td>0.00</td>
<td>0</td>
<td>[Devriendt 1996]</td>
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<tr>
<td>( \text{C}_2\text{H}+\text{O}_2 \leftrightarrow \text{CH}^*+\text{CO}_2 )</td>
<td>2.17E+10</td>
<td>0.00</td>
<td>0</td>
<td>[Devriendt 1996]</td>
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<td>( \text{CH}^* \rightarrow \text{CH} )</td>
<td>1.85E+06</td>
<td>0.00</td>
<td>0</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{CH}^*+\text{N}_2 \leftrightarrow \text{CH}+\text{N}_2 )</td>
<td>3.03E+02</td>
<td>3.40</td>
<td>-381</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{CH}^*+\text{O}_2 \leftrightarrow \text{CH}+\text{O}_2 )</td>
<td>2.48E+06</td>
<td>2.14</td>
<td>-1720</td>
<td>[Tamura 1997]</td>
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<tr>
<td>( \text{CH}^*+\text{H}_2\text{O} \leftrightarrow \text{CH}+\text{H}_2\text{O} )</td>
<td>5.30E+13</td>
<td>0.00</td>
<td>0</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{CH}^*+\text{H}_2 \leftrightarrow \text{CH}+\text{H}_2 )</td>
<td>1.47E+14</td>
<td>0.00</td>
<td>1361</td>
<td>[Tamura 1997]</td>
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<tr>
<td>( \text{CH}^*+\text{CO}_2 \leftrightarrow \text{CH}+\text{CO}_2 )</td>
<td>2.40E-01</td>
<td>4.30</td>
<td>-1694</td>
<td>[Tamura 1997]</td>
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<tr>
<td>( \text{CH}^*+\text{CO} \leftrightarrow \text{CH}+\text{CO} )</td>
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<td>0</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{CH}^*+\text{CH}_4 \leftrightarrow \text{CH}+\text{CH}_4 )</td>
<td>1.73E+13</td>
<td>0.00</td>
<td>167</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{CH}+\text{O}_2 \leftrightarrow \text{OH}^*+\text{CO} )</td>
<td>6.01E+10</td>
<td>0.00</td>
<td>0</td>
<td>[Porter 1966]</td>
</tr>
<tr>
<td>( \text{OH}^* \rightarrow \text{OH} )</td>
<td>1.45E+06</td>
<td>0.00</td>
<td>0</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{OH}^*+\text{N}_2 \leftrightarrow \text{OH}+\text{N}_2 )</td>
<td>1.08E+11</td>
<td>0.50</td>
<td>-1238</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{OH}^*+\text{O}_2 \leftrightarrow \text{OH}+\text{O}_2 )</td>
<td>2.10E+12</td>
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<td>-482</td>
<td>[Tamura 1997]</td>
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<td>( \text{OH}^*+\text{H}_2\text{O} \leftrightarrow \text{OH}+\text{H}_2\text{O} )</td>
<td>5.92E+12</td>
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<td>( \text{OH}^*+\text{H}_2 \leftrightarrow \text{OH}+\text{H}_2 )</td>
<td>2.95E+12</td>
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<td>( \text{OH}^*+\text{CO}_2 \leftrightarrow \text{OH}+\text{CO}_2 )</td>
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<td>0.50</td>
<td>-968</td>
<td>[Tamura 1997]</td>
</tr>
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<td>( \text{OH}^*+\text{CO} \leftrightarrow \text{OH}+\text{CO} )</td>
<td>3.23E+12</td>
<td>0.50</td>
<td>-787</td>
<td>[Tamura 1997]</td>
</tr>
<tr>
<td>( \text{OH}^*+\text{CH}_4 \leftrightarrow \text{OH}+\text{CH}_4 )</td>
<td>3.36E+12</td>
<td>0.50</td>
<td>-635</td>
<td>[Tamura 1997]</td>
</tr>
</tbody>
</table>

**Table 3.3.** Reactions added for \( \text{CH}^* \) and \( \text{OH}^* \) kinetics. Units for rate constants are cm, moles and sec, with the formulation

\[ k = A \text{E}^B \exp[-\text{E}_a/RT], \text{E}_a \text{ in cal mole}^{-1} \text{ and } R \text{ in cal mole}^{-1} \text{ K}^{-1}. \]
[Berman 1982], a branching ratio of 0.00185 for OH* can be inferred. Spontaneous emission and collisional quenching reactions were added to the mechanism in the same manner as described above. Note that chemi-excitation reactions I-III all have other channels which form ground state species, which are included in both kinetic schemes. The removal rate coefficients for major species in methane flames are known within 10% for OH* and 25% for CH* [Berman 1982].

3.13. Results and Discussion: CH* and OH*

The measured CH* distribution can be seen with the predictions of each kinetic mechanism in Fig. 3.14. Here the peak measured CH* mole fraction (1.2 x 10^-11) is a factor of 5 below that of the modified GRI Mech 2.11 and a factor of 10 below the predicted peak of the alternate (Yale) mechanism. In comparing Fig 3.14 with Fig. 3.7, CH* can be seen to peak in the flame anchoring region and exist along the flame-front, similar to measured and computed ground-state CH in this flame. Accordingly, the differences in flame length and lift-off height discussed earlier, in relation to CH, are indicated in Fig. 3.14. as well. However, differences can be seen in the spatial distribution of CH and CH* when examined at a finer scale, as seen in Fig. 3.15. Here the computed relative CH and CH* profiles, displayed at high resolution, are compared. The CH* distribution is more highly localized than that of CH, and exhibits a hook-like structure at the flame base which is not seen in CH. Furthermore, the CH* distribution peaks 400 µm closer to the burner surface than the corresponding CH peak.

The measured and predicted OH* distributions are similarly shown in Fig 3.16. The OH* profile is more highly localized than CH/CH*, peaking more sharply in the flame
Figure 3.14. Comparison of measured CH* profiles with the predictions of two kinetic mechanisms.
Figure 3.15. Structural differences in CH and CH* distributions.
Figure 3.16. Comparison of measured OH* profiles with the predictions of two kinetic mechanisms.
anchoring region. Since OH\(^*\) is formed from CH, OH\(^*\) can be seen to be spatially similar to CH\(^*\), most clearly at the flame base. Differences in lift-off and flame length can be seen among the three images, although not as clearly as those observed between CH and CH\(^*\) measurements and computations. The peak measured OH\(^*\) mole fraction (4 x 10\(^{-11}\)) lies between that of the Yale mechanism (2 x 10\(^{-11}\)) and the predicted peak of the modified GRI Mech 2.11 (5 x 10\(^{-11}\)). Both CH\(^*\) and OH\(^*\) occur in a very thin region on the order of 200 \(\mu m\) thick. To facilitate further comparison between experiment and computations, the CH\(^*\) and OH\(^*\) concentrations are integrated through the flame front, viewed according to the inset of Fig. 3.17 or 3.18; the integration is 100 \(\mu m\) along \(z\) by 100 \(\mu m\) into the page by 1 cm along \(r\). Note the excited radicals only are present in a narrow region along \(r\). This eliminates differences in spatial resolution and radial position between measurement and model, and expedites comparisons of measurement and predictions of excited state radicals versus height in the flame. Note that this integration produces an ordinate scale in Fig. 3.17 and 3.18 with units of molecules instead of the more familiar number density. The resulting integrated steady-state CH\(^*\) and OH\(^*\) profiles are shown as a function of height above the burner in Figs. 3.17 and 3.18, respectively. Both the CH\(^*\) and OH\(^*\) profiles peak at the base of the flame and steadily decline along the flame length to the tip of the flame 3 cm above the burner.

Figures 3.17 and 3.18 compare the measured data with the predictions of the model calculations using the Yale chemical mechanism and the augmented GRI-Mech 2.11. The model with augmented GRI-Mech 2.11 predicts concentrations of CH\(^*\) and OH\(^*\) that agree with the measurements within a factor of four and two, respectively. The shape of the variation with height above the burner of both the CH\(^*\) and OH\(^*\) steady-state...
Figure 3.17. CH* concentration, integrated across the flame front.
Figure 3.18. OH* concentration, integrated across the flame front.
concentrations are also well predicted by the augmented GRI-Mech 2.11. The calculation using the Yale mechanism does not predict the concentrations as well, over predicting the peak CH* concentration by a factor of six and under predicting the OH* by a similar factor of six. The calculation using the Yale mechanism does not match the variation of the CH* concentration as a function of height above the burner as well as the calculation with the augmented GRI-Mech 2.11. However, as noted earlier, the predictions using the Yale mechanism match the flame stand-off height, whereas calculations with the augmented GRI-Mech 2.11 find the flame 0.18 cm too close to the burner, as apparent in Fig. 3.17 and 3.18.

3.14. Conclusions

The results of this study indicate that GRI Mech 2.11 does an excellent job of predicting the peak CH concentration, considering the trace concentration and narrow spatial extent of the CH radical’s profile. Although the general properties of the flame shape and height are reasonable with GRI Mech, the large difference in lift-off height may be a cause for concern. The alternate mechanism provides a better prediction of flame onset, but agrees less well in peak CH concentration and in flame length.

As far as spatial distribution and overall characteristics are concerned, relationships similar to those observed between calculated and measured CH were correspondingly observed CH* and OH* as well. These results indicate that parameters such as flame shape and lift-off can be accurately measured with the modest experimental apparatus required to make flame chemiluminescence measurements. Although the quantitative comparisons between measured and predicted CH* and OH* concentrations are not out of the question, uncertainties in the production reaction rate coefficients for CH* and OH*
limit our ability to quantitatively predict the optical emission from methane flames to a factor of six. While CH* and OH* can conveniently provide flame structure information, the formation and destruction kinetics associated with excited-state species in flames require further research.
Chapter 4.
Effects of Buoyancy and Dilution on the Structure and Lift-off of
Coflow Laminar Diffusion Flames

4.1. Introduction

Combustion involves the production of high-temperature gases whose low density results in a buoyant acceleration, which can have a profound effect on flame properties. Quantities such as flame shape, lift-off, temperature, soot volume fraction, and blow-off limits, which are of importance in areas such as fire safety, pollutant production, and combustion efficiency, have been seen to be influenced by gravity [Cochran 1972, Ronney 1998, Lin 1999a, 1999b]. Microgravity flame studies allow gravity to be considered as an independent variable to understand combustion processes more fully.

For diffusion flames in particular, the ability to predict the coupled effects of complex transport phenomena with detailed chemical kinetics is critical in the modeling of turbulent reacting flows and in understanding soot formation and radiative transfer. In addition, the factors that affect extinction in diffusion flames are of practical importance in areas such as fire suppression and engine efficiency. The goal of our characterizations of coflow laminar diffusion flames is to bring to microgravity the multidimensional diagnostic tools available in normal gravity, and in doing so to provide a broader understanding of the successes and limitations of current combustion models. This is a first step toward a more detailed understanding of the interaction of convection, diffusion and chemistry in both buoyant and nonbuoyant environments.

