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THE UNIVERSITY OF OKLAHOMA

GRADUATE COLLEGE

**AN EXPERIMENTAL AND NUMERICAL INVESTIGATION ON
HYDROGEN-HYDROCARBON COMPOSITE FUEL
COMBUSTION**

A Dissertation

SUBMITTED TO THE GRADUATE FACULTY

in partial fulfillment of the requirements for the

Degree of

Doctor of Philosophy

By

AHSAN REZA CHOUDHURI

Norman, Oklahoma

2000

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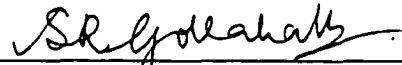
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AN EXPERIMENTAL AND NUMERICAL INVESTIGATION ON HYDROGEN-HYDROCARBON COMPOSITE FUEL COMBUSTION

A Dissertation APPROVED FOR
THE SCHOOL OF AEROSPACE AND MECHANICAL ENGINEERING

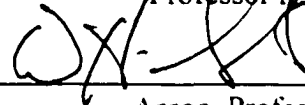
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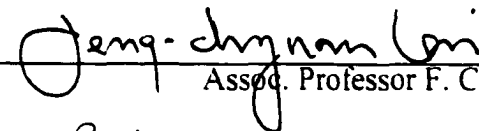
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*In the name of God, the most gracious and the most Merciful
"So glory to Him, in whose hand is the dominion of all things: and to Him will ye be all brought back"*

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**This dissertation is dedicated to
my loving wife Dr. Ruma Ahsan**

* Would you tell me, please which way I ought to go from here?* Asked Alice
That depends a good deal on where you want to get to. said the Chesire Cat.

Alice in Wonderland, Lewis Carroll, 1865

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Nomenclature

English

A	Placzek coefficient
A_{12}	Einstein spontaneous emission coefficient
a_i	Fit coefficient for collider species
B_H	Function of mass diffusivity of atom H
B_O	Function of mass diffusivity of atom O
B_{OH}	Function of mass diffusivity of radical OH
B_{ge}	Einstein absorption coefficient
C	Group of experimental constant. Constant of proportionality
c	Speed of the light
C_r	Concentration of the reactant mixture
D_H	Binary diffusion coefficient of H atom
D_O	Binary diffusion coefficient of O atom
D_{OH}	Binary diffusion coefficient of OH radical
d_i	Burner diameter
E_v	Vibrational energy of a molecule
f	Mixture fraction
F	Radiative heat loss factor
Fr	Froude number
Fr_f	Global flame Froude number
Fr_j	Jet exit Froude number
G	Vibrational term value
g_i	Nuclear Spin Degeneracy
h	Plank's constant
h	Static enthalpy (<i>Chapter Four</i>)
h	Flame length (<i>Chapter Five</i>)
H	Characteristics Flame Length (<i>Chapter Five</i>)
I_L	Incident laser energy
J	Rotational quantum number
k	Boltzman constant (<i>Chapter Three</i>)
k	Turbulent kinetic energy
k'	General rate of reaction for hydrocarbon fuel
l	Length scale (<i>Chapter Four</i>)
L^*	Nondimensional flame length
L_f	Visible flame length
L_H	Normalized lift-off height
L_n	Normalized blowout velocity
m	Mass
M	Mixture molecular weight

n_A	Number of molecules excited along the specific portion of the incident beam
P	Predissociation from the excited level
p	Pressure
P	Rate of production of turbulent kinetic energy
Q	Collisional quenching
q	Velocity scale (<i>Chapter Four</i>)
Q_R	Total flame radiation
Q_{rot}	Rotational partition function
Q_{vib}	Vibrational partition function
r	stoichiometric ratio (<i>Chapter Five</i>)
r_f	Burner radius
Re_H	Reynolds number based on characteristics flame length H
Re	Reynolds number
Re_j	Jet exit Reynolds number
S	Stimulated emission
$(S_L)_{max}$	Maximum laminar flame speed for pure fuel
S_f	Fluorescence signal
S_R	Raman signal
S_T	Turbulent flame speed
SV	Stern-Volmer Coefficients of fluorescence quantum yield
T	Local flame temperature
t	Time
T_∞	Surrounding temperature
T_{ad}	Adiabatic flame temperature
T_f	Flame temperature
U	Axial component of the mean velocity
u	Cartesian component of velocity in x-direction
u'	Axial turbulent fluctuation
U_{abs}	Absolute velocity
U_b	Blowout velocity
U_c	Centerline velocity
U_j	Jet Exit velocity
V	Radial component of the mean velocity
X_i	Species mole fraction
x	Cartesian coordinate
y	Cartesian coordinate
Y	Mass fraction
Y_∞	Stoichiometric mixture fraction

Greek

ν	Frequency
Ω	Collection solid angle

χ	Mole fraction of OH
ν	Vibrational quantum number
ρ	Density
μ	Molecular (laminar) dynamic viscosity
Γ	Turbulent diffusion coefficient
ε	Rate of turbulence dissipation (<i>Chapter Four</i>)
ϵ	Emissivity
ξ	Fuel and Air density ratio (ρ_F/ρ_A)
$\Delta\nu$	Absorption line width
ρ_A	Air Density
ρ_F	Fuel Density
η_D	Detection efficiency
μ_F	Dynamic viscosity of fuel
σ_{ge}	Absorption cross-section
ν_{ge}	Transition frequency
β_{ge}	Boltzman fraction
δ_{ij}	Kronekar Delta
Δl	Portion of the beam where molecules are excited
ν_L	Incident laser frequency
σ_q	Collision cross-section
η_T	Transmission efficiency
μ_t	Eddy (turbulent) dynamic viscosity
σ_t	Turbulent Prandtl number

Acronyms

BBO	Beta Barium Borate
CCD	Charged Coupled Device
CFI	Equivalence ratio based on CH ₄ only
EFI	Equivalent Equivalence ratio
EI	Emission Index
HFI	Equivalence ratio based on H ₂ only
ICCD	Intensified Charged Coupled Device
LIF	Laser Induced Fluorescence
OPO	Optical Parametric Oscillator
PAH	Polycyclic Aromatic Hydrocarbons
PLIF	Planar Laser Induced Fluorescence
PMT	Photomultiplier Tube

Abstract

An experimental and numerical study on the combustion characteristics of turbulent diffusion flames of natural gas-hydrogen composite fuel is presented. Three mixtures (90-10%, 80-20% and 65-35% by volume) of natural gas and hydrogen were used. The results are compared with the combustion characteristics of a pure natural gas flame. The following parameters were measured: (i) flame stability (blowout velocity, and lift-off height at blowout condition), (ii) temperature field (radial profiles at three axial locations), (iii) composition profiles of stable species (CO_2 , CO , NO , O_2), (iv) composition profiles of intermediate species (OH , CH , H and O), and (iv) visible flame length, flame radiation, emission indices and volumetric soot concentration. To study the flame stability five burners of 1mm, 2.3 mm, 3.8mm and 4.5 mm ID were used. Direct video photography, Schlieren imaging and acetone Planar Laser Induced Fluorescence (PLIF) imaging were used for flame stability and mixing study. For stable species concentration measurements, an uncooled quartz glass probe with chemiluminescence and infrared analyzers were used. Laser Induced Fluorescence (LIF) and Planar Laser Induced Fluorescence (PLIF) technique were used to measure radical concentrations. A combined LIF-Raman Spectroscopic procedure was carried out to quantify the LIF signals.

The following parameters were analyzed numerically: (i) cold jet mixing (axial and radial velocity, turbulent intensity, turbulent kinetic energy and local equivalence ratio), (ii) flame temperature (iii) stable species (CO_2 , CO , NO , O_2) and (iv) intermediate species concentrations (OH , CH , CN , H , and O). For the numerical computation, Favre-averaged Navier-Stokes equations with two-step reaction kinetics and the standard k - ϵ turbulence

model were used. The fuel jet exit Reynolds number was kept constant at 8700 for flame structure measurements, computation, and measurements of global characteristics. The corresponding flame Froude number ranged between 0.85-1.18 depending upon the mixtures of natural gas and hydrogen.

Both blowout velocity and lift-off height at blowout condition increase non-linearly with the increase of hydrogen concentration in the mixture for all burner sizes. A general relation is also presented correlating blowout velocity, burner diameter, and hydrogen content in the fuel mixtures. The lift-off height at blowout decreases with the increase of hydrogen concentration in the mixture. The numerical prediction shows that with an increase of hydrogen concentration in the fuel mixture, the axial velocity decays faster while the axial turbulent fluctuations and local turbulent flame speed increase.

Experimental measurements and numerical predictions show that the local flame temperature increases with the increase of hydrogen content in the mixture; consequently the NO production increases in the flame. On the other hand, an increase of hydrogen content in the mixture increases OH, H and O radical concentrations, which increase CO and soot oxidation. Also, the increase in OH, H and O concentrations enhances the flame stability by increasing the laminar flame speed of the composite fuel. The visible flame length, radiative heat loss, and volumetric soot loading decrease with the increase of hydrogen concentration in the fuel mixture. The CO emission index decreases and NO emission increases with the increase of hydrogen content in the fuel mixture.

Chapter One

Introduction

In the past few decades, hydrogen has received increased attention as a potential alternative for fossil fuel based power generation. Researchers around the world advocate hydrogen use because it can be produced from abundant sources and it is environmentally safe and clean. Moreover, hydrogen has 2.6 times the energy per unit mass compared to gasoline. From the point of view of combustion, hydrogen possesses superior characteristics to any other hydrocarbon fuel in terms of ignitability, low ignition delay, and higher flame stability. These inherent advantages turn it as the only potential fuel for hypersonic (SCRAM JET) as well as supersonic propulsion (RAMJET). However, its higher flammability and lower volumetric energy density make the on-board storage system complicated for both ground transportation and aircraft propulsion applications. Currently, two types of storage techniques have been proven technically feasible for the on-board storage of hydrogen: compressed H_2 and liquid H_2 . However, both these storage systems impose somewhat extreme conditions on fuel tanks. Compressed hydrogen needs 4 times the volume of gasoline for the same amount of energy. On the other hand, liquid storage of hydrogen extends this problem to cryogenic fuel transportation and metering. Since hydrogen application imposes severe volume and weight constraints, a strong motivation exists for the development of a hydrocarbon fuel-based airframe integrated SCRAM JET (Billing, 1993; Karagozian, 1992; Kay et al., 1969; Kay et al., 1971; Takahashi, 1995; Vinogradov et al., 1996; Waltrup, 1987). Dual Combustor Ramjets (DCR), which combine features of both

ramjet, and SCRAMJET are considered to offer a reasonable solution, especially when hydrocarbon fuels are employed. However, they also impose the limitation of heavier and complicated engine design. In this context, a hydrogen-hydrocarbon composite fuel might be a possible solution. The combustion characteristics of this mixed fuel might be extended toward both subsonic to supersonic regimes. The relatively slower reaction rate of a typical hydrocarbon fuel can be accelerated by mixing it with hydrogen, which results in an improved ignitability and flame holding (Bonghi et al., 1995; Burrows and Kurkov, 1996; Vinogradov, 1992). The above discussion prompts further investigations on the combinations of hydrogen-hydrocarbon hybrid fuels in high-speed propulsion systems. Besides, a recent investigation demonstrates the improved performance of internal combustion engines while burning a hydrocarbon fuel together with hydrogen than hydrocarbon alone (Shrestha and Karim, 1997). This study shows that the mixed fuel yields low CO and NO_x emissions and higher brake horsepower along with lower specific fuel consumption than the conventional fuel. However, none of the studies has reported physico-chemical mechanisms of the combustion of the mixed fuels causing these performance improvements. Although it might be assumed that the combustion characteristics of a mixed fuel lie between those of the two fuels components, the complex and nonlinear nature of chemical kinetics and multi-component diffusion mechanism makes it difficult to predict the performance of the mixed fuel.

The primary objective of the present study is to understand and quantify the combustion characteristics of hydrogen-hydrocarbon hybrid fuels. The previous studies done by the author were mainly focused to delineate the global combustion parameters such as

pollutant indices, flame structure, flame radiation etc.. (Choudhuri and Gollahalli, 1997, 1998a, 1998b, 2000a, 2000b; Choudhuri, 1997). In addition, intermediate radical and atom concentrations were measured using Laser Induced Fluorescence to observe the effects of hydrogen addition on hydrocarbon flame chemistry (Choudhuri and Gollahalli, 2000b, 2000c). A computational study with multi-step reaction kinetics was also conducted earlier to understand the effects of hydrogen addition on the hydrocarbon reaction mechanism (Choudhuri and Gollahalli, 2000b). To avoid the complexity of flow dynamics, all these previous studies were conducted in the laminar flame configuration. However, in most practical combustors flames are turbulent. In turbulent combustion, mixing is greatly enhanced. As a consequence, the combustion characteristics are significantly different from those of the laminar flame. The turbulent non-premixed combustion of the hydrogen-hydrocarbon hybrid fuel is investigated in the dissertation. The correlations between the flame stability and hydrogen content in the mixture at different burner diameters are studied. The effects of isothermal turbulent mixing on flame stability are also investigated.

1.1 COMPOSITE FUEL

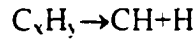
Generally, mixtures of two or more fuels are termed as *composite fuels*. Although in most cases the term *composite fuel* or *hybrid fuel* implies a mixture of two homogeneous phase fuels (gas-gas or liquid-liquid), the application of some heterogeneous composite fuels such as *cofiring of pulverized coal with natural gas* has wide practical applications. Most of the previous work on composite fuels concentrated on either liquid-liquid, such as gasoline-ethanol or gas-solid like coal-natural gas (Bayless et al., 1996), because of

the increased use of such fuels in practical combustors. Little information is available in literature on the combustion characteristics of composite gas fuels, presumably because it was thought that composite gas fuels have no practical significance. However, some recently developed techniques for storing natural gas (NG) as a liquid solution with other hydrocarbons at ambient temperature (Starling and Mallinson, 1995; Horstkamp & Mallinson, 1997) lead to the practicality of the combustion of composite-gas fuels. Furthermore, burning hydrogen with hydrocarbon fuels possesses some advantages such as low nitrogen oxides formation and improved control on flammability and flash back (Choudhuri 1997; Karim, et al., 1996; Karim, et al., 1995; Varde, 1981). Although it may be expected that the combustion characteristics of the hybrid fuel will fall between those of the two primary fuels, some of the previous investigations have revealed that composite fuels have radically different characteristics from those of the original fuels (Choudhuri and Gollahalli, 2000a). Hence a detailed study of the turbulent combustion characteristics of composite fuel is warranted.

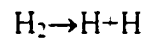
1.2 COMBUSTION OF HYDROGEN-HYDROCARBON COMPOSITE FUEL

In many aspects, the combustion of a composite fuel is different from that of its base fuels. The interactions of different chemical kinetics of each primary fuel of the composite fuel along with the flow dynamics significantly alter the combustion characteristics. For instance, hydrocarbon fuels have specific reaction routes to complete combustion. Although hydrogen chemistry is already embedded in hydrocarbon reaction mechanism, the presence of additional hydrogen as a fuel in composite mixture changes the order of importance or more specifically sensitivity of certain elementary reactions.

The hydrocarbon fuel reaction mechanism proposed by Peter (1993), is shown in Appendix-I. The reaction mechanism shows the importance of H atom concentration throughout the combustion process. In typical hydrocarbon fuel combustion, the H atom forms through an initial global reaction (Penko et al., 2000) given by



and then serves as a chain carrier. However, if hydrogen is present in the fuel along with other hydrocarbon fuels, another initial global reaction for hydrogen reaction becomes active



The addition of this reaction as an initial global reaction changes the priority of the reaction mechanism. For instance it influences the hydrocarbon reaction mechanism by accelerating the CH_3 consumption reaction (R43, R45, R48) without waiting to have sufficient H supplied from the initial hydrocarbon fuel breakup. Furthermore, the increase of CH_3 consumption gives an additional boost to fuel break up. Hence the effects of hydrogen addition become dominant because of the complex propagation of the reaction mechanism. This observation was evident in an earlier numerical and experimental investigation (Choudhuri and Gollahalli, 2000b). However, an important aspect that is prevalent in practical combustion system was not addressed in the earlier investigations namely, the effect of turbulence. Since flow configurations in most of the practical combustors are turbulent, information about turbulence-chemistry interaction is necessary to successfully employ the composite fuel in actual combustion environments. It can be expected that the presence of turbulence will significantly affect combustion. For instance, the addition of hydrogen tends to increase the stability of a turbulent flame.

To explain this phenomenon, information about both flame propagation and turbulent mixing are required. As mentioned earlier, very few fundamental studies are available in the literature describing the combustion mechanism of hydrogen-hydrocarbon fuels particularly effect of turbulence on chemistry in composite fuel flame. In this dissertation, an effort is made to address some of these issues.

1.3 OBJECTIVES

As pointed out earlier, flow-field turbulence and flame chemistry are highly interrelated. Three-dimensional velocity fluctuations cause rapid mixing in turbulent flames and thereby alter the production and destruction rates of intermediate radicals and atoms. The project is aimed at understanding the transient concentration of key intermediate radicals (OH, CH, H, O) in turbulent composite fuel flames. In fact, the rate of formation and destruction of these radicals dominates the combustion characteristics (flame stability, pollution emission, flame radiation etc.) of the composite fuels. The proposed study investigates the flame stability and its relation with cold jet mixing process. The global combustion characteristics such as pollution emission and flame radiation are also reported for different concentrations of hydrogen in a composite fuel mixture.

The specific objectives of the proposed project are:

- To quantify the stability criteria of hydrogen-natural gas composite fuel flames and to establish correlations between the flame stability and local fuel mass fraction based on cold jet mixing process.

- To understand the destruction and formation of the intermediate radicals (OH, CH and atoms (O, H) in hydrogen-hydrocarbon turbulent diffusion flames.
- To report the global combustion characteristics (pollution emission and flame radiation) of hydrogen-natural gas composite fuels.

1.4 PRACTICAL IMPLICATIONS OF THE PROJECT

The hydrogen-hydrocarbon fuel has many practical implications ranging from terrestrial to aerospace power generation. For instance, hydrogen-assisted burning of a hydrocarbon fuel can be used in lean burning gas turbine combustors for power generation. Also, as discussed earlier hydrogen-hydrocarbon composite fuel is an ideal candidate for hydrocarbon-based SCRAM jet engine. Besides, this composite fuel might be useful in microrocket applications in which fast combustion is required due to small combustion chamber length; however, hydrogen cannot be used alone because of storage problem. This investigation will provide an insight into the behavior of the combustion of a composite fuel from both the kinetics and the flow dynamics points of view. The stability behavior of hydrogen-hydrocarbon composite fuel presented here will certainly help in the design of combustors with increased flame stability. Also, the flame structure and pollution characteristics of the hydrogen-hydrocarbon flame studied here will establish a fundamental basis to understand and characterize environmentally safe combustion systems that employ composite fuels.

1.5 ORGANIZATION OF THE DISSERTATION

This chapter has introduced the concept of hydrogen-hydrocarbon composite fuel, and provided a preliminary indication of the potential benefits of such fuel. Also, various issues regarding the prediction of combustion performance of hydrogen-hydrocarbon composite fuels have been discussed.

Chapter Two presents an overview of the experimental technique used in the present investigation. The details of the experimental setup, instrumentation and laser diagnostic techniques such as Laser Induced Fluorescence and Raman Spectroscopy are discussed. The theoretical backgrounds of the spectroscopic technique, which are essential for employing LIF and Raman Spectroscopy in the present investigation, are also presented.

In *Chapter Three*, the spectroscopic modeling techniques for the quantification of LIF signal are presented. This chapter discusses the combined LIF and Raman Spectroscopy to quantify the collisional quenching encountered in LIF processes.

Chapter Four discusses the numerical methods used on the present investigation. The details of governing equations, turbulence modeling, reaction models, turbulence-combustion interaction and solution algorithm are presented. Also, this chapter describes the validation study of the numerical scheme used in the present study.

Chapter Five presents the stability of the flames for different concentration of hydrogen in the composite fuel mixture. The correlations of blowout velocity, burner diameter and the hydrogen concentration in the mixture are shown. Also, the stability mechanism is discussed in the background of cold jet flow field, which is computed numerically.

Chapter Six describes the thermo-chemical structure of the hydrogen-hydrocarbon turbulent diffusion flames. Measured and computed flame temperature, concentration of intermediate radicals (OH, CH, H, and O) and stable species (CO₂, CO, NO, O₂) are presented. The computed radial and axial profiles of mean absolute velocity, axial velocity and turbulent fluctuation for different concentrations of hydrogen in the fuel mixture are also presented.

Chapter Seven describes the global combustion characteristics such as visible flame length, pollution emission, soot formation and flame radiation of composite fuel flames. The emission indices of NO and CO are also presented for different mixture conditions.

In *Chapter Eight* a general discussion about the outcome of the dissertation is presented. Conclusions and future recommendations are also included in this chapter.

Chapter Two

Experimental Methods and Setup

Various combustion diagnostic techniques have been used in the present study, which includes Schlieren Deflectometry, Laser Spectroscopy, Infrared and Chemiluminescence Analyzers and Thermocouple based temperature measurement. In this chapter, the details of the experimental setup along with the background of measurement techniques are discussed. Also, the specifications of the instruments used in this investigation and the estimated measurement uncertainties (Appendix II) are given in Table 2.1 and Table 2.2 respectively. The nominal operating conditions and test matrix are shown in Table 2.3 and 2.4. Also, photographs of experimental setup and instrumentation are presented in PT2.1 and PT 2.3.

2.1 LABORATORY COMBUSTION CHAMBER

The circular fuel burners used in these experiments consisted of stainless steel tubes of 1 mm, 2.3 mm, 3.8 mm, and 4.5 mm ID, through which fuel was injected into an atmosphere of air. The burner was projected 14.5 cm above the chamber floor. The burner was located in a vertical steel combustion chamber (Fig. 2.1a) of 76 cm $\times$ 76 cm cross-section and 163 cm height. The chamber was fitted with rectangular windows of dimensions 20 cm $\times$ 20 cm $\times$ 145 cm on all of its four side-walls. Three of the windows were fitted with Pyrex plate glass and the fourth was fitted with a slotted metal sheet for introducing probes and a laser beam. Air was induced by natural convection into the test chamber through a 20 cm diameter circular opening in the base plate. Three layers of fine-wire-mesh screens were used to provide a uniform flow. The velocity distribution of

the incoming air inside the chamber at a height of 14.5 cm (burner exit) is given in Fig. 2.1(b). The velocity profile was measured with a hot wire anemometer while a natural gas flame issued from a 3.8 mm burner ($Re=8700$) was burning inside the chamber. The velocity profile indicates a uniform velocity in the central zone of the laboratory combustion chamber. The top of the combustion chamber was open to the atmosphere through an exhaust duct. A relay-operated butterfly valve was used to open the exhaust duct during experiments. The ambient pressure of the laboratory was maintained slightly above the atmospheric pressure to ensure positive draft inside the test chamber.

2.2 FUEL GAS SUPPLY TRAIN

The fuel gas supply train consisted of compressed natural gas (CNG), propane and hydrogen cylinders, pressure regulators, rotameters, a mixing chamber, inline filters and tubings. Figure 2.1(a) shows the fuel supply train with the laboratory combustion chamber. Natural gas was stored in a specially designed (fiber reinforced) CNG cylinder at a cylinder rated at 13 MPa. The hydrogen cylinder was rated at 16 MPa. By using two-stage pressure regulators, the downstream pressure in the fuel line was always kept constant at 0.3 MPa. The two fuels were mixed inside an annular mixing device in which secondary fuel was injected into the stream of primary fuel through a concentrically located injector. The length of the mixing device was sufficiently large (>150 hydraulic dia.) to ensure a homogenous mixture of fuels. The mixed fuels were then supplied to the burner through Teflon™ tubing. For a fixed jet exit Reynolds number, the volume flow rate of the hybrid fuel was calculated for different mixture conditions. The volume

flow rates of the primary and secondary fuels were then regulated with calibrated rotameters (Cole Parmer). The compositions of the base fuels are listed in Table 3.

2.3 SCHLIEREN DEFLECTOMETRY

Both color and gray scale Schlieren Deflectometry (RSD) (Shenoy et al., 1998; Al-Ammar et al., 1998; Rohmat et al., 1998; and Agrawal et al., 1998) were used for flame visualization. For sooty hydrocarbon flames, both flame boundary and schlieren boundary could be observed through this system. The Schlieren system is shown in Fig. 2.2a. Light (150-W continuous halogen light source connected with a 200 μm diameter fiber optic cable) from a slit aperture (50 μm) was collimated using an achromatic lens (diameter = 63 mm, focal length = 490 mm) and deflected while passing through the flame due to the variable density generated refractive index gradient. A decollimating achromatic (diameter = 63 mm, focal length = 490 mm) lens was used to refocus the light and form a deflected source image on the filter plane. The filter was a transparent film with a computer-generated continuous spectrum colored rectangular strip. The green part of the filter was used as the background (undisturbed) color in the system. Light was focused through the red region. Other colors appearing on the image showed the deflection of the incident light due to the refraction index gradient of the test medium. The filter was placed vertically to detect only the horizontal density-gradient field since the dominant mean density gradient of most of the flow field was horizontal. The image was acquired with a CCD detector. The image was acquired on-line using a high-speed color frame grabber (Matrox) in RGB format. In this experiment 25-ms time

width was used to grab each image frame. The schlieren setup and the filter are shown in Fig 2.2.

2.4 LASER SPECTROSCOPY

For combustion studies, laser spectroscopic techniques possess some unique advantages over other conventional experimental methods. Laser-based spectroscopic techniques are inherently non-intrusive and have high temporal and spatial resolution. Spectroscopic techniques such as Laser Induced Fluorescence (LIF), Raman Spectroscopy, Rayleigh Scattering can be used to measure temperature, species concentration and velocity fields of combustion systems. Hence, these techniques offer a unique opportunity to measure precisely temperature in laminar flows (by averaging over sufficient laser shots) and accurate probability functions (PDFs) in turbulent flows (by compiling single-shot data at one spatial location). In the present study, Laser Induced Fluorescence (LIF) and Raman Spectroscopy have been used for in-flame temperature and concentration measurements.

2.4.1 Laser Induced Fluorescence (LIF)

Laser Induced Fluorescence is one of the most developed and widely used resonant spectroscopic technique. This technique requires excitation of the tested species by a laser source that is resonant with a selected transition of interest. By measuring the strength of the fluorescence signal, the population density of the upper state is determined which actually bears some or all information of the thermodynamical properties of the tested medium. In LIF, a narrowband laser is tuned to a certain wavelength, which excites one or more transitions in the tested atom or molecule. Also,

the use of a spectrally narrow bandwidth allows individual rotational transitions that originate from certain vibrational and electronic levels to be excited. In the absence of competing energy loss mechanism, the population of the excited state, and hence the emitted fluorescence, are proportional to population at the ground state. And, at thermodynamic equilibrium, the population of an individual rotational state is proportional to the total density of the probed species and its temperature. Since the measured LIF signal is emitted by only the excited species an optical detection can be designed to detect signal from a particular wavelength, thus, the signal to noise ratio improves greatly. LIF is preferable among other spectroscopic techniques because of its excellent temporal (10 ns) and spatial resolution (0.1mm-1.0 mm) and high signal-to-noise ratio. Depending on the application, either single-point or multiple point measurements are possible. Pointwise LIF measurements provide information about flame structure and characteristics of combustion systems. Furthermore, by expanding the laser beam into a light sheet and using a camera to detect the fluorescence, imaging of the molecular properties within the laser light sheet is possible. Planar Laser Induced Fluorescence (PILF) can be used to quantitatively characterize a turbulent combustion system. Detailed reviews of LIF techniques can be found in references (Hanson and Setizman, 1990; Dyer and Crosley, 1985; Battles et al. 1994; and Hanson, 1997).

The energy levels that participate in typical LIF experiments are shown Fig. 2.3. A narrowband laser beam is tuned until its photon energy $h\nu_{ge}$ matches the energy difference between the ground state g and the excited (upper) state e . If the single-photon transitions between these states are allowed, a part of the population in the ground state is excited. Once in the upper state, the excited species may relax

spontaneously to any of the lower states. Alternatively, it may give up its energy by dissociation or ionization, or may be removed from this state by collisionally exchanging energy with another object. The latter is known as collisional quenching, and includes primarily collisions with other molecules and atoms. Figure 2.3 shows spontaneous emission to three energy levels. In some complex molecules the entire LIF process may involve tens or even hundreds of energy levels, however, each of these spontaneous transitions is decoupled from the other transitions. The entire LIF process can therefore be modeled as isolated radiative interactions consisting of separate two-level systems.

The successful implementation of LIF technique requires a definite measurement methodology for a particular species, properly designed laser source, detection system and control electronics. The LIF methodology and instrumentation used in the present investigation are discussed below. Figure 2.4a shows the schematic of the LIF measurement setup.

2.4.1.1 Measurement Methodology

For the LIF measurements of the radicals of interest, the spectral features of the transitions were carefully selected to avoid interference from the adjacent lines of the same species or underlying lines from other molecules. The details of the transitions (Table 2.5) and signal modeling used in this experiment are discussed below.

OH: For OH measurements, the excitation of (1.0) band of $A^2\Sigma^+ \leftarrow X^2\Pi$ system combined with detection of the strong (1.1) band has been used. The well-isolated $P_1(8)$

line at 285.265 nm was excited and the fluorescence signals were collected near 315 nm. In OH fluorescence measurement the laser energy was attenuated below 100 μ J/pulse to keep the OH fluorescence in the linear regime (Heard et al., 1992).

CH: The transition of (0,0) band near 431 nm of the $A^2\Delta \leftarrow X^2\Pi$ system has been used for CH measurement. In this case the transition is highly diagonal: so the excitation and the detection have been done on the same band. Since in the present investigation the detection was performed by a narrowband spectrograph, the signal was not contaminated by the on-resonance fluorescence signals of the nearby PAH (polycyclic aromatic hydrocarbons).

H: For H atom measurements, the two-photon excitation of the $1s \rightarrow 2s$ transition at 243 nm was used. The signal was collected near 656 nm ($2s \rightarrow 3p$).

O: For O atom measurements the two-photon excitation near 226 nm was used with the detection at 845 nm ($3s \rightarrow 3S$). The laser energy was kept below 200 μ J/pulse to avoid the interference from the strong adjacent NO band (1,1) of $A^2\Delta \leftarrow X^2\Pi$ system (Heard et al., 1992). However, at this excitation wavelength, it is not possible to avoid the interference of the photolytic production of O (Smyth and Tjossem, 1990).

2.4.1.2 The Laser System and Focusing Optics

The laser system used in this experiment consisted of a pulsed Nd:YAG laser pumped (GCR 200, Quanta Ray, Spectra Physics Inc.) Optical Parametric oscillator (OPO) with

frequency doubler option (MOPO-730, Quanta-Ray, Spectra Physics), which has a tunability range from 190 nm to 2000 nm (Fig. 2.4a). The tuning of laser beam through OPO entirely depends upon the angle of the OPO crystals, which gives a superior flexibility and convenience in LIF measurements of different radical species. The gain of an OPO system is derived from the nonlinear interaction between an intense optical wave and crystal having a large nonlinear polarizability coefficient. The 355 nm output of the Nd: YAG laser has been used to pump the OPO. The OPO used in this experiment is a coupled dual oscillator system where a high-energy power oscillator is injection-seeded with the narrow output from a master oscillator. This enables the coupled oscillator system to produce narrow bandwidth, high-energy coherent radiation. Type I phase matched Beta Barium Borate (BBO) crystal was used as the nonlinear gain medium. The BBO is a negative uniaxial crystal with intrinsic birefringence properties that are used to achieve critical phase matching. Tuning of OPO is accomplished by rotating the BBO crystal with respect to the optical resonator. The BBO crystals for the master and power oscillator are mounted on the opposite ends of a rotary shaft. The turning of the OPO crystals are controlled by a microprocessor-based automated control electronics. Although the OPO is very convenient to use, it needs a high precision optical alignment to obtain enough power in a desired wavelength. The output of the OPO was frequency doubled to access in the vacuum ultraviolet region. The output from the OPO was focused into the flame by using a 1 m focal length fused silica best-form laser focusing lens. For Planar Laser Induced Fluorescence (PLIF) measurement, the OPO output was converted to a laser sheet using a combination of cylindrical lenses. The sheet is 1 mm thick and 40 mm wide. The schematic of the PLIF setup is shown in Fig.

2.4 (b). A portion of the beam was deviated to a pyroelectric power meter to continuously monitor the incident beam energy, which was used to normalize the measurements for accounting pulse-to-pulse power variation.

2.4.1.3 The Detection System and Collection Optics

For LIF measurement, the fluorescence signals were collected at right angle of the incident beam using a photomultiplier tube (PMT) connected to a computer-controlled f/3.9 imaging spectrograph (monochromator). The monochromator has an input and output focal length of 220 mm and 257.36 mm respectively. The monochromator has two precision micrometer variable width slits attached before the input window and after the exit plane. In the present investigation the input slit width was kept constant at 0.1 mm and the output slit was kept wide open. The light optical layout of the monochromator is shown in Fig. 2.5. The fluorescence signal was detected through a narrow spectral window (0.1 nm) to reject the strong interference from luminous flame background and other banded emission (for instance interference from PAH signal). Although the spectrally narrow detection scheme yielded considerable signal loss, the problem was overcome by placing two low noise signal pre-amplifiers before acquiring the data. For OH measurement, the monochromator was tuned to 315 ± 0.1 nm. The PMT output was recorded using a gated integrator/boxcar averager (SRS 245, Stanford Research System). Both single shot and multiple laser shot measurements were made. The averaged output from the boxcar was acquired using a data acquisition software in conjunction with a Pentium Pro 200 MHz laboratory computer. For CH measurement the 431 nm output of the OPO was used. The pulse energy was kept close to 24 μ J/pulse

to ensure the linear fluorescence (Norton and Smith, 1991). The fluorescence signals were filtered through 433 ± 0.1 nm spectral window. The monochromator was mounted on a computer-operated stepper-motor driven two-dimensional traversing mechanism, which has a positional accuracy of 0.05 mm/m (Unislide). The detection optics were traversed along the width of the flames to measure the radial concentration profiles of the radicals.

For PLIF measurement, using the same focusing optics the OH fluorescence signal was focused on the filter plane placed in front of an ICCD (Intensified Charge Coupled Device) Camera. For OH measurement, the signal was filtered through a 2mm UG5 and WG305 Schott glass filter to keep the spectral detection window at 315 ± 8 nm.

2.4.1.4 Control Electronics and Gating System

For LIF experiments, the Gated Integrator/Boxcar Averager served as the primary control unit for signal gating. As mentioned earlier, the output signal from the Photomultiplier tube was amplified using two fast preamplifiers before sending to the integrator. The Gate generator was triggered externally by the Q-switch synchronization from the GCR. The Boxcar could provide variable gate delays from 5ns to 100ms with a detection gate width from 5ns to 100ns. It integrates the input signal during the gate, and the output from the integrator is then normalized by the gate width to provide a voltage, which is proportional to the average of the input signal during the sampling gate. The signal was further amplified according to the front panel sensitivity setting before sampled by a loop sample and hold amplifier, and outputting via a front panel BNC connector. Also, a portion of the pump laser beam was deviated using a beam splitter

and focused to a power meter to monitor the pulse-to-pulse pump power changes. The detected LIF signals were normalized by the pulse power to account for the pulse-to-pulse input energy variation.

For PLIF imaging, a dual channel digital delay with a gate pulse generator was used to control and synchronize the imaging process with laser pulse. The gate pulse generator was capable of producing variable gate pulse from 10 ns to 10ms with delays from 5ns to 10s whereas the digital delay further extended the delay capacity upto 10 seconds. This is essential to control the image data burst to camera controller, which has a maximum capability of handling data at 150 kHz rate. The gate pulse generator can be triggered optically or using Q-Switch advance synchronizing signal. Furthermore, a dual channel oscilloscope triggered by the Gate integrator was used to position the gate, which was opened only when the signal/noise ratio was favorable.

2.4.1.5 Timing Diagrams

The timing diagrams for LIF and PLIF imaging are shown in Figs. 2.6 (a) and (b). This figure shows how different control electronics and data acquisition systems were timed to synchronize with each other.

In LIF measurement synchronizing technique, the firing of the Q-switch initiates the sequence. The delay between Q-switch firing and output of OPO is around 16 ns and after emitting from the OPO the beam takes 3 ns to complete the optical path to reach the flame. So, after receiving signal from the Q-switch, the Boxcar Integrator delays 24 (19ns + 5ns) to open the detection gate for 5 ns. This additional delay was imposed to

ensure that the fluorescence signal was collected only from the peak power of the laser pulse. At the same time of opening the detection gate, the boxcar triggers the data acquisition signal to start acquiring data. The Fast mode enabled high speed data acquisition system to burst data in its onboard buffer memory before transferring to the memory.

For PLIF image acquisition, the sequence was also triggered by a Q-switch. The Q-switch triggers the gate pulse generator, and the gate pulse generator sends signal to the camera controller to open the electronic gate of the ICCD camera for 10 ns after 24 ns delays. Unlike LIF measurement, in PLIF measurement the image acquisition was always open and the shot by shot imaging was done through electronic gate control of the camera.

2.4.2 Raman Spectroscopy

Raman Scattering is the result of an inelastic, nonresonant interaction between an incident photon and a scatter (Laufer, 1996, Gardiner and Graves, 1989). The extent of scattering refers to certain properties of the probed species such as spectroscopic characteristics, density, and temperature. Hence, if the scattering characteristics of the probed species are known, its thermodynamics properties can be measured from Raman spectra.

The quantum theory approach to Raman Scattering recognizes that the vibrational energy of a molecule is quantized. A non-linear molecule will have $(3N-6)$ normal

vibrations and a linear molecule will have $(3N-5)$, where N is the number of atoms in the molecule. The energy of each of these vibrations will be quantised according to the relation

$$E_v = h\nu \left(v + \frac{1}{2} \right)$$

where ν is the frequency of the vibration and v is the vibrational quantum number controlling the energy of that particular vibration and having values of 0,1,2,3....etc. Perturbation theory is used to introduce quantisation into the Raman scattering theory. This approach applies perturbations to the ground state molecular wavefunctions until new wave functions are obtained which describe the vibrational excited state. The transition from ground state can then be regarded as being achieved via a perturbing wave function, which is the sum of the perturbation applied. This perturbing wave function has a corresponding energy and gives us a useful pictorial description of Raman scattering with the vibrational transitions occurring via this virtual energy level (Fig. 2.7). The Rayleigh scattering arises from transitions, which start and finish at the same vibrational energy level. Stokes Raman scattering involves a transition from a higher to lower vibrational energy level. At normal room temperatures, most molecular vibrations are in the ground, $v=0$ state and thus the anti-stoke transitions are less likely to occur than the Stokes transitions resulting in the Stokes Raman Scattering being more intense. This greater relative intensity becomes increasingly greater as the energy of the vibrations increases and the higher vibrational levels become less populated at any given temperature. For this reason the Stokes Raman scattering which is generally termed as Raman Scattering was used here. Since, Raman signals are inherently and highly affected by background signal and noise, it is often difficult to employ Raman

measurement successfully in the luminous part of the flame. In the present investigation, the Raman measurements were only used to develop a collisional quenching model to quantify the fluorescence signal. The details of the modeling techniques are discussed in Chapter Three.

2.4.2.1 Measurement Methodology and Setup

To measure the Raman spectra of the various species the laser is tuned to 248 nm to access the UV Raman spectra. The Raman signals were collected using the same monochromator and PMT combination used in fluorescence measurement. Although gate detection is not essential for Raman measurement, the signal gating along with narrow spectral window were still used to reduce noise level. The details of the Raman measurement setup are shown in Fig. 2.8. The wavelengths for the Raman signals used in this experiment for different species are summarized in Table 2.6.

2.5 Other Instrumentation

Various other measuring instruments such as standard gas analyzers, radiometer, low-energy He-Ne laser for soot concentration, high speed Cam Recorder were also employed in the study. The following subsections provide details of the various instruments used in the measurements.

2.5.1 Exhaust Emission Measurements

For measuring exhaust emissions, a quartz flue gas collector was mounted over the visible flame, and axially aligned with the burner (Fig. 2.9). A sample profile across the

collector diameter showed a variation of less than 1.5% in species concentration, and hence, the center point data were treated as the average representative values. Gas samples were collected from combustion products through an uncooled-quartz probe of tip diameter 1 mm and treated to remove particulate and moisture with a series of filters and an ice-chilled moisture trap. The sampling flow rate was adjusted such that the suction and local free stream velocities in the flow-field were close enough to ensure *quasi-isokinetic* sampling (Fristrom, 1975). A chemiluminescence analyzer (Thermo Environmental Inc. Model 2H) was used to measure the concentration of NO and NO_x. Two non-dispersive infrared (NDIR) analyzers (Rosemount Analytical Model 880A) were used to measure the concentration of CO and CO₂. Since the temperature of the exhaust stream at the sampling location was less than 1000 K, the measurements of CO mole fraction were not significantly affected by the lack of quenching (Gore and Zhan, 1996).

2.5.2 Flame Radiation Measurement

For measuring flame radiation, a wide-angle (150 °) highly sensitive pyroheliometer (Hy-Cal) was used. The wavelength sensitivity of the detector is shown in Appendix III. The output was digitized and sampled at a rate of 1 kHz and on-line averaged over 8 seconds by using the same data acquisition system mentioned earlier (Fig 2.10).