Laboratory studies performed on this flame, discussed in detail in Ch. 3, indicated that quantitative number densities of CH* and OH* could be measured in a short time (10 s) with a simple, compact optical setup that could be made compatible with existing microgravity facilities. Although reactions involving CH* and OH* are not included in
most kinetic mechanisms, the CH*, OH*, and ground-state CH distributions have spatially similar shapes and locations. Therefore, the ground-state CH distribution, which is easily computed, and the readily measured CH*/OH* distributions can be used to provide a convenient way of comparing lift-off height and flame shape in the diffusion flames under investigation. Furthermore, since CH* and OH* serve as a sensitive marker of flame structure, such measurements will be able to follow even small structural changes brought about by the absence of gravity. Given that the fuel composition affects flame chemistry and that buoyancy influences the velocity profile of the flow, we have the opportunity to study the chemistry and fluid dynamic interaction over a wide range of flame conditions. The microgravity environment consequently facilitates an extensive set of comparisons between experiment and computation.

Although various diffusion flames have been studied in a microgravity (µg) environment, this work contains advances in both diagnostics and modeling. Experimentally, many µg flame studies have relied on video camera measurements that are qualitative in nature [Haggard 1972, Edelman 1972, 1986, Bahadori 1990]. A comparison of a video image and a quantitative emission image can be seen in our standard flame in Fig. 4.1. Note that the structural features of the flame are much clearer on the emission image. Furthermore, the high signal-to-noise allows the quantitative image to be Abel-inverted to obtain detailed information about radical concentration along a highly-localized flame front. Additionally, many of the flames studied in µg are jet flames, which attach to the fuel tube and/or are burning in quiescent air [Sunderland 1996, Urban 1998]. Such flames are extremely difficult to model computationally, since the temperature and velocity boundary conditions are ill-defined. In this study,
quantitative, non-intrusive spatially-resolved flame measurements are made that can be compared directly with computational predictions.

4.2. Microgravity Venues

The goals of a microgravity flame study are linked to the gravity level and duration available at the chosen μg venue. The ground-based microgravity accommodations that are most widely used by researchers are the drop tower and the KC-135 reduced gravity aircraft. The drop tower provides 2.2 seconds of low-gravity test time for experiment packages consisting of up to 125 kilograms of hardware. The experiment package, constrained to fit in a 33” x 38” x 16” drop frame, experiences a gravitational acceleration of less than $10^{-5}$ g during the fall. The only external force acting on the falling experiment packages is the air drag associated with the relative motion of the package within the enclosure of the drag shield. At the end of a drop, the experiment is
decelerated with an airbag. The peak deceleration rate can be as high as 20gs. Eight to twelve tests can be performed at the facility in one day.

The KC-135 can provide periods of low gravity for up to 23-second intervals. The aircraft accommodates a variety of experiments and is often used to refine space flight experiment equipment and techniques and to train crew members in experiment procedures, thus giving investigators and crew members valuable experience in working in a weightless environment. Qualified researchers may fly with their experiment packages. The KC-135 obtains a low-gravity environment by flying a parabolic trajectory. Gravity levels twice those of normal gravity occur during the initial and final portions of the trajectory, while the brief pushover at the top of the parabola produces less than one percent of Earth’s gravity ($10^{-2} g_{\text{EARTH}}$). Local acceleration is influenced by air turbulence, engine noise, and other factors that make this $\mu$g acceleration unsteady. This noise in the acceleration profile aboard the KC-135 is termed “g-jitter,” and its effect on combustion processes is often a concern to experimenters. The number of low-g maneuvers on a given flight varies from 30-55, with an average of perhaps 40. Four flights are generally scheduled in a four day period.

The KC-135 was our venue of choice for several reasons. Most importantly, flames go through unstable behavior for some time after the influence of gravity is removed until the flame can equilibrate to its local acceleration environment. The 2.2 second drop tower seemed likely to provide only a short time, at best, for a steady flame to be measured. We therefore wanted to examine flame behavior when long low-g times were available and flame stability could be experimentally determined and assured. Next, the KC-135 is much more “data rich” than the drop tower; one day on the KC-135 provides (at worst) 300 s of low-g, compared to several seconds (at best) in the drop tower. Lastly,
flame measurements involving high-power, pulsed lasers were being planned at the time, which could be accommodated aboard the KC-135 but not in the drop tower.

4.3. Flame Parameters

The coflow burner, described in detail in Ch. 3, is used in this μg flame study. Recall that the standard flow conditions, examined in the last chapter and as well as in previous work, consist of fuel composed of 65% CH$_4$ diluted with 35% N$_2$ by volume (denoted 65/35 in later discussion). The exit velocity of both fuel and coflow was 35 cm/s. A wide range of flow conditions were measured in this study, with the CH$_4$/N$_2$ fuel composition varied from 100% CH$_4$ (denoted 100/0) to 30% CH$_4$ (denoted 30/70) in 5% increments, with fuel and air exit velocities held fixed at 35 cm/s. This was done to study flames with widely varying lengths, lift-off heights, and sootiness. For example, in 1 g the 100/0 flame is visibly sooting, 5 cm tall, and anchors 3 mm above the burner surface, while the 40/60 flame is weakly blue, 1 cm tall, and has a lift-off of 27 mm.

The role of buoyancy in a flame’s flow field can be roughly characterized by a dimensionless number called the Froude number (Fr) which is given by

$$ Fr = \frac{u^2}{g_{\text{EARTH}}z} $$

(4.1)

in which $u$ is the fuel exit velocity, $g_{\text{EARTH}}$ is 9.8 m/s$^2$, and $z$ is a characteristic length scale. In normal gravity, small Froude numbers ($<\sim1$) indicate buoyant flows while large Froude number flows are considered to be momentum-dominated. Alternatively, normal gravity flames acquire a buoyant velocity of order $(g_{\text{EARTH}}z)^{1/2}$ that can have an influence on the established flow-field. In our normal gravity 65/35 flame, for example, $u = 35$ cm/s and $z \sim 3$ cm, which provides a buoyant velocity of roughly 55 cm/s and Froude
number Fr \sim 0.4. These simple arguments indicate that buoyancy will play an important role in the behavior of the flames investigated in this study.

There were two goals of this initial round of microgravity experiments. First, we wanted to characterize the behavior of the coflow laminar diffusion flame in terms of its shape, stability, and propensity to soot for a variety of flow conditions, including our standard conditions for which detailed experimental and computational data are available at normal gravity. Careful attention to flame characteristics in high-g, “g-jitter”, and $\mu g$ can help optimize experimental techniques and develop intuition to guide future flame research. Tangibly, the mission deliverables were quantitative measurements of the CH*/OH* concentration distribution in the flame for a range of flow conditions.

In performing this microgravity study, improvements to the computational model have been made and new calculations performed for a range of gravity and flow conditions. Furthermore, modifications to the experimental approach were required as a consequence of the constraints imposed by the microgravity environment of the KC-135 reduced-gravity aircraft. Results from the computations and experiments are presented in the following sections.

4.4. Microgravity Flame Computations

The computational models used in this flame study included the chemical mechanisms of both GRI Mech 2.11 and the simple 26-species, $C_2$ hydrocarbon mechanism developed at Yale, discussed in the previous chapter. Preliminary computations, performed with GRI Mech 2.11 with different values for the gravitational constant, indicate that buoyancy plays a role in both the size and shape of the laminar diffusion flame. These computations are shown in Figs. 4.2. and 4.3.
Figure 4.2. Temperature computed at three different gravity levels.
Figure 4.3. Comparison of computed CH profiles at three different gravity levels.
Specifically, in Fig. 4.2 we illustrate the temperature isotherms for flames computed with \( g = 9.8, 4.9 \) and \( 0.0 \) m/s\(^2\). It is clear from the figures that, as the gravitational constant is lowered, the flames tend to become shorter and broader in appearance. This is also illustrated in Figure 4.3 where the corresponding CH isopleths are plotted. The influence of buoyancy on the flow-field, expected from the dimensional arguments made earlier, was confirmed computationally. This influence can be seen in Fig. 4.4., which displays the computed centerline velocity, utilizing the alternate mechanism, in both \( 0 \) g and \( 1 \) g. The peak velocity in these flames differs by more than a factor of two. Furthermore, the normal gravity flow velocity continues to increase with downstream distance, while the \( \mu g \) profile peaks near \( z = 15 \) mm and steadily declines.

![Computed Centerline Velocity Profiles](image)

**Figure 4.4.** Velocity profiles computed in \( 1 \) g and \( 0 \) g with the Yale mechanism.

While large variations in \( g \) are important in illustrating the differences between normal gravity and microgravity flame structure, small variations in \( g \) are important in determining whether the flame will be amenable for study in the NASA KC-135. During
the microgravity portion of the KC-135’s parabolic trajectory, the value of the gravitational constant can vary by as much as \( \pm 1\% \) of \( g_{\text{EARTH}} \). However, since this acceleration varies as a function of time, a time-dependent forced flame model would be required to follow possible flame instabilities. The computational model predicts flames resultant from steady accelerations only, so a weaker test of potential flame perturbations was performed. Numerical studies indicated the flame structure to be insensitive to steady changes in \( g \) of \( \pm 1\% \) \( g_{\text{EARTH}} \) about \( g=0 \) at the 65/35 flow condition. These computations indicated that the flame could be amenable to study aboard the KC-135, although the question of flame response to “g-jitter” remained unaddressed.

The remaining calculations performed in this study used the 26-species \( \text{C}_2 \) mechanism, which shows better agreement with measured lift-off than GRI Mech 2.11 at the 65/35 flow condition. Flame structure was calculated over a range of flow conditions in both \( \mu g \) and normal gravity. The results of the computed solution at standard flow conditions (65/35) and normal gravity were used as a starting point. In subsequent calculations, the value of the gravitational acceleration (\( g \)) was reduced by 10 cm/sec\(^2\) and a new solution calculated using Newton’s method. A range of flow conditions was reached by using the 65/35 flame as an initial condition and using a similar iterative procedure, varying the fuel mixture in 5% increments. Computations were performed for \( \text{CH}_4/\text{N}_2 \) fuel mixtures ranging from 30/70 to 75/25. However, calculations were not performed at flow conditions less dilute than 75/25 since these flames were observed to produce soot, which was not included in the computational model.
4.5. Experimental Setup

Microgravity facilities impose restrictions on experiment size, power consumption, and complexity. In order to be consistent with these requirements and still provide quantitative information that can be compared with models, the approach in the first phase of this work is to perform emission tomography on selected chemiluminescent flame radicals. The details of this technique were described in the previous chapter and will not be repeated here.