2.5.3 Visible Flame Height Measurement

To determine the visible flame height, a high-speed video camera was used. Strobe recording technique (1/6 seconds interval progression, 1/2000 S.S.) along with back-

light illuminating and DEIS (digital electronic image stabilization system) method were used to visualize the flame image in a dark background. The back-light illuminating method has the advantage of sharpening the image boundary of a very bright image in contrast with its dark background: also it helps to remove the pixel distortion due to imbalance in the pixel illumination of the CCD (charge couple device). A 2-second recording time was used to measure the visible flame height.

2.5.4 Soot Concentration Measurement

The technique proposed by Yagi and Iino (1966) was used to measure the soot concentration. A He-Ne laser (Spectra Physics) beam was passed through the flame (Fig 2.11). Due to the presence of soot the beam intensity was attenuated. The amount of attenuation was measured using a pyro-electric laser power meter placed on the other side of the beam. Yagi and Iino (1966) proposed a correlation between the soot concentration and the beam attenuation. The process is slightly modified by using a narrow-band laser line-filter to block the luminous flame background and the *Anti-Stoke* component of the Raman spectra due to light scattering from the soot particle. This method is fast and simple compared with the direct probing method. However, it results in the line-of-sight average concentration.

Table 2.1(a) Spectroscopic System Instrument Specifications

Instrument	Model/Manufacturer	Specifications
Pulsed Nd:YAG Laser	GCR 250-10 Spectra Physics	Repetition Rate: 10 Hz Wavelengths: 1064, 532, 355, 266 nm Line Width: 0.003 cm ⁻¹ Pulse Width: 10 ns
Optical Parametric Oscillator (OPO)	MOPO-730 Spectra Physics	Tuning: Signal: 440-690nm Idler: 735-1800nm Line Width: 0.2 cm ⁻¹ Pulse Width: 10 ns
Frequency Doubler (FDO)	FDO-970 Spectra Physics	Tuning: 198-440 nm Line Width: 0.2 cm ⁻¹ Pulse Width: 10 ns
Gated Integrator and Boxcar Averager	SR 250 and SR 280 Stanford Research Systems	Gate Width: 2ns-15μs Delay: 1ns-100ms
Photomultiplier Tube Cooled Housing	77345/77265 Oriel Instruments	Wavelength Range: 186-650nm (Appendix-IV) Biasing Voltage: -1000 V Photocathode size: 25 mm
Photomultiplier Power Supply	70705 Oriel Instruments	Voltage: -200 to -2000 V Current Load: 2 mA
Fast Preamplifier	SR 445 Stanford Research System	Bandwidth: DC-300Mhz Max. Gain: 125
Data Acquisition Hardware	Flash 12 Strawberry Tree Inc	14 bits Multichannel 1 MHz single channel sampling Noise: 5 μV RMS 8MB buffer memory
Data Acquisition Software	Work Bench PC for Windows Strawberry Tree Inc	
Pulsed Laser Power Meter	NOVA 30 Ophir Optonics Ltd.	Wavelength: 190-2000nm Power Range: 50mW-150W Energy Range: 100mJ-200J Power Density: 20kW/cm ²
Precision Power Meter	407A Spectra Physics	Wavelength: 250-1100nm Power Range: 5mW-20W Power Density: 20kW/cm
Data Acquisition Computer	P-200 Gateway	Pentium II, 200MHz 128 MB RAM

Table 2.1(a) Spectroscopic System Instrument Specifications (Contd.)

Instrument	Model/Manufacturer	Specifications
ICCD Camera (Intensified Charge Coupled Device Camera)	ICCD-576E Princeton Instrument	Full Frame ICCD Pixels: 576×384 Field: 12.9 × 8.6 mm CCD dark Current: 16 electrons/pixel/s Maximum Pixel rate: 1MHz Gating Speed: 5 ns FWHM
Camera Controller	ST-138 High performance Princeton Instrument	14 bits/pixel at 1 MHz 15 Linearity High Speed Image Burst
Digital Gate Pulse Generator	PG-200 Programmable Gate Pulse Generator Princeton Instrument	Pulse Width: 3.5ns-80ms Gate Delay: 20ns-80ms Jitter: 0.5ns
Image Acquisition Hardware	EISA Standard Interface Princeton Instrument	1 Megapixel per second
Image Acquisition Software	WinView Imaging Princeton Instrument	
Image Acquisition Computer	P-200 Gateway	Pentium II, 200MHz 128 MB RAM

Table 2.1(b) Schlieren System Specifications

Instrument	Model/Manufacturer	Specifications
Digital Color Video Camera	Model: 2222-2340 Cohu Inc.	Interline Transfer CCD Pixels: 768 × 494 Pixel Dimension: 8.4 × 9.8μm Signal/Noise Ratio: 40dB Format: RGB
Image Acquisition Hardware	Meteor Metrox	
Image Acquisition Software	Metrox Meteor	
Focusing Lenses	Melis Griot	Achromatic 63 mm diameter 760 mm focal length
Filter	In house made	
Image Acquisition Computer	PC+	Pentium II, 400MHz 128 MB RAM

Table 2.1(c) Other Instrumentation

Instrument	Type	Manufacturer	Range	Least Count
Digital Camera	Color CCD	Sony Mavica		
NO/NO _x Analyzer	Chemi-luminescence	Thermo Environment Model 42H	0-10 ⁴	0.01 ppm
CO ₂ Analyzer	NDIR	Rosemount Analyzer Model 880A	0-30%	0.1%
CO Analyzer	NDIR	Rosemount Analyzer Model 880A OFC Infrared Instruments Model 702D	0-2% 0-10 ⁶ ppm	0.1% 0.1 ppm
O ₂ Analyzer	Polarographic	Catalytic Research Model Miniox III	0-22%	0.1%
Thermocouple	Type R Type C	Omega Engineering Inc. Omega Engineering Inc.	350-1800 K 500 -2340 K	.01 K .01 K
Radiometer	Pyroheliometer	Hy-Cal		
Rotameter		Cole Parmer (12 different models were used)	10-20000cc/min (air)	0.1 cc/min (air)
Traversing Mechanism		Unislide/ Mycom Inc.	0-40 cm	0.5 mm

Table 2.2 Estimated Uncertainties

Measurements	% of Mean Value
Emission Index	1.7
Radiative Heat Loss	2
Visible Flame Length	1.8
Volumetric Fuel Flow rate	2.1
Flame Blowout Velocity	2.6
Liftoff Height	1.8
Concentration of NO	7.9
Concentration of NO _x	8.2
Concentration of CO ₂	8.6
Concentration of CO	8.8
Concentration of O ₂	4
Temperature	1.4
Soot Concentration	6.4
Concentration of OH	4
Concentration of CH	5
Concentration of O	5.5
Concentration of H	4.5

Table 2.3 Nominal operating Conditions

Burner diameter (ID)	1mm, 2.3 mm, 3.8mm, and 4.5mm
Fuel	H ₂ (98%+):0-100%* NG (95.8% CH ₄ , 1.19% C ₃ H ₈ , 1.15% C ₂ H ₆ , 0.50% CO ₂ , 0.10% N ₂ and remains are pentane, hexane and water)): 0-100%*
Ambient Temperature	295 K
Ambient Pressure	102 KPa

*By volume

Table 2.4 Test Matrix**(a) Flame Stability**

Burner Sizes (ID)mm	Mixture Conditions %NG-%H ₂	Measured Parameters
1, 2.3, 3.8, and 4.5	100-0	Blow-out Velocity Lift-off Height at Blowout condition
	95-5	
	90-10	
	85-15	
	80-20	
	75-25	
	70-30	
	65-35	
	0-100	

(b) Schlieren Photographs

Burner Sizes (ID) mm	Mixture Conditions %NG-%H ₂	Measured Parameters
3.8 mm	100-0	Images of the Flow-Field
	95-5	
	90-10	
	85-15	
	80-20	
	75-25	
	70-30	
	65-35	
	0-100	

(c) Flame Structure (Thermo-chemical) Measurement

Burner Size (ID)	Axial Locations % Flame Length	Reynolds number	Mixture Conditions %NG-%H ₂	Jet Exit Velocity	Jet Exit Froude Number	Global Flame Froude Number
3.8 mm	25% 50% 75%	8700	100-0	38.52	39785	0.868
			90-10	42.43	48300	0.934
			80-20	47.15	59638	1.011
			65-35	56.31	85068	1.154

(d) Emission Indices, Flame Radiation and Soot Formation Measurement

Burner Size (ID)	Axial Locations % Flame Length	Reynolds number	Mixture Conditions %NG-%H ₂	Jet Exit Velocity	Jet Exit Froude Number	Global Flame Froude Number
3.8 mm	25% 50% 75%	8700	100-0	38.52	39785	0.868
			90-10	42.43	48300	0.934
			80-20	47.15	59638	1.011
			65-35	56.31	85068	1.154

(e) Computation of Thermo-chemical Structure and the flow-field

Burner Size (ID)	Axial Locations % Flame Length	Reynolds number	Mixture Conditions %NG-%H ₂	Jet Exit Velocity	Jet Exit Froude Number	Global Flame Froude Number
3.8 mm	2-D Axisymmetric	8700	100-0	38.52	39785	0.868
			90-10	42.43	48300	0.934
			80-20	47.15	59638	1.011
			65-35	56.31	85068	1.154

Table 2.5 LIF Pumping and Detection Wavelengths

Species	Excitation	Pump	Detection
OH	$A^2\Sigma^+ \leftarrow X^2\Pi$	(1.0), 285.265	(1.1) P ₁ (8) 315 nm
CH	$A^2\Delta \leftarrow X^2\Pi$	(0.0) 431 nm	(0.1) 432.825 nm
H	1s → 2s → 3p	243 nm	656 nm
O	1s → 2s → 3s ³ S	226 nm	845 nm

Table 2.6. Species and their Raman Signal Wavelength

Species	Raman Signal Wavelength
N ₂	263.7 nm
CH ₄	253.8 nm, 267.8 nm and 268.5 nm
H ₂	277.1 nm
O ₂	258.4 nm
CO ₂	256.6 nm and 257.3 nm
CO	262.4 nm
H ₂ O	273.2 nm

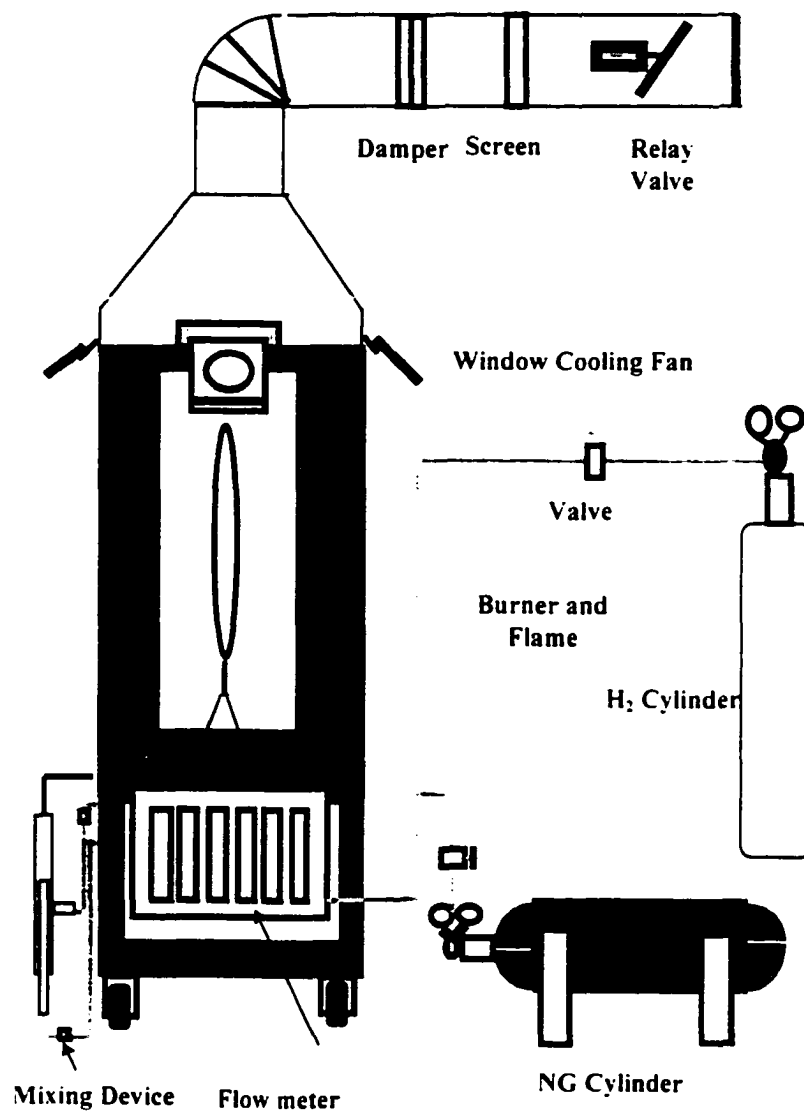


Fig. 2.1 (a) Laboratory Combustion Chamber and Fuel Supply Train

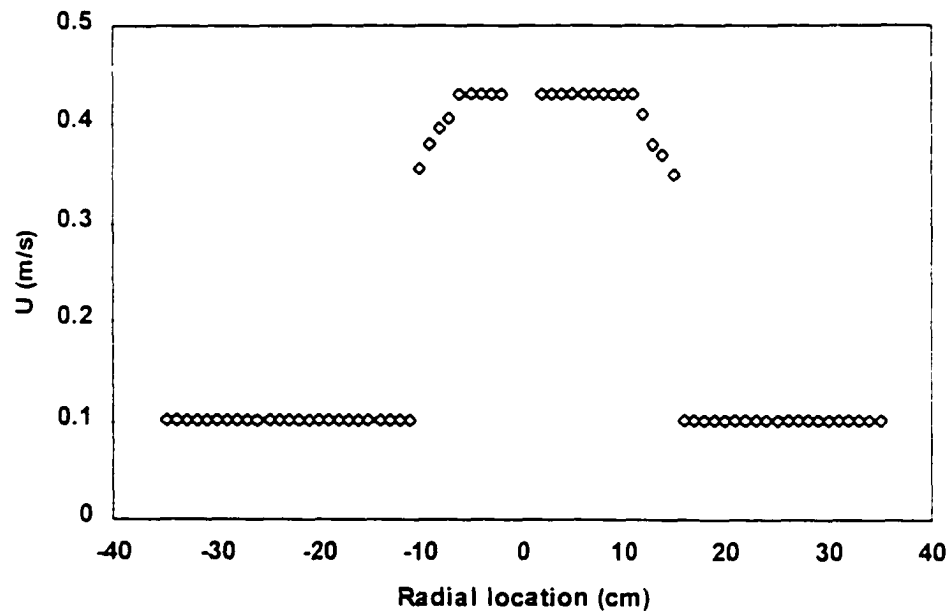
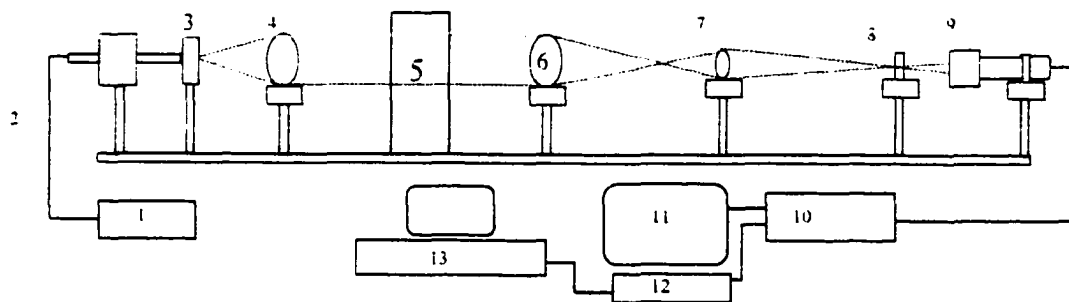


Fig 2.1 (b) Incoming Air Velocity Profile Inside the Chamber



1-Light Source, 2 -Fiber Optic Cable
 3-Source Aperture, 4 -Focusing Lens 5 -Test Chamber, 6 -
 Refocusing Lens 7 -Magnifying Lens, 8 -Rainbow Filter 9 -
 CCD Array, 10 -Camera Controller, 11 -Monitor, 12 -SVHS
 VCR, 13-Computer with Frame Grabber

Fig. 2.2 (a) Schematic of the Schlieren Setup



Fig 2.2 (b) Color Schlieren Filter

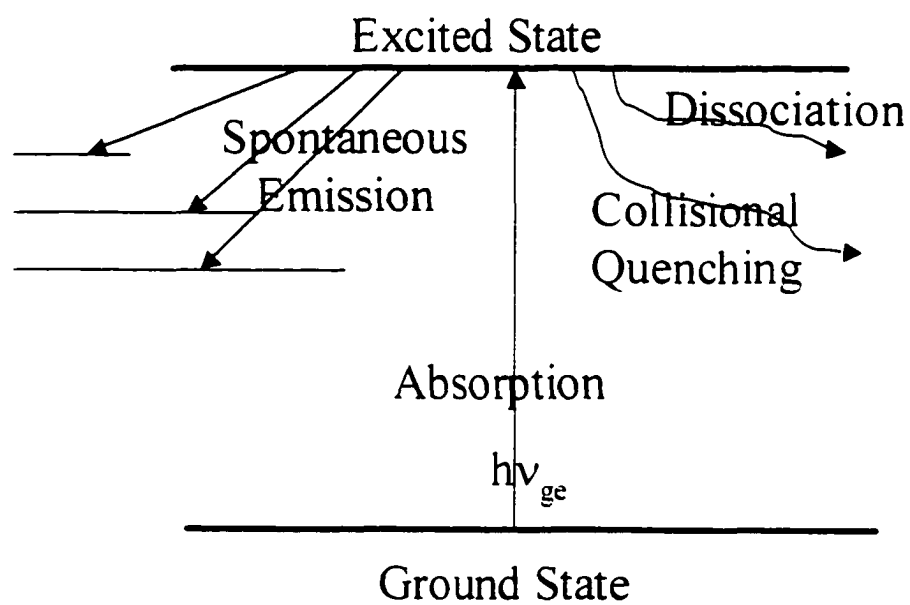


Fig 2.3 Energy Level Diagram For Laser Induced Fluorescence Measurement

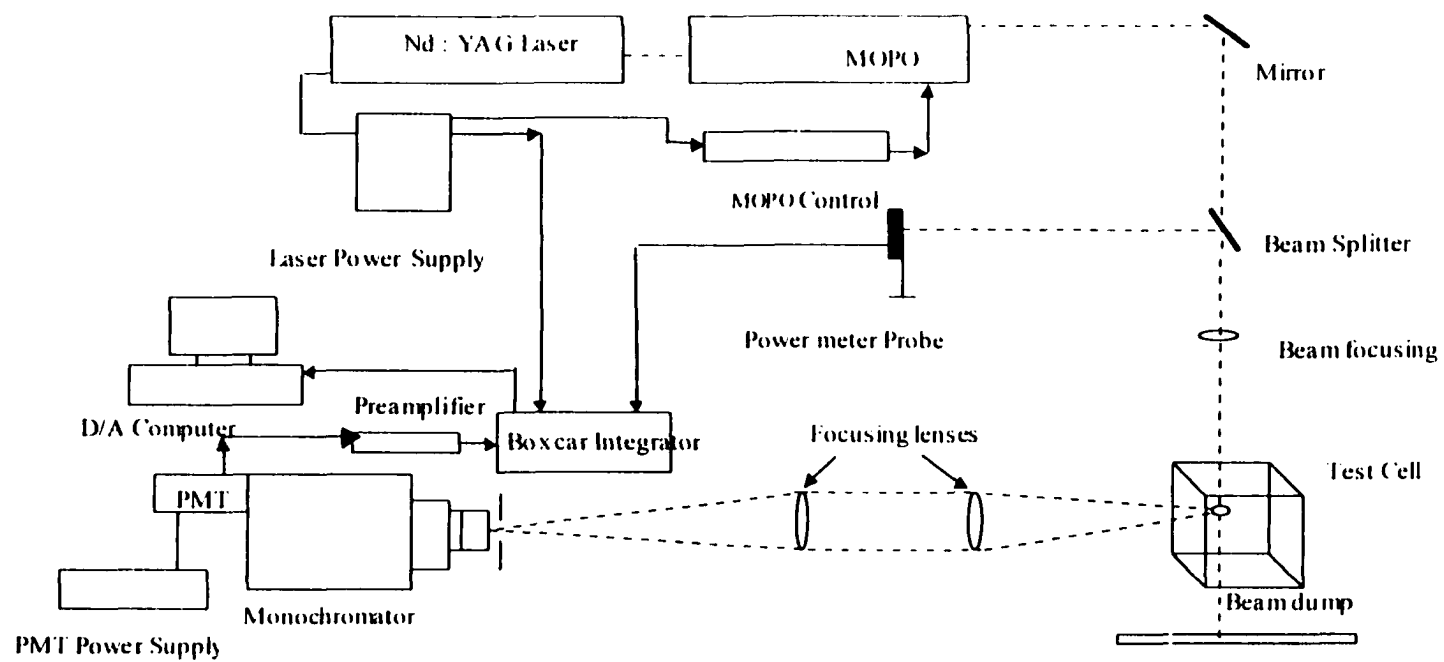


Fig. 2.4 (a) Schematic of LIF Setup

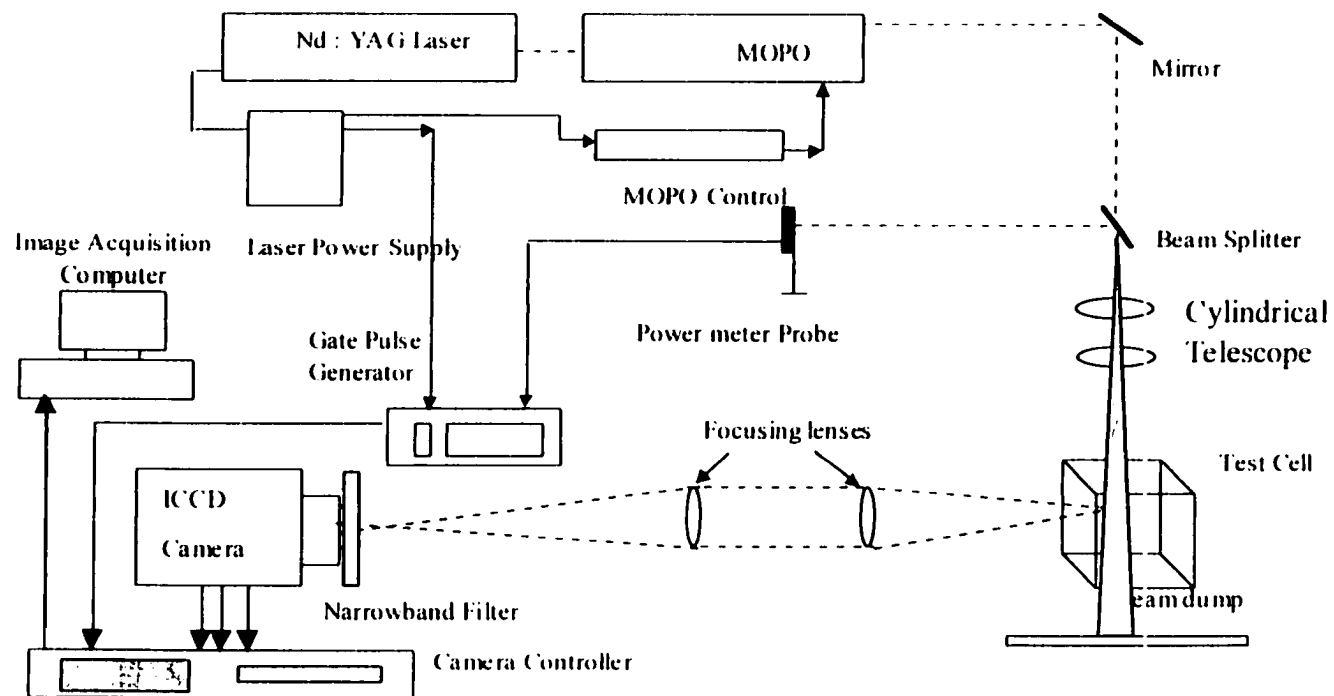
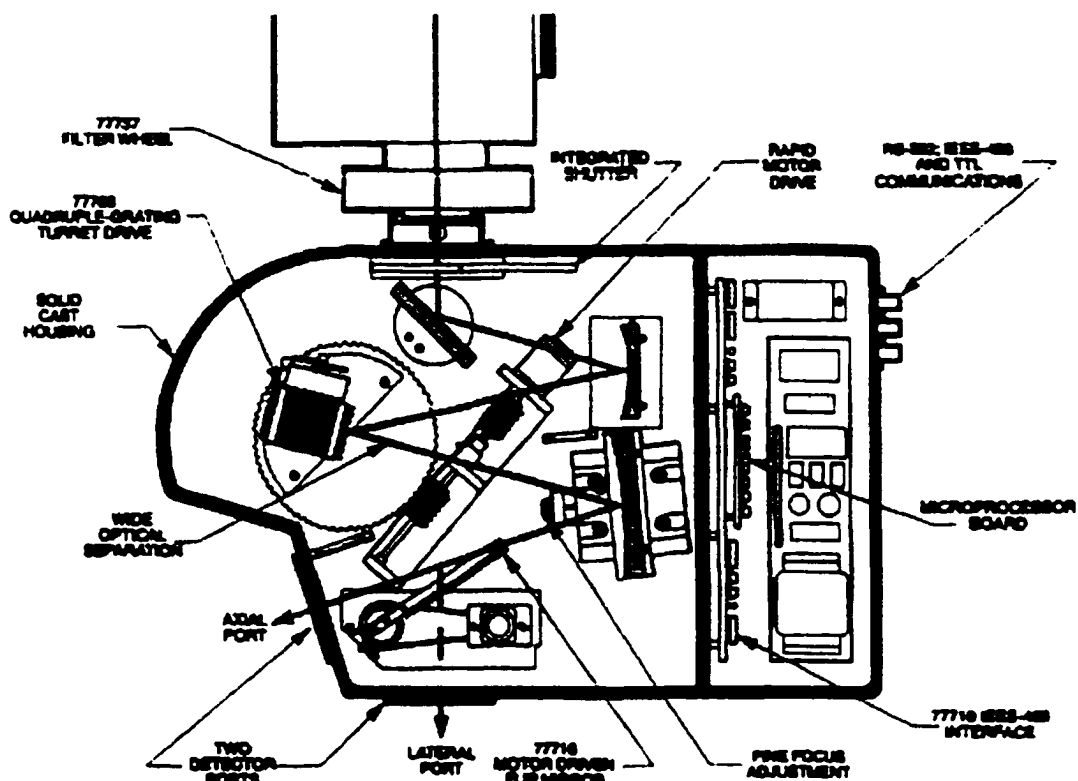


Fig. 2.4 (b) Schematic of PLIF Setup



Specifications: Model 257, Oriel Instrument
Imaging Spectrograph/Monochromator
 2400 l/mm Holographic, Blaze 250 nm, Range: 200-700nm
 Motormic driven variable slit: 4 μ m-2.0mm (15 mm High)
 Wavelength Resolution: 0.1 nm

Fig 2.5 Optical Layout of the Monochromator/Imaging Spectrograph

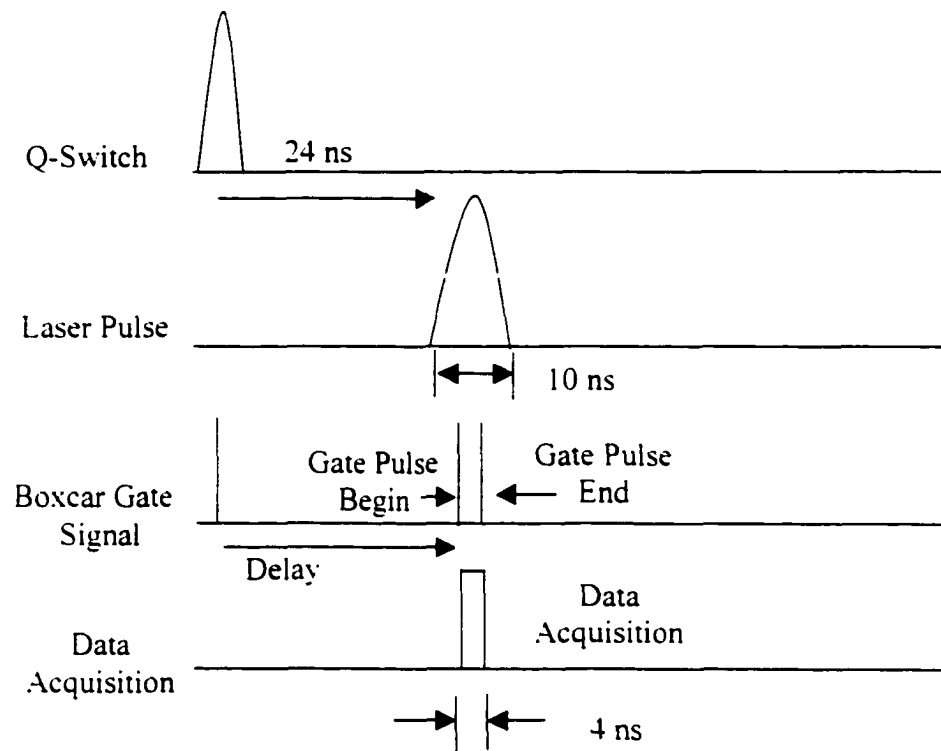


Fig. 2.6 (a) Timing Diagram For LIF Experiment

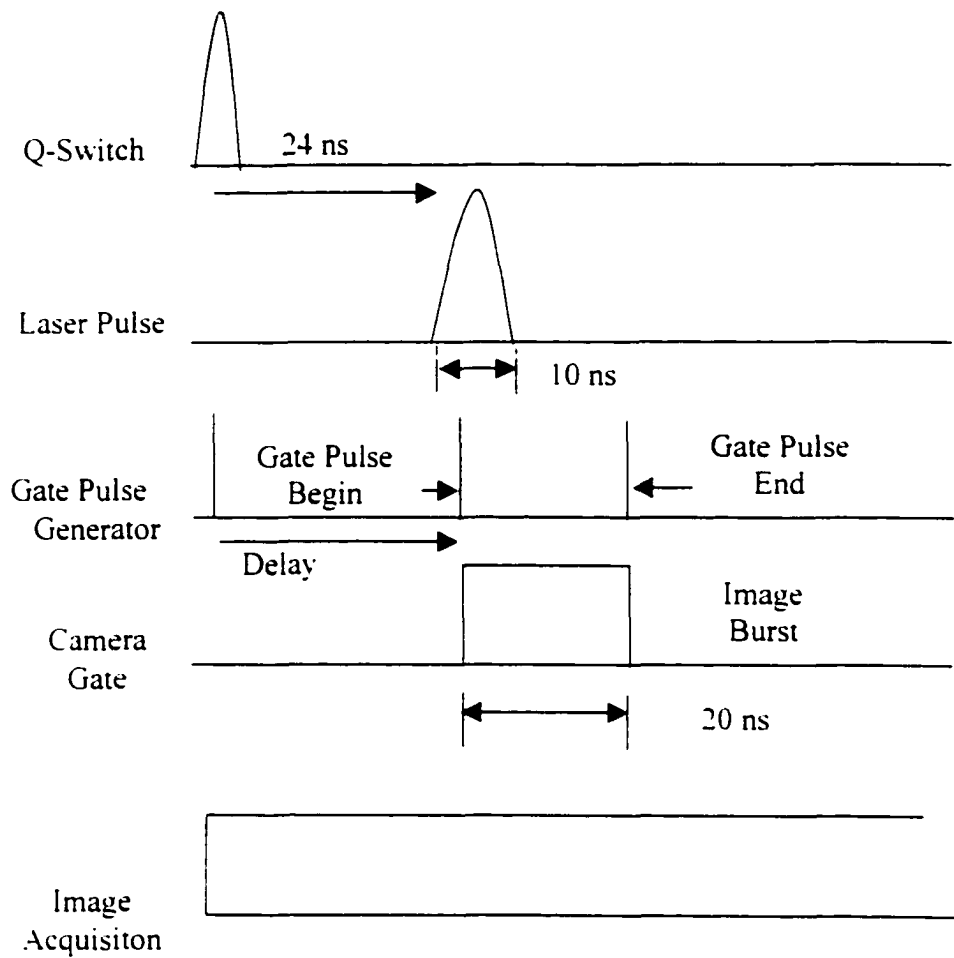


Fig. 2.6 (b) Timing Diagram For PLIF Experiment

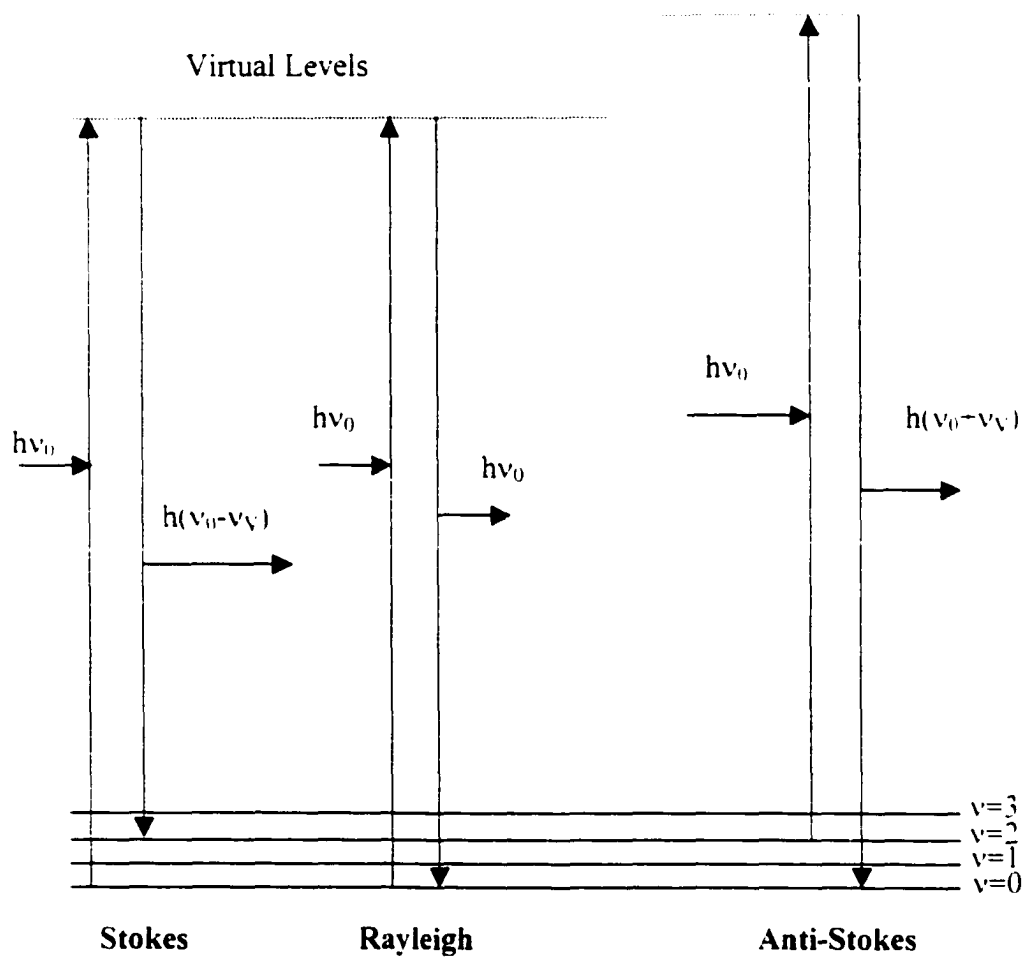


Fig. 2.7 Principles of Raman Spectroscopy

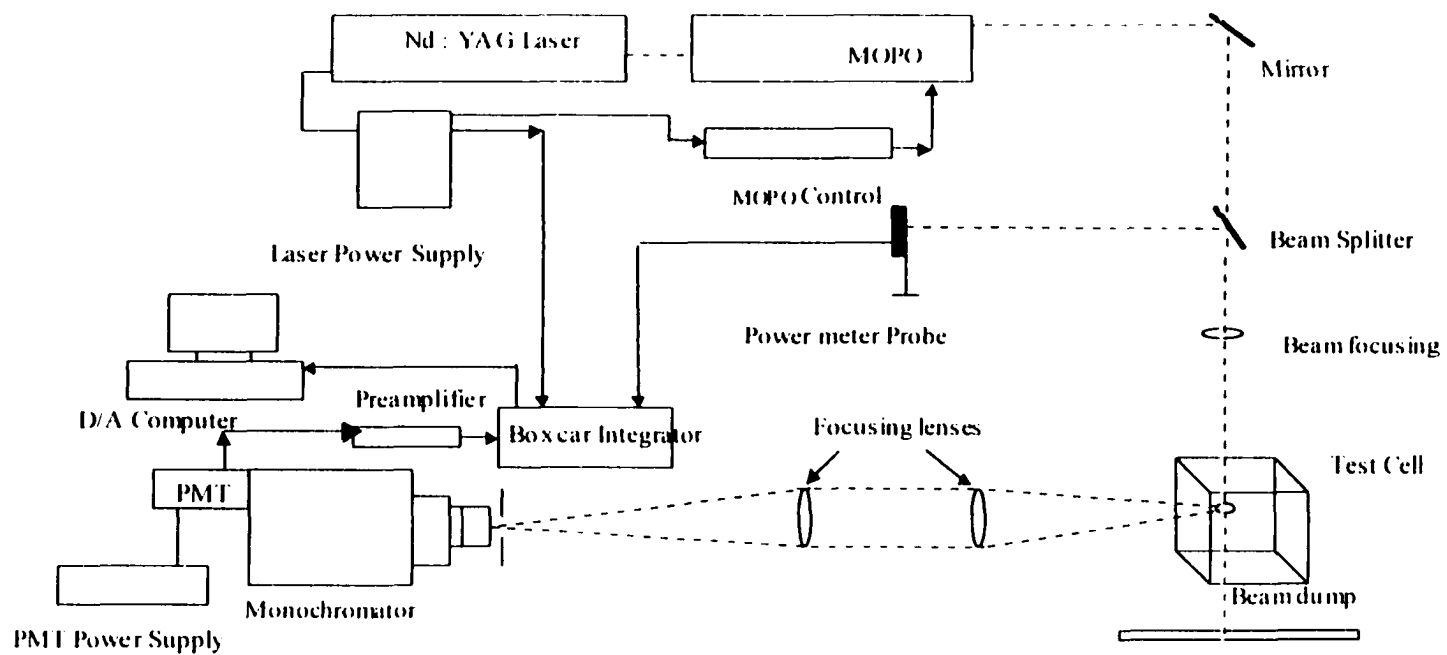
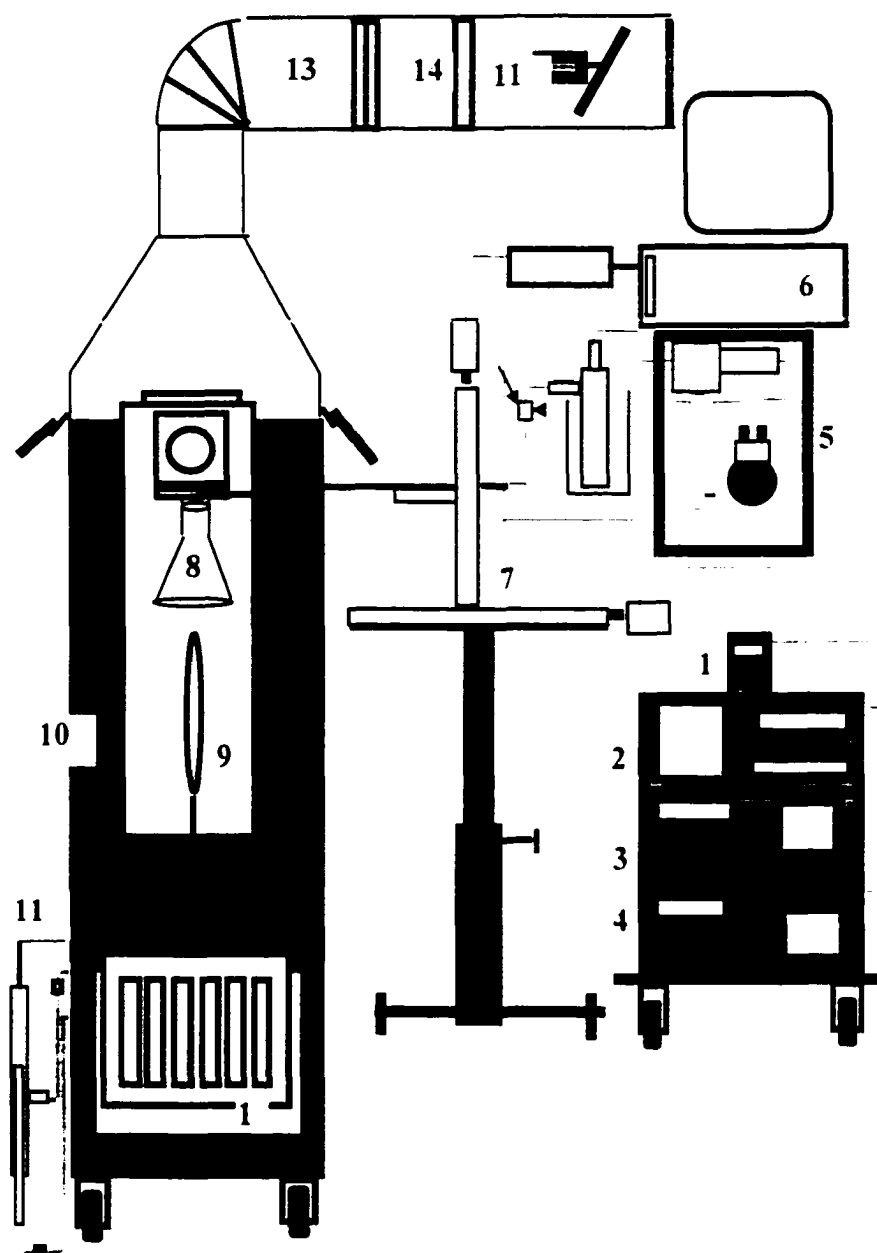