Experiments that are designed and built for the KC-135 must meet a stringent set of requirements. All research hardware must be able to withstand flight loads of 9 g in the forward direction, as well as 7 g down, 3 g aft, and 2 g’s lateral and up. Detailed equipment descriptions, structural and electrical load analysis, flammability calculations, and hazard analysis must be performed and every subsystem of the experiment approved before the experiment is allowed on the aircraft. Additionally, experimental procedures must be thoughtfully developed, refined, and documented, initially for safety reasons. Beyond safety assurance lies the fact that the experiment must be highly automated, due to the timing and acceleration profile of the low-g maneuvers. Before and after each parabola is a high-gravity period, during which movement and experimental adjustments are difficult to perform. Furthermore, six or seven parabolas are usually performed in rapid succession, allowing researchers only a minute or two of available time to adjust experimental equipment or software. Between flights, researchers generally have only a few hours to work on their experiment, which limits the ability to fix problems or re-define experimental goals during a flight week. Experimental issues are therefore a crucial part of this μg combustion study and will be discussed in some detail.
A schematic of the experimental equipment rack, for both side and top views, is shown in Figs 4.5 and 4.6. A combustion vessel and all measurement equipment are contained within this rack, which is a 33” x 38” x 16” experimental drop frame provided by NASA Glenn Research Center. The combustion vessel contains the burner and ignition system. A CCD detector, video camera, and all associated power supplies and cooling units are contained in this rack. Quantitative flame measurements are the spectrally filtered chemiluminescence images acquired by the Photometrics CH350 CCD detector. The Sony XC-999 color video camera is used to give qualitative insight into flame structure and soot production, as well as monitor the combustion chamber in real time. The experimental rack also houses the fuel supply, mass flow controllers and power supply, and relays allowing computer control of various components.

The external control rack, which contains equipment related to data storage, visualization, and experimental control, is diagrammed in Fig. 4.7. The SVHS VCR and video monitor are used to record and display real-time video information about the flame in microgravity conditions. A microcomputer, with keyboard, monitor, CPU, and trackball is used to control experimental systems and acquire and store quantitative flame emission images from the Photometrics CCD camera. Note that all monitors are placed behind a Lexan shield for safety reasons.

The burner and ignition system is housed inside a pressure vessel with 12” inner diameter and 25 1/2” of overall height. The pressure vessel is maintained at standard atmospheric pressure (14.7 psia) to mimic laboratory conditions and the parameters used in computational modeling. The pressure vessel has feed-throughs for an air line, a fuel line and electrical connections for the ignition system. There are two portholes sealed with aluminum plates that can be removed in a non-flight situation to perform laser
Figure 4.5. Experimental equipment rack, side view.
Figure 4.6. Top view of the three shelves of the experimental equipment rack.
Figure 4.7. Diagram of experimental control rack.
calibration. The third port of the vessel contains a 6” diameter, 1/2” thick, optical grade quartz window for flame imaging. The pressure vessel vents into the airplane’s overboard exhaust system.

Pressure at the vent of the pressure vessel can range from atmospheric pressure at sea level (when the aircraft is on the ground) to nearly 35,000 feet (aircraft at maximum altitude). To keep the combustion vessel at standard atmospheric pressure for all these conditions, an orifice plate and a computer-controlled servo valve are utilized. The orifice plate (1/16” hole diameter) exhausts the entire gas input under choked flow conditions. In parallel, a computer-controlled servo valve (1/4” diameter full open) allows the pressure in the vessel to remain constant when the vent pressure is higher than that necessary for choked flow -- up to atmospheric pressure (aircraft on the ground). The servo valve full-open diameter is chosen to provide a small (1%) pressure drop when venting to atmospheric pressure. As a redundant safety system, a third parallel vent path is provided by a pressure relief valve with a set point of 20 psig. A diagram of the exhaust system is shown in Fig. 4.8.

An ignition system is present in the sealed pressure vessel and must be controlled externally. The igniter consists of two parts: a 60° rotary solenoid and a 34 gauge kanthal hot wire. When the cue for ignition is given, an automated procedure begins. A signal is sent that triggers the rotary solenoid to swing the hot wire over the fuel jet. Current then flows into a single hot wire, which heats up and ignites the fuel. The wire is quickly retracted and the current supply cut off so the wire can cool down. The ignition system is illustrated in Fig. 4.9. The power for both the rotary solenoid and the hot wire is provided by the Tenma power supply on shelf 3 in the experimental equipment rack.
Figure 4.8. Diagram of exhaust system used to regulate combustion vessel pressure.
Figure 4.9. Diagram of remote ignition system used in microgravity flame study.
The methane fuel supply was “farmed out” in small quantities for safety reasons. Figure 4.10 shows the methane tank supply system. In this distribution system, six sampling cylinders (75 cc volume) are pressurized to 400 psig. Each cylinder contains 2 L of fuel and is individually selected with the seven-way valve. The amount of methane per cylinder is chosen such that if the entire contents of a fuel bottle were to empty out into the 44 L pressure vessel, the mixture would be below the lean flammability limit. The time taken to empty a cylinder depends on the flame conditions chosen -- at 65/35 it is 12 minutes. This distribution system provides an adequate methane supply that fits within safety constraints.

The microcomputer, running “in-house” software, was used to control and regulate various experimental components. In the foreground, digital signals from the computer control relays that enable/disable the coolant circulator and the ignition system. Also, analog voltages are generated to run the mass flow controllers which regulate nitrogen and methane fuel flows. The connections for these controls are shown in Fig 4.11. Combustion vessel pressure is controlled by monitoring a transducer and opening/closing the servo valve as necessary in a feedback loop. This process occurs in the background at 10 Hz. The connections to the auxiliary I/O board that controls these processes are shown in Fig. 4.12.
Seven way valve

Flow exiting into page.

1.5"

75 cc gas sampling cells @ 400 psi

Tubing is 1/8" OD thick walled stainless, valve fittings are 1/8" female swage

Figure 4.10. Methane fuel distribution system.
Figure 4.11. Computer connections for experiment control in the foreground.
Figure 4.12. Computer connections for experiment control in the background.
4.6. Measurement and Image Processing

Chemiluminescence measurements were made with the imaging system, integration time, and interference filters described in Ch. 3. The raw signals are similarly background corrected and Abel-inverted. Laser calibration occurred after the experiment was taken off the aircraft. A spectral overlap correction is again performed to obtain quantitative number densities of CH* and OH*.

At each flow condition the computed CH profile was used to define the flame sheet region where CH* and OH* coexist. The computations again indicated that the CH*/OH* radicals are present in a hot, thin zone at a constant temperature which varies from 1877 K at 50/50 to 1940 K at 75/25, independent of buoyancy. The absolute number densities that result from corrected measurements are combined with computed temperatures to provide CH* and OH* mole fractions. The peak mole fractions of CH* varied between 0.8 x 10^{-11} and 1.4 x 10^{-11}, and the corresponding peaks of OH* varied between 3.2 x 10^{-11} and 4.3 x 10^{-11} as the fuel composition was varied from 50/50 to 75/25. Uncertainties in these measurements are identical to those discussed in the previous chapter, although this study is more concerned with the structural information provided by CH*/OH* measurements rather than peak concentration values.

4.7. Results and Discussion

During each low-gravity maneuver on the KC-135, the combustion vessel pressure and airplane accelerometer signals were recorded simultaneously with the flame emission signal. The acceleration profile during a 25 second interval on the KC-135 is shown at the top of Fig. 4.13. When making flame emission measurements, care was taken to trigger the detection system after several seconds of low-g to give the flame time to
equilibrate. The acceleration during a typical 10 second data acquisition period is shown at the bottom of Fig. 4.13 to display the small-scale fluctuations present. A complete set of measured data, obtained from a single low-g maneuver, is shown in Fig. 4.14. Again, “g-jitter” can be seen in the acceleration plot at the bottom of this figure. Flame emission images analyzed in this study had a gravitational time-trace average of 0.01 g\textsubscript{EARTH}, with rms fluctuations of 0.02 g\textsubscript{EARTH}.

Understanding flame behavior in g-jitter is crucial to interpreting measurements on the KC-135. Measurements require a stable emission signal over the duration of a 10 second camera exposure. CH\(^*\) and OH\(^*\) concentrations peak strongly in the flame anchoring region and fall off rapidly along the flame length. During any given low-g maneuver, the flame base appears extremely stable on the video monitor, which allowed for careful emission measurements of both CH\(^*\) and OH\(^*\) for fuel compositions ranging from 50/50 to 100/0. The stability of the base of these flames is further supported by the fact that spatial features of the measured CH\(^*\)/OH\(^*\) profiles in \(\mu\)g are similar to those seen in 1 g. This implies that temporal fluctuations, which would have resulted in spatially-averaged measurements, did not occur. The flame tip, however, can behave differently than the flame anchoring region. In dilute fuel blends (40%-50% N\(_2\)), the entire \(\mu\)g flame appeared as stable as a normal gravity flame. For richer fuel mixtures, containing more than 75% CH\(_4\) by volume, the \(\mu\)g flames produced significant soot luminescence, which could be seen to fluctuate as a result of g-jitter. In the 100/0 flame, g-jitter causes the sooty region in the flame to “bounce”, translate, and change shape considerably.
Gravitational Acceleration aboard KC-135

Figure 4.13. A sample acceleration profile during a low-g maneuver on the KC-135.
**CH* Microgravity Emission Signal**

Standard Flow Conditions
65% CH<sub>4</sub> : 35% N<sub>2</sub> by volume

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**Pressure Transducer Signal**

Mean Pressure: 14.74 psia
RMS: 0.05 psia

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**Accelerometer Signal**

Mean Acceleration: 0.005 g<sub>EARTH</sub>
RMS: 0.02 g<sub>EARTH</sub>

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**Figure 4.14.** A full data set acquired during a single low-g parabola.
4.7.1. Flame Structure Measurements and Computations

Measurements were performed in 0 g and 1 g over a wide range of flow conditions. Some changes induced by buoyancy and fuel composition can be seen qualitatively in Fig. 4.15, which shows video images of 1 g and 0 g flames at three flow conditions. Quantitatively, measured flame shape, as indicated by the spatial distributions of the CH* and OH* radicals, can change significantly between normal gravity and microgravity. In general, a microgravity flame is shorter, wider, and has a higher flame front curvature relative to its normal gravity counterpart. Furthermore, since methane is lighter than air, density effects produce a normal gravity flame with a higher lift-off than the corresponding µg flame. These effects can be seen in the measured 1 g and 0 g CH* profiles shown in Fig. 4.16. The difference in lift-off is greatest for the 50/50 fuel mixture, where buoyant forces have the most time to operate before flame stabilization occurs. Note that the lift-off height of the 80/20 and 100/0 flames changes least when the influence of gravity is removed. Additionally, the 80/20 and 100/0 µg flames have soot luminescence visible at the flame tip within the CH* detection bandpass. Computations were not performed for these sooty flames, which makes the flame front temperature and therefore the measured peak mole fraction unknown. However, flame sheet temperatures were linearly extrapolated to these flow conditions to provide a peak mole fraction for visualization purposes. The OH* profile is more localized than that of CH*, although similar gravity-induced changes in flame structure can be seen. Soot luminescence is not visible within the bandpass of the OH* filter.
Figure 4.15. Diffusion flame video images at different flow conditions in 1 g and 0 g.
Figure 4.16 Normal gravity and microgravity CH* concentration measurements at different flow conditions.
Computed CH profiles, which utilized the alternate kinetic mechanism, are shown with measured CH* profiles at 65/35 in Fig. 4.17. Note that these comparisons should be made with the caveats discussed in Ch. 3. in mind -- the CH* profile is more highly localized than that of CH, and the location of the CH* peak concentration is 0.4 mm closer to the burner surface than that of CH. However, these spatial differences between CH and CH* are generally smaller than left/right asymmetries in the measured flames, and serve as an additional error source when comparing experiment and computation.

At standard flow conditions (65/35), the structural agreement is good for both of lift-off and flame shape, both in normal and microgravity, in that the differences in flame-front location between measurement and computation are comparable to measured flame asymmetries and spatial differences between CH and CH*. Although the computed flame length may appear to be over-predicted at this flow condition, CH* does not exist at the flame tip and as such is not a good indicator of flame length. As the fuel mixture is diluted further to the 50/50 flow condition, measurement and computation agree less well. This can be seen in Fig. 4.18, where normal gravity and µg measurements and computations are compared for this fuel mixture. In normal gravity, the flame lift-off height is considerably under predicted, while the µg predictions are in reasonable agreement with measurement.

4.7.2. Measured and Computed Lift-Off Heights

Figures 4.17 and 4.18 indicate that the accuracy of the computational model changes as the fuel composition is varied. To study this process systematically, we define the measured lift-off as the height above the burner where the CH*/OH* maximum occurs, and similarly for CH in the computations, utilizing the Yale mechanism. Since all three
65% CH₄: 35% N₂ by volume

Figure 4.17. Flame structure comparison utilizing computed CH (alternate mechanism) and measured CH* profiles.
Figure 4.18. Flame structure comparison utilizing computed CH (alternate mechanism) and measured CH* profiles.
peaks occupy similar spatial locations, we have the ability to plot measured and computed lift-off heights, both in µg and normal gravity, as a function of methane level in the fuel stream. This comparison is shown in Fig. 4.19. Uncertainties in measured lift-off arise from flame asymmetries at dilute flow conditions and are shown as error bars. At most fuel mixtures, the magnitude of these errors is smaller than the size of the data point.

The predicted lift-off height agrees well with measurement in 1 g at the 75/25 and 65/35 fuel mixtures. As the methane level decreases from 65% to 50%, with the total volumetric flow rate held constant, the computed and measured curves begin to separate. When the fuel mixture is diluted below 50% CH₄ in 1 g, the lift-off height becomes increasingly under-predicted, until the code computes a stable flame at fuel mixtures (35% and 30% CH₄) beyond the 1 g experimental blow-off limit (40% CH₄). Further, the difference between computed normal and µg lift-offs does not match the measured curves, which separate increasingly as the fuel mixture is diluted. The measured and computed 0 g lift-offs show reasonable agreement at the two points that allow direct comparison. Additionally, the variation in 0 g lift-off with fuel composition appears well predicted over the limit range of dilution levels seen here. In the following section, differences in measured and computed strain rates at extinction are investigated as a possible explanation for the observed discrepancies in lift-off.
Figure 4.19. Comparison of measured and computed lift-offs.
4.7.3. Strain Rate at Extinction

Many competing theories have been published to describe the characteristics of lifted diffusion flames and explain experimentally observed lift-off heights [Vanquickenborne 1966, Janicka 1983, Kalghatgi 1984, Peters 1983, 1985]. These theories of lift-off are assessed in [Pitts 1988], who concluded that none of the currently available theories for flame stabilization are fully satisfactory. However, two of these theories are discussed here in an attempt to relate differences in measured and computed lift-off to fundamental flame properties.

One possible explanation for flame stabilization is that the lift-off height of the coflow diffusion flame is related to the position where the local scalar dissipation (directly proportional to the strain rate) is comparable to the value obtained at extinction of a counterflow diffusion flame with the same fuel/dilution level. A higher counterflow scalar dissipation at extinction will imply a lower lift-off height as the strain rates in the coflow flame tend to be highest near the burner surface. In short, this theory suggests that flame stabilization occurs at a point where combustion extinction and propagation are balanced [Peters 1983]. Accordingly, to understand the reasons for the growing difference between the computed and measured coflow lift-off heights at high dilution levels, a series of experiments and computations were performed utilizing a counterflow diffusion flame burner and model. Fuel and oxidizer exit velocities were measured in a double-jet axisymmetric counterflow burner (diameter 19.5 mm) at two separation distances (L=9.5 mm, 20.8 mm) to compute a global strain rate at extinction. The CH₄/N₂ fuel composition was varied from 30/70 to 100/0 in 5% increments. At each flow condition, the exit velocities of fuel and oxidizer were carefully adjusted until the flame extinguished midway between the two burner surfaces. Error bars, which are at the
±10% level, are from the repeatability at the same fuel mixture measured at a different
time or burner separation distance. Arclength continuation computations were employed
to determine the exit velocities at extinction for methane-air diffusion flames at varying
levels of dilution [Giovangigli 1989]. The effective global strain rate, $K \text{ (s}^{-1})$, is given in
terms of the fuel exit velocity ($u_F$) and the oxidizer exit velocity ($u_O$) as [Kim 1992]

$$
K = \frac{2(-u_O)}{L} \left[ 1 + \frac{u_F \rho_F}{(-u_O) \rho_O} \right]^{1/2} 
$$

where $L$ is the separation distance between the two burners and $\rho_F$ and $\rho_O$ are the fuel and
oxidizer density, respectively. Note that this global strain rate is distinct from the local
strain rate obtained by fully characterizing the velocity field [Chelliah 1990]. Computed
and measured fuel and oxidizer exit velocities are input into equation (4.2) to the allow
comparison of strain rates at extinction, as shown in Fig. 4.20. This plot reveals that at
low dilution levels, the numerical extinction strain rates agree to within 5-10% of the
experimental values. However, as the nitrogen dilution mole fraction increases to 60%,
this difference increases to almost 100% -- as the dilution level increases, we obtain
increasingly larger differences between the numerical and experimental extinction strain
rates. This implies that there will be a growing difference between the experimental and
computational coflow lift-off heights. In addition, as the numerical counterflow
extinction strain rates were always larger than those determined experimentally, the
numerical lift-off heights should always be smaller than the measured values, exactly as
was observed. This explanation, while qualitatively reasonable, cannot explicitly relate
differences in extinction strain rate to differences in lift-off. For example, at the 65/35
flow condition, the measured and computed lift-offs agree while their corresponding
Global Strain Rate at Extinction

Figure 4.20. Comparison of measured and predicted extinction strain rates from a counterflow diffusion flame.
strain rates at extinction are 50% different. This study of extinction strain rates has indicated that discrepancies exist between measurement and computation on a fundamental level, and that these differences grow as the fuel mixture becomes more dilute. The strain rate at extinction likely has an influence on the stabilization behavior of the coflow flames under investigation, although a detailed computational study assessing these effects remains to be done.

Another theory, called the premixedness model, describes flame stabilization processes by considering the burning velocity of a locally-premixed flame [Kalghatgi 1984]. This explanation of lift-off suggest that lifted flames are stabilized near the stoichiometric contour, at a position where the local flame speed is equal and opposite to the velocity of unburned gas into the flame anchoring region [Vanquickenborne 1966]. These predictions could be tested in the coflow flames in this work by comparing the computed flow velocity at the stoichiometric contour to the results of laminar flame speed calculations. Unfortunately, laminar flame speed calculations have not been preformed as part of this work, so no such comparison can be made.

4.8. Conclusions

This work has revealed that many discrepancies exists between computation and experiment on a fundamental, visible level, namely in areas of flame lift-off and shape, that can be quickly revealed by flame emission experiments. Previous laboratory studies of this flame, which focused on a single flow condition (65/35) and gravity level, failed to notice the discrepancies between measurement and computation in weak, dilute flames. This disparity between measured and computed lift-offs indicates that the mechanisms that govern flame stabilization need to be more fully characterized computationally and experimentally.
Chapter 5.
Two-Dimensional Laser Diagnostics in Buoyant and Nonbuoyant Coflow Laminar Diffusion Flames

5.1. Introduction

Early laminar flame work in microgravity has focused largely on attached jet flames burning in quiescent air, and often involved video and thermocouple or probe measurements [Haggard 1972, Bahadori 1991, Urban 1998]. Many investigated flames could not easily be modeled, given the ill-defined temperature and velocity boundary conditions [Sunderland 1999, Lin 1999a]. Additionally, the qualitative nature of video images and the disrupting flow-field effects of thermocouples and probes present difficulties in interpreting experimental flame characterizations. In this work, a lifted axisymmetric laminar diffusion flame, which has been well-characterized previously both experimentally and computationally in normal gravity, is studied in a microgravity (µg) environment. Two-dimensional laser diagnostic techniques such as Rayleigh thermometry and Raman scattering, normally restricted to a laboratory environment, are extended to the µg environment of the KC-135 reduced-gravity aircraft. Additionally, laser-induced incandescence (LII) measurements are made in both µg and normal gravity. In sum, this work expands the scope of previous research by allowing comparisons of temperature, species, and soot over a range of flow conditions and gravity levels in a step toward a more detailed understanding of the interaction of convection, diffusion and chemistry in both buoyant and nonbuoyant environments.

The flame under investigation consists of nitrogen-diluted methane fuel surrounded by an air coflow, which is described in detail in previous chapters. In the work performed in Ch. 4, flame shape and lift-off were studied over a wide range of fuel mixtures, in which the computed CH profile and the measured CH* and OH* profiles were used to mark the
flame front and flame anchoring region. The large differences between measured and predicted lift-off heights seen for dilute fuel mixtures pointed to a need for further study. However, the chemiluminescence measurements provided structural information over an extremely narrow spatial region near the flame base; as such this work assessed global parameters such as flame shape and lift-off. Temperature measurements could allow for more definitive comparisons between experiment and computation over a larger spatial extent. Likewise, measurements of fuel and oxygen concentrations could provide insight into fuel/air mixing in the flame anchoring region, especially in cold regions where emission and temperature measurements provide little information.