Fig. 2.8 Schematic of Raman Setup



1. Oxygen Analyzer 2. NO-NO_x Analyzer 3. CO₂ Analyzer 4. CO Analyzer
5. Exhaust Treatment System: Particulate Filter, Ice-Bath, Vacuum Pump
6. Data Acquisition System 7. Stepper Motor Driven 2-D Traverse
8. Exhaust Collection Cone 9. Flame with Burner 10. Combustion Chamber
11. Mixing Device 12. Rotameters 13. Damper 14. Filter 15. Relay Valve

Fig 2.9 Emission Measurement Set up

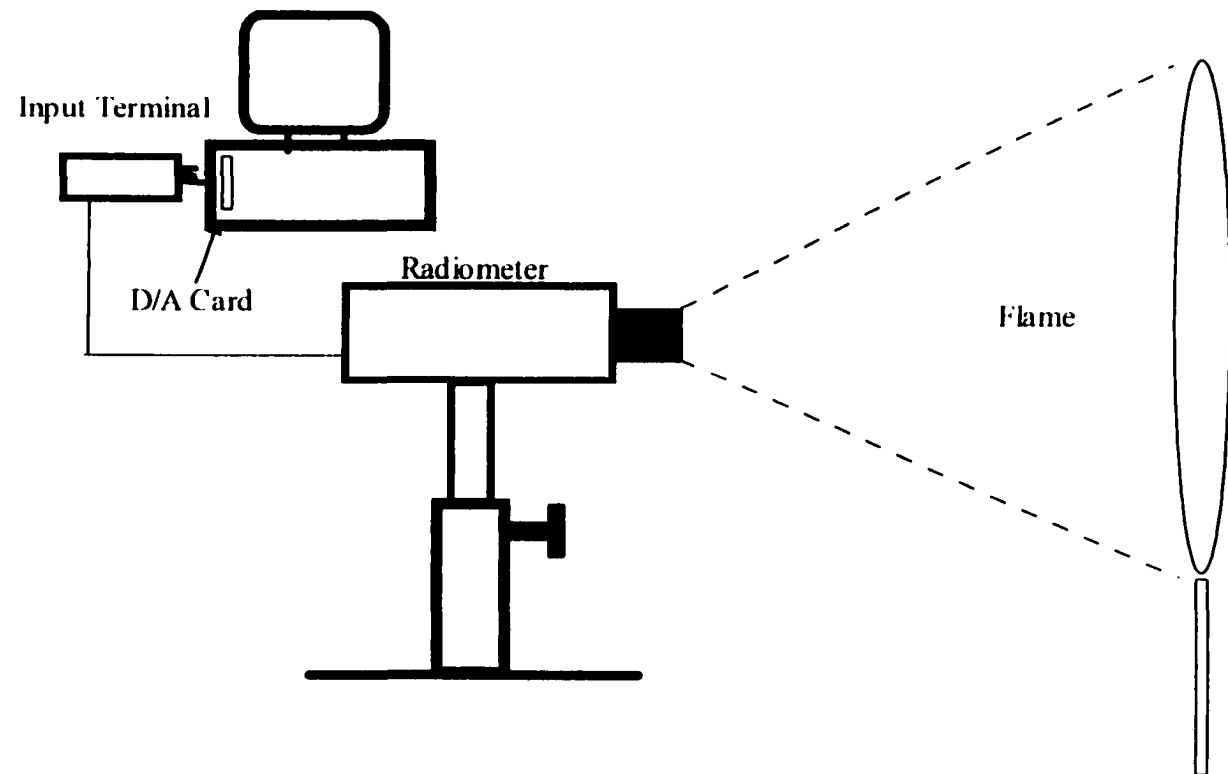


Fig. 2. 10 Instrumentation for Flame Radiation Measurement

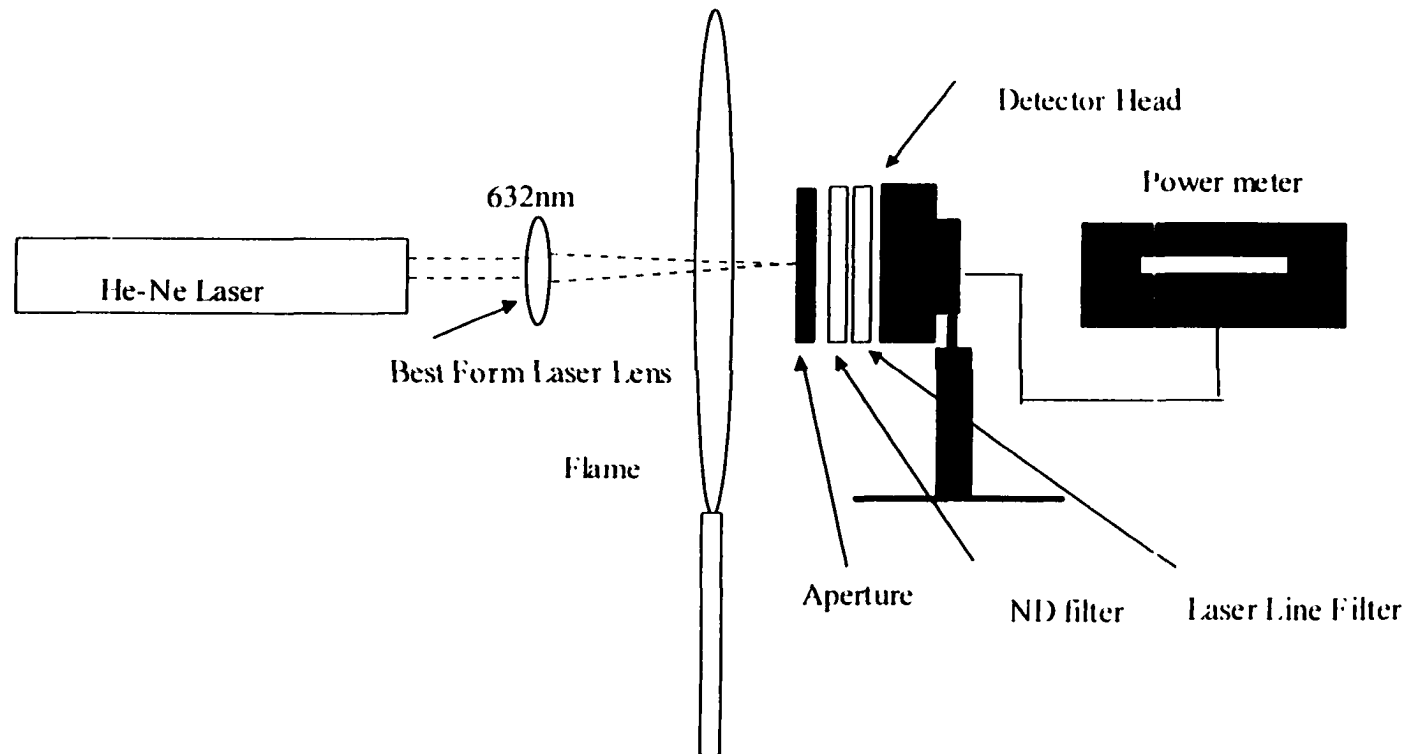
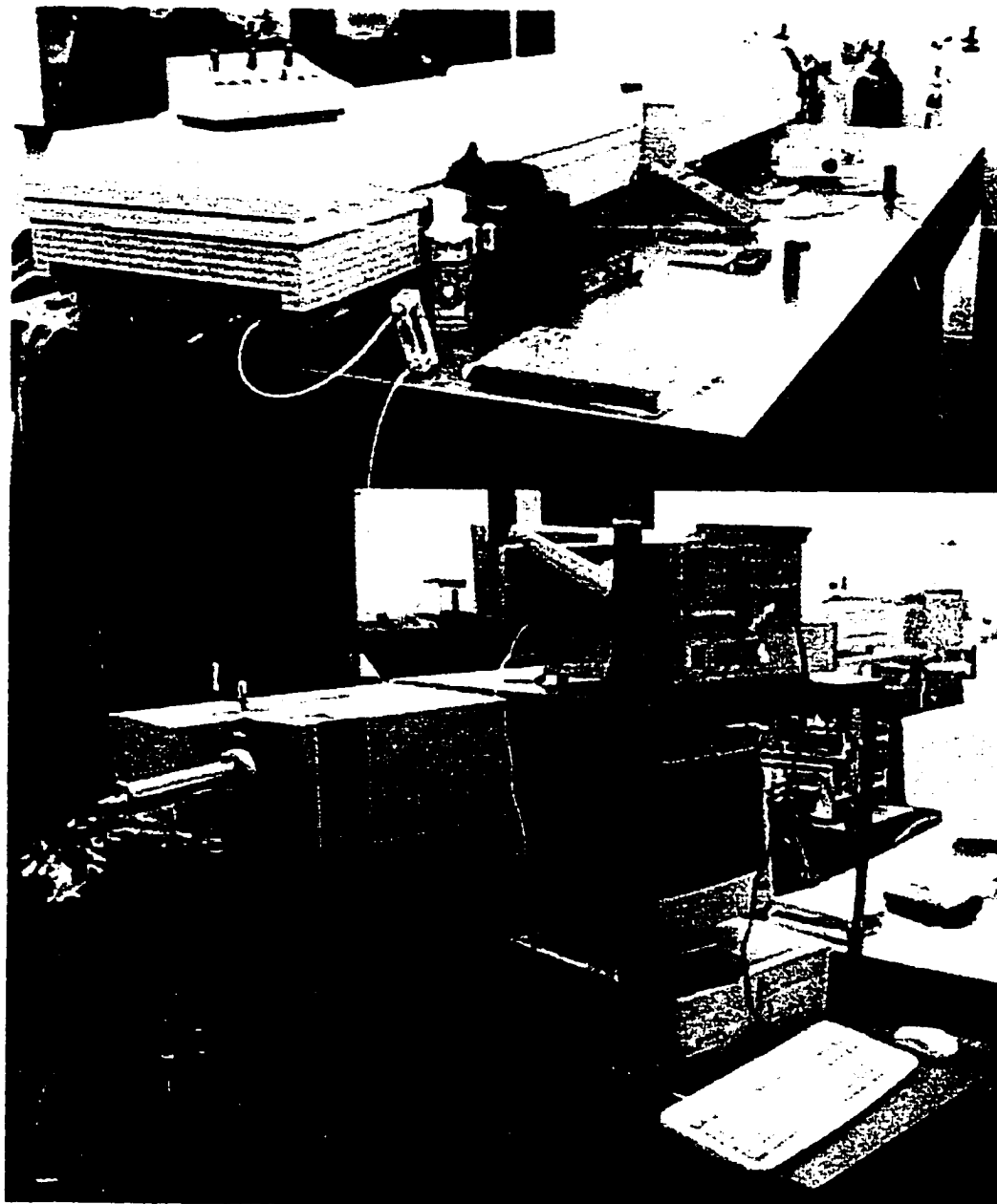


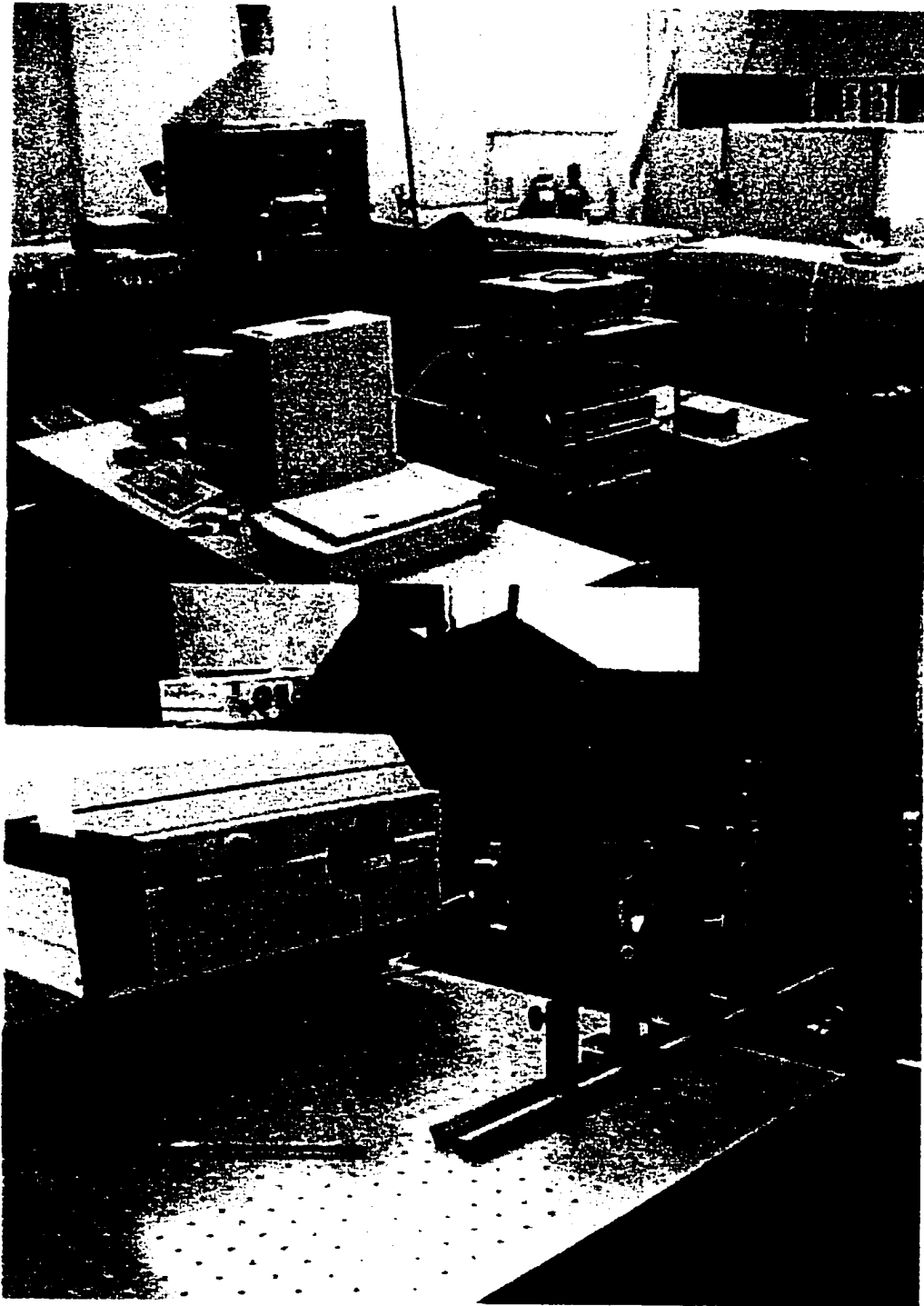
Fig. 2. 11 Instrumentation for Soot Concentration Measurement



PT 2.1 Laboratory Combustion Chamber



PT 2.2 LIF Setup



PT 2.3 PLIF Setup

Chapter Three

Spectroscopic Modeling

Quantification of LIF signals to absolute values of the concentration of molecules under investigation requires information about the fluorescence quantum yield. Fluorescence quantum yield is defined as the number of photons emitted per molecule pumped to the electronically excited state by absorption of the laser radiation. However, in combustion measurements the fluorescence quantum yield is dominated by a mechanism called collisional quenching. The collisional quenching is a process of non-radiative removal of electrons from its upper excited stage due to collision with other molecules and atoms. In fact, in atmospheric diffusion flames around 99% of the excited OH molecules are removed from its upper electronic excited stage due to the collision with adjacent molecules (Tamura et al., 1998; Setizman and Hanson, 1993; Smyth et al., 1990; Zizak et al., 1986; Paul et al., 1994). In certain cases, it is possible to minimize the effect of collisional quenching by carefully selecting the experimental technique. For instance, in low pressure flames, very short and quick detection gate width ($<10\text{ns}$) right after the laser pulse can be used to measure the fluorescence before a significant amount of quenching occurs. However, in atmospheric pressure flames the quenching is more rapid and it is extremely difficult to avoid it even with shortest detection gate width. Hence, it is often required to estimate, calculate or directly measure the rate of collisional quenching. Direct measurement of collisional quenching depends upon the fluorescence lifetime and requires picosecond spectroscopy, which is proven to be difficult in most of the experimental conditions. Estimation or calculation of collisional quenching requires information about collider specific quenching rate as a function of temperature at the

point of measurement. Hence, local temperature and species concentration information is required to calculate the collisional quenching. One usual practice is to assume equilibrium composition of the collider species based on the local measured (or computed) temperature (Setizman and Hanson, 1993, Smyth et al., 1990). However, this method imposes a certain amount of error in the quantification process of LIF signal, particularly in hydrocarbon diffusion flames where equilibrium does not exist in most regions of the flame. The other approach is to measure local temperature and collider species concentration directly and use them. However, this yields a high experimental complexity and often is very difficult and expensive to implement (Tamura et al., 1998). Nonetheless, a study comparing the estimation and direct computation of the collisional quenching will be highly beneficial for the implementation of the methods in future studies of diffusion flames where decision needs to be made between accuracy and complexity of the techniques.

In the present study, LIF measurements of OH radicals in natural gas turbulent flames ($Re=8700$) are presented. The collisional quenching data used to quantify OH fluorescence are derived using both estimation (using equilibrium composition) and calculation (using measured composition of the collider species) methods. Their differences are analyzed. The measurement of N_2 Raman spectra is used to calculate the local flame temperature whereas ultra-violet Raman spectroscopy is used to measure the concentrations of O_2 , CO_2 , CO , H_2O , H_2 , and CH_4 (Appendix-IV).

3.1 SPECTROSCOPIC SIGNAL MODELING

3.1.1 LIF Signal Modeling

The LIF signal of a species of interest can be modeled as (Laufer, 1996):

$$S_f = C \wedge n_A \wedge SV \quad [3.1]$$

where

S_f = Fluorescence Signal

C = Group of experimental constants including collection solid angle, transmission efficiency and quantum efficiency of the PMT.

SV = Stern-Volmer coefficient of Fluorescence quantum yield.

n_A = Number of molecules excited along the specific portion of the incident beam.

The experimental constants can be expressed as (Choudhuri and Gollahalli, 2000c)

$$C = \left(\frac{\Omega}{4\pi} \eta_T \eta_D \right) \quad [3.2]$$

where

Ω = Collection solid angle

η_T = Transmission efficiency

η_D = Detection or quantum efficiency of PMT

The number of excited molecules can be expressed as (Laufer, 1996)

$$n_A = C \frac{I_L}{h\nu_{ge}} \chi \beta_{ge} \sigma_{ge} \Delta l \quad [3.3]$$

where

I_L = Incident laser energy

χ = Mole Fraction of OH

σ_{ge} = Absorption cross section

ν_{ge} = Transition frequency

β_{ge} = Boltzman fraction

h = Plank's constant = 6.63×10^{-34} J-s

Δl = Portion of the beam where molecules are excited

subscript g and e denote the ground state and the excited state respectively. The absorption coefficient σ_{ge} can be expressed as

$$\sigma_{ge} = \frac{B_{ge} h \nu_{ge}}{\Delta \nu c} \quad [3.4]$$

where

B_{ge} = Einstein absorption coefficient

c = Speed of light

$\Delta \nu$ = Absorption linewidth

The Stern-Volmer coefficient or Fluorescence quantum yield can be written as (Choudhuri and Gollahalli, 2000c)

$$SV = \frac{A_{12}}{A_{12} + P + Q + S} \quad [3.5]$$

where

A_{12} = Einstein spontaneous emission coefficient for this transition

P = Predissociation from the excited level

Q = Collisional quenching

S = Stimulated emission

3.2 COLLISIONAL QUENCHING OF OH

The rate of collisional removal of x from its upper excited state can be expressed as (Tamura et al., 1998)

$$Q = \sum_i \chi_i q_{ix} \quad [3.7]$$

where

χ_i = Collider species mole fraction

q_{ix} = Rate of quenching of species x by collider species i

The collision cross section of OH of the state A has been measured over a wide range of temperature by several investigators (Tamura et al., 1998, Zizak et al., 1986, Paul et al., 1994). Paul et al. (1994) developed a model to fit the experimental findings.

$$q_{i\infty} = a_i \sigma_q T^{0.5} \quad [3.8]$$

where

a_i = Fit coefficient for collider species i

σ_q = Collision cross-section

T = Local flame temperature

The temperature dependence of the collisional cross section can be expressed as a two-parameter empirical equation

$$\sigma_q = \sigma_{q\infty} e^{\frac{\epsilon}{kT}} \quad [3.9]$$

The values of a , $\sigma_{q\infty}$ and ϵ/k for major collider species of OH compiled from (Tamura et al., 1998 and Paul et al. 1994) are summarized in Table 3.1.

3.3 COLLISIONAL QUENCHING OF CH

Quenching rate coefficients for CH at flame temperature are not so well established. For most of the important colliders found in flames, measurements were made in cooled and heated cells by Stuhl (1998), covering the temperature range 240 to 420 K and later extended to 950 K by Heinrich and Stuhl (1999). These measurements were combined

with laser pyrolysis/LIF measurements at 1300 K and upper limits from the flame studies to obtain an overall idea of the temperature dependence.

Unlike OH quenching, CH does not appear to be governed by attractive forces, except for strongly polar quenchers like H₂O and NH₃. Rather, the rate coefficient generally increases with temperature in an Arrhenius form, faster than would be the case for a constant cross section. Indeed a theoretical calculation indicates a barrier of potential surface by quenching H₂. Quenching by O₂ has a complex dependence suggesting perhaps a two-mechanism. Kenner et al. (1991) contains a comprehensive discussion of theoretical formulations of quenching of several hydride radicals including CH.

The data listed in Kenner et al. (1991) and Henrich and Stuhl (1999) were carefully examined to model the CH quenching cross section. However, the data presented in those studies are not quite consistent. Cross-sections for H₂ and CO are fixed to be the 1300 K value of Garland and Crosley (1986), the E_a values are used from Kenner et al. to form a three parameter rate coefficient

$$k_Q = AT^n \exp\left(-\frac{E_a}{kT}\right)$$

The cross section expression for CH quenching by CH₄ is based on 300 K measurement by Nokes and Donovan (1984). An E_a value for CH₄ is not available, instead the E_a value of C₂H₆ was used. Since the E_a (0.714 kJ) value is small for C₂H₆ and the exact value of E_a is not important for determining k_Q, this assumption did not critically affect the accuracy of the model.

For O_2 , N_2 and CO_2 the three-parameter Arrhenius fit given by Henrich and Stuhl (1999) was used. Since the values of N_2 and CO_2 have large values of the important coefficient n , uncertainty is considerable in their extrapolation to values above the highest measured temperature i.e. 1300 K. Stuhl (1998) indicated that uncertainties are on the order of 25% in the fitted values of these parameters.

Information about Quenching of CH by H_2O measured above 415 K is not available in the literature. In this situation two possibilities could be considered based on theoretical reasoning (quantum structure and collision orientation, comparing with similar quencher such as NH_3): first, a constant cross-section with the rate coefficient increasing as the square root of temperature; second, a constant cross-section with the rate coefficient decreasing as the square root of temperature. Since the first consideration is more consistent with the analogy of similar polar quencher NH_3 , in the present study the rate coefficient of H_2O was assumed to vary as $T^{1/2}$.

Also, no information was available regarding the quenching of CH by H and OH. For convenience the quenching rate coefficients were set same as H_2 . Although, the theoretical reasoning behind this assumption is not very strong, with no available data it is the closest plausible assumption that can be made. Table 3.2 summarizes the cross-section and rate coefficient of different collider species used in the present study.

3.4 CALCULATION METHODOLOGY

As mentioned earlier, there are three methods available to evaluate the rate of collisional quenching (i) direct measurement of fluorescence lifetime using picosecond spectroscopy (ii) estimation from models using the local flame temperature and the equilibrium composition of the collider species based on the flame temperature (iii) calculation from models by using the local flame temperature and measured collider species concentration. In the present study, a relative study of methods (ii) and (iii) is presented for their application in atmospheric pressure diffusion flames.

To evaluate the rate of collisional quenching through methods (i) and (ii) the local flame temperature and collider species concentration needs to be known since Eq. 6, 7 and 8 requires input of collider species and temperature at the point of measurements. However, this often contradicts the purpose of certain experiments. A common practice is to use the equilibrium composition of the collider species by simply assuming that the error due to this approximation is acceptable. Although this approximation simplifies the experimental complication, quantitative information about the error still requires to interpret the absolute concentration of the probed species.

3.5 RAMAN SPECTROSCOPY

Raman scattering is the result of an inelastic, nonresonant interaction between an incident photon and a scatterer (Cheng et al., 1998, Laufer, 1996, Gardiner and Graves, 1989). The extent of scattering refers to the certain properties of the probed species such as spectroscopic characteristics, density, and temperature. Hence, if the scattering

characteristics of the probed species are known, its thermodynamics properties can be measured from Raman spectra (Gardiner and Graves, 1989).

$$S_R(\nu, J) \propto \frac{g_J(2J+1)(\nu+1)\nu_s^4 A_{\Delta J=0, J}}{Q_{rot} Q_{vib}} e^{\left(\frac{hc [-G_v(\nu) - F_v(J)]}{kT} \right)} \quad [3.10]$$

The Raman signal from Stokes Q-branch transition can be expressed as

S_R = Raman signal

ν = Vibrational quantum number

J = Rotational quantum number

g_J = Nuclear spin degeneracy

A = Placzek coefficient

Q_{rot} = Rotational partition function

Q_{vib} = Vibrational partition function

G = Vibrational term value

F_v = Rotational term value

k = Boltzman constant

T = Local temperature

The exponential dependence of S_R on temperature is the basis of Raman scattering based temperature measurements.

3.6 CONCENTRATION MEASUREMENT USING RAMAN SPECTROSCOPY

The Raman signal due to pulsed laser illumination of the probed area

$$S_R = C \times n_A \quad [3.11]$$

where

C = Group of experimental constant [Eq. 3.2]

n_A = Number of molecules of the probed species

Number of molecules of the probed species can be determined as (Gardiner and Graves, 1989)

$$n_A = \frac{I_L}{h\nu_L} \chi \beta_g(T) \left(\frac{d\sigma_s}{d\Omega} \right) \Delta l \quad [3.12]$$

where

I_L = Incident laser energy

ν_L = Incident laser frequency

χ = Mole fraction of the probed species

$\beta_g(T)$ = Temperature dependent Boltzman fraction

h = Plank's constant = 6.63×10^{-34} J-s

Δl = Portion of the excited beam where molecules are excited

$\left(\frac{d\sigma_s}{d\Omega} \right)$ = Raman differential cross-section

For several molecules of interest in combustion diagnostics, the model proposed by Bischel and Black (1983) from experimental findings is a reasonable approximation.

$$\left(\frac{d\sigma_s}{d\Omega} \right) = A \frac{\nu_s^4}{(\nu_i^4 - \nu_L^2)^2} \quad [3.13]$$

where

A = Placzek coefficient

ν_i = Experimental coefficient

ν_s = Stoke's frequency

ν_L = Incident laser frequency

The coefficients A and ν_i extracted from Bischel and Black (1983) are summarized in Table 3.3.

3.7 TEMPERATURE MEASUREMENT USING RAMAN SPECTROSCOPY

The temperature measurement can be done by the spectral fit of the N_2 -Q branch. The theoretical N_2 spectra obtained by Eq. 3.10 is compared with N_2 spectrum. The N_2 Raman spectra are preferable for temperature measurement because its omnipresent behavior in diffusion flames.

3.8 RESULTS AND DISCUSSIONS

Radial profiles of measured flame temperature and species concentration measured 8mm above the burner tip are shown in Fig. 3.1. The values represent an averaged measurement over 20s, which is equivalent to 200 laser pulses. The flame temperature data were calculated from the spectral fitting of the N_2 raman spectra (Eq. 3.10). The

axial location of the measurement was approximately 8 mm above the burner height. The peak flame temperature occurred at the fuel-air interface, which was approximately 2.5 diameters from the central axis of the flame. Close to the fuel-air interface, measured flame temperature showed a comparable agreement with previous thermocouple measurements (Choudhuri and Gollahalli, 2000a), and 10-15% higher than those measured by OH spectral fitting (Choudhuri and Gollahalli, 2000b). The error imposed by the approximated collisional-quenching model in OH LIF measurement might be the reason of this discrepancy. It is apparent that N_2 -based Raman spectroscopic technique provides a more accurate measurement of temperature than those of fluorescence spectroscopy in diffusion flames, where wide ranges of flame stoichiometry exist and it is difficult to accurately estimate collisional removal of excited molecules due to lack of sufficient information about collider species and their collision cross-section. However, the signal-to-noise ratio in LIF technique is much higher and is preferable where significant scattering from non-probed molecules occurs such as in the soot burning regions of hydrocarbon diffusion flames. The radial concentration profiles of the major collider species at the same axial location are also shown in Fig 3.1. The mole fractions of the species were calculated from their Raman scattering using Eq. 3.11 and Eq. 3.12. The radial profile of the CO_2 mole fraction followed the temperature profile. The CH_4 mole fraction decreased as fuel was consumed in the fuel-air interface located 2.5 diameters from the flame axis. The O_2 concentration decreased towards the central axis of the flame. The peak concentration of H_2O occurred in the air-side of the flame.

The rate of quenching of OH, calculated using the measured temperature and collider species concentrations (Eq. 3.7 and Eq. 3.8) is shown in Fig. 3.2. The σ_{qx} values were taken from Paul et al. (1994). The calculation shows that the maximum possible quenching is due to CH₄. The collisional removal by other major species is in the same order. Furthermore, the quenching of OH due to CH₄ is significant on the axis of the flame where significant amounts of CH₄ exist.

The rate of quenching of OH, estimated from the equilibrium composition (Reynolds, 1986) of collider species at the measured flame temperature is shown in Fig. 3.3. The estimated rate of collisional removal of OH from equilibrium composition was lower than the measured rate. The radial profile of OH mole fraction calculated from LIF signal using both equilibrium composition and measured collider species concentration is shown in Fig. 3.4. The peak of the OH radical concentration occurred in the fuel-air interface where the flame temperature has its maximum value. The OH concentration decays quickly on both sides of the fuel air interface since most of the reactions involving OH formation usually occurs close to the peak temperature region. In fact, OH radicals serve as a marker of the reaction zone. The differences of the OH mole fraction computed by using estimated and calculated quenching models are significant on both sides of the fuel-air interface.

The rates of collisional quenching of CH for different major colliders calculated from measured collider concentrations are shown in Fig. 3.5. The collisional quenching rate of CH is a factor of 10 lower than that of OH quenching. As mentioned, earlier the most

dominant quenchers are polar molecule H_2O and CH_4 . The H_2O quenching is higher in fuel-oxidizer region due to formation of high H_2O in this area. Similarly, the CH_4 quenching is higher close to the central axis where maximum concentrations of CH_4 exist. The estimated collisional quenching rate based on equilibrium values of collider concentration are shown in Fig. 3.6. The rates of estimated quenching are quite inconsistent with the measured counterpart. As it can be seen that, the methods of estimation yield higher quenching by H_2 and CO . Due to the equilibrium assumption both H_2 and CO concentrations are over predicted which lead to high values of collision rates by these molecules. Since the equilibrium calculation is solely temperature dependent, it does not account for several low temperature H_2 and CO consumption mechanisms in hydrocarbon chemistry and thereby predicts high concentrations of H_2 and CO . So it is readily evident that the equilibrium assumption for collisional removing estimation seriously affects the accuracy of CH fluorescence quantification process. The radial profiles of CH concentration based on method of estimation and method of calculation are shown in Fig. 3.7 and it can be seen that unlike OH radical, use of method of estimation leads to higher CH concentration measurement in flame.

Comparative studies of OH and CH quenching quantified through two different approaches are presented in this study. Albeit the calculation process from measured collider species is superior to the estimation from equilibrium process, however, the purpose of the study is to quantify the amount of error if a equilibrium composition based collisional model is used. In fact, it is not always possible to measure the collider species concentration due to the complexity and cost of the experiments. Although

Raman Scattering technique is a robust spectroscopic technique. its application is quite challenging in practical combustion situations due to its very low signal-to-a noise ratio. On the other hand, the LIF based measurement is a quite attractive technique with very high signal-to-noise ratio and the fluorescence signal can be quantified with a reasonable accuracy even with the approximated quenching model.

Table 3.1: Parameters for OH Collisional Quenching

Collider	σ_{qr} (Å ²) Tamura et al.	σ_{qr} (Å ²) Paul et al.	ϵ/k (K)	a
N ₂	0.351	0.4	624	4.47
O ₂	8.00	8.0	243	4.37
CO ₂	11.87	11	488	4.16
CO	12.30	12.0	397	4.47
H ₂ O	17.87	20	434	4.92
H	14.29	14.5	84	15.0
OH	14	20	384	4.99
H ₂	4.24	4.5	224	10.88
CH ₄	13.68	11	488	4.16

Table 3.2: Parameters for CH Collisional Quenching

Collider	Quenching Cross Section, Å ² $\sigma_q = AT^B \exp(-C/T)$			a
	A	B	C	
N ₂	1.03E-9	2.9	-192	4.88
O ₂	8.61E-6	1.64	-867	4.79
CO ₂	8.67E-13	3.8	-854	4.59
CO	8.31	0	0	4.88
H ₂ O	166	-0.5	0	5.30
H	221	-0.5	686	15.09
OH	221	-0.5	686	5.36
H ₂	221	-0.5	686	11.02
CH ₄	52.8	-0.5	84	5.43

Table 3.3 Coefficients for Bischel and Black's (1983) Raman Differential Cross-section model

Molecule	$A \times 10^{-28} \text{ cm}^2/\text{s}$	$\nu_1 \times 10^4 \text{ cm}^{-1}$
CH ₄	10.4	7.23
H ₂	8.74	8.48
N ₂	3.02	8.95
O ₂	0.459	5.69
CO ₂	1.4	1.38
CO	1.0	2.145

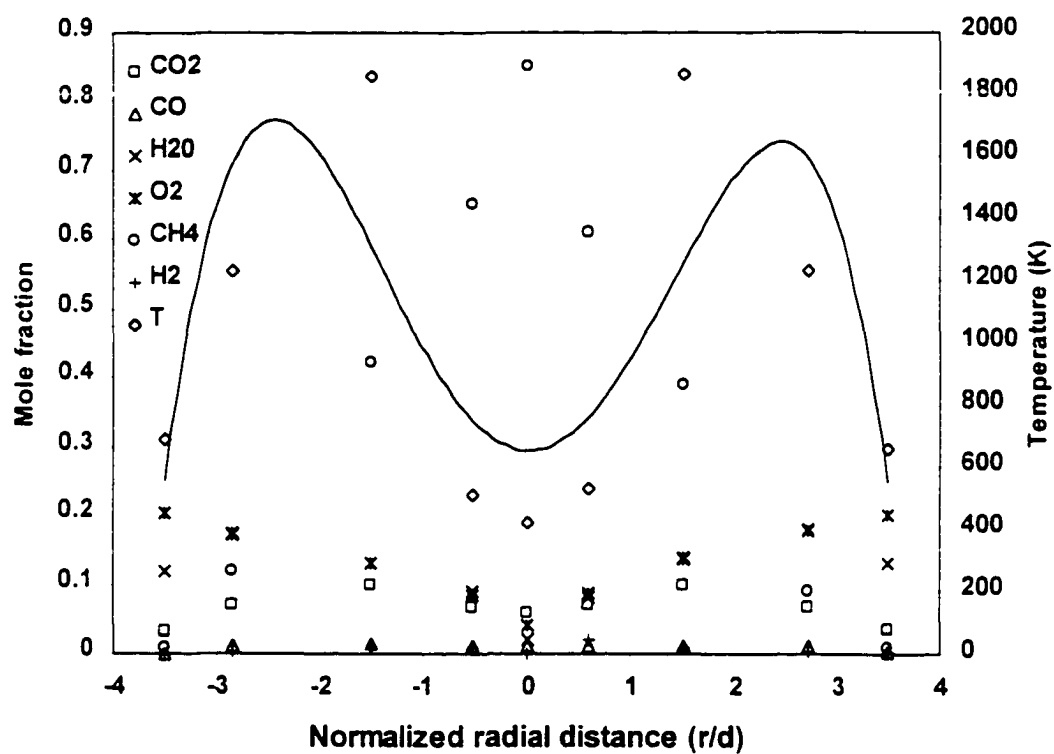


Fig. 3.1 Radial Profiles of Time-averaged Flame Temperature and Collider Species Concentration

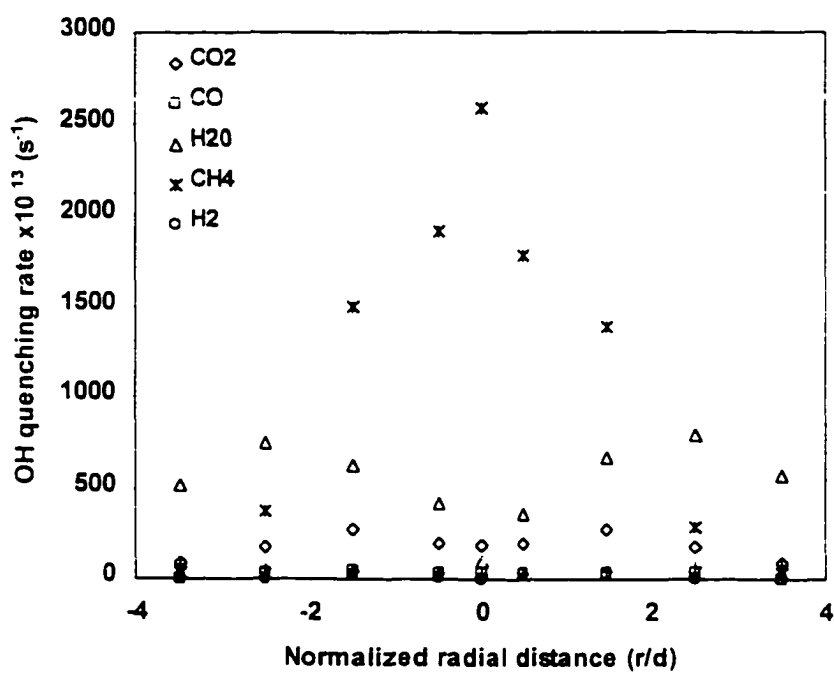


Fig. 3.2 Radial Profiles of OH Quenching Rate Calculated From Measured Collider Species Concentrations

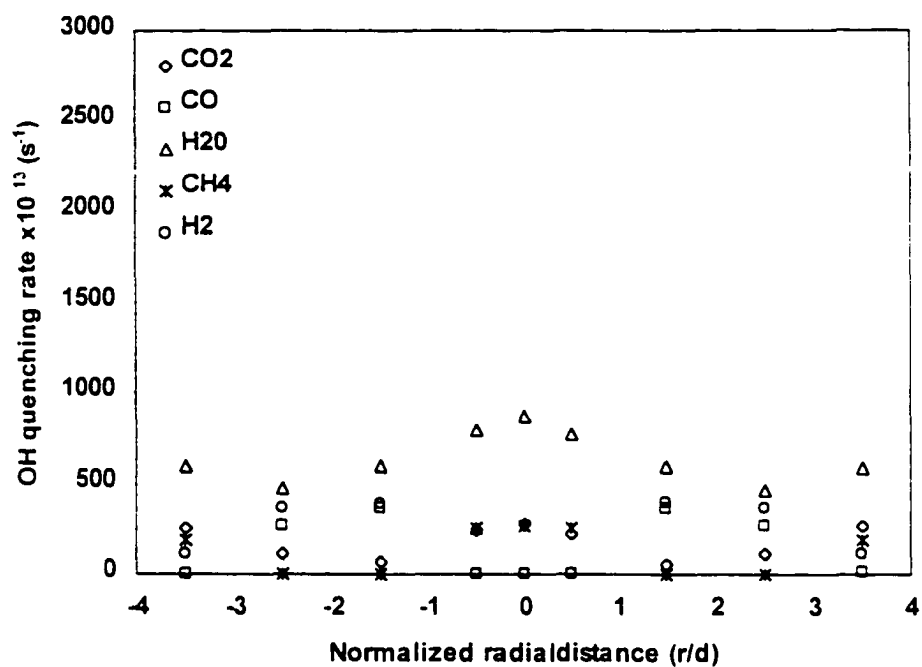


Fig. 3.3 Radial profiles of OH quenching rate estimated from local equilibrium composition

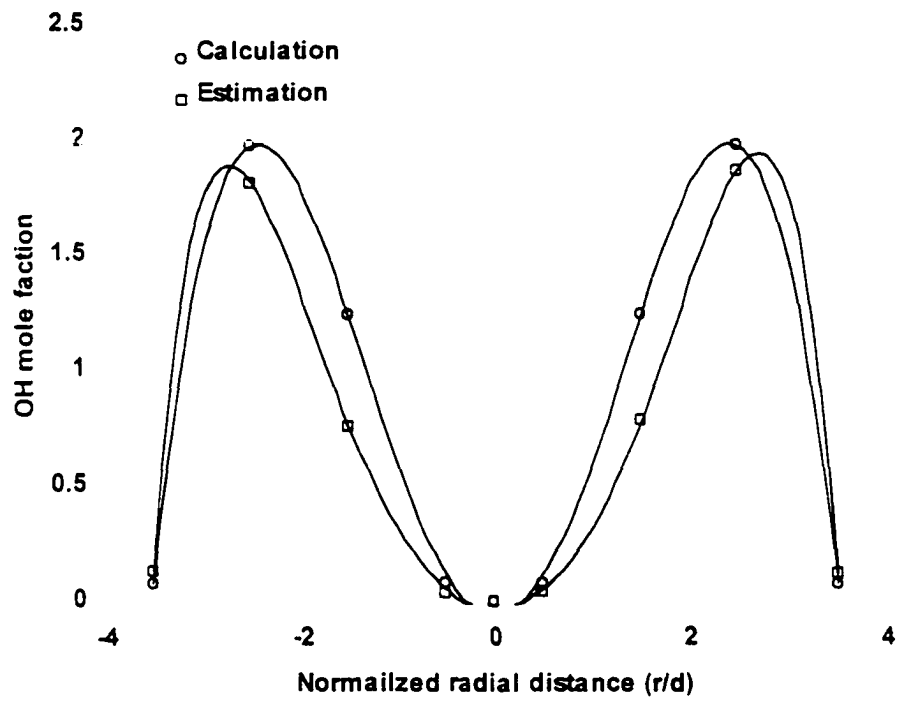


Fig. 3.4 Radial Profiles of OH Concentration

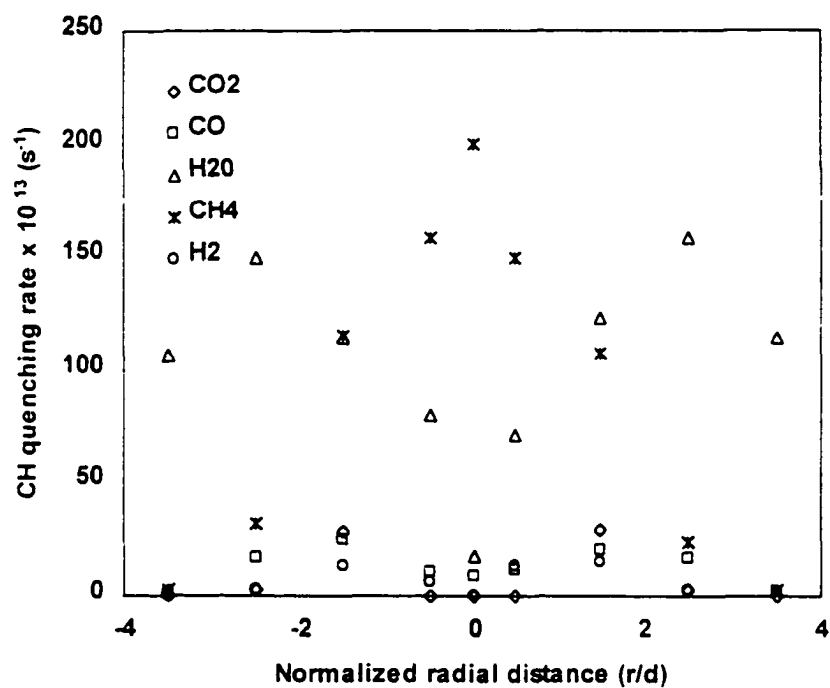


Fig. 3.5 Radial Profiles of CH Quenching Rate Calculated From Measured Collider Species Concentrations

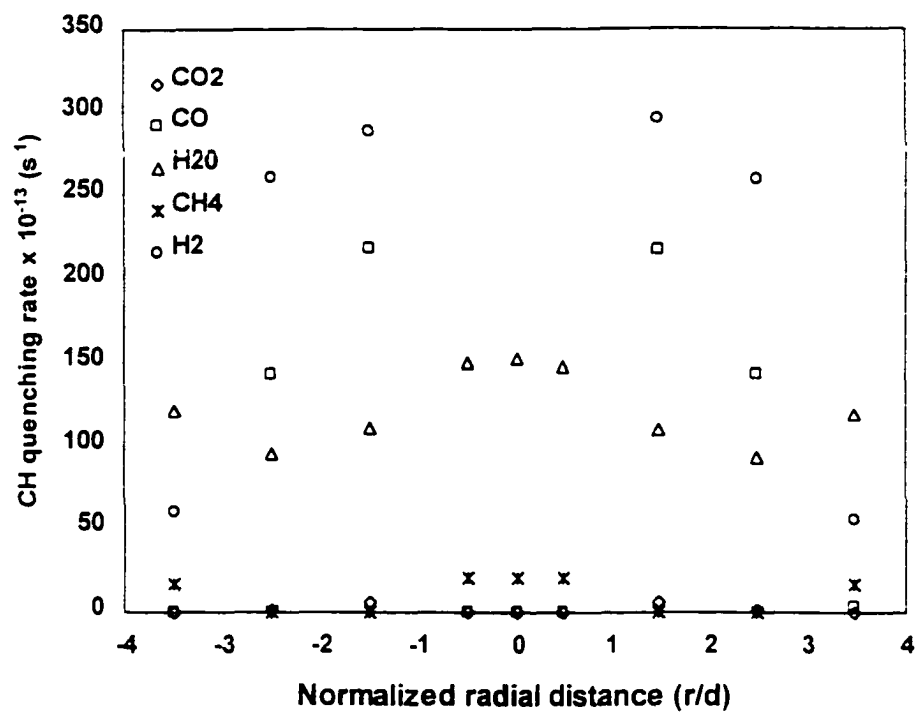


Fig. 3.6 Radial profiles of CH quenching rate estimated from local equilibrium composition

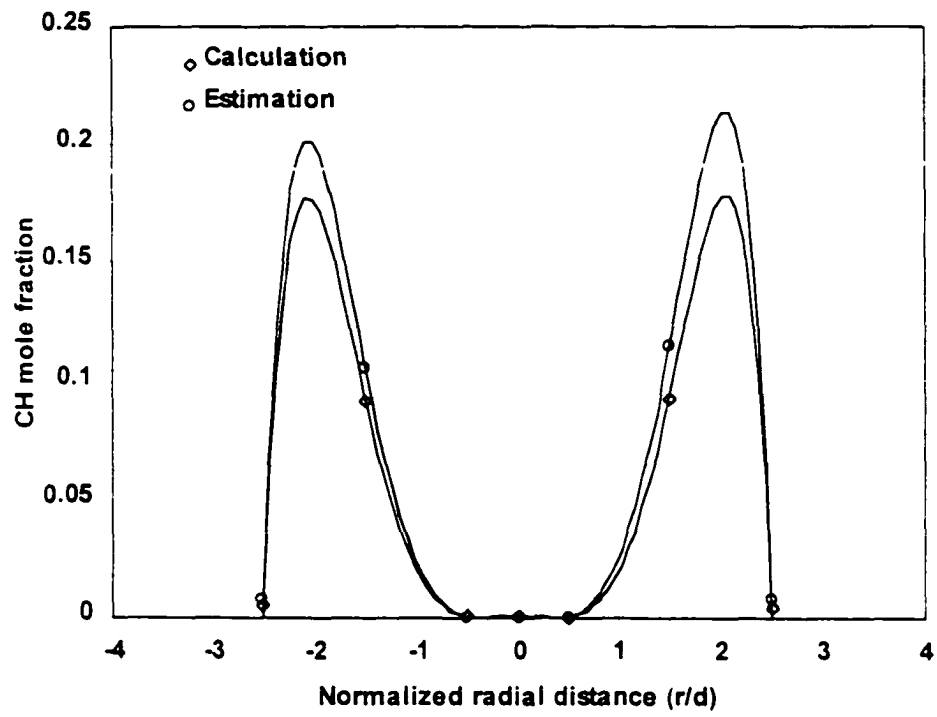


Fig. 3.7 Radial Profiles of CH Concentration

The flow-field of the hydrogen-hydrocarbon composite fuels issued from a circular burner in quiescent atmosphere was numerically computed. Both cold and burning jets were simulated. Turbulence model and turbulence-chemistry interaction were employed in the computation. A single step global reaction mechanism was used to represent the kinetics. The numerical computations were conducted using the CFDRC-ACE™* computational fluid dynamics software which contains a Structured/Unstructured Flow Solver (CFD-GUI™), an Interactive Geometric Modeling and Grid Generation module (CFD-GEOM™) and a 3-D Computer Graphics and Animation Software (CFD-VIEW™). The code was run on Windows NT Platform with Gateway GP6-400 (Pentium II 400 MHz and 256 MB RAM) PC.

4.1 Geometric Grid Generation

The flow field was modeled using a multi-domain approach with a body-fitted coordinate system and structured grid system. The axisymmetric computational domain (half of the flame) was extended 85.6 cm in the axial direction and 14 cm in the radial direction and had a burner (3.8 mm ID) protruding 2 cm inside the domain. The whole computational domain was divided into three blocks for smoother grid generation. A total number of 13845 (195×71) cells were generated using non-uniform grid spacing to provide an adequate resolution near the jet axis and close to the burner where gradients were large. The grid spacing increased in the radial and axial directions to save the CPU

time since gradients were small in the far-field. During grid generation and geometric modeling, important factors such as right-handed grid, smooth transition from small to large cells (grid tolerance) and grid orthogonality were also taken into account. Furthermore, the computational results were checked for grid independence and it was found that increasing the total number of cells to 19565 (215× 91) produced only a change of 0.8% in mixture fraction for the base line conditions. The computational domain was bounded by the axis of symmetry and free-symmetry in the radial direction and by inflow and outflow in the axial direction. The outer-boundaries were located far from the burner exit (=220 burner diameters) and the symmetry axis (=37 burner diameters). Jet diffusion flame is usually anchored to the outside edge of the burner tip. Hence, the near burner flame structure is usually affected by the boundary-layer development on the fuel tube and the heat transfer between the burner tip and the flame. In the present computation a no-slip boundary condition was imposed on the outer wall of the burner to account for the effect of boundary layer. However, the heat transfer between the burner and flame was neglected to avoid the complexity of a conjugate heat transfer problem. The details of the computational domain are summarized in Table 4.1 and shown in Fig. 4.1.

4.2 Governing Equations

For solving the flow field, Favre Averaged Navier Stokes Equations (FANS) were used. In this type of averaging technique a flow quantity is decomposed as its mean and fluctuating value

$$\theta = \tilde{\theta} + \theta'' \quad \text{where } \tilde{\theta} = \overline{\rho\theta} / \bar{\rho} \quad [4.1]$$

Applying the Favre averaging procedure to continuity equation, the Favre-averaged continuity equation can be obtained

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j) = 0 \quad [4.2]$$

Similarly, the Favre-averaged Navier Stokes Equations are

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{u}_i) + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_i \tilde{u}_j) = -\frac{\partial \bar{p}}{\partial x_i} + \frac{\partial}{\partial x_j} \left[\bar{\mu} \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \frac{\partial \tilde{u}_m}{\partial x_m} \delta_{ij} \right) \right] + \frac{\partial}{\partial x_i} (-\rho \tilde{u}_i \tilde{u}_i'') + \bar{\rho} f_i \quad [4.3]$$

In the present computation, the Boussinesq eddy viscosity concept was used in which the Reynolds stress $-\rho \tilde{u}_i \tilde{u}_i''$ is treated as a linear function of the mean strain rate

$$-\rho \tilde{u}_i \tilde{u}_i'' = \mu_t \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \frac{\partial \tilde{u}_m}{\partial x_m} \delta_{ij} \right) - \frac{2}{3} \bar{\rho} k \delta_{ij} \quad [4.4]$$

Where μ_t is the turbulent eddy viscosity and k is one half the trace of the Reynolds stress tensor.

$$k = \frac{1}{2} (\tilde{u}_k \tilde{u}_k'') \quad [4.5]$$

By substituting the Reynolds stress term in the Navier Stokes Equation

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{u}_i) + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_i \tilde{u}_j) = -\frac{\partial \bar{p}}{\partial x_i} + \frac{\partial}{\partial x_j} \left[(\bar{\mu} + \mu_t) \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \frac{\partial \tilde{u}_m}{\partial x_m} \delta_{ij} \right) \right] - \frac{2}{3} \frac{\partial}{\partial x_i} (\bar{\rho} k) + \bar{\rho} f_i \quad [4.6]$$

Similarly, when averaging technique applied to the static enthalpy equation, the averaged equation also contains a fluctuating enthalpy and velocity term, which also requires modeling through Boussinesq concept.

$$-\widetilde{\rho u''h''} = \Gamma_t \frac{\partial \tilde{h}}{\partial x_i} \quad [4.7]$$

The term Γ_t is the turbulent or eddy diffusivity and is related to the turbulent viscosity through the turbulent Prandtl number σ_t ,

$$\Gamma_t = \frac{\mu_t}{\sigma_t} \quad [4.8]$$

The mean enthalpy equation is derived by adding the mean kinetic energy equation to the mean static enthalpy equation.

$$\frac{\partial \tilde{p}h}{\partial t} + \frac{\partial}{\partial x_i} (\tilde{\rho} \tilde{u}_i \tilde{h}) = \frac{\partial}{\partial x_i} \left\{ (\Gamma + \Gamma_t) \frac{\partial \tilde{h}}{\partial x_i} \right\} + \frac{\partial \tilde{p}}{\partial t} + u_i \frac{\partial \tilde{p}}{\partial x_i} + (\bar{\mu} + \mu_t) \frac{\partial \tilde{u}_i}{\partial x_i} \left(\frac{\partial \tilde{u}_i}{\partial x_i} + \frac{\partial \tilde{u}_i}{\partial x_i} - \frac{2}{3} \frac{\partial \tilde{u}_k}{\partial x_k} \delta_{ii} \right) \quad [4.9]$$

4.3 Turbulence Modeling

In the computational methodology applied in the present investigation, the effect of turbulence is accounted for via turbulent or eddy viscosity. Following the kinetic theory of gases, the eddy viscosity is modeled as the product of a velocity scale q and a length scale l .

$$\mu_t = C \bar{\rho} q l \quad [4.10]$$

where C is a constant of proportionality. Various turbulence models differ in the way q and l are estimated. In the present study, standard k - ϵ model based on Launder and Spalding (1974) was used. This model employs two partial differential equations to estimate the velocity and length scales and hence commonly known as the two-equation

model. These equations are the k and ε equations, which govern the transport of the turbulent kinetic energy and its dissipation rate respectively. The modeled equations are

$$\frac{\partial(\bar{\rho}k)}{\partial t} + \frac{\partial}{\partial x_j}(\bar{\rho}\tilde{u}_j k) = \bar{\rho}P - \bar{\rho}\varepsilon + \frac{\partial}{\partial x_j} \left[\frac{\bar{\mu} + \mu_t}{\sigma_k} \frac{\partial k}{\partial x_j} \right] \quad [4.11]$$

$$\frac{\partial(\bar{\rho}\varepsilon)}{\partial t} + \frac{\partial}{\partial x_j}(\bar{\rho}\tilde{u}_j \varepsilon) = C_{\varepsilon_1} \frac{\bar{\rho}P\varepsilon}{k} - C_{\varepsilon_2} \frac{\bar{\rho}\varepsilon^2}{k} + \frac{\partial}{\partial x_j} \left[\frac{\bar{\mu} + \mu_t}{\sigma_\varepsilon} \frac{\partial \varepsilon}{\partial x_j} \right] \quad [4.12]$$

The production term P defined as

$$P = \bar{\rho}\mu_t \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} - \frac{2}{3} \frac{\partial \tilde{u}_m}{\partial x_m} \delta_{ij} \right) \frac{\partial \tilde{u}_i}{\partial x_j} - \frac{2}{3} k \frac{\partial \tilde{u}_m}{\partial x_m} \quad [4.13]$$

The square root of k is taken to be the velocity scale while the length scale is obtained from

$$l = \frac{C_\mu^{1/4} k^{3/4}}{\varepsilon} \quad [4.14]$$

The expression for eddy viscosity

$$\mu_t = \frac{\bar{\rho}C_\mu k^2}{\varepsilon} \quad [4.15]$$

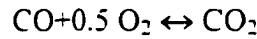
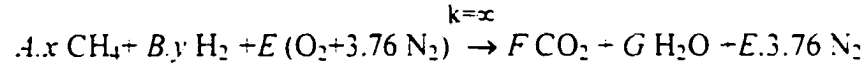
The five constants used in the model are (Launder and Spalding (1974))

$$C_\mu = 0.09; \quad C_{\varepsilon_1} = 1.44; \quad C_{\varepsilon_2} = 1.92; \quad \sigma_k = 1.0; \quad \sigma_\varepsilon = 1.3$$

4.4 Reaction Model

In the present computation, a two-step chemistry model was used to represent the chemical kinetics. The kinetics model consists of a stoichiometrically correct reaction

step describing the oxidation of methane and hydrogen and a reversible CO oxidation reaction (Brink et al., 1999).



The stoichiometric coefficients A , B , E , F and G were determined depending on the value of x and y which are the mass fraction of natural gas and hydrogen in the fuel mixture.

The forward and reverse rate constants used in CO oxidation reaction are $1.178 \times 10^9 \text{ cm}^3/\text{gmol}\cdot\text{s}$ and $4.132 \times 10^{11} \text{ cm}^3/\text{gmol}\cdot\text{s}$ respectively (Howard et al., 1972).

The reaction can be generalized as

$$\sum \alpha_i R_i = \sum \beta_j P_j \quad [4.16]$$

where R_i and P_j are the reactants and products respectively while α_i and β_j are the corresponding moles. The equation can be rewritten as mass unit

$$\sum \alpha_i M_i R_i \rightarrow \sum \beta_j M_j P_j \quad [4.17]$$

by normalizing both sides with $\alpha_i M_i$

$$\sum r_i^* R_i \rightarrow \sum r_j^* P_j \quad [4.18]$$

The quantity r_i^* stands for the required mass of R_i to stoichiometrically react with a unit mass of R_i . Similarly r_j^* represents the mass of P_j produced per unit mass of R_i . The stoichiometric mass fraction is defined as

$$Y_i^* = r_i^* Y_i^0$$

now, if $Y_i^* < Y_i^0$ it implies that R_i is in excess and if $Y_i^* > Y_i^0$ it implies that R_i may be fully consumed. If R_m is the reactant species which satisfies the following relation

$$Y_m^* - Y_m^0 = \max(Y_i^* - Y_i^0) \quad i = 1, 2, \dots, N_R$$

then R_m will be fully consumed and the mass fractions of the remaining reactant and product species can be determined from the following relations

$$Y_i = Y_i^0 \left(1 - \frac{Y_m^0}{Y_m^*} \right) \quad [4.19]$$

$$Y_i = Y_i^0 + Y_i^* \frac{Y_m^0}{Y_m^*} \quad [4.20]$$

The mixture molecular weight, density and species concentration were calculated from mass fraction using the following relations

$$x_i = \frac{Y_i M}{M_i} ; \quad M = \frac{1}{\sum \frac{Y_i}{M_i}} ; \quad c_i = \frac{\rho Y_i}{M_i} \quad [4.21]$$

4.5 Turbulence Combustion Interaction

Through the fast chemistry assumption, the mixture fraction is used as conserved scalar and it can be used to model turbulence-combustion interaction. With this assumption, the reaction time is assumed to be negligibly short in comparison with the turbulent mixing time. In the limit of fast chemistry, the instantaneous species concentrations and temperature are functions only of the instantaneous mixture fractions. To solve for the instantaneous mixture fraction, a probability density function (pdf) needs to be defined. The computation code used in the present study assumes a *top hat* function for mixture fraction pdf with possible delta function at 0 and 1.

$$p(f) = \begin{cases} P_0, & f = 0 \\ c, & 0 < a \leq f \leq b < 1 \\ P_1, & f = 1 \end{cases} \quad [4.22]$$

To determine the parameters P_0 , P_1 , a , b and c , the following moments are used.

$$\int_0^1 p(f) df = 1$$

$$\int_0^1 f p(f) df = \tilde{f}$$

$$\int_0^1 (f - \tilde{f})^2 p(f) df = \tilde{g}$$

The mean, $\tilde{f}$, and variance $\tilde{g}$, of the mixture fraction are determined by solving the following transport equations

$$\bar{\rho} u_i \frac{\partial \tilde{f}}{\partial x_i} = \frac{\partial}{\partial x_i} \left\{ \frac{\mu_i}{\sigma_i} \frac{\partial \tilde{f}}{\partial x_i} \right\} \quad [4.23]$$

$$\bar{\rho} u_i \frac{\partial \tilde{g}}{\partial x_i} = \frac{\partial}{\partial x_i} \left\{ \frac{\mu_i}{\sigma_i} \frac{\partial \tilde{g}}{\partial x_i} \right\} + 2 \frac{\mu_i}{\sigma_i} \left(\frac{\partial \tilde{f}}{\partial x_i} \right) - C_D \frac{\bar{\rho} \varepsilon}{k} \tilde{g} \quad [4.24]$$

C_D has a value of 2.

So the algebraic relations derived from pdf are followings

$$P_0 + P_1 + c(b - a) = 1 \quad [4.25]$$

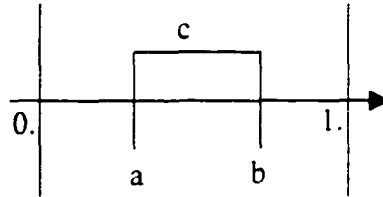
$$P_1 + \frac{1}{2} c(b^2 - a^2) = \tilde{f}$$

$$P_1 + \frac{1}{3} c(b^3 - a^3) = \tilde{g} + \tilde{f}^2$$

since the limit of f brings in two additional constraints, there are only 3 equations with 5 unknowns. The expression of $p(f)$ for $0 < f < 0.5$ are shown for three different cases

$$\text{If } 3\tilde{g} < \tilde{f}^2 \quad [4.26(a)]$$

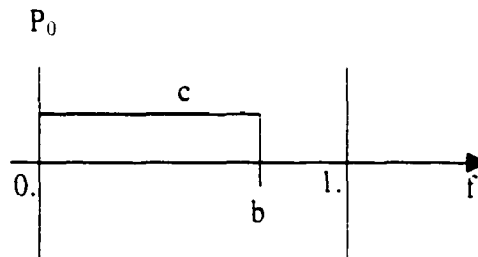
$$\begin{aligned}
 P_0 &= P_1 = 0 \\
 a &= \tilde{f} - \sqrt{3\tilde{g}} \\
 b &= \tilde{f} + \sqrt{3\tilde{g}} \\
 c &= \frac{1}{2\sqrt{3\tilde{g}}}
 \end{aligned}$$



$$\text{if } \tilde{f}^2 \leq 3\tilde{g} < 2\tilde{f} - 3\tilde{f}^2$$

[4.26(b)]

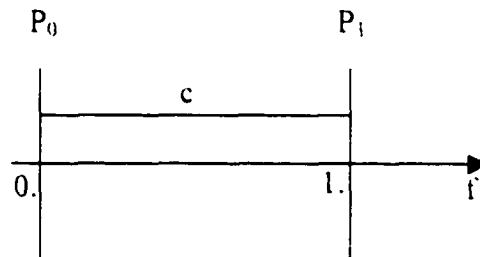
$$\begin{aligned}
 P_0 &= \frac{3\tilde{g} - \tilde{f}^2}{3(\tilde{f}^2 + \tilde{g})} \\
 P_1 &= 0 \\
 a &= 0 \\
 b &= \frac{3(\tilde{f}^2 + \tilde{g})}{2\tilde{f}} \\
 c &= \frac{8\tilde{f}^2}{(\tilde{f}^2 + \tilde{g})^2}
 \end{aligned}$$



$$\text{if } 3\tilde{g} \geq 2\tilde{f} - 3\tilde{f}^2$$

[4.26(c)]

$$\begin{aligned}
 P_0 &= (1 - \tilde{f}) + 3\tilde{f}(1 - \tilde{f}) + 3\tilde{g} \\
 P_1 &= \tilde{f} + 3\tilde{f}(1 - \tilde{f}) + 3\tilde{g} \\
 a &= 0 \\
 b &= 1 \\
 c &= 6\tilde{f}(1 - \tilde{f}) - 6\tilde{g}
 \end{aligned}$$



4.6 Thermodynamic Variables

From first chemistry assumption, the thermodynamic variables can be directly evaluated from mixture fraction. The mean values of mass fraction density and temperature can be

obtained by averaging the instantaneous values at the given mixture fraction. with the pdf over the sample domain

$$\begin{aligned}\tilde{Y}_i &= \int_0^1 Y_i(f)P(f)df \\ \tilde{T}_i &= \int_0^1 T_i(f)P(f)df \\ \tilde{\rho}^{-1} &= \int_0^1 \frac{P(f)}{\rho(f)} df\end{aligned}\quad [4.27]$$

4.7 Discretization Method

All governing equations, except the continuity equation can be expressed as a general combination of transient, convection, diffusion, and source terms.

$$\frac{\partial}{\partial t}(\rho\phi) + \frac{\partial}{\partial x_j}(\rho u_j \phi) = \frac{\partial}{\partial x_j} \left(\Gamma \frac{\partial \phi}{\partial x_j} \right) + S_\phi \quad [4.28]$$

$$\text{Transient} + \text{Convection} = \text{Diffusion} + \text{Source}$$

where S_ϕ is the source term. Γ is the transport coefficient and ϕ is the dependent variable. In the present computation, a cell-centered control volume approach is used. In this method the finite difference equations (FDE) are formulated by evaluating and integrating fluxes across the faces of the control volume (Fig 4.2) in order to satisfy the conservation equation. To evaluate the convective fluxes over a control volume a special technique known as smart scheme (also hybrid scheme) is used. Use of this method minimizes false diffusion. Also, the source term is linearized. The smart scheme and source term linearization technique used in the present technique are discussed later. The two-dimensional form of the discretized equations can be written as follows

$$a_P \phi_P = a_W \phi_W + a_E \phi_E + a_S \phi_S + a_N \phi_N + S \quad [4.29]$$

where P, W, E, N are the locations where the values are computed and a_P , a_W , a_N etc are the link coefficients. The equation is known as the finite difference equation (FDE), which is in general nonlinear because the link coefficient themselves are function of ϕ_P , ϕ_N etc. When the FDE is formulated for each computational cell, it results in a set of coupled nonlinear algebraic equations. An iterative solution scheme is employed which solves each dependent variable sequentially and repeatedly with the target of improving solution at each iteration. The iterations are under relaxed to avoid numerical oscillation.

4.7.1 Smart Scheme

The conventional central difference scheme may give raise to non physical oscillation in the numerical solution. Furthermore, the iterative method tends to be unstable when central differencing scheme is used to evaluate convection terms. So, some damping coefficient is needed for stability. In the present computation the damping coefficient was constructed as

$$C_e = d_e C_e^{UP} + (1 - d_e) C_e^{CN} \quad [4.30]$$

where d_e is a blending factor that combines the central scheme with first order upwind scheme to produce stable scheme. The coefficient d_e is also known as damping coefficients. The above equation can be rewritten as

$$C_e = G_e \frac{\phi_P + \phi_E}{2} - d_e |G_e| \frac{\phi_E - \phi_P}{2} \quad [4.31]$$

The scheme has an accuracy between 1 and 2. If $d_e = 0$, the scheme is second order accurate center difference scheme and if $d_e = 1$ the scheme is first order accurate upwind scheme. Also, in the present computation, a special technique termed as smart scheme

was utilized to evaluate d_c at different locations of the flow-field depending upon the local variation of ϕ . A new function known as the Minimize limiter is defined as follows

$$\text{Minimize}(\alpha, \beta) = \text{sign}(\alpha) \max[0, \min(|\alpha|, \beta)] \quad [4.32]$$

The smart scheme with adaptive damping coefficient can also be expressed as

$$d_c = 1 - \text{Minimize}(1, r_c)$$

where r_c is a parameter defined to measure the variation of ϕ . If $I-1$, I , $I+1$ denote the grid points W, P, and E respectively

$$r_c = \frac{\phi_{I-1-u} - \phi_{I+u}}{\phi_{I-1} - \phi_I}$$

where $u = \text{sign}(G_c)$

Thus u is -1 or 1 . The smart scheme reduces to the first-order upwind scheme when $r_c < 0$, to central difference scheme when $r_c > 1$, and to the second-order upwind scheme for $0 < r_c < 1$.

4.7.2 Source Term Linearization

The source term which is a function of ϕ is linearized as (Patankar, 1980)

$$S_o = S_o^p \phi + S_o^u \quad [4.33]$$

such that S_o^p is negative. S_o^p and S_o^u can be functions of ϕ . These coefficients are evaluated using the latest available value of ϕ i.e. the value of ϕ at the end of the last iteration. The linearized source term is integrated over the control volume, which results

in

$$S = S_p \phi_p + S_u \quad [4.34]$$

the coefficients are given as

$$S_p = \forall S^p$$

$$S_U = \forall S^U$$

4.7.3 Linear Equation Solver

In the present computation, the conjugate gradient solver (CGS) was used to simultaneously solve the linear equations. This method possesses some advantages over conventional iterative method like TDM (Tri-Diagonal Matrix) method. The CGS algorithm applied to the system $A\phi = S$ is shown below

$$\text{Initialization (n=0): } \begin{aligned} r_0 &= S - A\phi \\ q_0 &= p_{-1} = 0; \rho_{-1} = 1 \end{aligned} \quad [4.35]$$

$$\begin{aligned} \rho_n &= r_0^T r_n; \beta_n = \frac{\rho_n}{\rho_{n-1}} \\ u_n &= r_n + \beta_n q_n \\ p_n &= u_n + \beta_n (q_n + \beta_n p_{n-1}) \\ v_n &= A p_n \end{aligned}$$

$$\begin{aligned} \text{Initialization (n}\geq 0\text{): } \quad \sigma_n &= r_0^T v_n; \alpha_n = \frac{\rho_n}{\sigma_n} \\ q_{n+1} &= u_n - \alpha_n v_n \\ r_{n+1} &= r_n - \alpha_n A(u_n + q_{n+1}) \\ \phi_{n+1} &= \phi_n + \alpha_n (u_n + q_{n+1}) \end{aligned}$$

The convergence rate of conjugate gradient algorithms depends on the spectral radius of the coefficient matrix.

4.8 Solution Algorithm

The SIMPLEC (Semi Implicit Pressure Linked Equations Consistent) proposed by Van Doormaal and Ruthbay (1985) are used in the present computation. The overall solution procedure for the SIMPLEC algorithm is shown in Fig 4.3. All the parameters in the solution procedure are dictated by the number of times a procedure is repeated, and can be specified by the user. These are the number of iterations (NITER), the number of continuity iterations(C_ ITER) and, in the case of a transient simulation is simply repeated at each time step of a transient simulation. The number of iterations to be performed is dictated by the overall residual reduction obtained. At each iteration, the program will calculate a residual for each variable, which is the sum of the absolute value of the residual for that variable at each control cell. The descriptions of the procedure are as follows

1. All solution values are initialized. For steady state flows in present computation, the initial condition is only a starting point of the iterative procedure.
2. All terms needed for the u, v, and w momentum equations are evaluated for each cell. The linked coefficients and the source term are calculated. When the terms for all cells are assembled, they form a linear system.
3. The linear systems for the u, v, and w momentum equations are solved using CGS solver.

4. The mass imbalance is evaluated and an equation for the pressure correction is posed at each cell. All pressure correction equations are then assembled into a linear system and solved using the CGS solver.
5. The pressure correction is applied to the previous step applied to the pressure field. The velocity field is corrected by using an algebraic relationship between pressure correction and velocity correction.
6. The mixture fraction equations are solved.
7. Steps 2 to 6 are repeated until convergence is obtained.

The following assumptions have been made to simplify the numerical analysis

1. Two-dimensional incompressible axisymmetric flow
2. Newtonian Fluid
3. Two-step finite chemical kinetics for fuel oxidation
4. Non-stiff kinetic system
5. Soot free diffusion flame (no heterogeneous combustion)
6. Negligible radiation losses from the flame

4.9 Validity of the Numerical Solution

The validity of the numerical code was verified by comparing computed results with experimental data presented by Katta and Roquemore, 1998. The experimental conditions used by Katta and Roquemore, 1998 are listed in Table 4.2. The code along with the detailed chemical kinetics of methane combustion (Appendix I, Choudhuri and Gollahalli, 2000b) was used to compute the mass fraction of methane and oxygen in a laminar methane jet which has same initial conditions as those used in the experiments

of Katta and Roquemore, 1998. Fig. 4.4 and Fig. 4.5 show the radial mass fraction of methane and oxygen respectively at an axial location 50 mm downstream. The computed mass fractions of methane and oxygen show good agreement (within 6% of the experimental values) with the experimental measurements.

Table 4.1 Details of the Computational Domain

General Features	Axial Dimension: 85.6 cm Radial Dimension: 14 cm Number of Grids: 13854 (195 × 71) Grid Clustering: Hyperbolic Tangent		
Boundary Conditions		Inflow	Outflow
Flow	Velocity	Fixed Velocity $U_{air} = 2 \text{ m/s}$ $V_{air} = 0$ $U_{fuel} = 55 \text{ m/s}$ $V_{fuel} = 0$	Extrapolated
	Pressure	$P(\text{rel}) = 0$	Extrapolated
	Temperature	$T = 300 \text{ K}$	Extrapolated
Turbulence	Kinetics Energy (k/U_c^2)	0.002 (air) 1 (fuel)	3
	Dissipation	0.1 (air) 0.1 (fuel)	400
Initial Conditions			
Flow Turbulence	Velocity	$U = 5 \text{ m/s}$ $V = 0$	
	Pressure	$P(\text{rel}) = 0$	
	Kinetic Energy (k/U_c^2)	2	
	Dissipation	300	
k-ε Model		Turbulent Prandtl No = 0.73 Turbulent Schmidt No.=0.73	

Table 4.2 Experimental Conditions Used by Katta and Roquemore, 1998

Burner Type	Tubular
Burner ID	1.2 mm
Burner OD	2.1 mm
Fuel	98% CH ₄ , 1% N ₂ , 0.35% C ₃ H ₈
Burner Exit Reynolds Number	270
Burner Exit Velocity	3.78 m/s

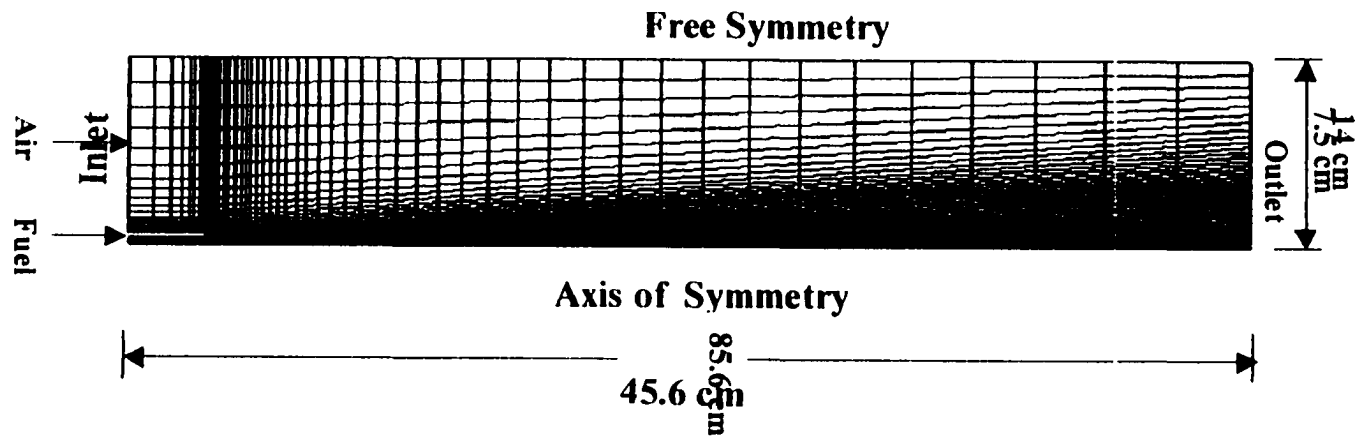


Fig. 3.1 Computational Domain

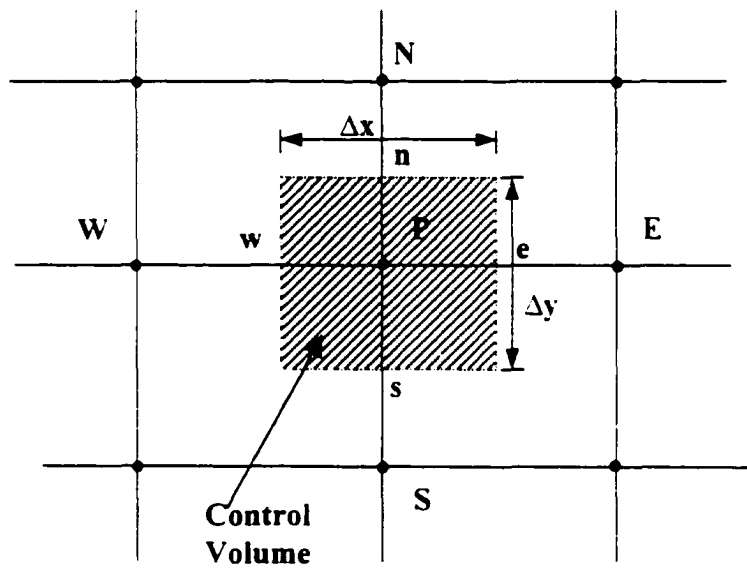


Fig. 4.2 Control Volume for Two-dimensional Grid

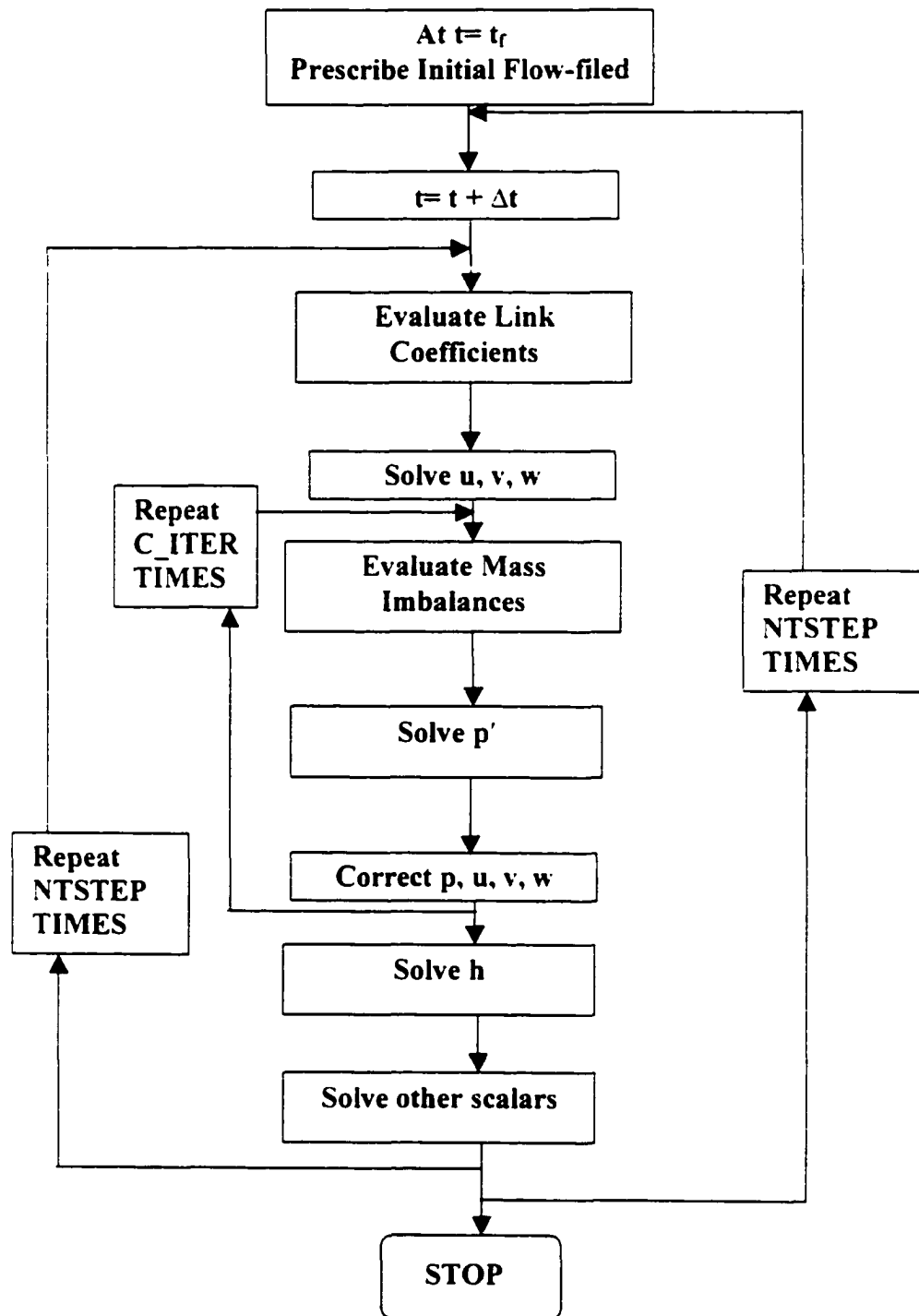


Fig. 4.3 Solution Flowchart for SIMLEC Algorithm

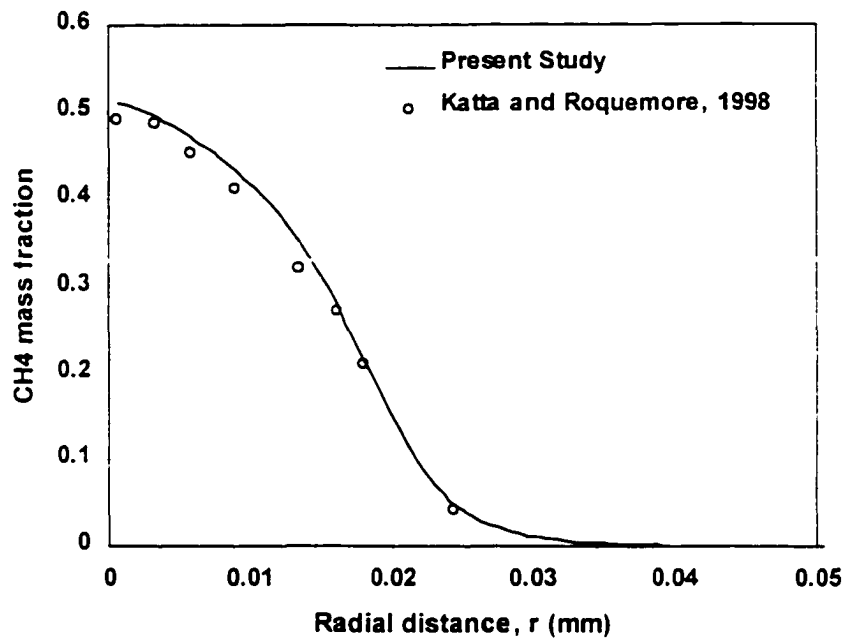


Fig. 4.4 Comparison of Measured and Computed CH₄ Mass Fraction for Code Validation

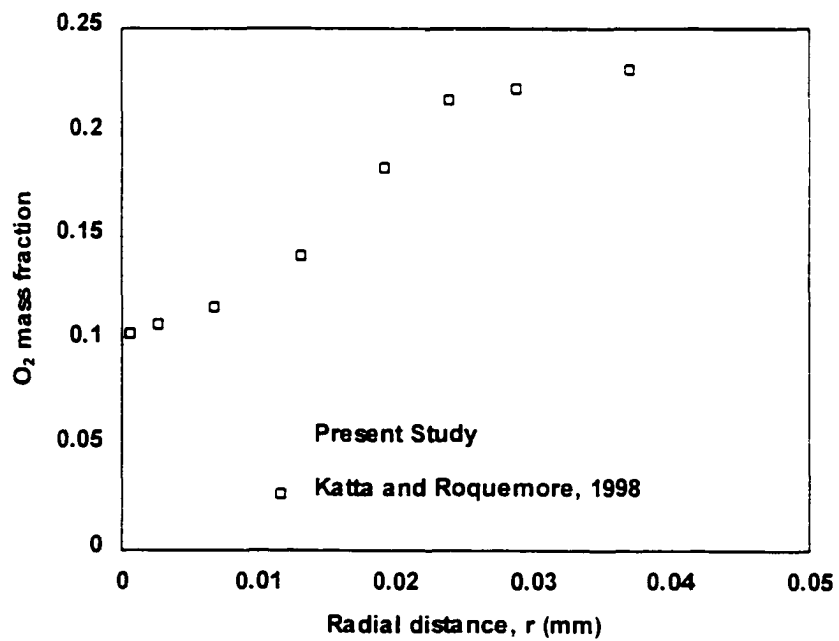


Fig. 4.5 Comparison of Measured and Computed O₂ Mass Fraction for Code Validation

The stabilization mechanism of turbulent jet diffusion flames is a long-time interest in combustion research due to numerous practical applications. In addition, stability of jet diffusion flames is an important fundamental aspect to explore. Previously, several investigators have suggested that the isothermal mixing process of a non-burning fuel jet and an oxidizer determines the stability behavior of their turbulent jet diffusion flames (Broadwell et al., 1984, Cha and Chung, 1996, Pitts, 1989). These studies have also shown that it is possible to develop empirical expressions from known mixing behavior of the flow to determine the lift-off and blowout characteristics of the flame. Although these investigations have included a wide range of fuels and flow conditions, very little information is available regarding the stability mechanism of composite or mixed fuels. Kalghatgi (1981) studied the blowout velocity of gas mixtures such as methane/air, methane/carbon dioxide, propane/air, and propane/carbon dioxide. Broadwell et al. (1984) extended that study to derive an expression for the blowout velocity of gas mixtures. However, none of these studies included the stability mechanism of hydrogen-hydrocarbon mixtures. Also, to the best of the author's knowledge, no study has yet been published to demonstrate the validity of the theoretical and empirical expressions for hydrogen-hydrocarbon hybrid fuels. Furthermore, several investigators identified that, although the prediction of blowout velocity through these models is good for hydrocarbon fuels, its agreement with measurements is poor for pure hydrogen fuel. In this context, further investigations are required to understand the stability mechanism of jet diffusion flames of hydrogen-hydrocarbon mixtures.