Two-dimensional laser imaging techniques such as Rayleigh and Raman scattering are difficult to perform in microgravity facilities due to experimental size, weight, and power requirements. In the last few years, highly compact, pulsed Nd:YAG lasers have become available to alleviate some of these problems. However, many practical issues remain in making such measurements on the KC-135 reduced-gravity aircraft, among them spurious light scattering in Rayleigh measurements and possible flame fluctuations as a result of g-jitter. In addition, although a computational soot model does not currently exist for this flame, two-dimensional laser-induced incandescence (LII) measurements are made, both in normal and microgravity, to quantify changes in soot concentration and distribution in an unsteady low-g field. Although LII measurements have been performed in microgravity by other researchers, a systematic study of unsteady soot-field behavior in g-jitter has not been attempted [Megaridis 1996, Vander Wal 1997].

In the following sections, the scope and goals of this microgravity study are defined. The experimental modifications required to implement laser diagnostics are detailed. The two-dimensional temperature and major species measurements made are described and
compared with normal gravity and $\mu$g flame computations, described in detail in Ch. 4. Finally, LII measurements and results are discussed to gain insight into steady and unsteady laminar soot processes.

5.2. Flame Parameters

The coflow burner, described in detail in Ch. 3, is used in this $\mu$g flame study. A wide range of flow conditions were again examined, although in this study particular attention is paid to experimentally determining blow-off limits, which is defined as the most dilute fuel mixture, given as a CH$_4$/N$_2$ percentage, for which sustainable combustion can occur. Accordingly, more dilute fuel compositions were investigated in this study, in which mixtures from 100% CH$_4$ (denoted 100/0) to 30% CH$_4$ (denoted 30/70) were investigated.

The goals of this advanced microgravity study divided into several categories. First, single-shot Rayleigh measurements were done over a range of flow conditions to assess flame stability in the presence of g-jitter. Since the gravitational field is measured simultaneously with these time-resolved Rayleigh measurements, this study allowed flame properties to be correlated with local acceleration to determine the flow conditions and gravity levels under which a given flame is amenable to multi-shot averaging. Two-dimensional temperature information was then extracted from multi-shot Rayleigh measurements made in validated, stable flames.

In the next set of measurements, Raman scattering was utilized to measure CH$_4$ and O$_2$ number densities in the flame anchoring region of two flames that were previously determined to be stable. Although the Raman signal is acquired along a line during a single parabola, two-dimensional profiles can be constructed from multiple measurements and carefully compared with computation.
Finally, for the flow conditions that were found to produce significant soot, soot volume fractions were mapped out with laser-induced incandescence (LII). For these measurements, the time resolution provided by single-shot measurements is needed to characterize fluctuating soot distributions in a time-varying acceleration field. Again, the measurements made can be indexed within the $g$ profile of a given parabola to allow the role of $g$-level and $g$-history to be examined in soot processes.

5.3. Experimental Setup

In this second-round $\mu g$ combustion study a rugged, modular experimental system is developed to be capable of performing a range of two-dimensional laser diagnostics to fully characterize our combustion system. Achieving the desired experimental capabilities required new equipment and a host of modifications to existing $\mu g$ research hardware. Recall that the initial $\mu g$ experiment, described in Ch. 4., was housed in two racks -- an experimental rack and an external control rack. The experimental rack held a windowed combustion vessel that contained the burner and ignition system, along with cameras to make flame measurements. The external control rack contained a microcomputer system to control and monitor the experiment, along with a VCR and video monitor to view the flame in real time. To perform laser measurements in $\mu g$, the experimental rack and external control rack are modified somewhat. However, the most significant change to the previous experiment was the addition of a high-power pulsed Nd:YAG laser, which requires its own mounting rack and associated equipment. This laser produces up to 220 mJ per pulse in the green (at 532 nm) and operates at 10 Hz.
New safety issues for this experiment relate to the use of a Class IV laser. The beam path was entirely enclosed to prevent physical contact and stray reflections, and a special mounting system was built to couple the three experiment racks and ensure proper beam alignment.

A schematic of the experimental equipment rack, for both side and top views, is shown in Figs 5.1, 5.2 and 5.3. A three-axis accelerometer was incorporated into the experiment, mounted to the top of the drop frame to provide a measure of gravitational fluctuations. The only change in the optical setup is the addition of a micro-channel plate image intensifier and corresponding power supply. The image intensifier is used to acquire signal only when the laser is firing, to minimize background contributions from flame luminosity. The pressure vessel now contains three windows, for laser beam entrance and exit as well as flame imaging. Inside the combustion vessel, the burner is mounted to a stepper motor to allow the flame to be translated relative to the laser beam. A light, mounted outside the vessel, was added to verify the position of the burner as it was translated on the stepper motor. Flexible plastic hoses are used to deliver air and fuel to the burner over its full range of motion. A diagram of the pressure vessel interior is shown in Fig. 5.4. A two-way valve is added to the experiment rack to act as a gas-switching manifold, to allow calibration or background gasses to flow through the burner coflow. A cantilevered platform is added to hold a laser power meter, which also acts as a beam stop. Note that the laser beam enclosures on the platform are not shown in these diagrams.
Figure 5.1. Side view of experiment rack used in μg laser diagnostic experiments.
Figure 5.2. Top view of the top two shelves of the modified experiment rack.
Figure 5.3. The bottom shelf of the modified experiment rack, top view.
The ignition system was redesigned in these experiments. First, another hot wire was added such that the flame could be lit with either wire, in which the wire is selected with a switch. With this design, a back-up ignition source exists in the event of breakage. Second, the ceramic tubes are mounted vertically to a 90° rotary solenoid to avoid blocking the laser beam. This igniter is diagrammed in Fig. 5.5.
Figure 5.5. Modified ignition system, utilizing a 90° solenoid and a back-up hot wire.
A laser and optics rack represents the primary addition to the previous experimental setup. The laser used is a Big Sky CFR 400, which is a flashlamp-pumped Nd:YAG capable of producing 220 mJ per pulse at 532 nm and 10 Hz, with a pulse width less than 9 ns and a beam diameter of 7 mm. An optical rail is mounted above the laser head, upon which various lenses and irises can be mounted for beam shaping. This rack is detailed in Figs 5.6 and 5.7, without laser optics. The laser beam, shown in green, is steered through two turning mirrors to increase the path length before entrance into the combustion chamber. This improves the beam quality for measurements. Please note that the laser beam is entirely enclosed for safety reasons, although the beam enclosures are not shown here. Also included in this rack is a restraining bar that prevents the laser power supply from coming forward in the event of a 9 g crash. A fan is installed in
Big Sky Laser
Power Supply
and
Cooling
Unit

CONTROL PANEL

Optical Rail

33.06 "

9 g
Forward
Support

Turning
Mirrors

Bottom Shelf

36.00 "

Figure 5.7. Top view of each shelf of the laser rack.
the bottom shelf to cool the laser head and assure stable laser operation. The second shelf of the laser rack contains a timing unit, a stepper motor controller, fuses for the fan, and a digital logic circuit. The timing unit is a delay generator, which combines with the logic circuit to allow the triggering of the image intensifier and power meter through the computer.

The laser rack and experiment rack must now be mated so the beam can be steered through the entrance window and brought to a focus over the burner centerline. The arrangement of these two racks is shown in Fig. 5.8.

![Diagram](image.png)

**Figure 5.8.** Relative positioning of laser rack and experiment rack aboard the KC-135.
The external control rack, fuel supply, and exhaust system were unchanged from the previous \( \mu \mathrm{g} \) study. With the expansion of equipment and power requirements in the modified experiment, the DC power needs consumed a three-channel power supply, as diagrammed in Fig. 5.9.

![DC Power Diagram](image)

The symbol indicates that the circuit is normally open and is controlled by a relay channel.

**Figure 5.9.** DC power diagram. Relay control not shown.
The direct laser beam is surrounded by opaque or highly absorbing material in all locations. On the laser rack, the beam was blocked on the left and right by shear plates and on the front and top by 1/4” thick Plexiglas (#2423 red) that attenuates stray laser light by more than a factor of $10^4$. Similarly, a box-like beam enclosure was built around the laser power meter on the cantilevered platform.

Several hardware and software modifications were made to expand data acquisition capabilities as well as regulate increasingly intricate experimental systems. These changes are discussed rather than diagrammed in the interest of brevity. Control of the laser and stepper motor took place through the computer’s serial connections in the printer and modem ports. An analog input was added to monitor the laser energy from the power meter, while digital output channels were added to trigger relays capable of gating the image intensifier and turning the vessel light on and off. Additionally, the software was modified to acquire all three acceleration components at 100 Hz, with a 2 ms delay between channels.

5.4. Measurements and Image Processing

Temperature measurements are inferred from Rayleigh scattering, which is an elastic light scattering mechanism that can be used to measure number density. The Rayleigh signal ($S_{\text{Ray}}$) at a given point is directly proportional to number density and Rayleigh cross section ($\sigma_{\text{Ray}}$) of the gas mixture at that point, as discussed in Ch. 2. Since these flame measurements take place at atmospheric pressure, the ideal gas law can be used to relate number density to temperature (T), which gives us an expression for the measured Rayleigh image as
\[ S_{Ray}(x,y) = k_0 \frac{\sigma_{Ray}(x,y)}{T(x,y)} \] (5.1)

where \( k_0 \) is a constant determined from the Rayleigh signal in the ambient air region of the image, where \( T = 300 \) K and \( \sigma_{Ray} \) is known. Computed major species profiles are utilized to correct for the spatial variation in the Rayleigh cross section in a procedure that will be described shortly.

Single-shot Rayleigh measurements were performed for three fuel mixtures, in a 5 mm region located 50 mm above the burner surface. Since instabilities occurring anywhere in the flame are likely to propagate downstream, flame fluctuations are likely to be observed here. Additionally, there is no soot present in this region at any flow condition to interfere with Rayleigh measurements. The 50/50, 65/35, and 100/0 flow conditions were investigated. The single-shot measurements obtained had poor signal to noise, so the background and response-corrected Rayleigh signals were integrated axially. These radial signal profiles from each shot were found to be highly repeatable for the 50/50 and 65/35 flow condition while exhibiting small (~500 \( \mu \text{m} \)) spatial fluctuations in the 100/0 flame.