In this dissertation, an experimental and numerical study was conducted to investigate the effects of hydrogen addition on the stability of hydrocarbon jet diffusion flames. Flame stability was determined for different contents of hydrogen in natural gas-hydrogen fuel mixture. The result was explained in the context of cold isothermal mixing process of a hydrogen-hydrocarbon hybrid fuel jet issued into quiescent air. The turbulent mixing rate of non-reacting hybrid fuel was solved numerically and the local concentrations of hydrogen and natural gas were calculated for different inlet mixture conditions. The effects of the local concentration on the flame stability mechanism were analyzed.

5.1 THEORETICAL BACKGROUND

Isothermal mixing process of a non-reacting turbulent jet with global density variation has a significant importance for predicting flame stability such as lift-off and blowout. The lift-off and blowout phenomena of jet flames depend on the mixing of the fuel and air in the non-reacting region downstream of the jet exit. The shadowgraphs presented by Savas and Gollahalli, 1986 clearly showed the turbulent mixing region below the base of a lifted flame.

5.1.1 Lift-off and Blowout of Turbulent Jet Diffusion Flames

Usually, the reaction zone of a turbulent jet flame is located at a certain distance from the burner exit where the average concentration of the fuel in its isothermal mixture with surrounding air renders its flame speed maximum (Pitts, 1989). Hence, an estimation of the local concentration of an isothermal fuel jet issued into air yields a good prediction

of the lift-off height. Kalghatgi, 1981 proposed a semi-empirical correlation to calculate the lift-off height based on the maximum laminar flame speed.

$$\frac{\rho_F (S_b)_{\max} L_b}{\mu_F} = 50 \left(\frac{U_b}{(S_L)_{\max}} \right) \left(\frac{\rho_F}{\rho_A} \right)^{1.5} \quad [5.1]$$

This expression predicts lift-off height for hydrocarbon fuels with considerable accuracy. however, the previous studies showed that it did not work well for hydrogen (Pitts, 1989).

Several theories exist regarding the blowout of turbulent diffusion flames. Some of these theories emphasize that the imbalance between the local flow velocity and turbulent flame speed is the primary cause of flame blowout. When the local flow velocity increases to a critical value, combustion can not be sustained and flame extinction occurs. According to these concepts flame blowout is due simply a flow equilibrium processes. Other investigators have explained flame blowout behavior based on the mixing of large-scale turbulent structures. These models are based primarily on the competition between the turbulent mixing time and combustion/ignition time in large-scale turbulent structures. Kalghatgi, 1981 proposed the following correlation to estimate blowout velocity of jet flames:

$$\frac{U_b}{(S_b)_{\max}} \left(\frac{\rho_F}{\rho_A} \right)^{1.5} = 0.017 Re_H \left(1 - 3.5 \times 10^{-6} Re_H \right) \quad [5.2]$$

The Reynolds number Re_H is given by

$$Re_H = \frac{\rho_F (S_b)_{\max} H}{\mu_F} \quad [5.3]$$

The characteristic length, H , is the distance along the burner axis where the mean fuel concentration falls to its stoichiometric value and can be estimated by

$$H = 8 \left[\frac{Y}{Y_s} \left(\frac{\rho_F}{\rho_A} \right)^{0.5} - 5.8 \right] r_i \quad [5.4]$$

The predictions showed a qualitative agreement with measurements for most hydrocarbon fuel mixtures (Cheng and Chiou, 1998), however yielded a poor agreement for hydrogen-air mixtures.

5.1.2 Prediction of Lift-off and Blowout of Composite Fuel Turbulent Flames

The analytical and semiempirical models for lift-off and blowout of turbulent diffusion flames presented earlier can predict the stability behavior of most hydrocarbon fuels with considerable accuracy. Several studies showed that, experimental results agreed well with the predicted value (Broadwell et al., 1984; Pitts, 1989; Cha and Chung, 1996). On the otherhand, due to some crucial assumptions, in case of hydrogen flame, the model predictions do not match well with the experimental findings. Furthermore, the validity of these models has never been investigated for hydrogen-hydrocarbon mixtures. To use these models for hydrogen-hydrocarbon flames several parameters such as *maximum laminar flame speed* (S_{Lmax}), *stoichiometric mass fraction of fuel* (f_s), and *equivalence ratio* of composite fuels need to be properly defined and estimated. In

the next few sections. the approach used in the present investigation to define and estimate these parameters are discussed

5.1.3 Laminar and Turbulent Flame Speed of Composite Fuels

As mentioned earlier, the values of maximum laminar flame speeds of mixed fuels are required to use the previously discussed models for predicting the lift-off and blowout velocities of composite fuels. However, experimental or theoretical studies in literature to predict the maximum laminar flame speed of a mixture of fuels are severely limited. Although several studies have delineated the effects of inert diluents on flame speed, those results are inapplicable to the present study since both components of the mixture are reactive. A study with moist carbon monoxide has shown that the flame speed is raised considerably by an addition of a very small amount of hydrogen or hydrogen-containing fuels (Pitts, 1989). Then, the question is how do we estimate the maximum laminar speed of hydrogen-hydrocarbon composite fuels. One way might be to assume a linear dependence of the flame speed with the relative concentration of each constituent of the mixture. Yu et al., 1986 have shown that their measured values of laminar flame speed vary quasi-linearly with a specially defined parameter, R_H to indicate the contribution of hydrogen in the composite fuel. However, R_H varies nonlinearly with fuel mole fraction, and thus the laminar flame speed has a nonlinear relation with hydrogen content in the mixture. Furthermore, two crucial assumptions, i.e., complete oxidation of hydrogen and non-existence of competitive reactions, made in deriving R_H make this concept less attractive for use in diffusionally-controlled flames. In this context, a diffusion theory of flame speed might be a good approximation for a mixture.

The theory proposed by Tanford and Pease, 1947 assumed that the rate of diffusion of active radicals into the unburned gas determines the magnitude of the flame velocity. Their results also indicated that the equilibrium concentration of hydrogen atoms is an important factor in determining the flame velocity. Later, Simon, 1951, modified the theory in which the concentrations of hydroxyl radicals and oxygen atom were taken into account and derived the expression

$$S_b = \sqrt{\frac{rC_r}{X_P} k' \left(\frac{D_{H^*} p_{H^*}}{B'_{H^*}} + \frac{D_{OH^*} p_{OH^*}}{B'_{OH^*}} + \frac{D_{O^*} p_{O^*}}{B'_{O^*}} \right)} \quad [5.5]$$

Simon found that for most of the hydrocarbon fuels the value of k is $1.4 \times 10^{11} \text{ cm}^3/\text{mole sec}$. Later Dugger et al., 1955 combined *Tanford-Pease theory* and *Zel'dovich-Frank-Kamenetsky-Semenov theory* together and formed a unique correlation of maximum laminar flame speed (S_{Lmax}) and a combination of partial pressure, $6.5p_{H^*}+p_{OH^*}+p_{O^*}$. The maximum laminar flame speed of fuel blends (methane and hydrogen) calculated from Dugger correlation and extracted from the experiments of Yu et al., 1986 are shown in Fig. 5.1. The chemical kinetics computational code CHEMKIN (Kee et al., 1993) was used to compute the equilibrium compositions of H, O, and OH for different hydrogen-natural gas mixtures. Although many different theoretical and empirical formulas have been developed to predict turbulent flame speed (S_T), there appears to be a general agreement that turbulent flame velocity only depends on r.m.s value of axial fluctuations and laminar flame speed. In the present investigation the following expression which relates the turbulent flame velocity with its laminar counterpart and axial velocity

fluctuations in the wrinkled flame region is used to calculate the flame velocity (Turns, 1996).

$$S_T = 3.57S_L \left(\frac{u'_{rms}}{S_L} \right)^{0.7} \quad [5.6]$$

5.1.4 Equivalent Fuel Formula and Stoichiometric Fuel Mass Fraction

An equivalent representation of hydrogen-hydrocarbon composite fuels in terms of fuel formula C_xH_y is required to calculate the stoichiometric fuel mass fraction ($Y_{F,s}$) from stoichiometrically corrected chemical reaction. This leads to the concept of deriving an equivalent fuel formula presented in Choudhuri and Gollahalli, 2000a. For different hydrogen contents in hydrogen-natural gas mixture, the value of x and y and corresponding Y_s were calculated and tabulated in Table 5.1. The calculation procedures are shown in Appendix V.

Table 5.1 Equivalent Fuel Formulas

%NG-H2%	x	y	Y_s
100-0	1	4	0.0548
95-5	0.99	3.98	0.0547
90-10	0.986	3.972	0.0547
80-20	0.969	3.938	0.0546
65-35	0.937	3.938	0.0544

5.1.5 Equivalence Ratio for Composite Fuel

For meaningful data reduction, an effective equivalence ratio for composite fuels need to be developed. If the mole fractions of the hydrocarbon fuel (methane in the present study), hydrogen, and air are, respectively C_{H_2} , C_{CH_4} , and C_A , then in order to oxidize C_{H_2} amount of hydrogen totally, $C_{H_2}/(C_{H_2}/C_A)_s$ of air will be needed. If the remaining air

is used to oxidize the hydrocarbon fuel, an effective equivalence ratio (EFI) can be defined as follows and a sample calculation is included in Appendix V.

$$EFI = \frac{\frac{C_{CH_4}}{C_A} - \frac{C_{H_2}}{(C_{H_2} + C_A)_s}}{\left(\frac{C_{CH_4}}{C_A} \right)_s} \quad [5.7a]$$

Two other equivalence ratios based on pure methane (CFI) and pure hydrogen (HFI) are also defined by considering only methane or hydrogen in the mixtures. The CFI and EFI values are calculated using standard equivalence ratio formula (Turns, 1996).

$$CFI = \frac{\frac{C_{CH_4}}{C_A}}{\left(\frac{C_{CH_4}}{C_A} \right)_s} \quad [5.7b]$$

$$HFI = \frac{\frac{C_{H_2}}{C_A}}{\left(\frac{C_{H_2}}{C_A} \right)_s} \quad [5.7c]$$

5.1.6 Significance of Isothermal Mixing on Lift-off and Blowout Velocities

Several investigators have shown that the lift-off and blowout behavior of turbulent diffusion flames could be predicted through the isothermal mixing process of cold fuel jet and air [5. 9]. Turbulent jet flames blow out at velocities at which the flame base is located well downstream and where the centerline concentration reaches a value (Y_{max}) at which the flame propagation velocity is equal to $(S_b)_{max}$ (Broadwell et al., 1984, Pitts,

1989, Kalghatgi, 1981). In his paper, Pitts, 1989 found that blowout occurred at a downstream location where the flame base reached its maximum radius. In fact, when the local velocities of the base of a lifted turbulent jet flame exceeded a certain critical value, blowout occurred. Therefore, by computing the local velocities of an isothermal cold mixing jet it is possible to predict its flame blowout limit.

5.2 RESULTS AND DISCUSSIONS

5.2.1 Flow Structure

The near-field schlieren images of lifted NG-H₂ flames before blowout at different mixture conditions are shown in Fig. 5.2 (a). The corresponding color schlieren pictures are also shown in Fig 5.2 (b). The flames are issued from a 3.8 mm ID burner and fuel jet exit velocity is set to a certain value which is just before blowout condition of a specific NG-H₂ mixture (100% NG: 37 m/s, 90-10% NG-H₂: 52 m/s, 80-10%: 55 m/s, and 65-35% 78 m/s) . The lifted portions of the flames are turbulent and resemble the appearance of cold (isothermal) turbulent jets. As discussed earlier, mixing of fuel and oxidizer in this region plays an important role to determine the stability of the flame. The formation of turbulent structures is evident in all mixture conditions. However, the lift-off height at blowout condition decreases as hydrogen content increases in the mixture. This is because of the wide flammability limit of hydrogen fuel. The presence of even a small amount of hydrogen initiates combustion and subsequently supplies enough heat and chain carrier radicals or atoms (especially H atoms) to sustain a natural gas flame. Hence, the effects of hydrogen addition in natural gas flame stability are two-fold: first, the burning of hydrogen prevents flame blow-out, second the increase of

chain carrier radical concentrations enhances the burning rate of natural gas. Consequently, the combined effects of these two phenomena improve the stability of natural gas flames. Then, the question arises how flame stability correlates with hydrogen addition in the mixture, and if there is any correlation, how does it change with burner size. This will be discussed in later sections along with the technique of using existing theoretical models to predict the lift-off height and blowout velocity of a hydrogen-natural gas composite fuel. The corresponding color schlieren pictures are shown in Fig 5.2 (b). The color pictures also reveal the turbulent mixing of the lifted part of the flames.

Acetone PLIF images which describe the mixing of fuel and air in cold jets are shown in Fig 5.3. These two non-burning jets of 100% NG and 65-35% NG-H₂ mixture were issued from a 3.8 mm burner at a constant jet exit velocity of 55 m/s. The corresponding jet exit Reynolds number are 12425 and 10203 respectively. The acetone vapor was mixed with the incoming natural gas fuel and the fluorescence image of the acetone serves as the tracer of fuel air mixing. The images are false color composites and different slices are taken at different times. Each slice is an average of hundred images at that location. The red color shows highest concentration of acetone or otherwise higher mixing of fuel and air while green and blue correspond to lower mixing of fuel and air. The black color represents minimum mixing i.e. either pure fuel or pure air. The near-field large coherent structures are prominent in the 100% NG fuel jet and it can be seen that the fuel mostly follows the vortex pattern. As the jet grows in the mid and far-downstream the mixing is intense and smaller structures start forming. The formation of roll up eddies, their coalescence, the development of secondary instabilities, and their

fine-scale disintegration are quite similar to those observed by Savas and Gollahalli, 1986. Hence, for pure natural gas, the fuel air mixing is primarily governed by vortex pattern and shear layer dynamics. However, as 35% hydrogen was added in the hydrogen-natural gas composite fuel mixture the flow field changes drastically. The breakup of the large eddies occurs earlier and the development of secondary instabilities is quicker in the 65-35% NG-H₂ mixture jet. This might be caused by two possible reasons. First, The effect of density; second the effect of multicomponent diffusion of gas mixtures. With the increase of hydrogen concentration in the fuel the air to fuel density ratio (ρ_A/ρ_F) increases from 1.8 to 2.6. The increases density gradient causes change in large structure and thereby enhances mixing process. Similar observation has been made by Brown and Roshko (1974) in turbulent mixing layer. In a study they demonstrated that the large structures of turbulent mixing layers are changed when the density gradient between two layers increases. They have also shown that the spreading angle of the mixing layer, which represents the mixing rate, increases as the density difference increases. However, in the present case the effect is higher than what expected. This can be attributed towards the increase of relative diffusion velocity due to presence of three gases, which is sometimes comparable with the turbulent mixing process (Warntz et al., 1996). Primarily, the presence of hydrogen, which diffuses seven times faster than other hydrocarbon fuels and has eight times lower density than methane, plays the crucial role of changing the mixing pattern of the flow-field. The multi-component diffusion mechanism of gas mixtures is still an active area of research, and more theoretical and experimental development is required to understand the mixing mechanism of multiple gas mixtures. However, from the present investigation one

conclusion can be drawn, i.e., the addition of hydrogen greatly enhances the mixing of air and fuel.

5.2.2 Blowout Velocity

The variation of blowout velocity as a function of hydrogen content in the mixture for different burner diameters is shown in Fig. 5.4. As mentioned earlier, blowout velocity increases with the increase of hydrogen content in the mixture. For the 1 mm ID burner, the blowout velocity of the 100% natural gas is 9.6 m/s, which increases to 12.8 m/s with the addition of 5% hydrogen to the mixture. The increase in blowout velocity is slow initially for the 1 mm burner; however, the rate increases as the hydrogen concentration is past above 20% of the fuel mixture. The effect of hydrogen addition on flame stability is also pronounced for burners with higher diameters. The blowout velocity of the 100% NG with the 2.3 mm burner is 23.9 m/s, which increases to 30.3 m/s as 5% H_2 is added to the fuel mixture. In general, an addition of 5% H_2 to the fuel mixture causes a 26-30% rise in flame blowout velocity. However, the rate of increase is higher for higher concentrations of hydrogen in the mixture. The rate of increase of flame blowout velocity is 38-40% with the increase of additional 5% H_2 in the 70-30% NG- H_2 composite fuel. Clearly, these trends indicate the dominance of hydrogen in the stability mechanism of hydrogen-hydrocarbon composite fuel. Theoretical predictions of blowout velocities based on Eq. 5.2-4 are also shown in Fig. 5.4 with solid and dashed lines. The maximum laminar flame velocities are calculated using Eq. 5.5 and Dugger correlation. The equilibrium compositions are calculated using the chemical kinetics equilibrium kinetic code CHEMKIN (Kee et al., 1993). It can be seen that the experimental measurements agree fairly well with the theoretical predictions. Within

experimental uncertainties, the predicted values are slightly higher for fuel mixtures with 35% H₂ concentration. However, it was observed that the accuracy of theoretical predictions largely depends on the proper estimation of maximum laminar flame speed. In the absence of adequate experimental data, the diffusion theory proposed by Tanford and Pease, 1947 seems to be a reasonable choice for computing maximum laminar flame speed, since it actually considers the effects of H, O, and OH radical concentrations for estimating flame speed which are important in hydrogen-hydrocarbon composite fuel combustion. The variation of normalized blowout velocity, $\frac{U_b}{S_{L,max}} \left(\frac{\rho_F}{\rho_A} \right)^{1.5}$, with specially defined Reynolds number, $R_H = \frac{HS_{L,max}\rho_F}{\mu_F}$, is shown in Fig. 5.5. The normalized blowout velocity data from 1mm, 2.3 mm, 3.8mm, and 4.5 mm burners collapse on to a single line, and are uniquely correlated with the parameter R_H . However, it is to be noted that this correlation is only possible when the maximum laminar flame speed, stoichiometric mass fraction, and equivalence ratio of the composite fuels are properly estimated.

5.2.3 Lift-off Height at Blowout Conditions

Fig. 5.6 shows flame lift-off height as a function of hydrogen content in the hydrogen-natural gas mixture for different burner sizes. The measured lift-off height for 100% NG agrees well with the data in the literature for all burner sizes. For the 2.3 mm ID burner, the lift-off height is 6.0 cm, which is within $\pm 4\%$ of those measured by Pitts, 1989. Also, it agrees well with the theoretical prediction (6.1 cm) using Eq. 5.1. As shown in conjunction with flow-field structures, flame lift-off height at blowout condition

decreases with the increase of hydrogen content in the mixture. For 1mm burner the maximum lift-off height at blowout condition is 2.5 cm, which decreases to 2.4 cm with a 5% increase in hydrogen content in the fuel mixture. The mixture flame speed increases with the increase of hydrogen content in the mixture, which eventually reduces the flame lift-off height by moving the flame base towards the burner. Also, as discussed earlier, due to wide flammability limits, even the presence of a small amount of hydrogen keeps the flame close to the burner exit by directly and indirectly altering the flame characteristics. The addition of 5% H₂ to 100% NG causes on an average 3-5% decrease in lift-off height for all burner conditions. However, at higher concentrations of hydrogen in the fuel mixture the rate of decrease is around 9-13%. The variation of the normalized lift-off height, $L_H = \frac{\rho_F S_{L,max} L_b}{\mu_F}$, as a function of normalized blowout

velocity, $L_n = \frac{U_b}{S_{L,max}} \left(\frac{\rho_F}{\rho_A} \right)^{1.5}$, is shown in Fig. 5.7. It can be seen that it is possible to correlate the blowout velocity and lift-off height at blowout condition with a unique relation. However, once again the validity of this correlation is related to the composite fuel parameters mentioned earlier.

5.2.4 Isothermal Cold Jet Mixing

Isothermal mixing of a non-reacting turbulent fuel jets issuing from a 3.8 mm ID tube for 100-0%, 90-10%, 80-10%, 65-35% and 0-100% NG-H₂ gas mixture, with 55 m/s jet exit velocities is numerically investigated. This condition is well above the blowout velocity of the 100% NG flame, slightly higher than the blowout velocity of 90-10%

NG-H₂ flame and below the blowout velocity of 80-10%, 65-35% and 0-100% NG-H₂ flames. Hence, setting the exit jet velocity to 55 m/s results in the following conditions:

Table 5.2 Flame Conditions Corresponding to 55 m/s Jet Exit Velocity

%NG-%H ₂	Flame Conditions	Re _j
100-0	No Flame (Well above blowout velocity)	12425
90-10	No Flame (Just after blowout)	11883
80-20	Lifted Flame	11299
65-35	Attached Flame	10203

Therefore, the computations of non-reacting at these configurations will reveal information about how the local flow-field variables affect the stability mechanism of jet flames.

5.2.4.1 Local Equivalence Ratio

The local equivalence ratio is calculated using the computed local air and fuel concentrations. As mentioned earlier, three equivalence ratios are defined to demonstrate the effect of equivalence ratio on the flame stability mechanism. The CFI is defined considering only methane in the fuel mixture whereas the HFI considers only hydrogen in the fuel mixture (Eq. 5.7 b, c). The equivalent equivalence ratio EFI is calculated based on Eq. 5.7a. The two-dimensional contours of the three equivalence ratios for different mixtures of hydrogen and natural gas are shown in Figs. 5.8-10. The contour plots represent a two-dimensional area, which extends to 20d_j in the axial direction and 4d_j in the radial direction. For all fuel mixtures CFI has its maximum value on the centerline, which decays in the outwards radial direction. The contour plots of CFI show that with the increase of hydrogen concentration in the mixture the CFI decreases in the near-field region. At an axial location of 15d_j the CFI values are between 1.2-1.4 (Fig.

5.8). As mentioned earlier, the 100% NG flame does not exist in this location for 55 m/s jet exit velocity. The CFI value decreases in this location as hydrogen concentration increases in the mixture. For the 90-10% NG-H₂ flame, the CFI values in this location range between 0.5-1.0. However, the HFI value increases because of the addition of hydrogen to the mixture. The HFI values (Fig. 5.9) lie between 1.8-2 for the 90-10% NG-H₂ flame. The corresponding EFI values (Fig. 5.10) are 0.2-0.5; and as mentioned earlier, flame also does not exist in this location for the 90-10% NG-H₂ mixture. However, unlike the 100% NG flame, the condition is just after the blowout of 90-10% NG-H₂ flame. As more hydrogen is added to the mixture, the CFI value decreases, and both HFI and EFI increase. For the 80-20% NG-H₂ mixture, a stable lifted flame can be obtained. The corresponding CFI, HFI and EFI values for this condition are 1.2, 3.5 and 0.75 respectively. As seen HFI=3.5 keeps the flame anchored at this location despite a low CFI. The corresponding EFI=0.75 indicates the condition of natural gas-hydrogen mixture at which the mixed fuel flame exists. A further increase in hydrogen concentration (65-35% NG-H₂) results stable attached flame. The CFI, HFI and EFI values at the same axial location are 1.1, 3.9 and 1.0 respectively. Clearly, the EFI indicates the condition where a stable attached flame is possible for the mixed fuel. The axial profiles of CFI, HFI and EFI for different mixture conditions are shown in Figs. 5.11-13. As discussed in relation with the contour plots, the value of CFI decreases outside the potential core as hydrogen concentration increases in the mixture. Throughout the potential core of the cold fuel jet, the fuel concentration remains almost constant and then starts decaying at a distance $5d_j$ from the jet exit. Inside the potential core, the CFI values for all mixture conditions are approximately constant, however, CFI

decreases outside the potential core, as hydrogen concentration increases in the mixture. Similar trends are observed for HFI concentrations, except HFI increases with the increase of hydrogen concentration in the fuel mixture. However, the EFI shows a clear distinction of hydrogen addition to the fuel mixture. The EFI increases as hydrogen concentration increases in the mixture. The centerline value of EFI decreases after the potential core and for the 90-10% NG-H₂, EFI diminishes after 10d_j. The radial profiles of CFI, HFI and EFI at axial locations of 8d_j are shown in Figs. 5.14 –16. As expected, CFI, HFI and EFI have maximum value on the centerline and decrease radially outwards. However, it is interesting to note that at this location although centerline EFI has almost similar values for all mixture conditions, the EFI values away from the centerline also serve as the determinant factor for stability mechanism.

5.2.4.2 Local Velocity

The centerline profiles of the normalized axial velocity for different natural gas-hydrogen mixtures are shown in Fig. 5.17. It can be seen that the potential core of the fuel jet is extended up to six-burner diameters. Right after the potential core the axial velocity of the fuel jet, which has a higher hydrogen concentration decays faster than those with a lower hydrogen concentration. The radial profiles of axial, and radial velocity at an axial location of 8d_j for different fuel mixture are shown in Figs. 5.18-19. The axial velocity is maximum on the centerline, which decreases with radial distance. Close to the central axis the fuel mixture with a lower hydrogen concentration has a higher axial velocity (Fig. 5.18). However, away from the burner axis, the axial velocity of fuel mixture with higher hydrogen concentration decays faster than in the mixtures of

lower hydrogen concentration. The radial profiles of the normalized radial velocity for different fuel mixtures are shown Fig. 5.19. The centerline radial velocity is almost zero, which increases with the increase of radial distance. At far outwards location the negative radial velocity indicates the inward infusion of the air inside the fuel jet. Initially, fuel mixture with higher hydrogen concentration has a higher radial velocity. However, the scenario reverses at more outward radial locations. This is an interesting observation, which indicates that the mechanism of flame stabilization over the radius of the jet (Pitts, 1989). At a higher hydrogen concentration, although the centerline velocity is higher for the fuel mixture with higher hydrogen concentration, the lower velocity at larger radii prevents the local extinction of the flame.

5.2.4.3 Turbulent Fluctuation

The axial profiles of axial turbulent fluctuation for different fuel mixtures are shown in Fig. 5.20. Inside the potential core, the axial fluctuation remains constant and does not change appreciably with the mixture composition. The fuel mixture with a higher hydrogen concentration has a higher turbulent fluctuation outside the potential core. Since, most theories predict that the turbulent flame velocity increases with the increase of axial fluctuation, the increased turbulent fluctuation directly contributes towards the enhanced flame stability of fuel mixture with higher hydrogen concentration. The radial profiles of the axial fluctuation for different fuel mixtures at 8d_j axial location are shown in Fig. 5.21. The amount of fluctuation is minimum close to the centerline, which increases and reaches a maximum value and then gradually decays. Away from the central axis, mixture with a higher hydrogen concentration has a higher turbulent

fluctuation than those with a lower hydrogen concentration, which certainly indicates the increase of turbulent flame velocity in this location.

5.2.4.4 Turbulent Flame Speed

The axial and radial profiles of the calculated turbulent flame velocity using Eq.5.6 for different natural gas-hydrogen mixture are shown in Figs. 5.22 and 5.23. As mentioned earlier, turbulent fluctuation is directly correlated with axial fluctuation and the maximum laminar flame velocity, and increases with the increase of axial fluctuation. The turbulent flame velocity increases with the increase of hydrogen concentration in the fuel mixture since both laminar flame speed and axial fluctuation increases with the increase of hydrogen concentration. Also, it can be seen that turbulent flame speeds of the mixtures with higher hydrogen content decays slowly than the local axial velocity which prevents the flame to become extinct in that location.

5.2.4.5 Turbulent Kinetic Energy

The axial and radial profiles of turbulent kinetic energy for different fuel mixtures are shown in Figs. 5.24 and 5.25. Along the fuel jet axis, turbulent kinetic energy increases after the potential core. Also, mixtures with higher hydrogen content have higher turbulent kinetic energy, which explains the influence of turbulent kinetic energy of cold jet over the flame stability mechanism. The radial profiles of turbulent kinetic energy show the increase of turbulent energy increases and reaches its maximum value at a location away from the centerline, which decreases with a further increase in radial distance.

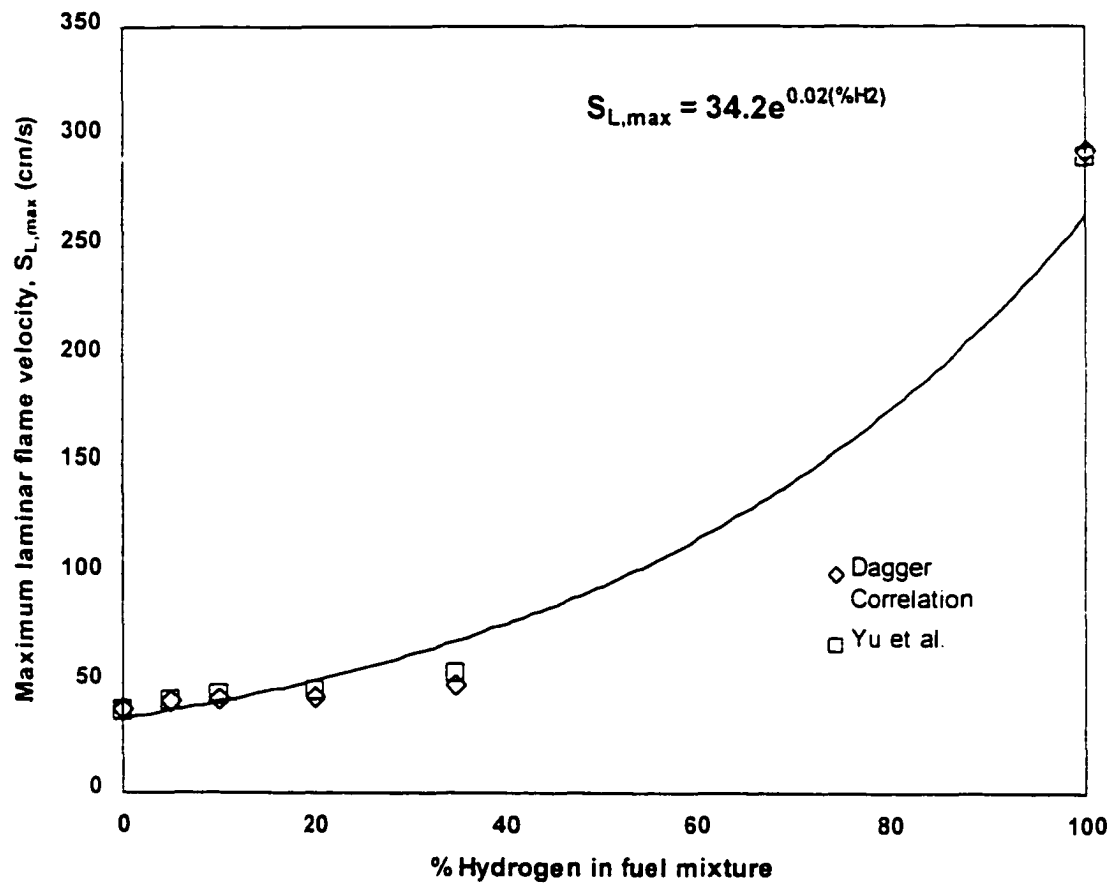


Fig. 5.1 Variation of Maximum Laminar Flame Speed ($S_{L,max}$) with %Hydrogen in the Fuel Mixture

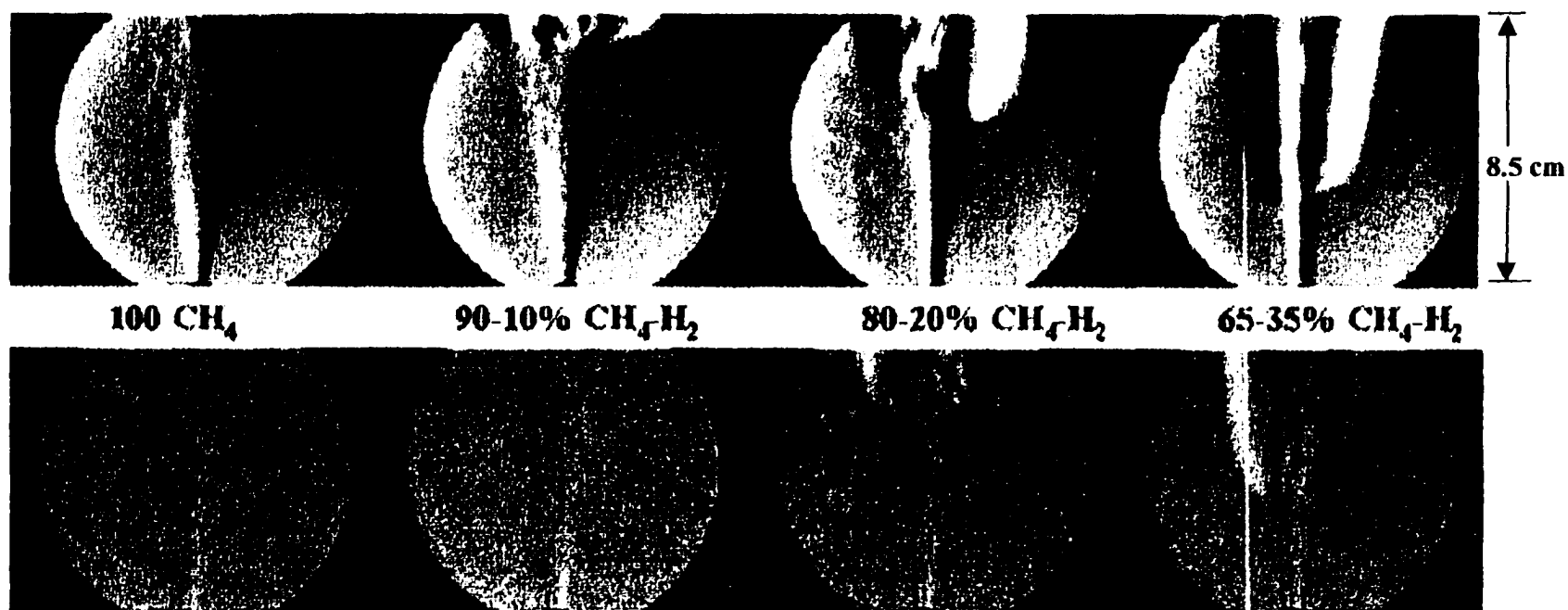


Fig. 5.2 Near-Burner Schlieren Images (a) Black white (b) Color

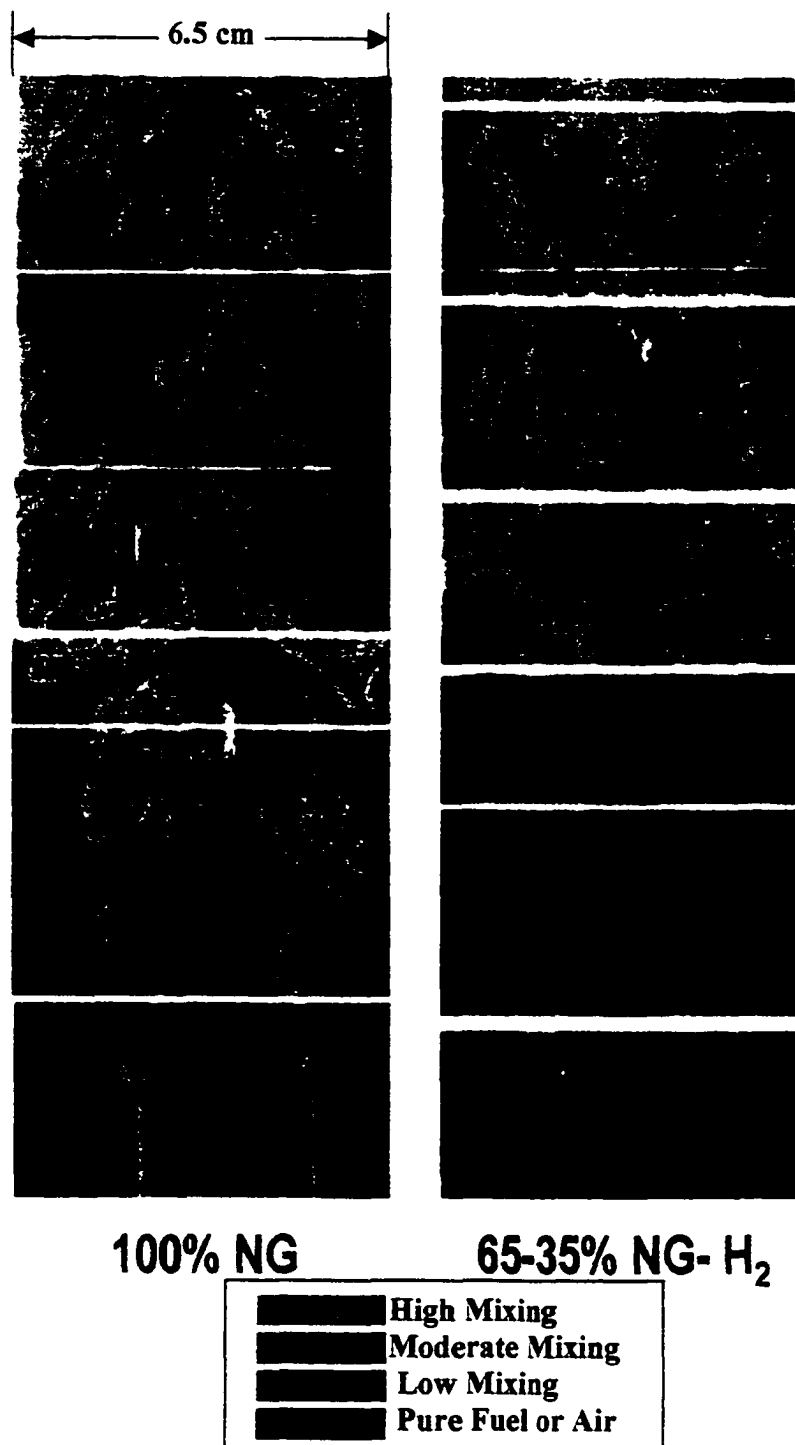


Fig. 5.3 Composite Acetone PLIF Images of NG-H₂ Flames

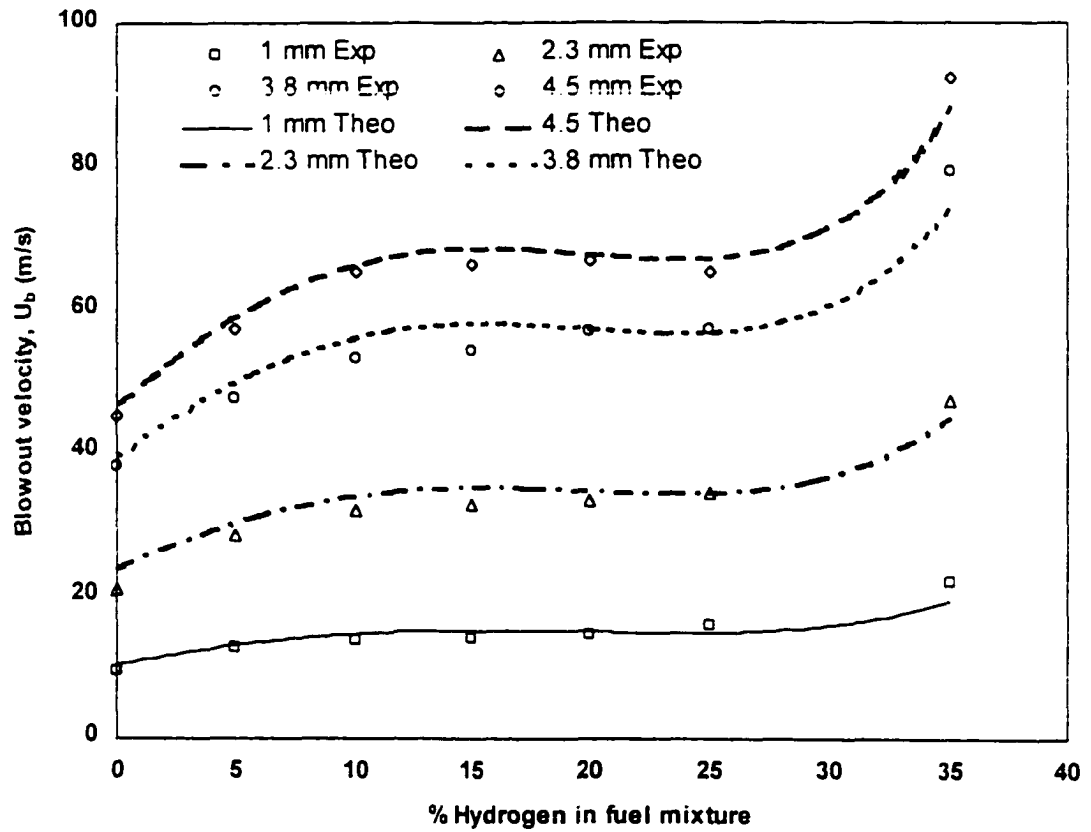


Fig. 5.4 Variation Blowout Velocity with %Hydrogen in the Fuel Mixture

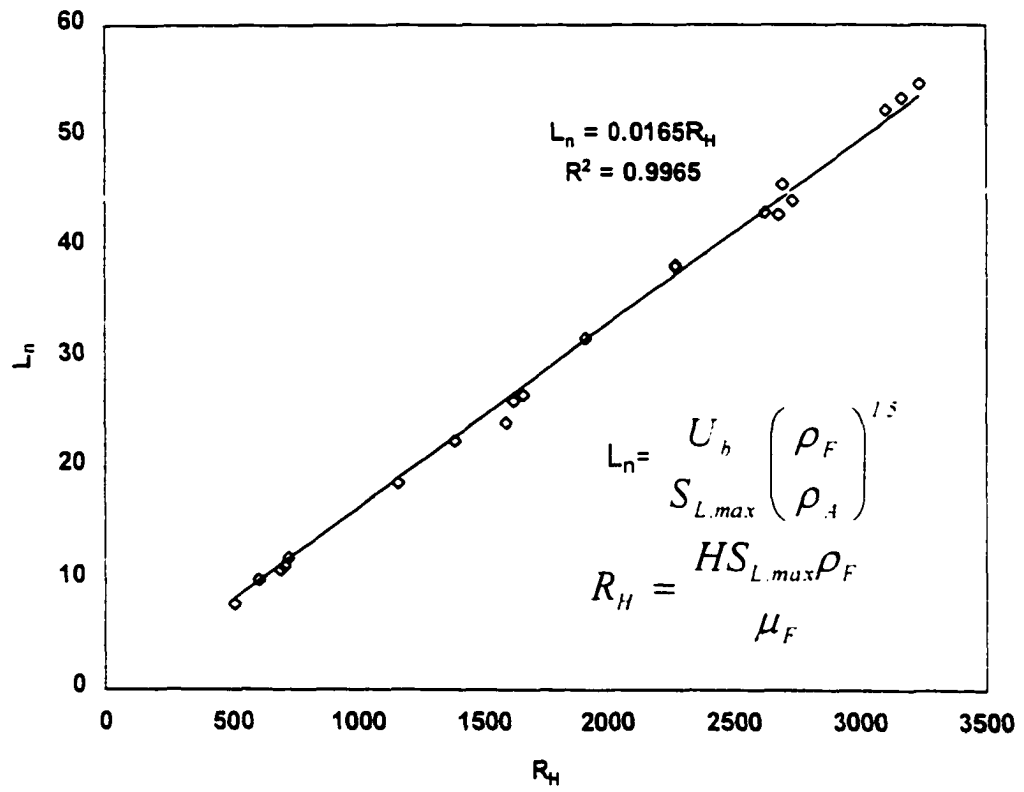


Fig. 5.5 Variation Normalized Blowout Velocity with R_H

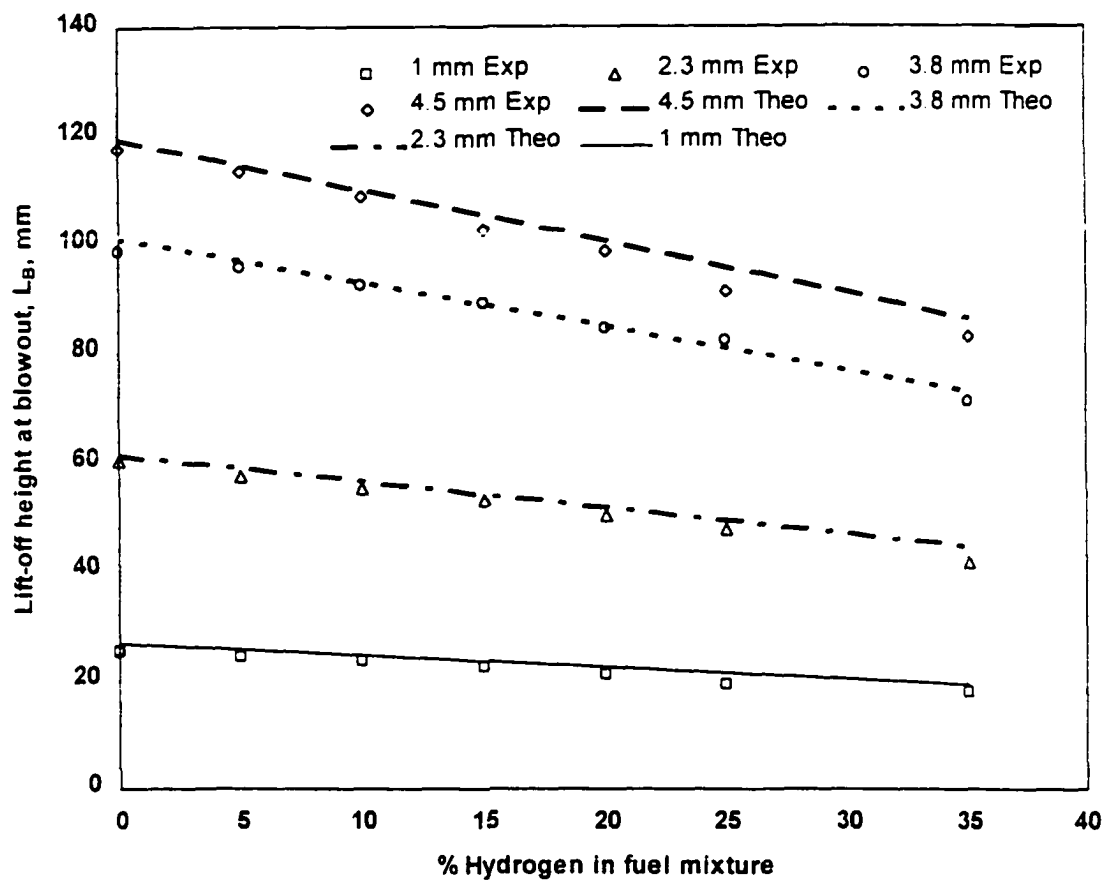


Fig. 5.6 Variation Lift-off Velocity with %Hydrogen in the Fuel Mixture

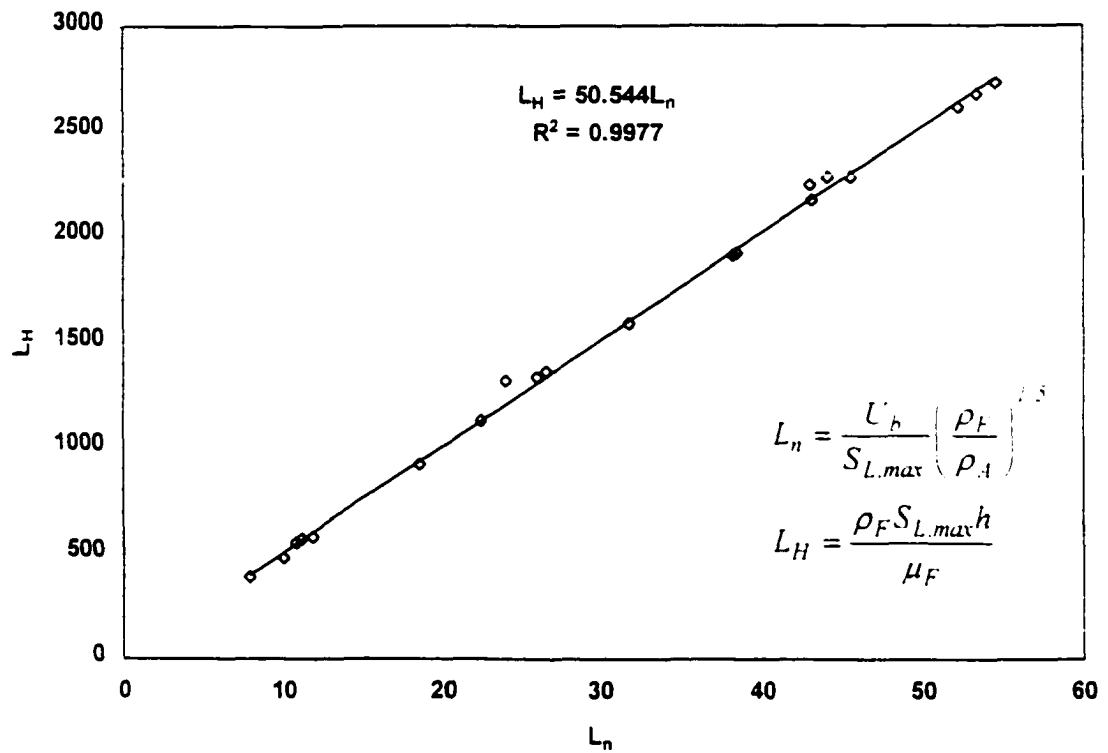


Fig. 5.7 Variation Normalized Lift-off Height with Normalized Blowout Velocity

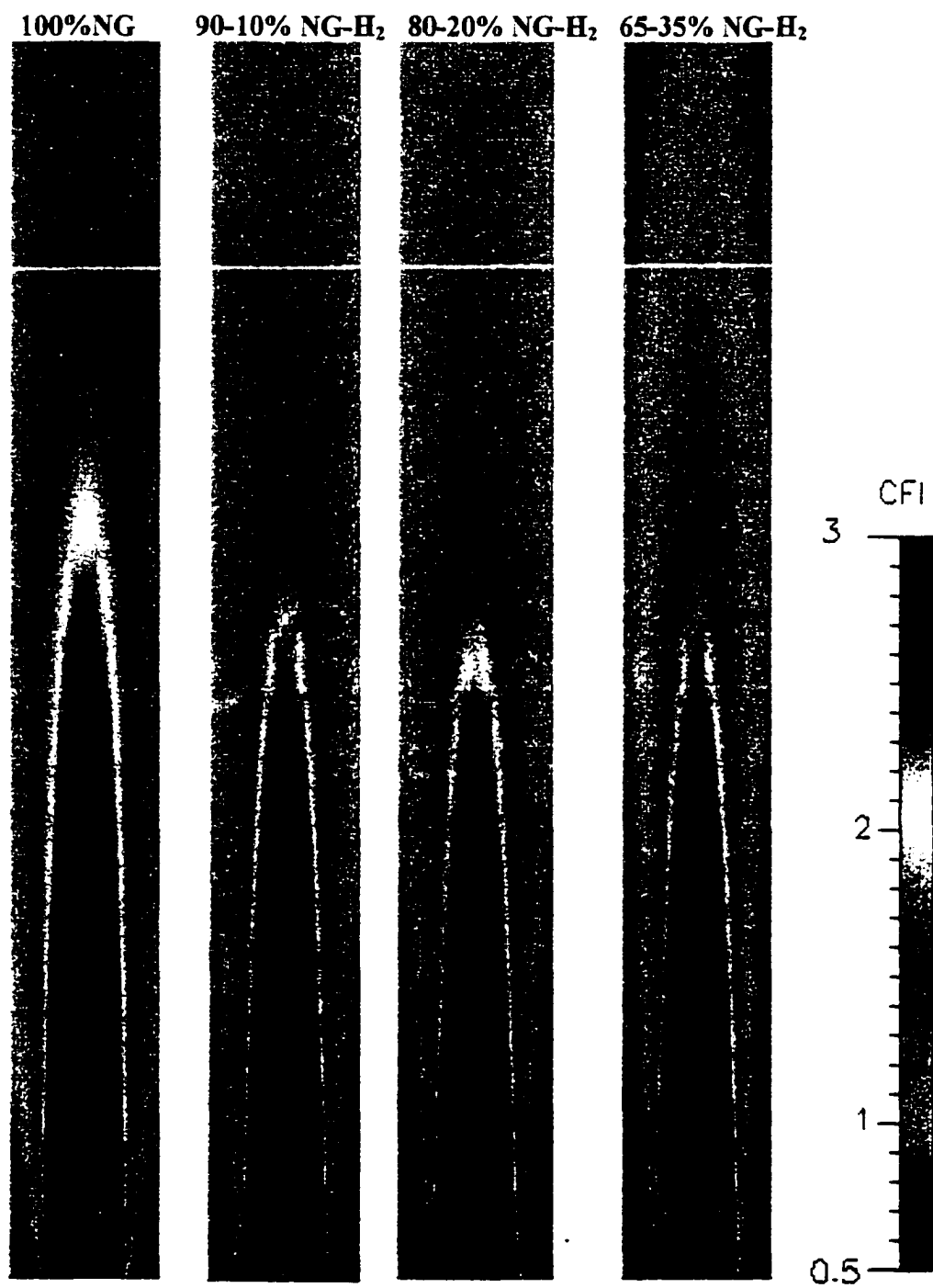


Fig. 5.8 Computed CFI Contour of Cold Fuel Jets at Various Mixture Conditions

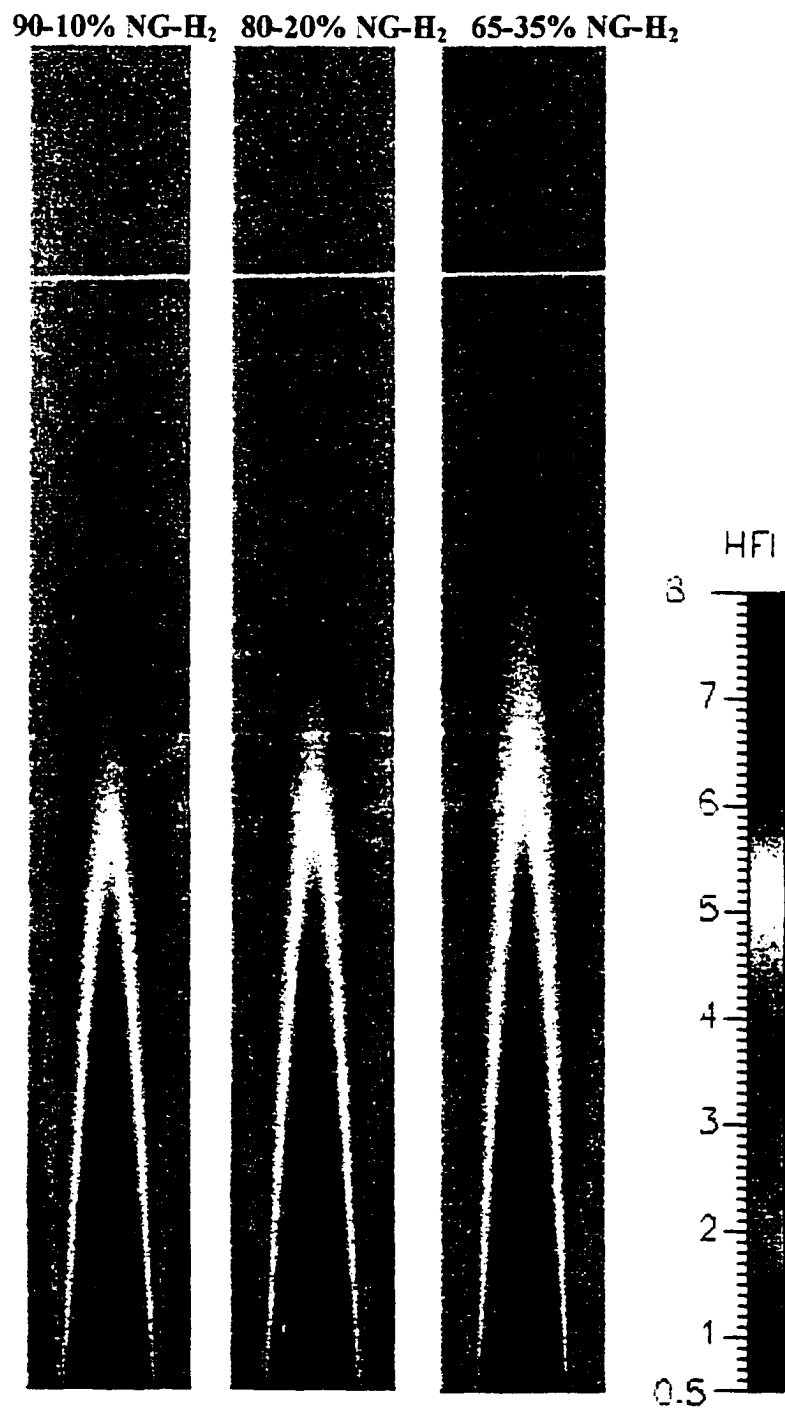


Fig. 5.9 Computed HFI Contour of Cold Fuel Jets at Various Mixture Conditions

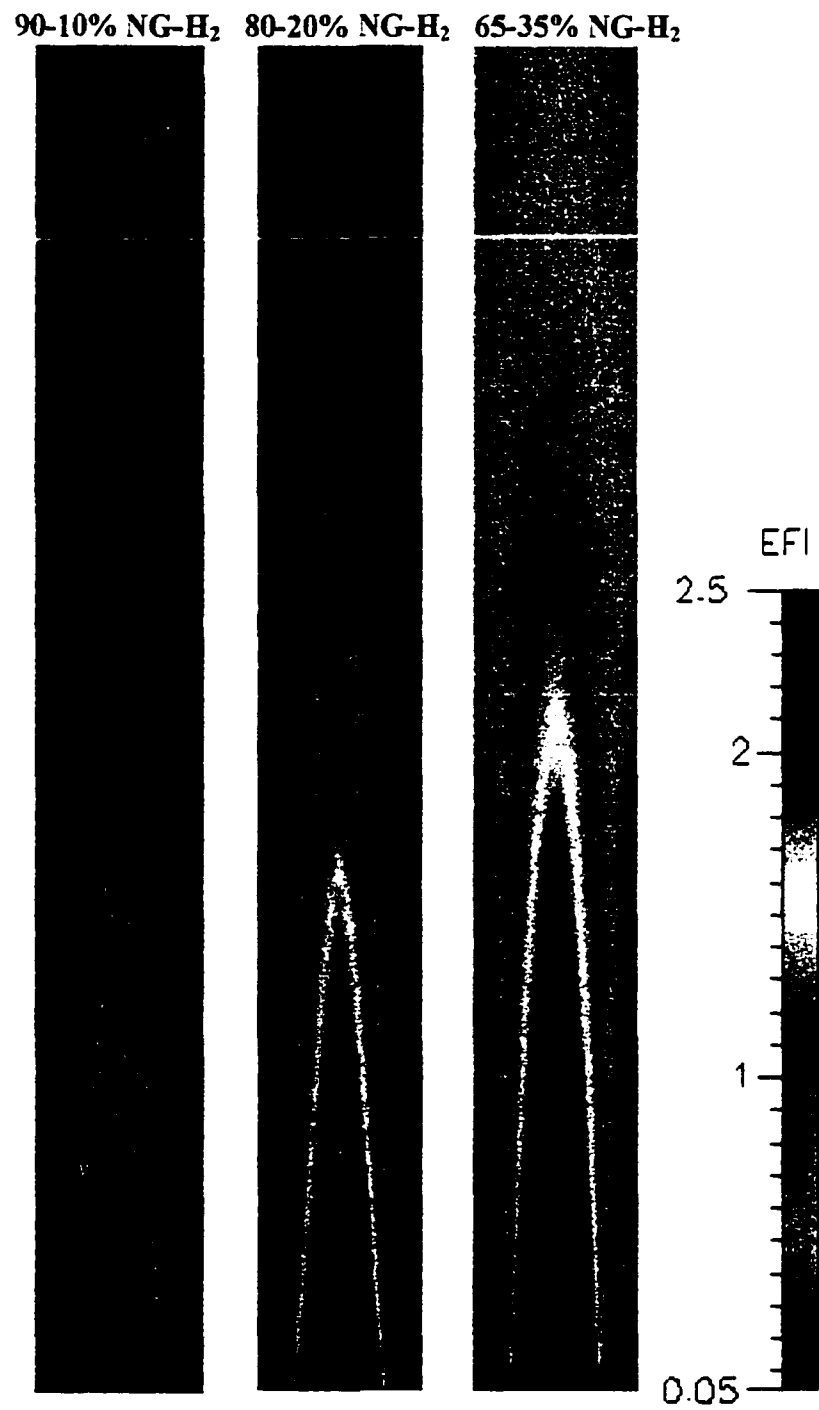


Fig. 5.10 Computed EFI Contour of Cold Fuel Jets at Various Mixture Conditions

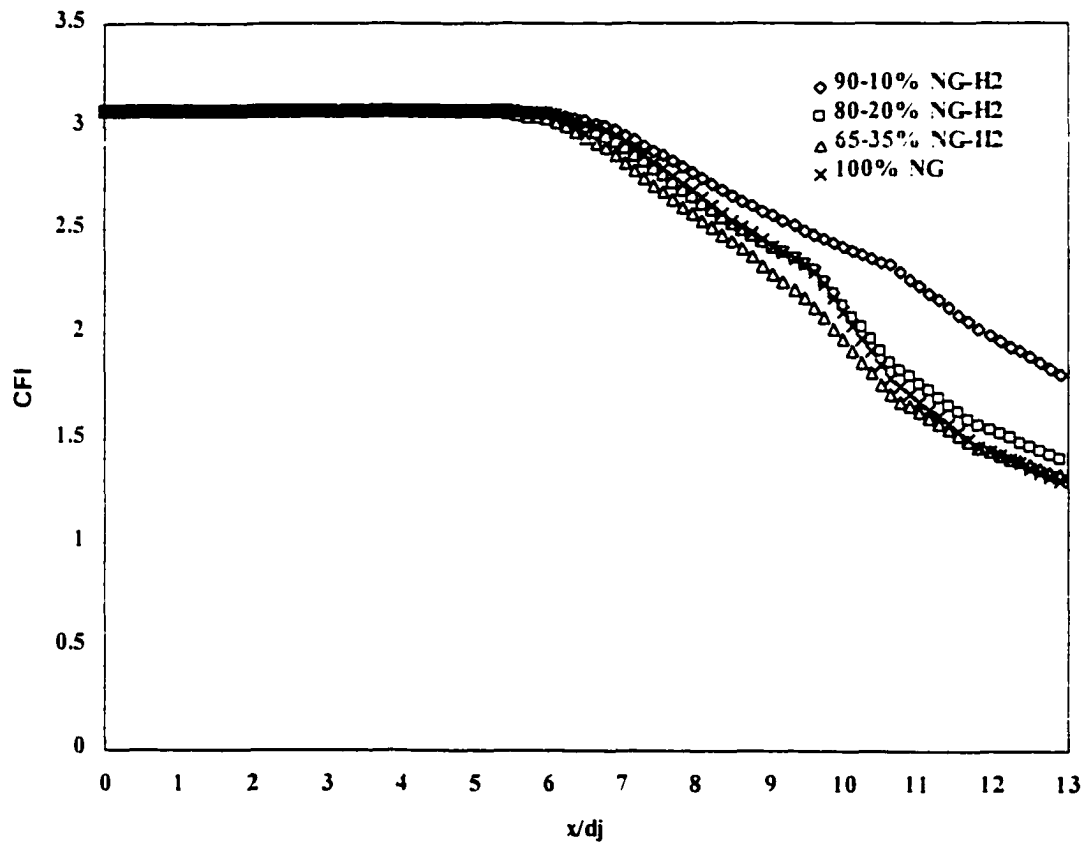


Fig. 5.11 Computed Axial EFI Profiles of Cold Fuel Jets at Various Mixture Conditions

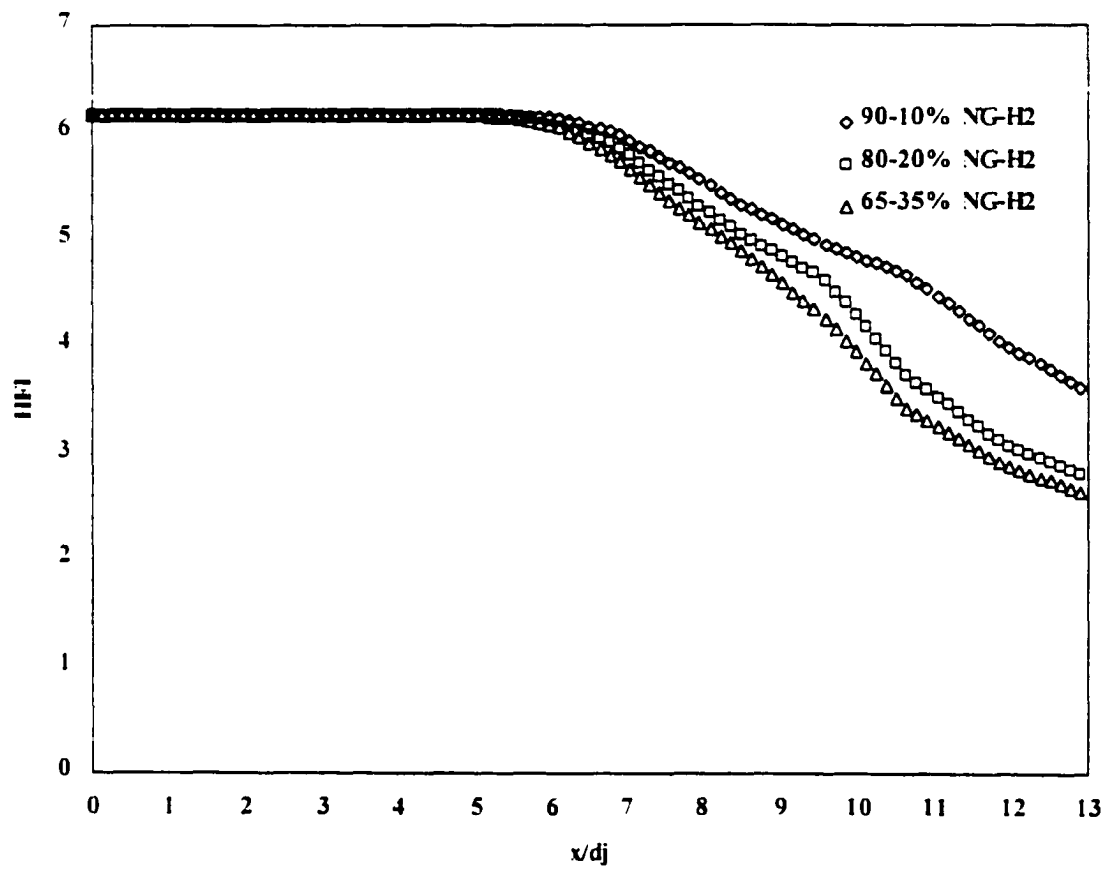


Fig. 5.12 Computed Axial HFI Profiles of Cold Fuel Jets at Various Mixture Conditions

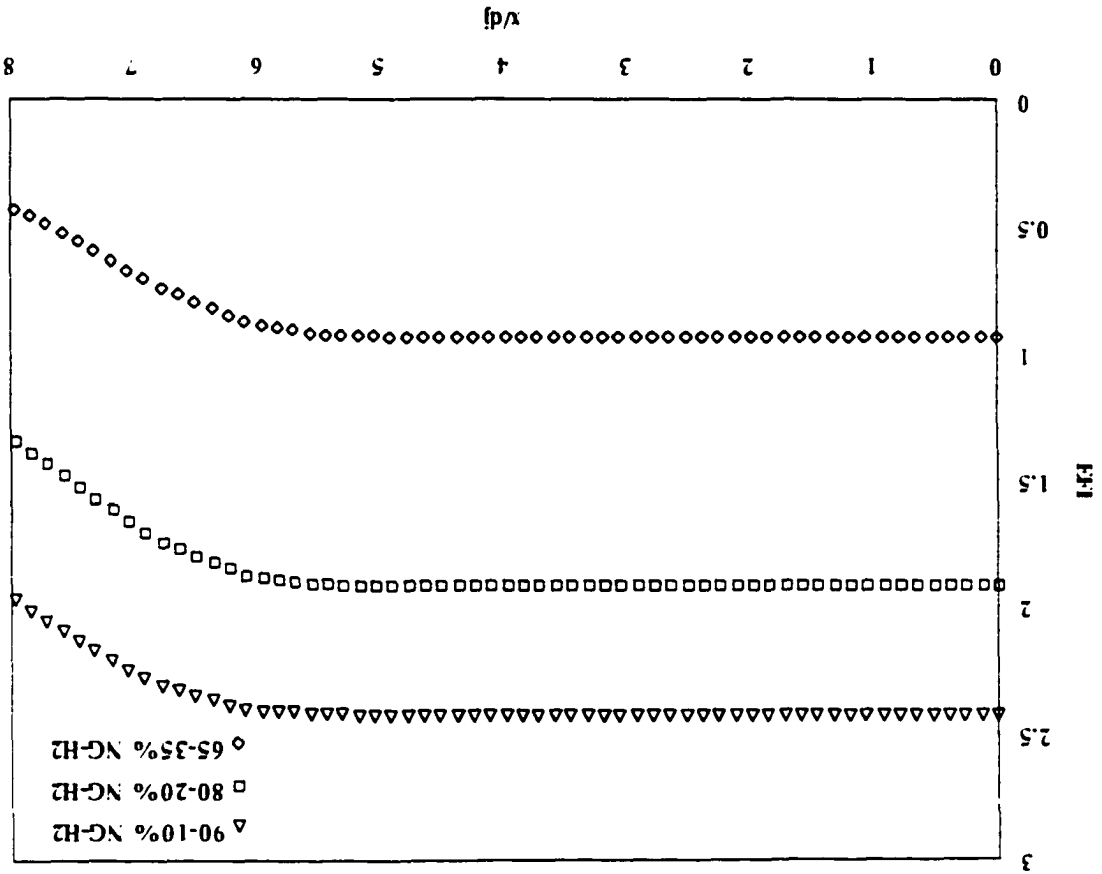


Fig. 5.13 Computed Axial EFI Profiles of Cold Fuel Jets at Various Mixture Conditions

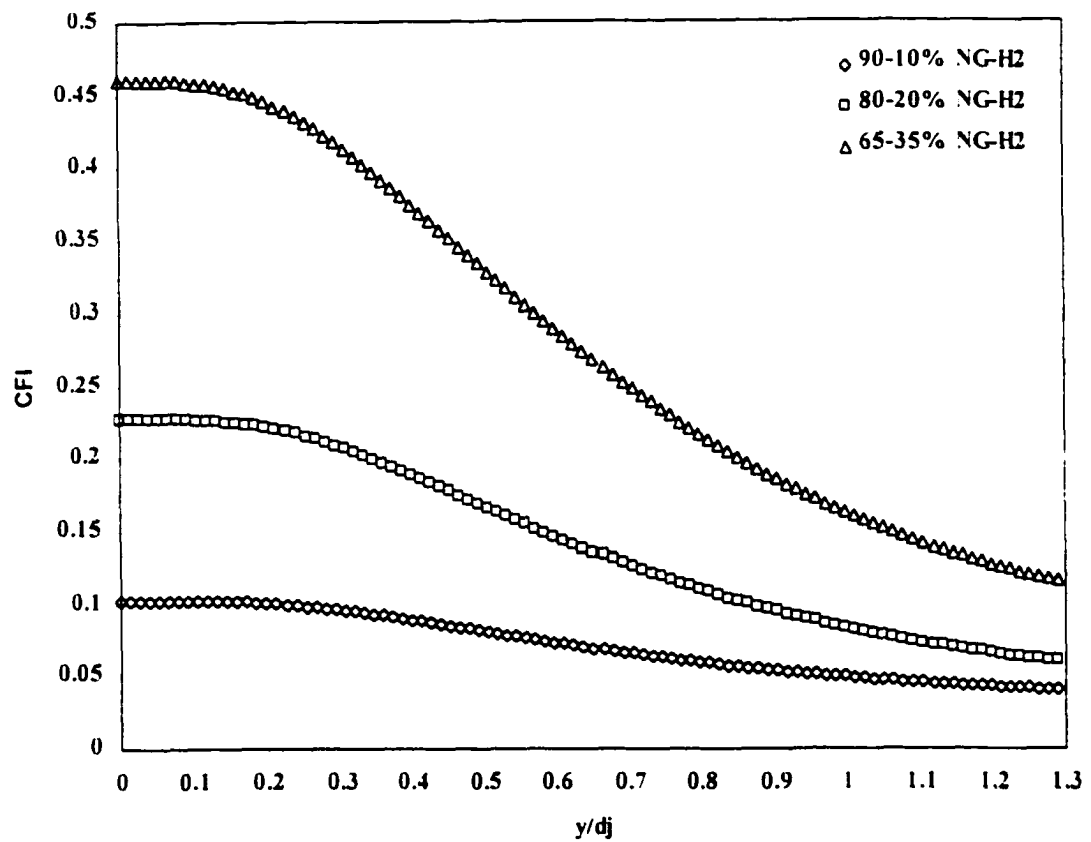


Fig. 5.14 Computed Radial CFI Profiles of Cold Fuel Jets at Various Mixture Conditions

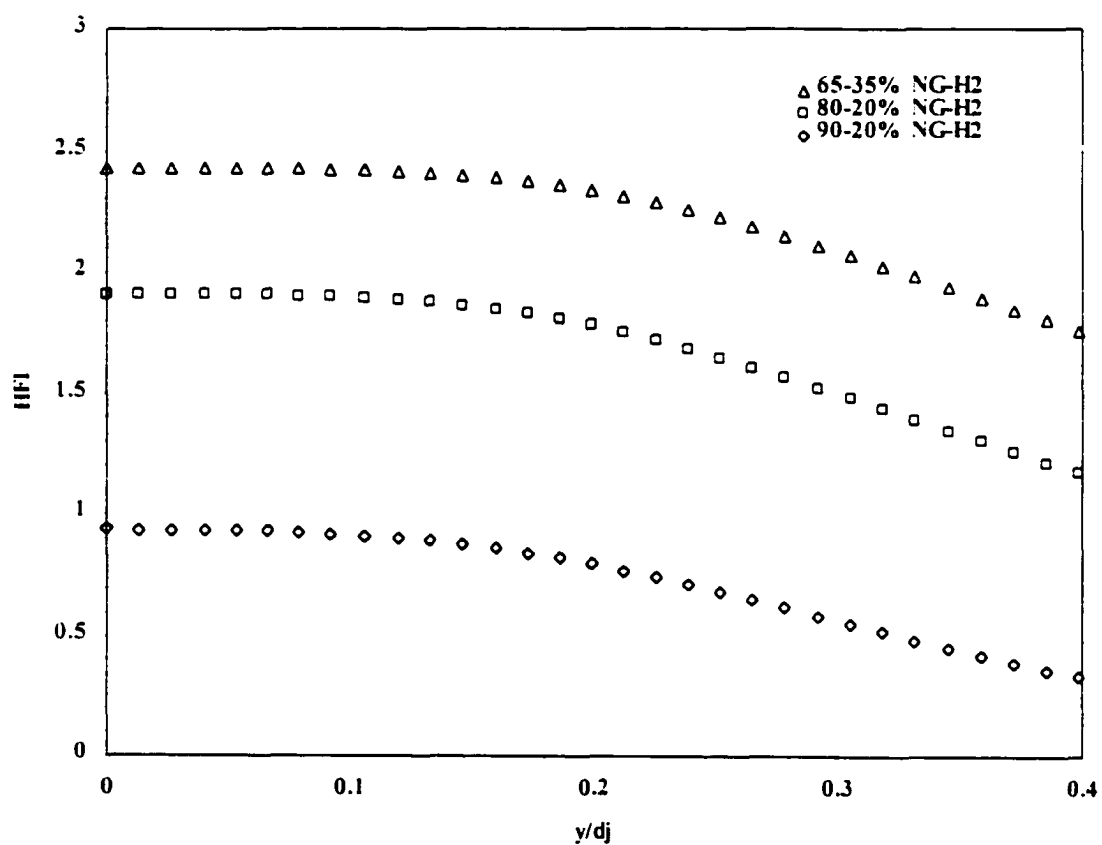


Fig. 5.15 Computed Radial HFI Profiles of Cold Fuel Jets at Various Mixture Conditions

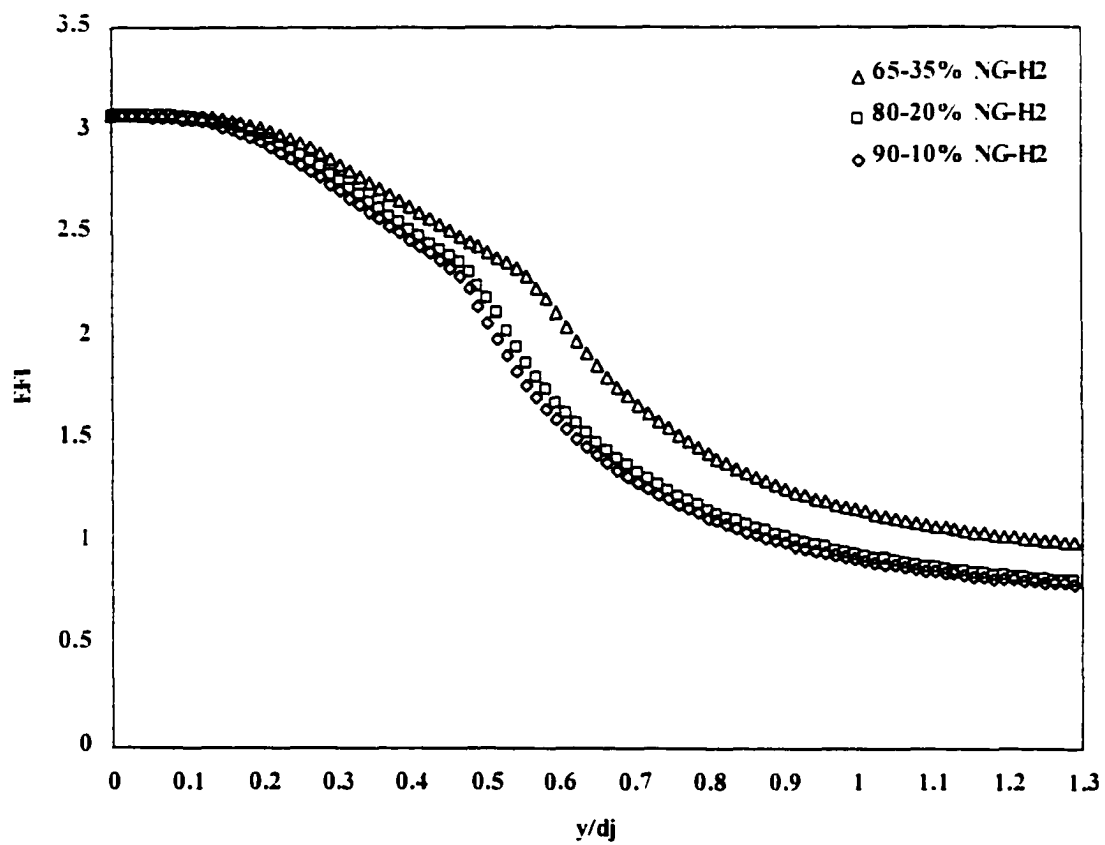


Fig. 5.16 Computed Radial EFI Profiles of Cold Fuel Jets at Various Mixture Conditions