These characterizations determined the temperature field in non-sooting flames, which were not near the \( \mu \text{g} \) blow-off limit, to be stable under the influence of “g-jitter” at the flow conditions measured. Accordingly, the temperature field could be reliably measured with a 100 shot integration. Beam heights were 7 mm and 10 mm in different flight weeks, each with a beam waist of 300 \( \mu \text{m} \). Laser energies were between 80-100 mJ per pulse, and the intensifier gate time was 2 \( \mu \text{s} \). The imaged region extended radially from the jet centerline out to ambient air. An interference filter (532 nm center, 10 nm bandwidth) was used in the collection optics to discriminate against flame luminosity.
At a given height above the burner surface, three measurements are required to produce a corrected Rayleigh signal. Scattering from air is recorded to account for detector response and vignetting, and a signal from a uniform concentration of helium is acquired to evaluate spurious laser scattering. The background image (RayBG) at this location is then formed from a linear combination of the helium (RayHe) and air shots (RayAir) according to [Long 1993]

$$\text{Ray}_{\text{BG}} = \text{Ray}_{\text{He}} - \frac{\sigma_{\text{Ray},\text{He}}}{\sigma_{\text{Ray},\text{Air}} - \sigma_{\text{Ray},\text{He}}} (\text{Ray}_{\text{Air}} - \text{Ray}_{\text{He}})$$

where $\sigma_{\text{Ray},\text{He}}$ and $\sigma_{\text{Ray},\text{Air}}$ are the Rayleigh cross sections in helium and air, respectively. This ratio in equation (5.2) is equal to 0.017. The Rayleigh signal in the appropriate flame is then imaged (RayFlame) and the corrected Rayleigh image at a given height is

$$S_{\text{Ray}}(x,y) = \frac{\text{Ray}_{\text{Flame}} - \text{Ray}_{\text{BG}}}{\text{Ray}_{\text{Air}} - \text{Ray}_{\text{BG}}}$$

The central portion of the beam was selected at each height and the corrected images from different downstream locations were tiled together to form the full measured field. This process is illustrated in Fig. 5.10. The images shown of the left-hand side were acquired close to the burner, and surface scattering can be seen in the lower right corner of each of the measured images. The corrected image obtained from these three measurements is shown in the top right corner, and the full-field, tiled image is shown beneath it.

A certain amount of structure or “stripiness” can be seen in the tiled image in Fig. 5.10, which is due to non-uniformities in the laser beam. These corrected Rayleigh signals are now inverted to form an initial temperature image, where the Rayleigh cross section is assumed to be spatially constant and the temperature in ambient air is set to
Figure 5.10. Illustration of background and response correction on Rayleigh images.
300 K. In μg especially, changes in beam structure throughout an experiment can make these initial temperature images discontinuous, as can be seen in Fig. 5.11. Since the measured temperature field is known to be continuous, some type of “stripe correction” can be done to account for variations in beam structure. In previous work, signal variations in ambient air were used to correct measured images [Fourguette 1986]. This “ambient” stripe correction is performed here and shown in the middle column of Fig. 5.11. In the upper half of the flame, the edge of the imaged region contains air warmer than 300 K. The resulting ambient stripe-corrected image is seen to have artificially cool temperatures in this region. Furthermore, considerable structure remains in the lower half of this temperature image. This disappointing performance prompted the development of
A new technique, called a radially-integrated stripe correction. This is done by summing
the initial temperature profile radially and fitting this “noisy” curve with a fourth order
polynomial to provide a smoothly varying profile. The “noisy” profile is then scaled and
divided by the smooth profile to provide a “stripe” image that contains beam structure
information. The initial temperature measurement is then scaled and divided by the stripe
image, resulting in a smooth, continuous temperature field, as seen on the right hand side
on Fig 5.11. Note that this procedure does not change the mean values of these
measurements.

The spatial variation of the Rayleigh cross section must now be taken into account.
Major species profiles have been computed over a wide range of flow conditions and
gravity levels, which allow the Rayleigh cross section to be determined as a function of
temperature in these flames. The relative Rayleigh cross section, in which $\sigma_{\text{Ray},N_2=1}$, is
shown for the 65/35 flow condition in Fig 5.12. Since the Rayleigh cross section of
methane is more than twice that of air, the curve’s double-valued nature arises from the
presence of fuel and air at 300 K. The fuel and products are represented in the top branch
of this curve, where $\sigma_{\text{Ray}}$ varies rather steeply, while the bottom branch, the air side,
Before iterative procedure, $\sigma_{\text{Ray}} =$ constant

After iterative procedure, $\sigma_{\text{Ray}}$ varies spatially

1920 K

300 K

Figure 5.13. Effects of iterative correction for spatially-varying $\sigma_{\text{Ray}}$.

varies more slowly. The fuel/products branch was fit to a fourth-order polynomial and the air side was fit with a line to correct the temperature measurements made at each fuel mixture. With these fits, the initial temperature image can be turned into a spatial map of the Rayleigh cross section, which can be combined with the Rayleigh signal according to equation (5.1) to obtain a new temperature field. This process is iterated until a self-consistent temperature field is obtained, which typically takes five or six iterations. The highly visible effects of this iterative procedure are shown for the 65/35 flow condition in Fig. 5.13, which is typically a 10% effect in the high temperature zone.

Measurements were made over a wide range of flow conditions and buoyancy levels, from nearly-sooting flames (75/25) to the normal gravity blow-off limit (40/60) down to the $\mu$g blow-off limit (30/70). Note that aboard the KC-135, flames more dilute than 50/50 will strain out during the high-g pullout portion of each parabola, requiring the
flame to be lit at the start of each low-g portion of the maneuver. Thermometry measurements were made in highly sooting flames (100/0) as well but are not easily interpreted, since computed Rayleigh cross sections as a function of temperature are not available in these flames and soot scattering overwhelms the Rayleigh signal in certain areas of the flame.

Raman scattering, a weak, inelastic process, was used to measure major species concentrations. For these measurements the laser beam was focused into line (300 µm beam waist) with the laser at full power, 220 mJ per pulse at 532 nm. Recall from Ch. 2 that the number density $N$ inferred from the one-dimensional measured Raman signal ($S_{\text{Ram}}$) of a species $i$ can be written as

$$\left[N_i(x)\right]_{\text{Flame}} = \left[N_i\right]_{\text{Re f}} \frac{I_0,\text{Re f}}{I_0,\text{Flame}} \frac{[S_{\text{Ram,}i}(x)]_{\text{Flame}}}{[S_{\text{Ram,}i}(x)]_{\text{Re f}}} \frac{\alpha_{\text{Ram,}i}[T_{\text{Re f}}]}{\sigma_{\text{Ram,}i}[T_{\text{Flame}}(x)]}$$  (5.4)

where the “ref” subscript refers to reference-condition calibration measurements, $I_0$ the laser intensity, and $\sigma_{\text{Ram}}$ the cross section of the Raman-active molecule.

The Raman signal was imaged along the laser line in two distinct spectral regions at separate times. The first measurements used a 630 nm center, 10 nm bandpass filter to measure methane Raman. The second set utilized a 590 nm center, 10 nm bandpass filter to measure oxygen Raman. The 10 nm spectral region centered on the oxygen Raman peak also includes a significant interference from methane Raman scattering. After measurements are made in both spectral regions, the appropriately scaled methane measurement can be subtracted from the $(O_2+CH_4)$ measurement to result in a measured oxygen profile. In both measurements, a uniform concentration of nitrogen was imaged at each height to determine background levels, while a full field of methane or air (as appropriate) was used to determine detector response and to calibrate flame
measurements. The temperature variation of the Raman cross section is not accounted for in these number density determinations, since O₂ and CH₄ exist over a modest temperature range in the flame anchoring region and the spectral window is broad (10 nm). Aboard the KC-135, signals were collected at 30 heights with an integration of 100 shots. For laboratory measurements, data was taken at 150 heights, with the (O₂+CH₄) signal integrated over 600 shots and the methane signal over 200 shots to improve signal-to-noise.

Laser-induced incandescence (LII) measurements were made with the 532 nm laser shaped into different heights with a 300 µm beam waist. A blue-additive filter, which transmitted roughly 90% between 400 nm and 450 nm, was used in the detection system. For all sooty flames studied in normal gravity and microgravity, the LII signal was measured as a function of laser intensity to ascertain an optimum per pulse energy for the determination of soot volume fraction. Since the soot field was observed to fluctuate during “g-jitter” on the KC-135, the LII images taken in µg are single-shot, with a 1 µs gate time. These time-resolved images are indexed with the local acceleration to assess the behavior of these flames in an unsteady gravitational field. Flame luminosity background measurements were made in µg but could not be subtracted from measured LII signals due to the unsteadiness present in the flames. Single-shot LII measurements were made successfully in the 100% CH₄ (100/0) flame, where five measurements could be made during a given low-g parabola. For normal gravity measurements, the soot levels present in methane flames were insufficient to allow for single-shot measurements, so a 100 shot integration was used.
5.5. Results and Discussion

5.5.1. Temperature Profiles

The first temperature measurements of these flames focused on the “standard” flow conditions, 65/35, to investigate the role of buoyancy in this extensively characterized flame. Measured and computed temperature distributions in the 65/35 flame are directly compared in Fig. 5.14. Excellent agreement in flame structure and lift-off height can be seen in the normal gravity flame. In the \( \mu \)g flame, we see the computations successfully predict that when the influence of gravity is removed, the high temperature zone becomes shorter and wider. Stray light reflections prohibited measurements less than 4 mm above the burner surface, but that was sufficient to resolve the flame anchoring region. Measured and computed temperatures are compared more quantitatively in Fig. 5.15, where the centerline temperature profiles are plotted for this 65/35 flame in both 1 g and 0 g. In the normal gravity flame, agreement in lift-off and peak temperature is good, although the flame length is somewhat over-predicted. Agreement in the nonbuoyant flame is excellent -- flame lift-off, the lower peak temperature, and shorter flame length are all well predicted.
65% CH₄:35% N₂ by volume

Figure 5.14. Measured and computed temperature profiles at standard flow conditions.
Figure 5.15. Centerline temperature profiles in 1 g and 0 g for 65/35 flow condition.
As nitrogen is added to the fuel stream in 5% increments (by volume), the normal gravity blow-off limit is reached at the 40/60 flow condition. Temperature fields were then measured and computed at this experimental limit, as can be seen in Fig. 5.16. The normal gravity flame is highly lifted, in strong disagreement with computational prediction. This discrepancy exists to a lesser extent in the µg flame. Figure 5.17 illustrated that in both normal gravity and µg, the computed peak centerline temperature is higher than measurement by 40 K, which is within measurement error. As the fuel mixture is further diluted to 35/65, the flame cannot be experimentally established in normal gravity, including attempts where a less dilute flame was lit and the dilution level was gradually increased. In µg, however, this flame ignites and its temperature profile can be measured. The measured and computed temperature distributions can be seen in Fig. 5.18. Examination of the µg temperature profiles again reveals a large difference in lift-off but reasonable agreement in the location of the flame tip. Visually, the computed µg flame looks hotter than its measured counterpart. These suspicions are verified in Fig. 5.19, where the measured and computed centerline temperature profiles is shown. The computed peak flame temperature is 150 K hotter than measurement. This significantly lower temperature is logically consistent with the occurrence of a small amount of spatial averaging due to flame fluctuations, which would noticeably flatten out temperature peaks in the flame anchoring region. Although the measured flame was visually stable, single-shot Rayleigh measurements were not performed on flames this dilute; accordingly some amount of flame instability in g-jitter is possible at this flow condition.
Figure 5.16. Measured and computed temperature profiles at the normal gravity blow-off limit.
Figure 5.17. Centerline temperature profiles in 1 g and 0 g for 40/60 flow condition.
35% CH₄:65% N₂ by volume

Figure 5.18. Measured and computed temperature profiles at the 35/65 flow condition, beyond the normal gravity blow-off limit.
Figure 5.19. Centerline temperature profiles in 0 g for 35/65 flow condition.
Blow-off limits were explored further on the KC-135. Figure 5.20. illustrates how the 30/70 flame is ignited and fluctuates in the time-varying gravitational field of the KC-135. The 28/72 flow condition was tested as well – for this mixture a flame attached to the hot wire during ignition but did not sustain itself after the ignition source was removed. The 30/70 flame was therefore established as the $\mu$g blow-off limit aboard the KC-135. As seen in Fig. 5.20., the flame moved 1 to 2 mm during a given low-g maneuver. Temperature measurements were performed with shorter integration times (50 shots) in the hope of capturing the flame during a stable period. Unfortunately, the measured 30/70 profile is highly spatially averaged, which yields artificially low flame temperatures. This measurement, shown to illustrate mean spatial characteristics, can be seen in Fig. 5.21. This flame is extremely flat, faint, and highly lifted, anchoring roughly 37 mm above the burner surface, in contrast to the computational prediction. At this flow condition, the low-noise $\mu$g environment of the Space Station may be required to provide a flame stable enough to allow for quantitative, multi-shot measurements.