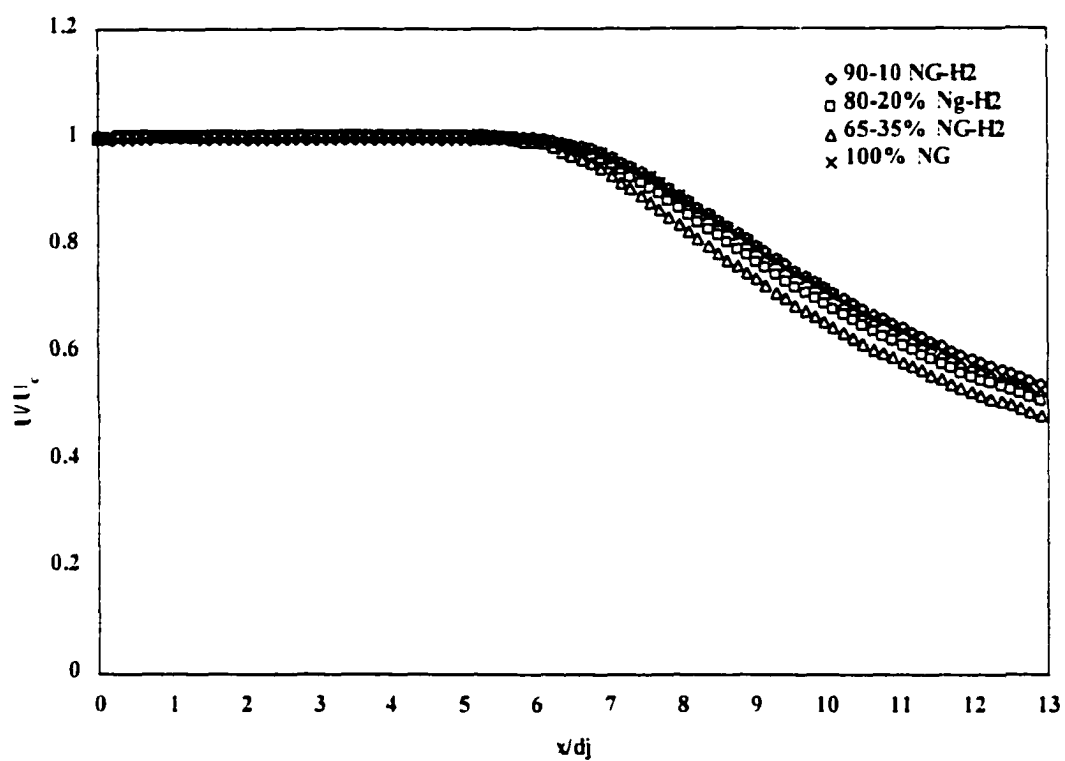


Fig. 5.17 Computed Axial Profiles of the Normalized Axial Velocity of Cold Fuel Jets at Various Mixture Conditions

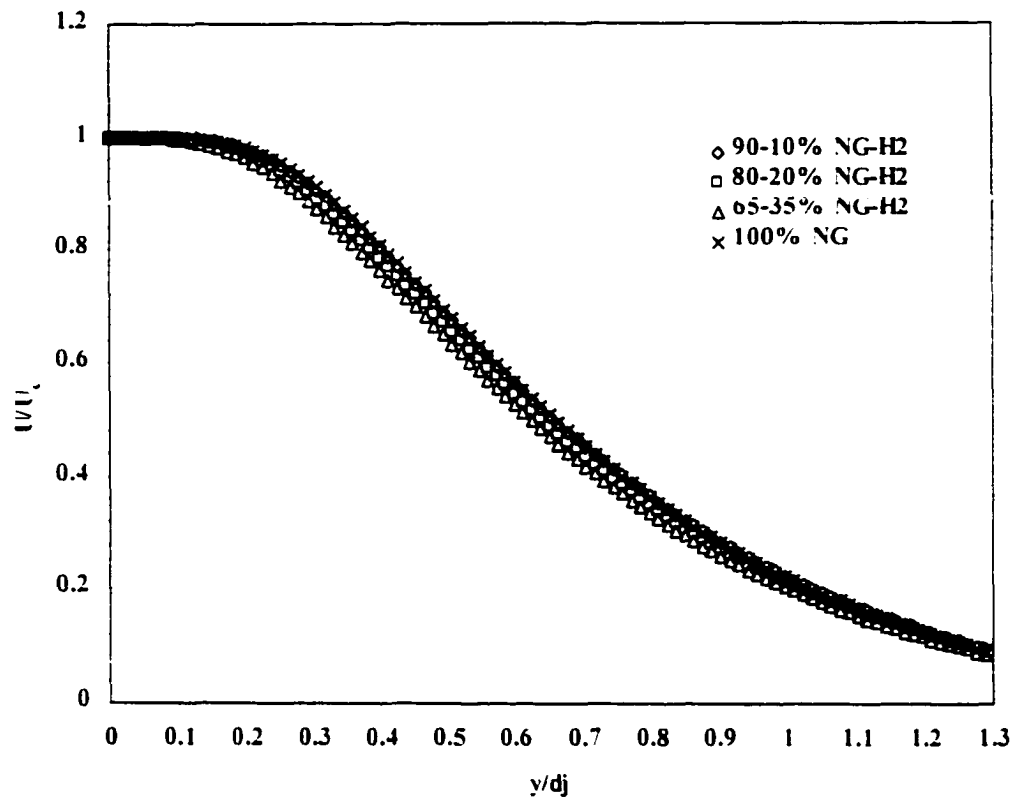


Fig. 5.18 Computed Radial Profiles of the Normalized Axial Velocity of Cold Fuel Jets at Various Mixture Conditions

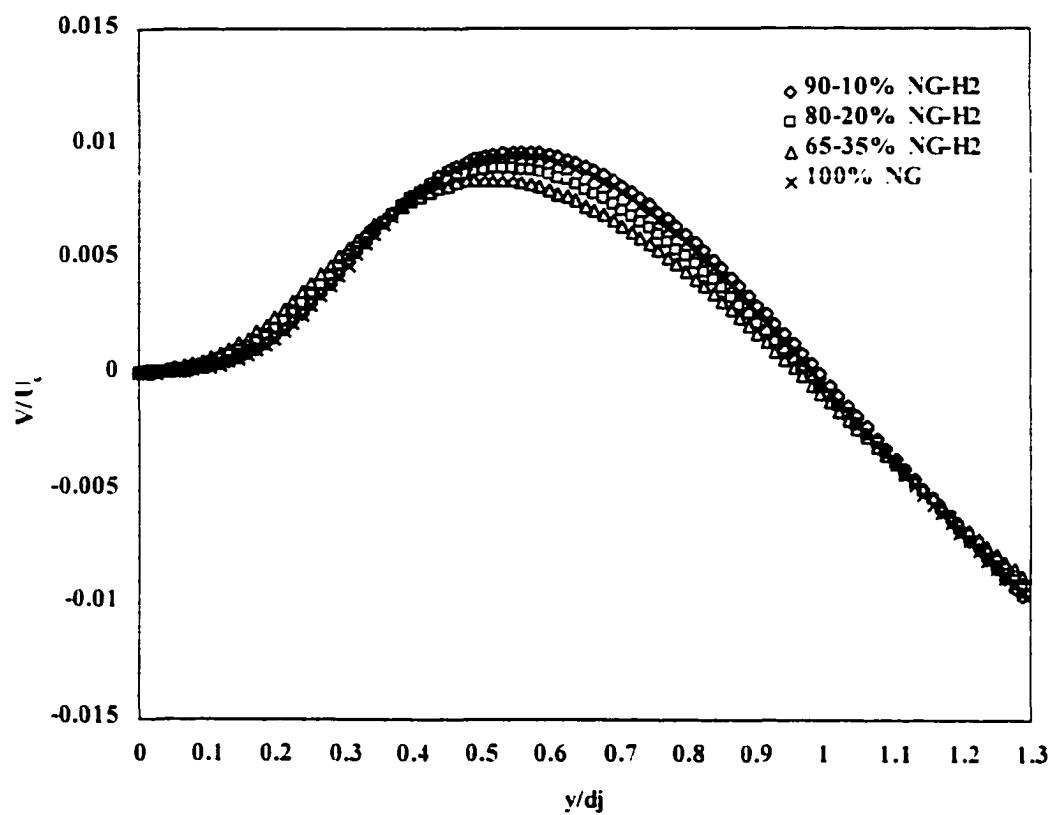


Fig. 5.19 Computed Radial Profiles of the Normalized Radial Velocity of Cold Fuel Jets at Various Mixture Conditions

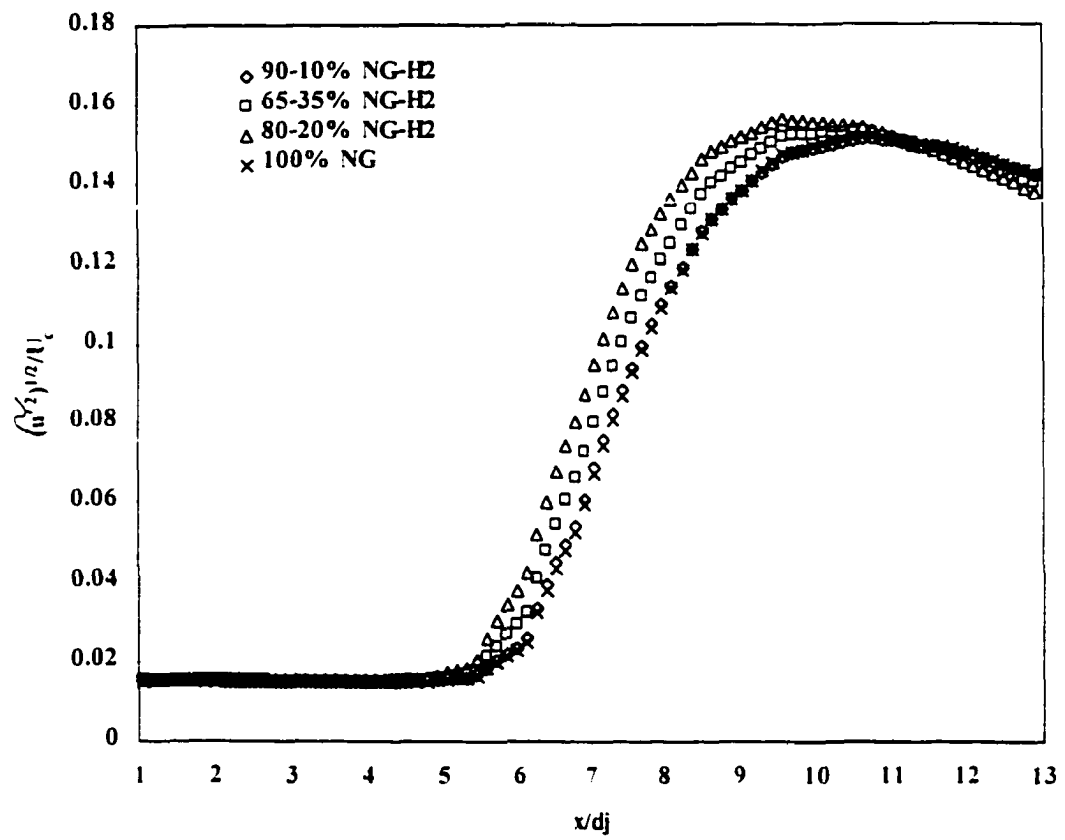


Fig. 5.20 Computed Axial Profiles of the Normalized Axial Fluctuation of Cold Fuel Jets at Various Mixture Conditions

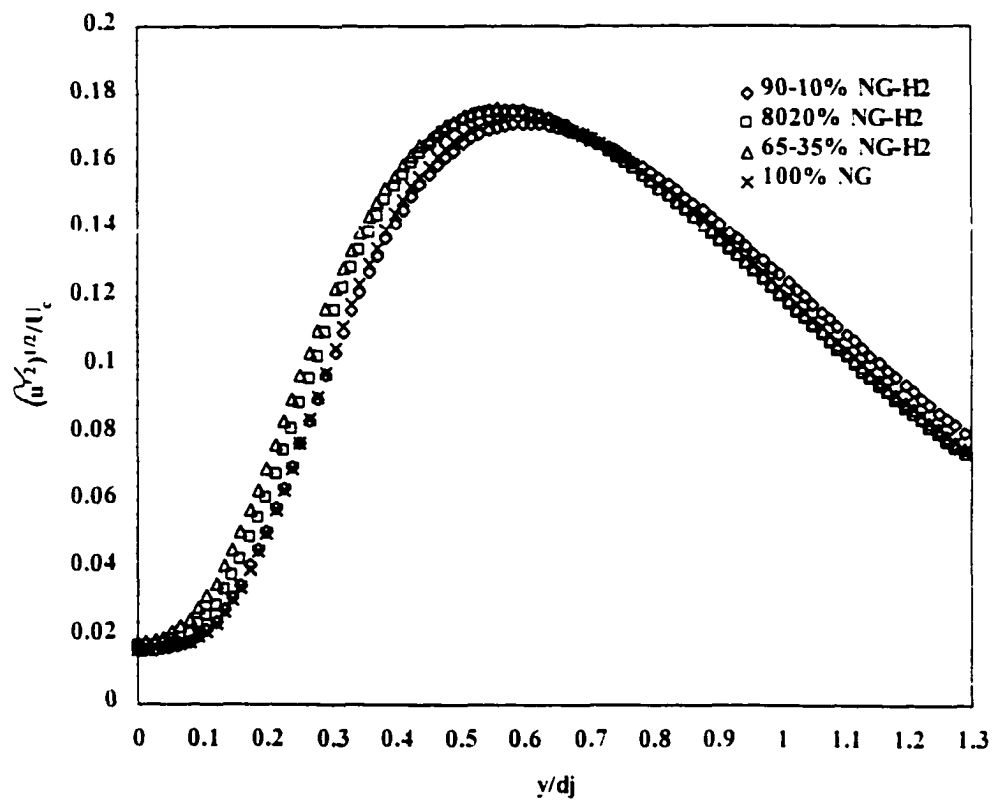
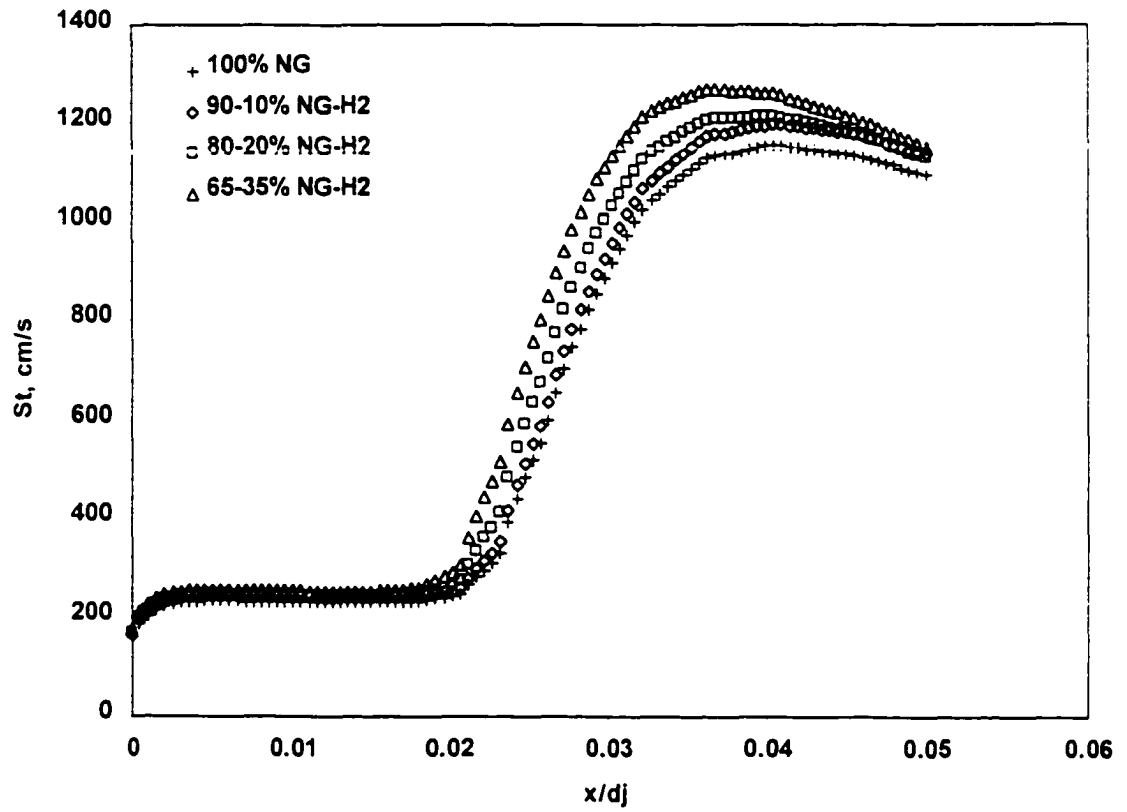


Fig. 5.21 Computed Radial Profiles of the Normalized Axial Fluctuation of Cold Fuel Jets at Various Mixture Conditions



**Fig. 5.22 Computed Axial Profiles of the Turbulent Flame Speed
(Calculated Using $S_{L,max}$) at Various Mixture Conditions**

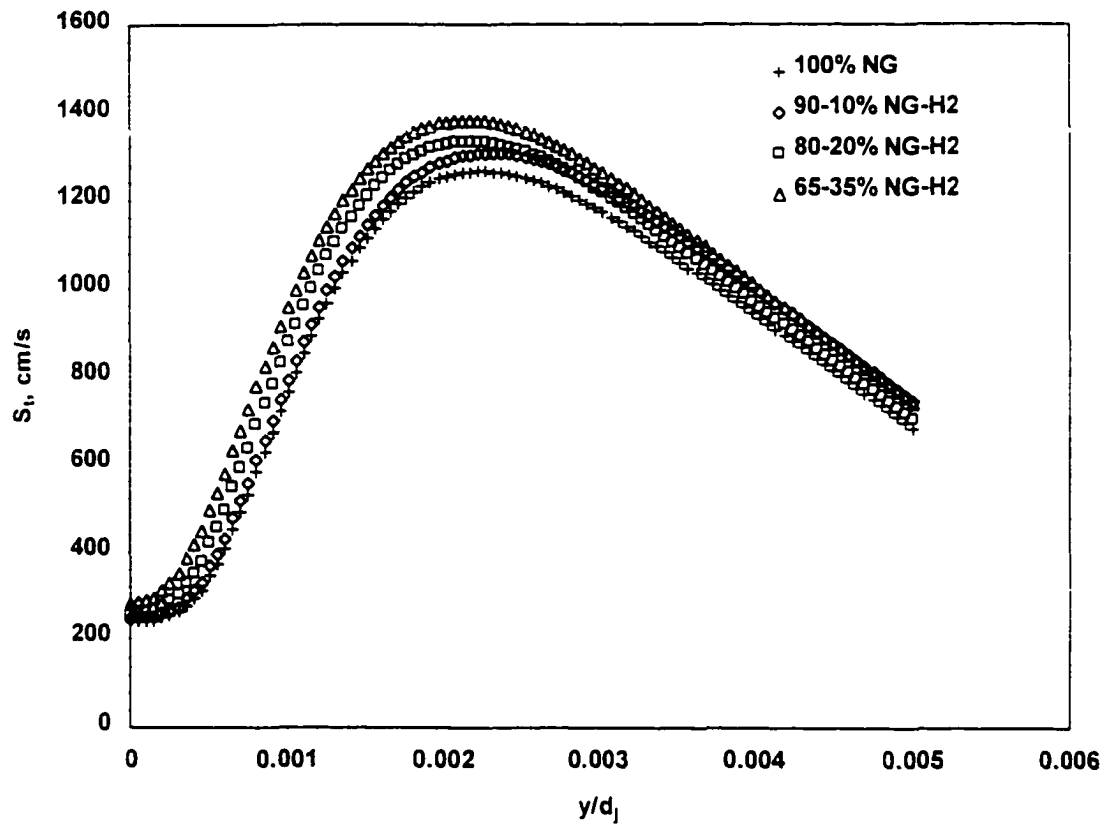


Fig. 5.23 Computed Radial Profiles of the Turbulent Flame Speed (Calculated Using $S_{L,max}$) at Various Mixture Conditions

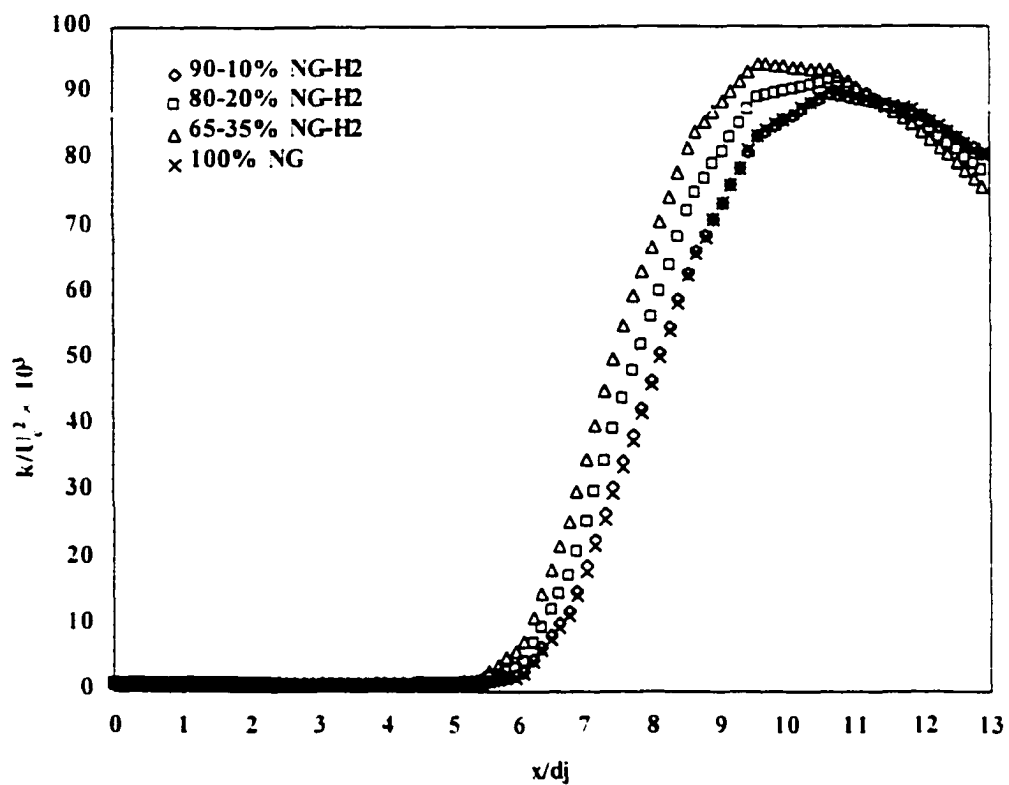


Fig. 5.24 Computed Axial Profiles of Turbulent Kinetic Energy of Cold Fuel Jets at Various Mixture Conditions

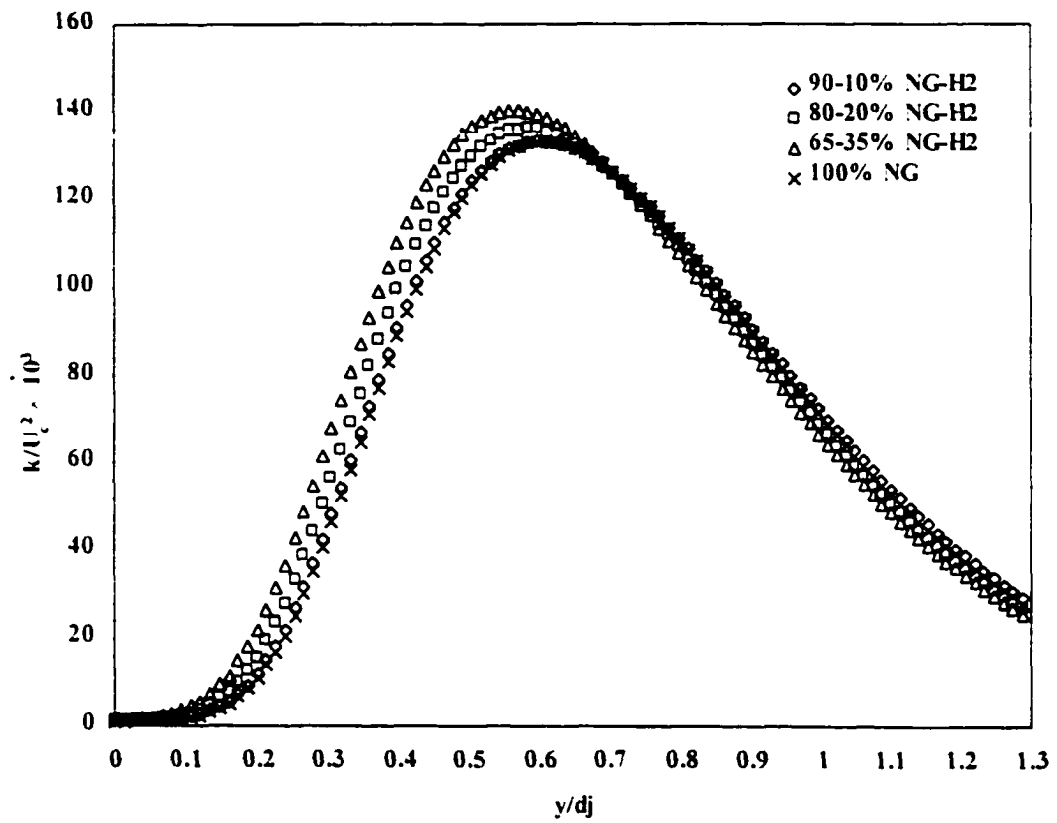


Fig. 5.25 Computed Radial Profiles of Turbulent Kinetic Energy of Cold Fuel Jets at Various Mixture Conditions

Chapter Six

Flame Structure

The combustion mechanism of natural gas becomes complex with the addition of hydrogen. Since the combustion chemistry of hydrocarbon fuels is significantly different from that of hydrogen, the flame structure of the composite fuel largely depends on the interaction of the two mechanisms. A detailed knowledge about the production and destruction of the stable species and intermediate radicals is necessary to understand the fundamental combustion chemistry of the composite fuels. The radicals such as OH, CH and CN are important intermediates in all aspects of combustion chemistry. In combustion environment hydroxyl radical (OH) is a reliable indicator of flame zones, flow structure, and temperature (Battles et al., 1997; Hanson et al., 1990; Hanson, 1997; Smyth et al., 1990). Also, OH radical has been implicated as a dominant oxidizer of soot particles in flames (Smyth et al., 1990, 1994). On the other hand, CH radical is involved in a wide range of combustion chemistry due to its rapid addition and subtraction reactions with numerous species (Miller and Taylor, 1987; Norton and Smith, 1991). For example, CH radical plays an important role in the inception stage of soot formation in hydrocarbon flames through the chemi-ionization reaction of ground state CH with atomic oxygen and electronically excited CH ($A^2\Delta$) with acetylene. The CH radical can easily enter into carbon-hydrogen bonds, which is an important process for hydrocarbon growth chemistry leading to soot particles. In addition, the reaction of CH with N_2 is believed to be the main source of *Prompt NO* formation. Regardless of the local flame stoichiometry, the CH radical usually encounters a wide variety of reactive collision partners. On the other hand, H and O bear a tremendous importance in relation with the

local laminar and turbulent flame speeds. As it was seen in Chapter Five, the H and O concentrations are some of the key factors, which determine local flame speed and thereby control the stability mechanism. Information about local flow-field velocity and its interaction with H and O formation/destruction will be highly beneficial to explain the increased flame stability of natural gas-hydrogen mixtures.

In this chapter, probe measurements of CO₂, CO, NO and O₂ and LIF measurements of the concentration profiles of OH, CH, H and O radicals/atoms in the near-burner, mid-flame and far burner regions of turbulent flames burning different mixtures of natural gas and hydrogen are presented. The local flame temperatures are calculated from N₂ Raman spectra through spectral fitting techniques described in Chapter Three. The LIF signals are also quantified for specific concentrations using the spectroscopic modeling technique discussed in the same chapter. The near-field planar images of OH concentration are also shown. The measured flame temperature and species concentrations are compared with computed values. The computed values of the local axial, radial velocity and turbulent fluctuations are also reported. The fuel jet exit Reynolds number has been kept constant at 8500.

6.1 Flame Temperature

The measured and computed radial temperature distributions in near-burner ($0.2 L_f$), mid-flame ($0.5 L_f$) and far-burner ($0.75L_f$) regions for different compositions of natural gas and hydrogen are shown in Figs 6.1-6.3. The temperature distributions were calculated using N₂ Raman signals through a spectral fitting method. The near-burner

radial temperature profiles show dual humps, which are typical for diffusion flames. The peak of the temperature lies on the *stoichiometric contour* of the flames where most of the reactions take place. The stoichiometric contour is a radial location where outwardly moving fuel meets incoming air. Experimental results show that for the 100% natural gas flame the stoichiometric contour is relatively flat and is located relatively far from the burner axis. With the increase of hydrogen content in the mixture, the peak temperature increases and the stoichiometric contour moves inward and becomes steeper. This result indicates that the overall reaction rate increases as the hydrogen content in the mixture increases. Since hydrogen diffuses and reacts faster, it encounters the incoming oxidizer first and releases heat quickly, which further enhances the burning of the natural gas. The peak temperature in the 100% natural gas flame is 1600 K. The measured peak flame temperature in the natural gas flame has a poor agreement with equilibrium temperature, 2225 K¹ (Choudhuri, 1997). First, the concept of equilibrium does not hold well for hydrocarbon reaction chemistry, second the effective flame temperature in a hydrocarbon flame is low due to heat losses both in the form of banded radiation from carbon dioxide and continuous radiation from solid soot particles. The computational results using the two-step simplified chemical kinetics show a similar trend for the same mixture condition. Compared to experimental results, predicted temperatures at the stoichiometric contour are higher and the shape of the contours are steeper for all mixtures. However, the trend showing the inward movement of the stoichiometric contour agrees quite well with the experimental findings. Since the effect of flame radiation was neglected in computations the discrepancy in the peak temperature values is understandable. Furthermore, due to the inherent uncertainties in

the reaction kinetics model, the computation predicts a faster fuel-burning rate than its experimental or rather physical counterpart, which ultimately results in a steeper stoichiometric contour. The computed results show that, for flames with a higher hydrogen concentration in fuel mixture, the temperature remains low value close to the burner axis which implies the amount of reactions is small in this region. However, the measured values of temperature show a significant increase in temperature at the corresponding locations. This is possibly due to the formation and destruction of heavier hydrocarbons (C_2 and higher) near the fuel jet axis for flames with higher hydrogen concentration. As reaction proceeds on fuel oxidizer interface (stoichiometric contour), the fuel starts pyrolyzing inside the fuel-rich zone due to heat convection and radiation from the flame surface, and hence species with C_2 or higher number of carbon atoms form near the fuel core. However, due to their higher mass they are unable to diffuse faster and again undergo changes inside the fuel-rich zone. Therefore, reactions involving heavier hydrocarbons are mostly confined close to the burner axis, which significantly contribute to the temperature changes at that location. Since, all the elementary reactions were neglected in the reaction mechanism and radiation effects were omitted, the computational scheme was unable to resolve this phenomenon.

In the mid-flame region, the radial temperature profile of the 100% NG flame shows a peak on the central axis (Fig. 6.2). However, with the increase in hydrogen concentration, the peak shifted symmetrically in two radial locations. Also, the peak flame temperature increases with the increase of hydrogen concentration in the mixture. It is evident that the reaction zone of a pure NG flame is distributed over the radial locations in mid-flame region. Also, the formation of heavier Polycyclic Aromatic

Hydrocarbons (PAH) in the 100% NG flame keeps the reactions confined close to the central axis. In contrast, the increase in hydrogen concentration in the mixture increases the overall mass diffusivity of the mixtures, which ensures faster movement of the hydrogen outside the jet core. Hence, even in the mid-flame region, the flames with a higher hydrogen concentration have distinct stoichiometric contours. The radial profiles of the computed flame temperatures at the mid-flame region are also presented. The measured and the predicted trends are not similar in these locations: in fact the disagreement is severe. These discrepancies can be attributed to two fundamental reasons: first, the simplified mechanism used for computation does not account for any intermediate polycyclic hydrocarbons; second, formation of solid soot particles, which contributes to heterogeneous combustion and flame radiation, is not included in computation. As discussed earlier, formation of heavier hydrocarbons as an intermediate product keeps the reactions active on the central axis. Hence, experimental measurements show a high flame temperature in this location. On the other hand, the numerical computation does not account for this phenomenon. As combustion gases move to far-downstream locations (far-burner region, $0.75L_f$), most of the fuel pyrolyzing reactions already cease, and CO and soot oxidation reaction dominates. It can be seen that the flame temperature profiles at the far-burner locations for all mixture conditions have a peak on the central axis (Fig. 6.3). Similar to near-burner and mid-flame regions, fuel mixtures with higher hydrogen concentration have higher peak flame temperature in the far-burner location. The trends of the computed flame temperature show good agreement with measured values. As mentioned earlier, in this location CO oxidation is the dominant mechanism. Since the global CO reaction is embedded in the

simplified reaction mechanism used for numerical computation, the flame temperature prediction is good in the far-burner region. Yet, the predicted values are higher than the measured values.

6.2 Stable Species Concentration Profiles

Radial concentration profiles of CO_2 , CO, NO, O_2 at different axial locations for different mixture conditions are shown in Figs 6.4-6.16. The computed radial profiles of CO_2 and CO at these locations are also presented for comparison.

6.2.1 Carbon dioxide (CO_2)

Since the stoichiometry and the flame temperature affect the formation rate of CO_2 , the radial profile of CO_2 concentration is known to follow the radial temperature profile of the flame. The radial profiles of CO_2 concentration (volumetric percent) in different axial locations for various mixtures of natural gas and hydrogen are shown in Figs.6.4-6.6. Similar to the temperature profile, the measured CO_2 radial profiles in the near-burner region, exhibit the prominent double-hump structure, which becomes flatter in the mid-flame region and disappear in the far-burner region. At the same Reynolds number, the increase in hydrogen concentration decreases the total carbon input into the system and thereby reduces the total CO_2 formation. The experimental measurements show the CO_2 formation over a wide range of radial locations of the flame. However, the computed values show the reactions are confined to a small region around the stoichiometric contour. Also, in the near-burner location, the computation does not show a significant amount of CO_2 formation close to the burner axis. The predicted values are

an order of magnitude higher than the measured values. Similar to flame temperature the trend of the computed CO₂ concentration profile in the mid-flame region does not match with the measured profiles. However, in the far-burner the agreement is reasonable. In the far-burner region, the computed values show sharp decreases of CO₂ concentration with the increase of hydrogen concentration in the mixture.

6.2.2 Carbon monoxide (CO)

The measured and computed radial concentration (volumetric percent) profiles of CO at different axial locations are shown in Figs. 6.7-6.9. Carbon monoxide is formed in the fuel-rich zone of the flames as an intermediate product of combustion due to fuel pyrolysis. This reaction is very rapid as it only involves the radicals. The measured CO profiles show that in the near-burner region the double hump is present for all mixture conditions. However, in the downstream region the single axial peak of CO reveals that the reactions are not confined to the flame sheet. The CO is formed over the entire cross-section of the flame and accumulates around the flame axis. Also, it has been observed that a higher concentration of hydrogen in fuel decreases the CO concentration in the flame. This is understandable since the increase of Carbon/Hydrogen ratio and a decrease of the effective flame temperature increase CO production. Also, the addition of hydrogen in the mixture increases OH formation, which further prompts the reduction of CO concentration by increasing CO oxidation ($\text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H}$). In near burner region, the peak CO values occur at the same locations despite an increase in hydrogen concentration. The inward movement of the stoichiometric contour does not alter the peak CO concentration. It is evident that the CO formation mechanism is not highly

sensitive to the peak flame temperature, rather it largely depends on the concentration of the intermediate radicals such as OH and H. It can be seen that with most mixture conditions (except 100% NG) the CO peak occurs on the fuel lean side of the stoichiometric contour. This is interesting, since it is expected that CO concentration should be higher on the fuel rich side. However, it is evident that in the composite fuel flames of the present investigation, the CO forms close to fuel-rich side of the stoichiometric contour and is rapidly oxidized on the stoichiometric region due to the abundance of OH radicals. The remainder of CO accumulates in the fuel-lean side of the stoichiometric contour. The near-burner profiles of the computed CO concentration show a similar trend, which reconfirms the argument presented in conjunction with the experimental measurements. However, the computation predicts a lower CO than the experimental measurement. The mid-flame radial concentration of CO shows a dual hump profile with peaks at 5d_j location. The computed values also show a similar trend. In the mid-flame region, the magnitudes of measured and computed values of CO concentration are comparable. In the far-burner region, the peak CO concentration occurs close to the burner axis. As expected, CO concentration decreases with the increase of hydrogen concentration in the fuel mixture. However, the rate of decrease becomes sharper very sharp as hydrogen concentration increases from 20% to 65%. As mentioned earlier, CO formation and destruction mechanisms primarily depend on the concentration of OH radicals. However, both soot and CO compete for OH for oxidation. As hydrogen concentration exceeds 20% concentration the total soot loading of the flame suddenly decreases, which makes more OH available for CO oxidation.

This is a possible explanation of the sharp decrease of CO concentration for mixtures with more than 20% hydrogen concentration.

6.2.3 Nitric Oxide (NO)

The measured concentrations (ppm) of NO in the near-burner, mid-flame and far-burner regions for different mixtures of natural gas and hydrogen are shown in Figs. 6.10-6.12. In diffusion flames, the radial profiles of NO concentration are generally expected to follow the temperature profiles since the formation of NO through Thermal-Zeldovich route is highly temperature dependent (the activation energy required to initiate the reaction chain is high). In the present study, at mixture conditions of natural gas-hydrogen composite fuel, the peak temperature is high on the stoichiometric contour and in the fuel-rich zone. Hence, it can be expected that the thermal route is the primary means of NO production. It has been found that in all of the present experiment, the NO concentration profiles closely follow the temperature profiles. In all three locations, NO formation is higher for fuel mixtures with hydrogen content. As it is mentioned earlier, the increase of hydrogen concentration in the fuel mixture increases the local flame temperature, which causes an increase in NO production through thermal route.

6.2.4 Oxygen (O₂):

Pure diffusion flames of fuels issuing into surroundings do not contain any oxygen at the burner exit. As the flame entrains air, the concentration (volumetric percent) of O₂ along the flame axis increases gradually. On the air-side of the reaction zone, in the radial

direction, the O_2 concentration reaches its local molar concentration in air, whereas on the fuel side, the O_2 concentration varies from very low to a maximum, depending upon the location of the radial plane. Thus, the radial profiles are 'V' shaped. In this study, the above mentioned trends have been verified. Also, the increase of natural gas concentration slightly decreases the O_2 concentration inside the flame. The radial profiles of O_2 concentration at different axial locations for different mixtures of natural gas and hydrogen are shown in Figs. 6.13-6.15. In the near-burner location, flames with higher hydrogen content in the fuel mixture show higher O_2 concentration than those of flames with lower hydrogen content. Again, this shows increases mass diffusivity of the fuel mixture with the increase in hydrogen content. However, this observation is not prominent in mid-flame and far-burner region.

6.3 Intermediate Radicals/Atoms Concentration

6.3.1 OH Radical Concentration

The instantaneous PLIF images of OH concentration in the near-burner region for different hydrogen concentrations in the fuel mixture are shown in Fig. 6.17. It can be clearly observed that OH radical concentration increases sharply with the increase of hydrogen concentration in the mixture. The instantaneous OH PLIF images of the 80-10% NG- H_2 flames are superimposed on the computed mean velocity field, and are shown in Fig. 6.18. This qualitative representation of velocity and chemistry interaction indicates how local flow-field velocity is related with formation and destruction of intermediate radicals. As seen from the figure, at the location of maximum OH concentration, the fuel is rushing towards the reaction zone. The near-burner profiles of

measured and computed OH concentrations for different mixtures of natural gas and hydrogen are shown in Figs 6.18-6.20. The OH radical concentration typically shows peaks on stoichiometric contour, as it is a reliable indicator of the reaction zone. The measured peak volumetric concentration of OH in the 100% NG flame at the near-burner location is 7.1×10^{-3} which is in close agreement with the findings of other investigators (Smyth and Tjossem, 1990). The concentration of OH radical increases as the hydrogen content increases in the mixture. This is expected since with the increase of hydrogen content in the fuel mixture, the production of OH through the reaction set [R1, R3, R5, R7, R11 and R15] increases while the destruction through [R25] decreases (Appendix I). However, in the near-burner region close to burner axis, 90-10% and 80-20% NG-H₂ flames shows an area of constant OH concentration. However, as hydrogen concentration increases to 65% the region of OH formation largely increases. The computed profiles OH concentration in the near-burner region show a trend similar to that of flame temperature. The computed results show that with the increase of hydrogen concentration in the fuel mixture, the OH mole-fraction increases and the locations of peak OH concentration move closer to the burner axis. The computation shows a higher OH concentration than measurement. In the mid-flame region both the 100% NG and 90-10% NG-H₂ flames show peaks on the burner axis. However, the 80-20% and 65-35% NG-H₂ fuel mixtures show peaks at 2.5d_j and 1.5d_j locations. As discussed earlier, faster dispersion of reaction zone accompanying the increase of mixture mass diffusivity is primarily responsible for this trend. However, the computed profiles fail to resolve this trend at this axial location. The measured trends of OH concentration in the far-burner regions are random, and do not show any definite radial variation. Also, OH

concentration is low in this region. However, the overall trend shows that an increase in hydrogen concentration in the mixture increases OH concentration in the far-burner region. The computed values do not confirm to this random profile, but show a gradual increase in OH concentration close to the burner axis.