After these temperature measurements were made at blow-off limits, we now have the ability to compile lift-off heights measured with temperature as well as flame emission over a greater range of flow conditions than shown in Fig. 3.19. This complete compilation of measured and computed lift-off heights is shown in Fig. 5.22. For clarity, error bars are not shown on this plot, although all measurements made with both temperature and emission are within error (based on flame asymmetries) of each other at the 65/35 and 50/50 flow conditions. This Figure highlights the large discrepancies between measurement and computation in dilute flow conditions, both in 1 g and 0 g.
30% CH₄:70% N₂ by volume

The flame attaches to the hot wire igniter at the start of a low-g parabola. The gas mixture continues to burn as the hot wire igniter is retracted.

The flame anchors downstream of the ignition point. Although stable, the flame bounces and changes shape during g-jitter until it strains out after a high-g pullout.

Figure 5.20. Video images of 30/70 flame, the µg blow-off limit aboard the KC-135.
Figure 5.21. Spatially-averaged temperature measurement compared with computation.
Figure 5.22. Complete comparison of measured and computed lift-offs.
5.5.2. Oxygen and Methane Concentrations

Fuel and oxygen Raman measurements were performed in both µg and normal gravity at the 65/35 and 40/60 flow conditions. These conditions were chosen to study air/fuel mixing in a flame that is well predicted (65/35) and at the normal gravity blow-off limit where the computational model has less predictive ability. The predicted and measured oxygen profiles are shown for the 65/35 flame in Fig. 5.23. In the measured normal gravity image, a left-right flame asymmetry can be seen -- the flame anchors in a slightly different location on either side of the jet centerline, while the computations assume perfect axisymmetry. In comparing measured and computed O₂ profiles, the characteristics of the “horn-like” structure of air being entrained into the fuel stream are of particular interest. At this “standard” flow condition in normal gravity, this structure compares well on one side of the flame but not the other. On the right side of the measured profile is a plume of oxygen, which is more concentrated and extends further downstream than any predicted by the computation. In µg, however, the measured flame is nearly symmetric, with the behavior at the fuel/oxygen interface well predicted by computation. The different integration times used in 1 g and 0 g resulted in distinct detection limits, which correspond to the number density equivalent to 1.5% at room temperature in 1 g and 3.5% at room temperature in 0 g. Signal levels beneath these detection limits are displayed as zero in the color scales of the images shown in Fig. 5.23. Fluorescence interferences can be seen in the O₂-free “hot zone” region of each measured oxygen image. In this study, we are interested in regions of the flame where fuel and oxygen coexist, which is distinct from the (T > 1400 K) region where these interfering species are present [Gomez 1987]. Accordingly, the sophisticated measurement procedures and correction schemes available to reduce these interferences are not used.
Figure 5.23. Measured and computed oxygen profiles at standard flow conditions.
65% CH$_4$:35% N$_2$ by volume

$g=9.8\ m/s^2$

$g=0.0\ m/s^2$

Figure 5.24. Measured and computed methane profiles at standard flow conditions.
Figure 5.25. Centerline profiles of measured and computed CH₄ number density.
here [Dibble 1990, Marran 1996]. Figure 5.24 shows the measured and computed methane profiles in normal gravity and $\mu g$. Again a small asymmetry can be seen in the normal gravity profile; otherwise, agreement is excellent. This assessment is confirmed in Fig. 5.25, where the centerline methane profiles are shown. In normal gravity, the measured and computed profiles are separated by a small distance (~500 $\mu m$), with excellent overall agreement. The $\mu g$ centerline plot shows the measured methane curve decaying slightly more quickly than predicted by computation.

For the 40/60 flow condition, fuel/air mixing discrepancies between measurement and computation are greatest in normal gravity. This can be first seen in Figs. 5.26 and 5.27, where the measured and computed oxygen distributions and methane distributions, respectively, are shown. In the measured 1 g flame a significant amount of oxygen mixes with the fuel before ignition occurs -- the fuel travels far downstream until flame stabilization occurs radially away from the jet centerline. In this normal gravity flame, Raman scattering measures a relatively uniform methane concentration just before the flame anchoring region, where the computations predict steep radial concentration gradients. The 1 g computations reveal no evidence of partial premixing or the high degree of liftedness. Microgravity measurements show no oxygen along the jet centerline in the flame anchoring region, in agreement with computational prediction. Additionally, the character of the fuel/oxygen interface appears reasonably well predicted on each side of the flame, despite the measured flame asymmetry. This set of comparisons between measurement and computation further illustrates the experimental sensitivity of these dilute flames to slight flow asymmetries and the computational difficulty of modeling flame ignition and stabilization at these flow conditions. Note that the flame asymmetries seen in the 65/35 and 40/60 flame are not seen in thermometry
40% CH₄:60% N₂ by volume

\[ g = 9.8 \text{ m/s}^2 \quad \text{and} \quad g = 0.0 \text{ m/s}^2 \]

\[ 5.5 \times 10^{18} \text{ cm}^{-3} \]

Figure 5.26. Measured and computed oxygen profiles at the normal gravity blow-off limit.
40% CH₄:60% N₂ by volume

\[ g = 9.8 \text{ m/s}^2 \quad \text{and} \quad g = 0.0 \text{ m/s}^2 \]

Figure 5.27. Measured and computed methane profiles at the normal gravity blow-off limit.
Figure 5.28. Centerline profiles of measured and computed CH$_4$ number density.
measurements since the Rayleigh signal is acquired on one side of the centerline and reflected to fill the domain. The measured and computed methane centerlines are shown in Fig. 5.28. The normal gravity curves reflect the measured partial premixing and the under predicted lift-off height. In the $\mu$g profiles, the fuel decay rate is well predicted, the two curves are separated by a sizable distance (~5 mm). Note that the abrupt changes in the slope of these curves, which occurs near $z = 25$ mm in 1 g and $z = 13$ mm in 0 g, are due to flame asymmetry.

5.5.3. Laser-Induced Incandescence

In making laser-induced incandescence (LII) measurements on the KC-135, single-shot measurements were repeated at multiple times to assess the fluctuations in soot volume fraction and distribution in relation to the time-varying local acceleration. The shot-to-shot variations in measured LII signals in the 100/0 flame, with a 15 mm beam height, are shown in Fig. 5.29. for two different parabolas. In this figure each acquired image can be matched to its g-level and g-history by number. The measured soot concentration and distribution is repeatable for measurements made during similar g-levels and g-histories – for example in images 3, 4, and 8. Therefore, the distribution which results after a long (>= 3 s) period of “smooth” g = 0 is considered to be the “best available” soot measurement in the noisy gravitational field available aboard the KC-135. LII images acquired after a “bumpy” change in g are distinct from the “best available” images. After a gravitational discontinuity, the size of the soot containing region may decrease (images 2 and 6) or increase (image 7). Additionally, the steady rise in acceleration at the end of each parabola tends to decrease the peak soot volume fraction, as seen in images 5 and 10. Flame luminosity images can be acquired in $\mu$g, but due to
Figure 5.29. Single-shot LII measurements performed in 100/0 flame.
flame fluctuations cannot be used to correct measured LII signals; the luminosity background was measured to be less than 5% of the peak LII signal in all cases.

While a 15 mm beam is best for revealing spatial variations over a large spatial area, this expanded beam can produce irregular or asymmetric LII signals, as seen in Fig. 5.29. When measurements are made in a laboratory setting, the beam profile can be measured and used to correct the measured LII profile; this could not be done aboard the KC-135. Accordingly, a smaller beam size (5 mm) was used to produce a cleaner LII signal.

Measured LII signals were tiled together to produce the images shown in Fig. 5.30. On the left are three images taken when g is decreasing linearly, as shown in the acceleration profile in the lower left corner. On the right hand side is the “best available” $\mu g$ soot measurement, with its corresponding gravitational trace in the lower right hand corner of Fig. 5.30. The peak LII signal differs by 50% between these two flames. This “best available” $\mu g$ measurement is shown with the normal gravity LII measurement, which has been corrected for optical throughput and beam non-uniformities in Fig. 5.31. When the influence of gravity is removed, the (uncalibrated) peak soot volume fraction increases by a factor of 15 while the soot-containing region contracts axially and expands radially. LII measurements were made in normal gravity for the 80/20 flow condition, and found the peak soot volume fraction to be a factor of 7 less than that measured in the 100/0 flame in normal gravity. The 80/20 flame was examined in $\mu g$ but produced insufficient soot for viable single-shot measurements. Once probe measurements are made to provide an absolute calibration of measured signals, these LII measurements can be compared with the predictions of computations to assess the validity of existing soot models. Additionally, a low-noise $\mu g$ environment with long time scales may be
Figure 5.30. Tiled LII images: g decreasing (left) and the “best available” g=0 image (right).
100% CH$_4$ by volume

\[ g = 9.8 \text{ m/s}^2 \]

\[ g = 0.0 \text{ m/s}^2 \]

Figure 5.31. Normal gravity and $\mu g$ relative soot volume fraction profiles.
necessary to allow the flame to reach steady-state, as observed in sooty flames by other researchers [Urban 1998].

Recommendations for making LII measurements in diffusion flames were discussed in Ch. 2. Chief among these were laser excitation at 1064 nm, and detection coincident with the laser pulse at a wavelength greater than 600 nm. Not one of these guidelines was followed for practical reasons related to the μg environment in which these experiments were performed. The laser source at 532 nm was selected to be capable of performing Rayleigh thermometry and Raman scattering as well as LII. The time gate and spectral window of detection were severely constrained by the need to make effective single-shot measurements. Spectrally-resolved LII measurements made by other researchers show very little incandescence intensity to exist in the red (> 600 nm) relative to the blue (400-450 nm) [Vander Wal 1994]. Future laboratory-based work could involve making LII measurements with multiple excitation/detection schemes to assess the extent to which large particles biased the relative soot volume fraction determinations of this study. This refined knowledge may become increasingly important as detailed soot computations are performed in this flame.

An experimental challenge for future work will be to make non-intrusive temperature and soot measurements in laminar flames such as these. The processes by which temperature information was extracted from measured Rayleigh signals used in this investigation are likely to be ineffective in sooty flames. This is illustrated in Figure 5.32, which shows measurements made in the 100/0 flame. Here an initial Rayleigh temperature profile, in which $\sigma_{Ray}(x,y) = \sigma_{Ray,Air}$, is shown with the measured relative soot volume fraction. Elastic scattering from large soot particles produces a “hole” in the measured temperature profile at roughly 37 mm above the burner surface along the jet.
**100% CH₄ by volume, normal gravity**

Figure 5.32. Influence of soot particulates on Rayleigh measurements, 100/0 flame.
centerline. Additionally, the presence of soot and soot precursors gives the narrow inner region of the flame an initial temperature that is likely far below its actual temperature. Note that particulate scattering influences the measured Rayleigh signal in upstream regions (z < 30 mm) where little or no laser-induced incandescence is seen. This distinction between the optical properties of soot and soot precursors has been seen by other researchers [Vander Wal 1996b]. While the influence of soot particles on Rayleigh measurements are seen readily in Fig. 5.32., a more subtle effect can be seen in Fig. 5.33. in the 80/20 flame. In this lightly-sooting flame, the initial Rayleigh temperature profile is free of obvious interferences. However, when the uncorrected temperature is compared with the measured LII signal, the inner core of the flame can be seen to have a low initial temperature, as in Fig. 5.31. These comparisons illustrate that a full computational and experimental study of sooting flames require the development of a procedure to correct for the spatial variation of the Rayleigh cross section due to the presence of soot particulates and precursors as well as changes in the local gas mixture.
80% CH$_4$: 20% N$_2$ by volume, normal gravity

Figure 5.33. Influence of soot particulates on Rayleigh measurements, 80/20 flame.
5.6. Conclusions

In conclusion, temperature, oxygen and methane concentration, and relative soot volume fraction were successfully measured in coflow laminar diffusion flames in both normal gravity and \( \mu g \). When steady, low-soot flames can be established in \( \mu g \), the temperature measurements made are as accurate in \( \mu g \) as in 1 g. Although \( \mu g \) constraints limit the spatial region measured and signal-to-noise relative to laboratory major species measurements, the \( \mu g \) images obtained allow many features of air/fuel mixing to be compared with computation. In sum, these temperature and major species measurements afford the most extensive set of comparisons with flame computations to date. Soot measurements have also been made in 1 g and 0 g that await calibration and comparison with future computational predictions.
Chapter 6.
Summary and Conclusions

A range of laminar, axisymmetric, nitrogen-diluted methane/air diffusion flames were successfully characterized with flame emission tomography, Rayleigh and spontaneous Raman scattering, laser-induced fluorescence, and laser-induced incandescence. Measurements were performed in a microgravity (\(\mu g\)) environment as well in normal gravity to study the influence of buoyancy on properties such as flame shape and lift-off, temperature field, fuel/air mixing, and relative soot concentration and spatial distribution. Experimental determinations of flame structure, temperature, and \(\text{CH}_4/\text{O}_2\) concentrations are compared directly with the predictions of state-of-the-art computational models in both normal gravity and \(\mu g\).

Initial experiments measured CH, CH\(^*\), and OH\(^*\) in the well-characterized, normal-gravity, “standard” coflow diffusion flame where the fuel was comprised of 65\% \(\text{CH}_4\) : 35\% \(\text{N}_2\) by volume (denoted 65/35). CH concentration was measured with linear laser-induced fluorescence, while flame emission tomography was used to characterize the CH\(^*\) and OH\(^*\) distribution. Rayleigh scattering was used to calibrate all measurements, resulting in quantitative number densities. The measured peak CH mole fraction was within 20\% of the prediction of GRI Mech 2.11, which is within measurement error. The corresponding agreement with the alternate mechanism was 40\%, a bit outside of measurement uncertainties. However, the spatial distribution of the CH radical was seen to be highly sensitive to choice of kinetic mechanism, and neither prediction matched both the measured flame lift-off height and flame length. Although uncertainties in the production reaction rate coefficients for CH\(^*\) and OH\(^*\) limit our ability to quantitatively predict the optical emission from methane flames to a factor of six, the measured and
computed CH* and OH* distributions are spatially coincident with ground-state CH. Accordingly, detailed measurements of flame structure can be made with the modest experimental setup required for chemiluminescence measurements. These flame emission measurements were made as quantitative as possible through a detailed investigation of optical collection effects and data analysis and interpretation of tomographic inversions.

This initial normal gravity work illustrated that flame emission measurements could be made in a short time (10 s) and in a light weight, compact experimental setup that could be made compatible with existing µg facilities. Therefore, a combined normal gravity / microgravity flame study was conducted, utilizing CH* and OH* as a marker of changes in flame structure occurring when the influence of gravity is removed. In this work a wide range of fuel mixtures were studied, ranging from undiluted, highly sooting flames to dilute, blue and highly lifted flames at the other end of the continuum. This was done to study flames with widely varying lengths, lift-off heights, and sensitivity to the presence of gravity. The results indicated that agreement in measured and computed lift-off and flame shape was excellent at moderately dilute fuel mixtures, such as 65/35. As the fuel is diluted, however, the computations begin to underpredict both the normal gravity lift-off height and the change in lift-off between buoyant and nonbuoyant environments. Previous studies of this flame, which focused on a single fuel mixture and gravity level, failed to realize these discrepancies.

The µg flame structure study, discussed above, increased our experimental expertise and understanding of µg diffusion flame behavior aboard the KC-135 reduced-gravity aircraft. This initial work therefore facilitated the addition of a high power, pulsed laser to our experimental setup to allow for the measurement of temperature, major species,
and soot distributions. A wide range of flow conditions were again examined, although in this research particular attention is paid to determining flammability limits and characterizing the most dilute flame that can be experimentally established.

The first temperature measurement of these flames focused on the “standard” flow conditions, 65/35, to investigate the role of buoyancy in this extensively characterized flame. Excellent agreement in flame structure and lift-off height can be seen in both the normal gravity and µg flame. The corresponding centerline temperature profiles revealed good agreement in peak temperature in 1 g, although the flame length is somewhat over-predicted. Centerline agreement in the nonbuoyant flame was excellent -- flame lift-off, the lower peak temperature, and shorter flame length are all well predicted. As nitrogen is added to the fuel stream in 5% increments (by volume), the normal gravity blow-off limit is reached at the 40/60 flow condition. Here the normal gravity flame is highly lifted, in strong disagreement with computational prediction. This discrepancy exists to a lesser extent in the µg flame. In µg, a temperature profile can be measured at 35/65 as well as 30/70, the µg blow-off limit aboard the KC-135. Though the 35/65 flame was stable, the 30/70 flame moved 1 to 2 mm during a given low-g maneuver. Accordingly, the measured 30/70 profile is spatially averaged, which yields artificially low temperatures. This flame is extremely flat, faint, and highly lifted, anchoring roughly 37 mm above the burner surface. At this flow condition, the low-noise µg environment of the Space Station may be required to provide a flame stable enough to allow for quantitative, multi-shot measurements.

Fuel and oxygen Raman measurements were performed in both µg and normal gravity at the 65/35 and 40/60 flow conditions. These conditions were chosen to study air/fuel mixing in a flame that is well predicted (65/35) and in a dilute flame where the
computational model has less predictive ability. In comparing measured and computed O₂ profiles, the characteristics of the “horn-like” structure of air being entrained into the fuel stream are of particular interest. At this “standard” flow condition in normal gravity, this structure compares well on one side of the flame but not the other. On the right side of the measured flame is a plume of oxygen which is more concentrated and extends further downstream than any predicted by the computation. This fuel-air mixing study has therefore revealed disparities between measurement and computation not seen previously, even in the otherwise well-predicted 65/35 flame. In µg, however, the measured flame is nearly symmetric, with the behavior at the fuel/oxygen interface well predicted by computation. Measurements at the 40/60 fuel condition revealed that partial premixing occurs in the normal gravity flame, which was not predicted by computation.

Laser-induced incandescence (LII) measurements were made on the KC-135, in which single-shots were repeated at multiple times to assess the fluctuations in soot volume fraction and distribution in relation to the time-varying local acceleration in the 100/0 flame. In a given 15 mm region above the burner surface, the peak soot volume fraction can vary by as much as 50% over the course of a low-g parabola. However, the measured soot concentration and distribution is repeatable for measurements made during similar g-levels and “g-histories.” Therefore, the distribution which results after a long (3 s) period of “g = 0” is considered to be the “best available” soot measurement in the noisy gravitational field available aboard the KC-135. When the influence of gravity is removed, the (uncalibrated) peak soot volume fraction increases by a factor of 15 while the soot-containing region contracts axially and expands radially. These measurements can soon be compared with the predictions of computations to assess the validity of existing soot models. Additionally, a low-noise µg environment with long time scales
may be necessary to allow the flame to reach steady-state, as observed in sooty flames by other researchers.

Perhaps the most important conclusion of this work is that many discrepancies exist between computation and experiment on a fundamental, visible level, namely in areas of flame lift-off and shape, that can be quickly revealed by flame emission experiments. A single flame model should be able to predict the behavior of all of the non-sooting flames investigated here -- accordingly this research has found problems in existing flame models which were not seen previously. Future fundamental coflow diffusion flame studies should involve fully characterized boundary conditions, which include velocity measurements, as well as computational sensitivity analysis to these boundary conditions in an attempt to better compare measurement and computation. Additionally, the sensitivity of computed lift-off to the strain rate at extinction should be calculated, as well as laminar flame speeds, to further evaluate flame stabilization mechanisms.

An experimental challenge for future work will be to make complementary non-intrusive temperature and soot measurements in highly sooting flames such as the 100/0 flame. Future soot studies could involve making LII measurements with multiple excitation/detection schemes to assess the extent to which large particles are biasing the relative soot volume fraction determinations of this study, while the role of soot particulates and precursors on the Rayleigh signal must be accounted for to make temperature measurements in these flames.
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