6.3.2 CH radical concentration

The measured CH concentration profile along with the computed values at different axial locations in the flame of different mixtures of natural gas and hydrogen are shown in Figs 6.21-6.23. In the near burner region, the CH concentration profiles have peaks in the fuel-rich side of the fuel-oxidizer interface (Fig. 6.21). One implication of this observation is that CH is a marker of the zone where reaction is occurring, while OH indicates where reaction has occurred (Dyer and Crosely, 1985). Compared to experimental values, computed values show a much narrower region where CH is forming, however the peak computed values are three times higher than of measured peak values. Since the post processing technique of computed CH concentration ignores most of the CH destruction mechanism and over emphasizes the local flame temperature, the computation yields a high CH concentration. Also, in the near-burner region computation predicts the CH formation confined to a very narrow zone just outside the stoichiometric contour. Both measurement and computation show the decrease of CH concentration with the increase of hydrogen content in the mixture. In the mid-flame region, the peak of CH concentration occurs on the burner axis for the 100% NG flame (Fig. 6.22). However, the CH concentration profiles in the flames of natural gas-hydrogen composite fuels still show the dual hump nature in this location. The trend predicted by computation is similar to the near-burner profile and the

predicted values are an order of magnitude higher than measured values. Similar to OH profile, the far burner CH concentration radial profiles are also random and do not resemble any systemic variation. However, similar to near-burner and mid-flame CH concentration the overall CH concentration increases with the increase of hydrogen concentration in the mixture. The far-burner profiles of computed CH concentration show a peak on the central axis (Fig. 6.23)

6.3.3 H Atom Concentration

The measured and computed profiles of H atom at different mixture conditions are shown in Figs. 6.24-6.26. In the near-burner location, the peak H atom concentration occurs on the rich side of the stoichiometric contour. The maximum measured H-atom concentration in the 100% natural gas flame is 9×10^{-3} , which is comparable to the equilibrium value 7.9×10^{-3} (Choudhuri 1997). However, the value does not agree well with the present computation (29×10^{-3}) and computed value (18.5×10^{-3}) presented in Smyth and Tjossem (1990). This discrepancy can be attributed to the simplified chemistry model used in the present computation and equilibrium of R5 (Appendix 1) used by Smyth and Tjossem (1990). Certainly, this simplification might underpredict the H₂O formation through the reaction sequence R5-R6 and thereby overshooting the H-mole fraction. With the increase of hydrogen content in the fuel mixture, the H radical concentration increases in all axial locations. The computed profiles of H concentration also yield a similar trend. In the mid-flame region, the peak H concentration occurs on the burner axis for all fuel mixtures except the 65-35% NG-H₂. For the 65-35% NG-H₂ mixture, the concentration profile still shows the symmetric peaks outside the burner

axis. The computed values show the peak H concentration on the central axis for all mixtures. However, the computed and measured values of peak H concentration are comparable. Similar to OH and CH radical concentrations, the far burner concentration of O radicals also varies randomly and does not show any particular trend (Fig. 6.26). However, in far-burner region the overall H concentration increases with the increase of hydrogen content in the mixture. The computed H concentration profiles for different natural gas-hydrogen mixtures show peaks on the central axis.

6.3.4 O Atom Concentration

The radial distribution of O atom concentration in the near-burner region for different mixtures of natural gas and hydrogen are shown in Fig. 6.27. The highest O concentration lies on the lean side of the stoichiometric contour. Both measured and computed values of the O concentration also show a significant amount of O production in the fuel-rich side. The computation yields a peak value of 3.1×10^{-3} O-mole fraction in the 100 % natural gas flame, which is in an excellent agreement with the results (average 2.9×10^{-3}) of other investigators (Smyth and Tjossem, 1990). This value is comparable with the equilibrium value 2.6×10^{-3} (Choudhuri, 1997). Hence, a simplification of the reaction mechanism by assuming the partial equilibrium of O atom is feasible. This is completely opposite compare to what occurs in a laminar (Choudhuri and Gollahalli, 2000b) where O atom mole fraction does not equilibrate due to super equilibrium O concentration. However, in a turbulent flame the O radical concentration does reach equilibrium. In axial locations, the mole fraction of O atom increases with the increase of hydrogen content in the fuel mixture. This is reasonable since the recombination

reaction (R24, Appendix I) slows due to the decrease in third-body collisions as the amount of hydrogen increases in the mixed fuel. The agreement of the trends of computed and measured O concentration in the mid-flame region is poor (Fig. 6.28) although the magnitude is comparable. The measured values show peaks in locations outside the central burner axis, whereas computed values show peaks on the burner axis. The measured radial profiles in the far-burner regions show nearly constant value of O concentration along the burner radius. However, the computation predicts a peak O concentration on the burner axis. Similar to in the near-burner and mid-flame regions, in the far-burner region O concentration increases with the increase of hydrogen content in the mixture.

6.4 Local Velocity Field

The computed axial profiles of the normalized absolute velocity ($U_{abs}^2 = U^2 + V^2$) for different concentrations of natural gas-hydrogen mixtures are shown in Fig. 6.30. Axial velocity decays with the increase of axial distance. However, the axial velocity decays faster with the increase of hydrogen content in the mixture. The radial profiles of the normalized axial velocity at three axial locations for different mixtures of natural gas and hydrogen are shown in Figs. 6.31-6.33. The axial velocity has a peak on the burner axis, which decays along the radial direction. Similar to axial profiles, the radial profiles at all three locations show a faster decay of axial velocity with the increase of hydrogen concentration in the mixture. The centerline axial velocity reduces with the increase of axial distance. The radial profiles of axial turbulent fluctuations at different locations for different mixture conditions are shown in Figs. 6.34-6.36. Similar to that in the cold jet,

the turbulent fluctuation increases with the increase of hydrogen content in the mixture. Close to the burner axis, the axial turbulent fluctuation has a higher intensity, which reduces as the radial distance increases. The increase in hydrogen concentration in the mixture increases the formation of OH radicals and H, O atoms. Simultaneously, an increase in hydrogen concentration causes a faster decay of axial velocity in both axial and radial directions. Also, it was shown earlier that the mean flow field velocity changes direction close the radical formation zone.

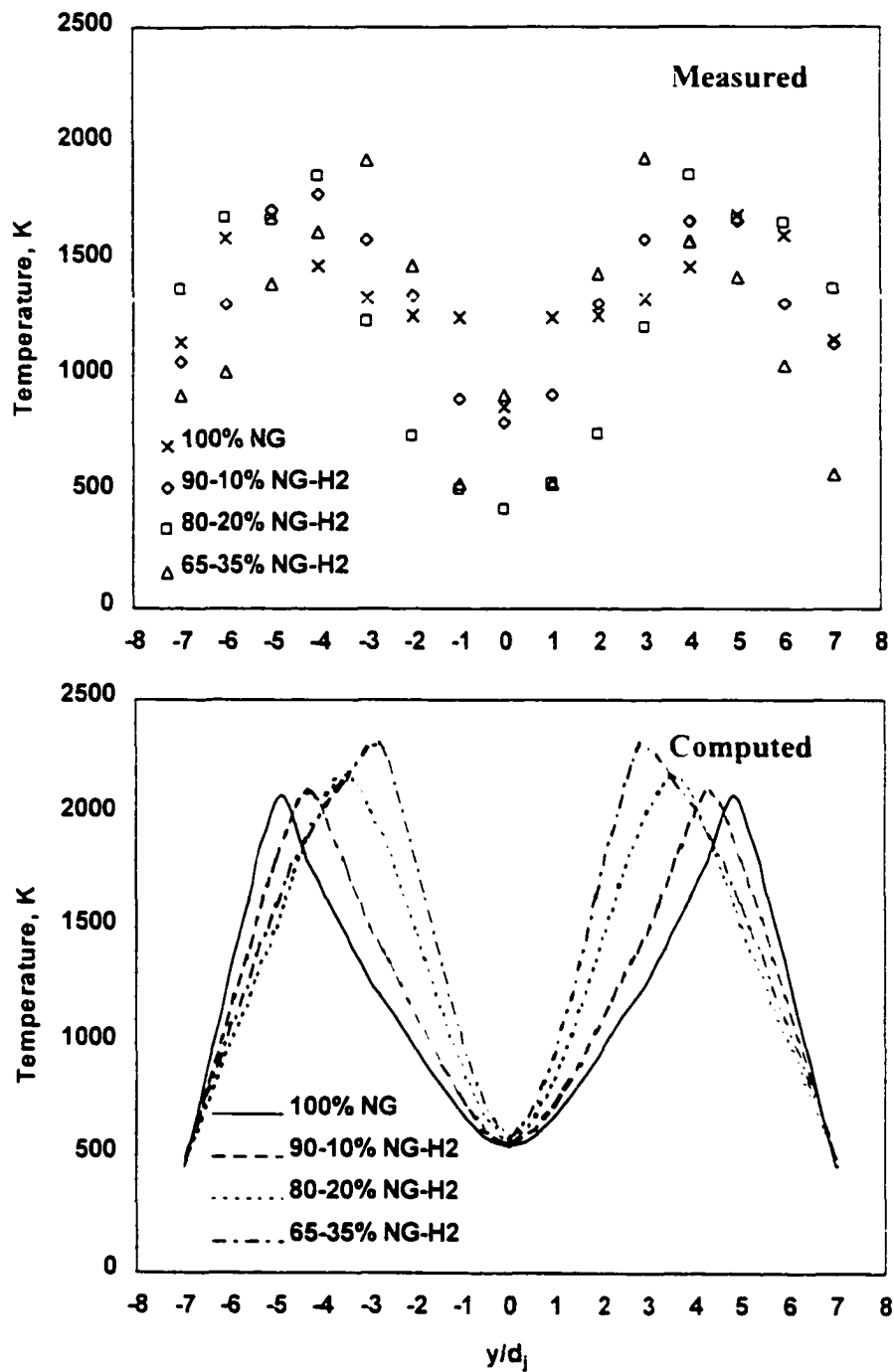


Fig. 6.1 Measured and Computed Radial Flame Temperature Profiles at a Near Burner ($0.25 L_f$) Location

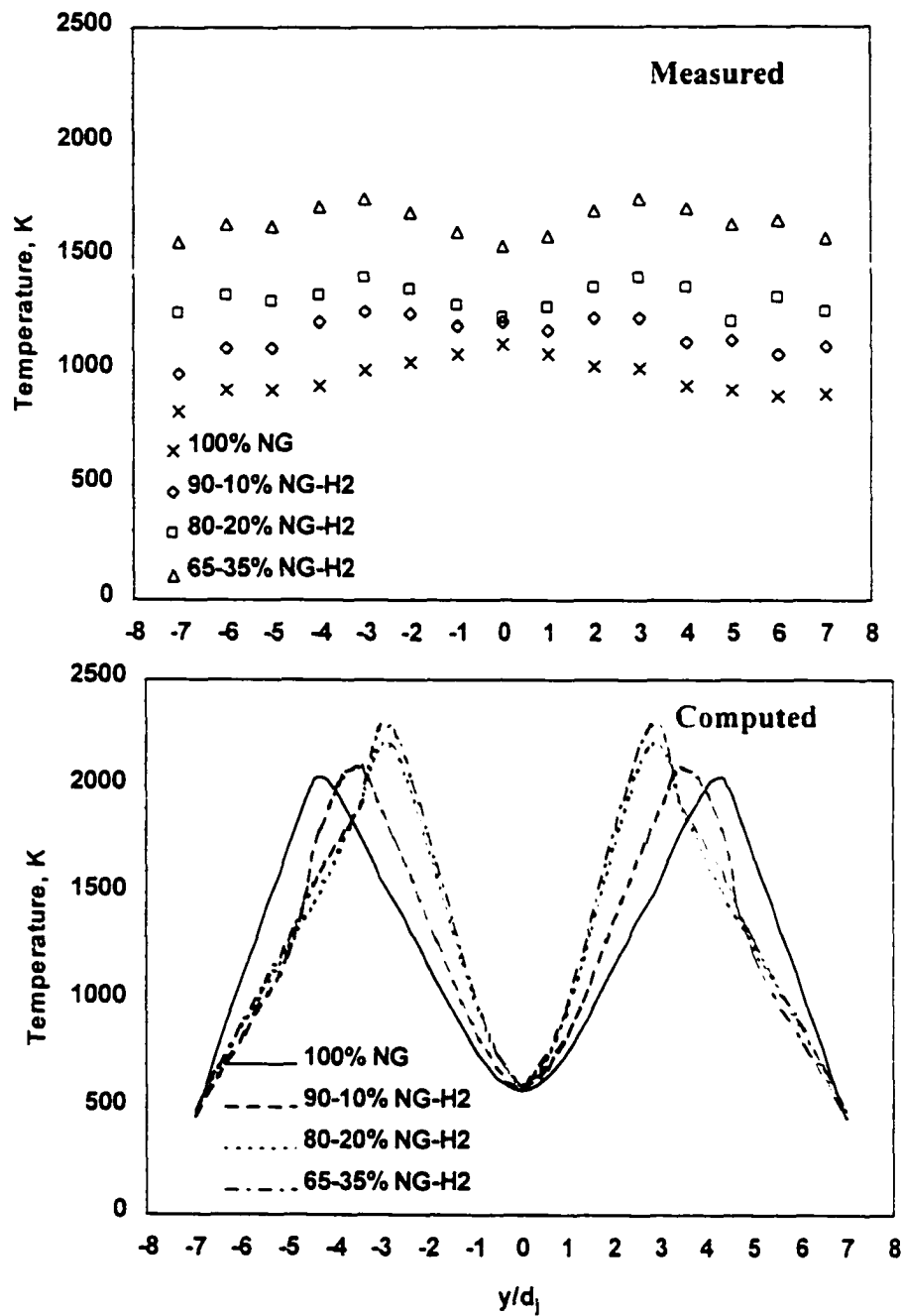


Fig. 6.2 Measured and Computed Radial Flame Temperature Profiles at a Mid-flame ($0.50 L_f$) Location

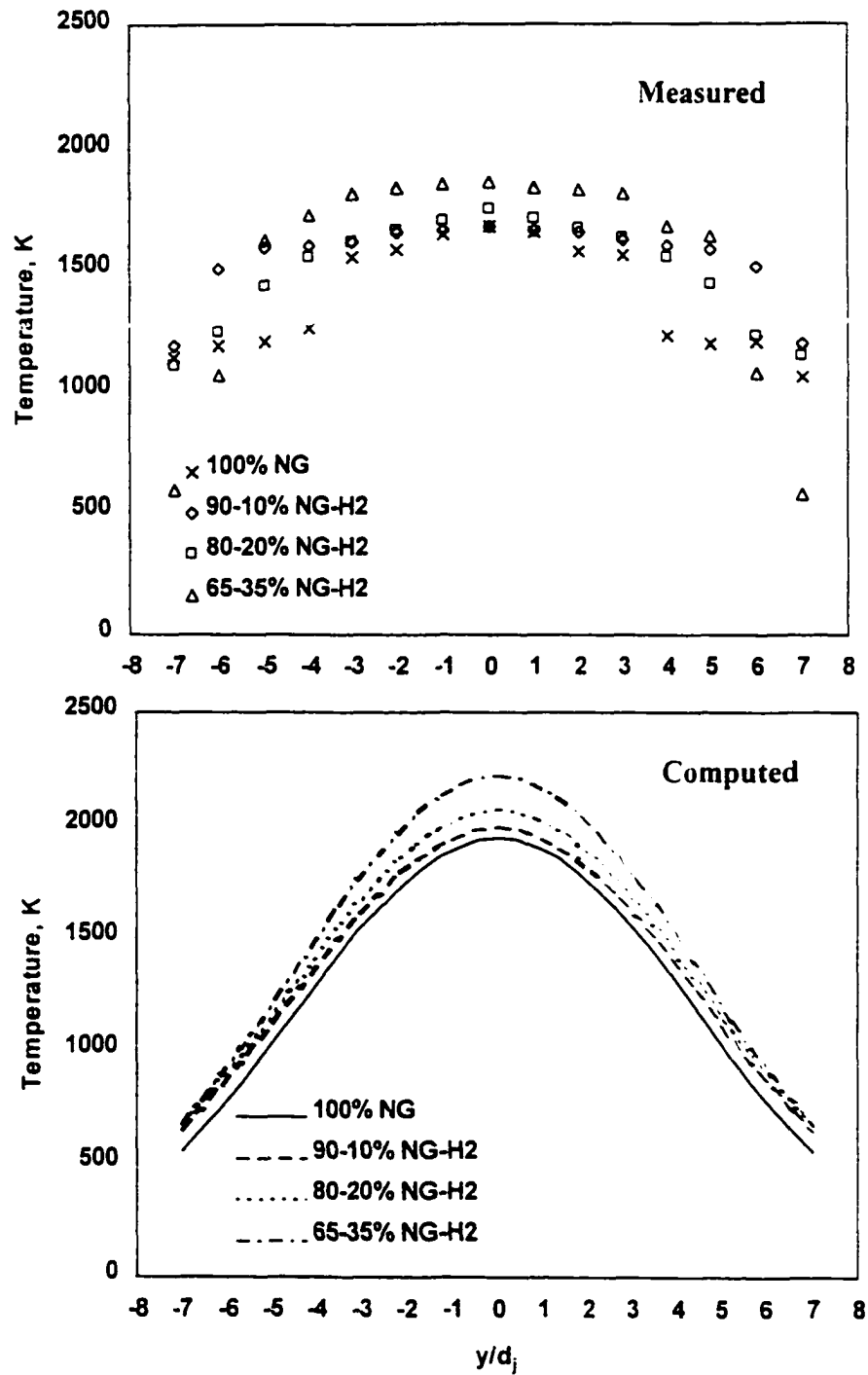


Fig. 6.3 Measured and Computed Radial Flame Temperature Profiles at a Far-Burner ($0.75 L_f$) Location

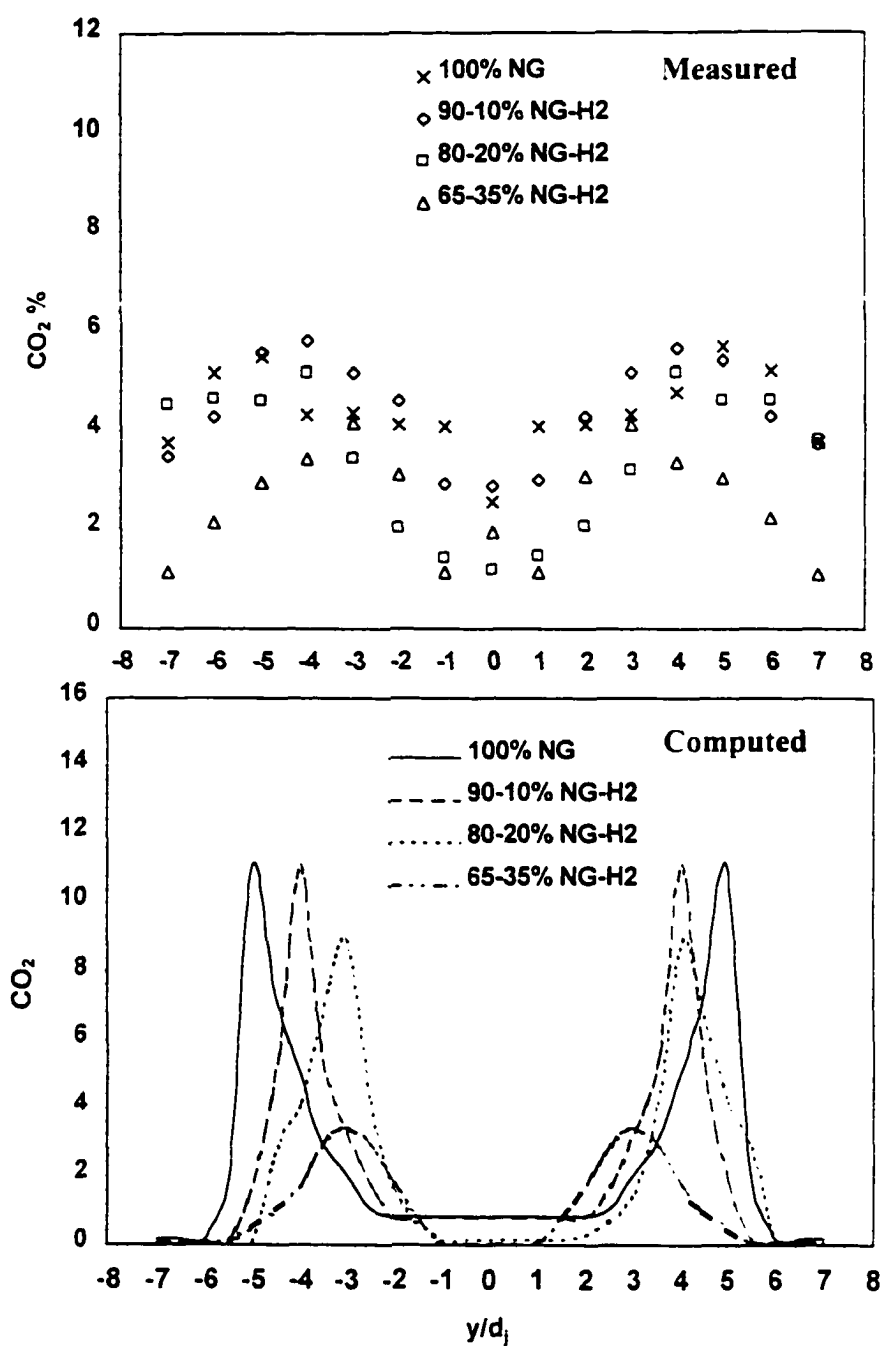


Fig. 6.4 Measured and Computed Radial CO₂ Profiles at a Near-Burner (0.25 L_f) Location

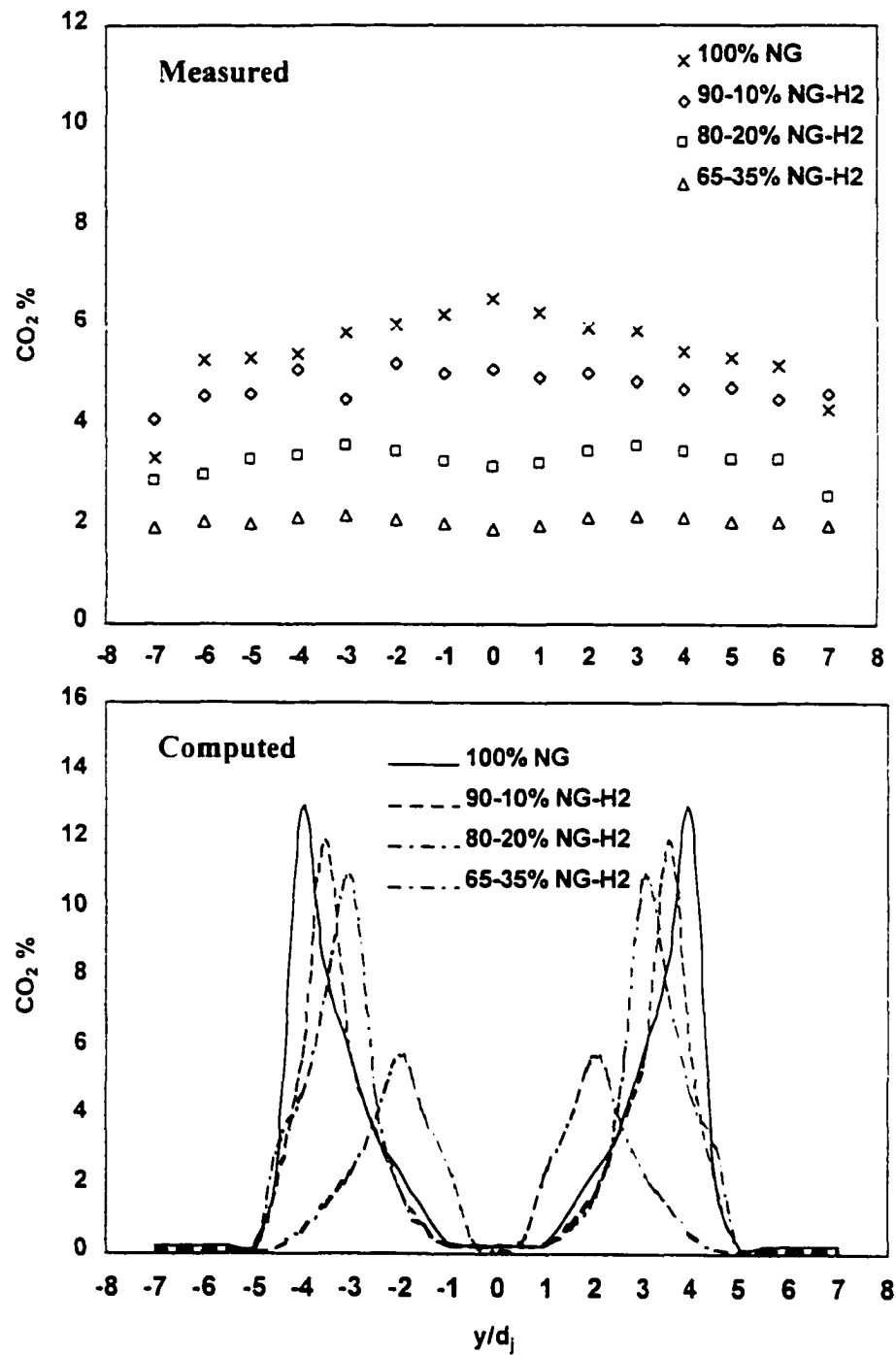


Fig. 6.5 Measured and Computed Radial CO₂ Profiles at a Mid-Flame (0.50 L_f) Location

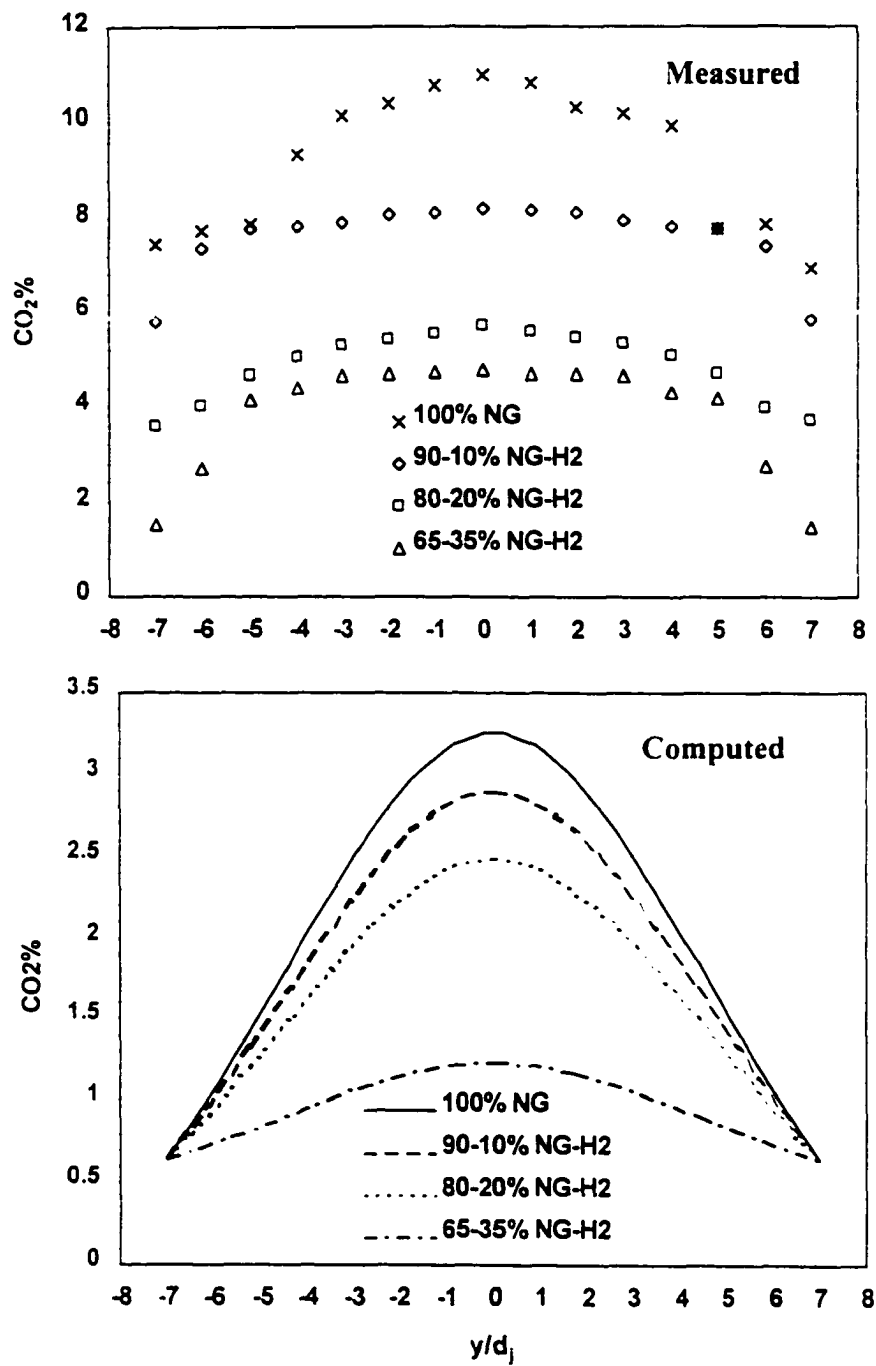


Fig. 6.6 Measured and Computed Radial CO₂ Profiles at a Far-Burner (0.75 L_f) Location

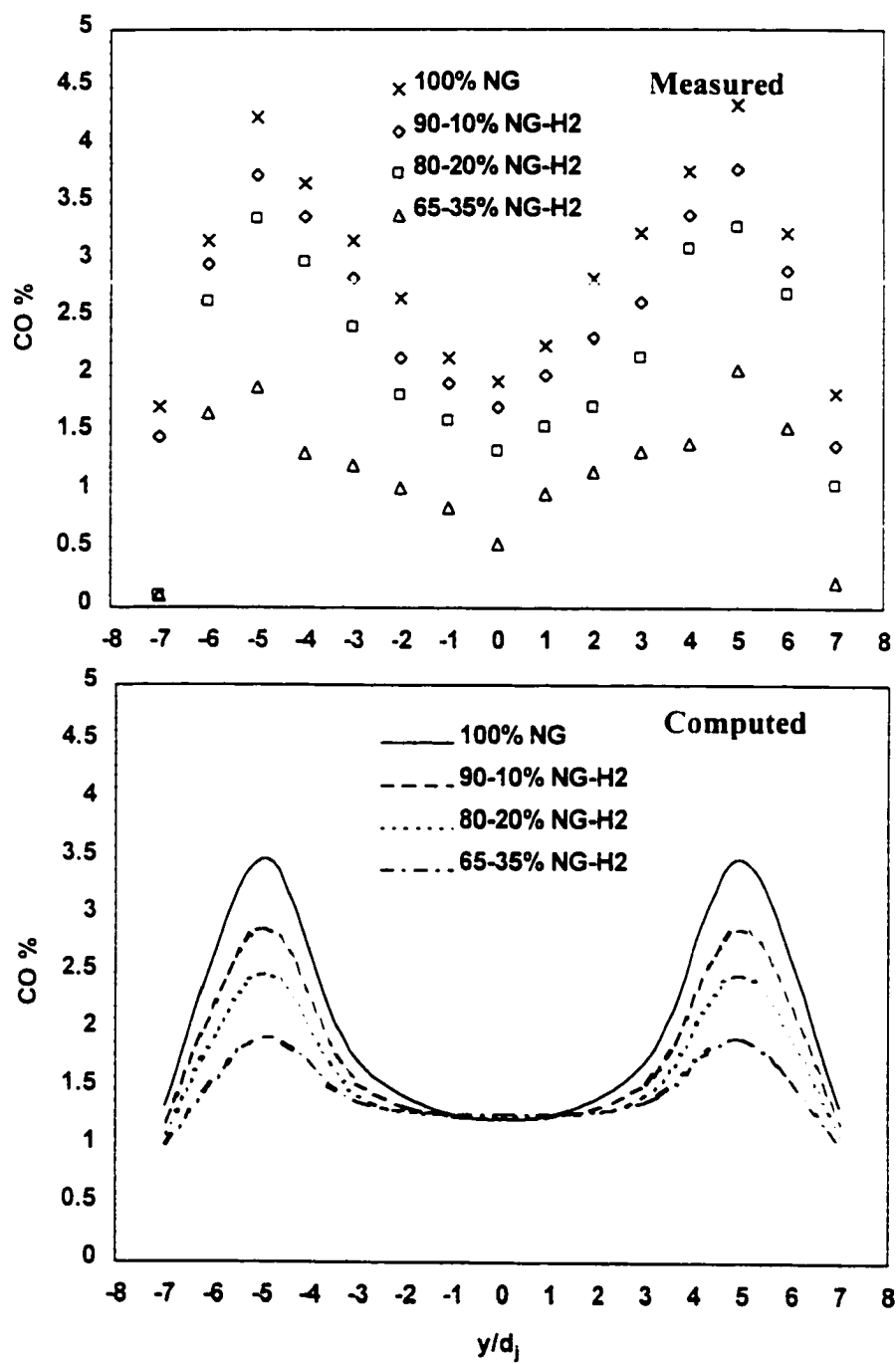


Fig. 6.7 Measured and Computed Radial CO Profiles at a Near-Burner ($0.25 L_f$) Location

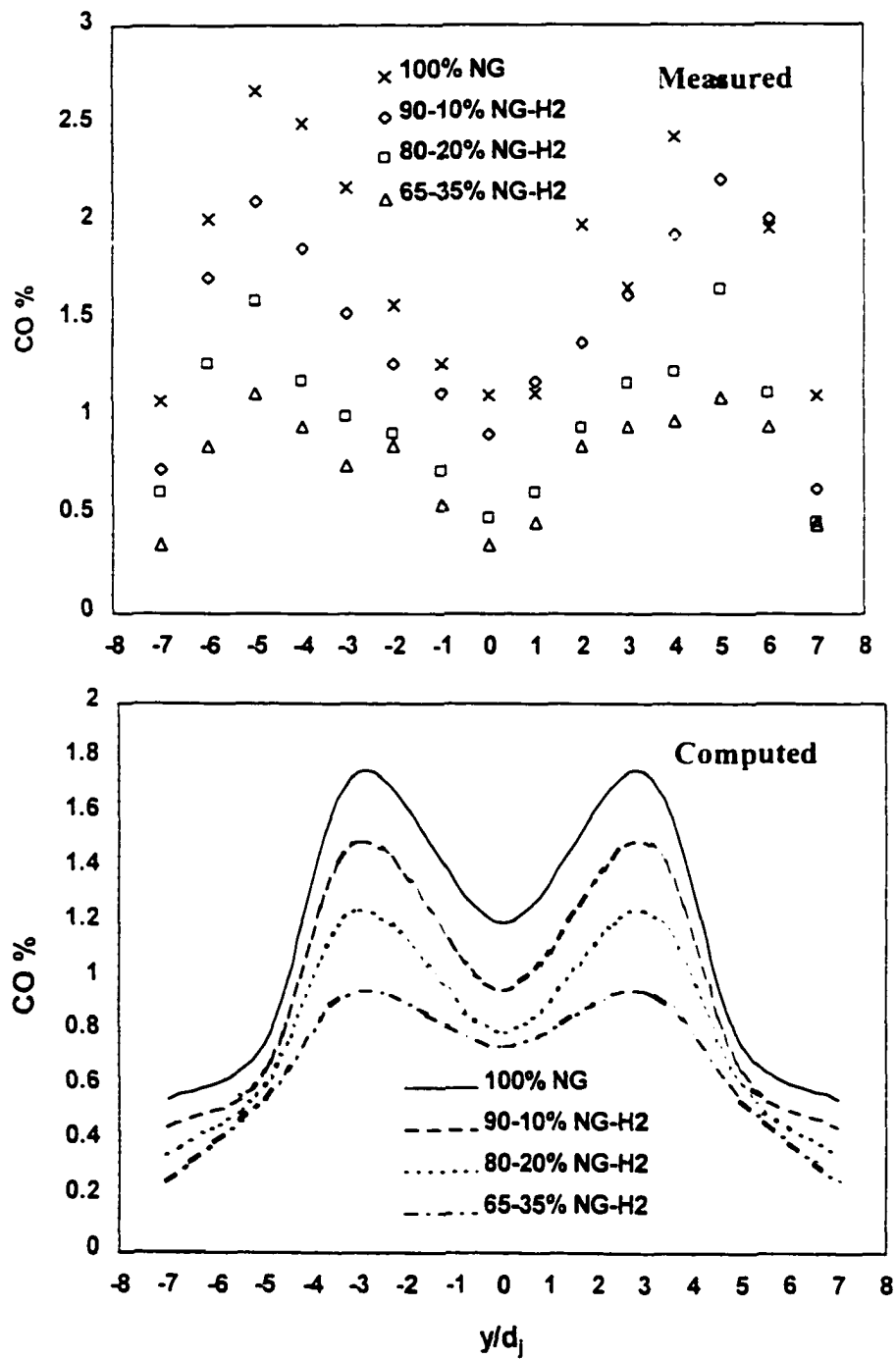


Fig. 6.8 Measured and Computed Radial CO Profiles at a Mid-Flame ($0.50 L_f$) Location

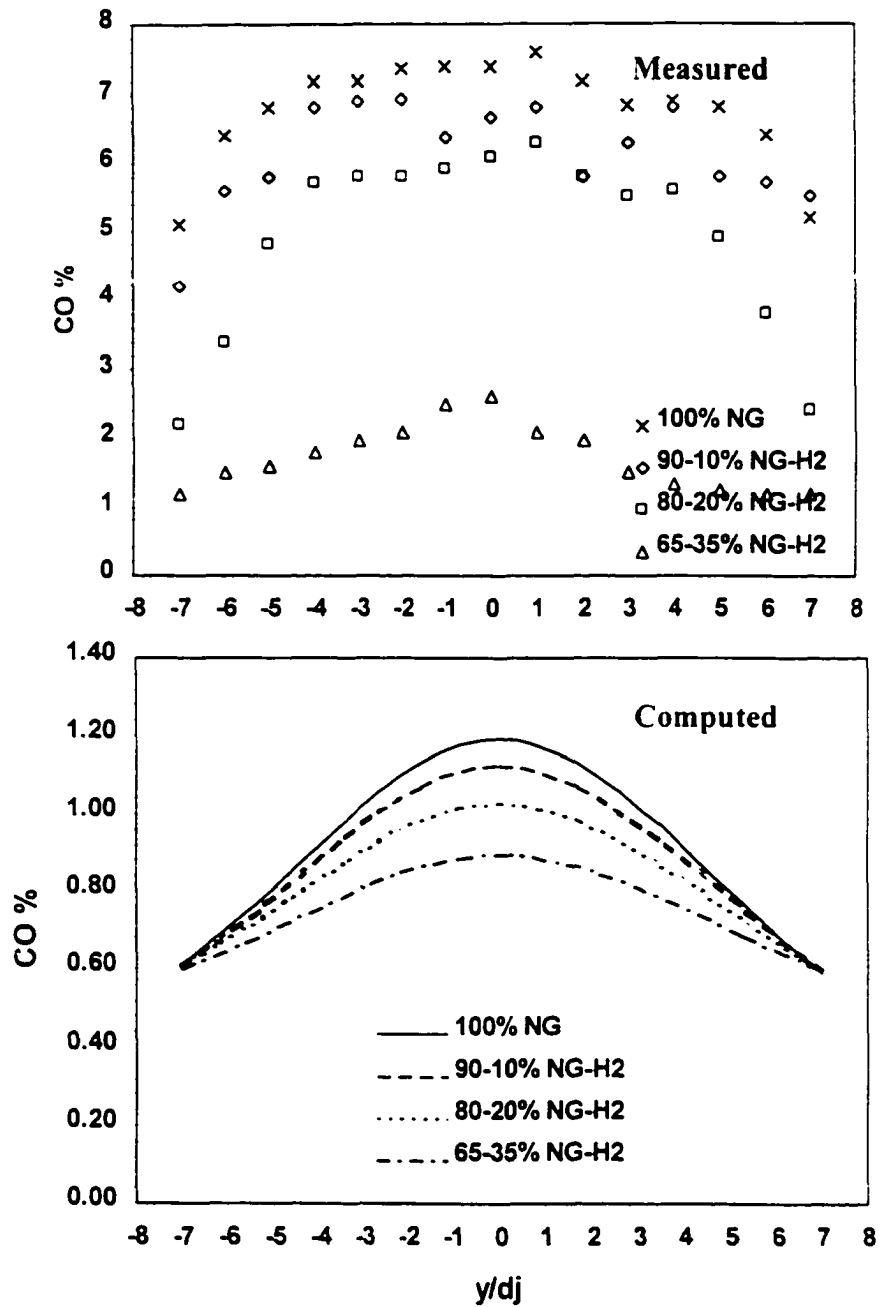


Fig. 6.9 Measured and Computed Radial CO Profiles at a Far-Burner ($0.75 L_f$) Location

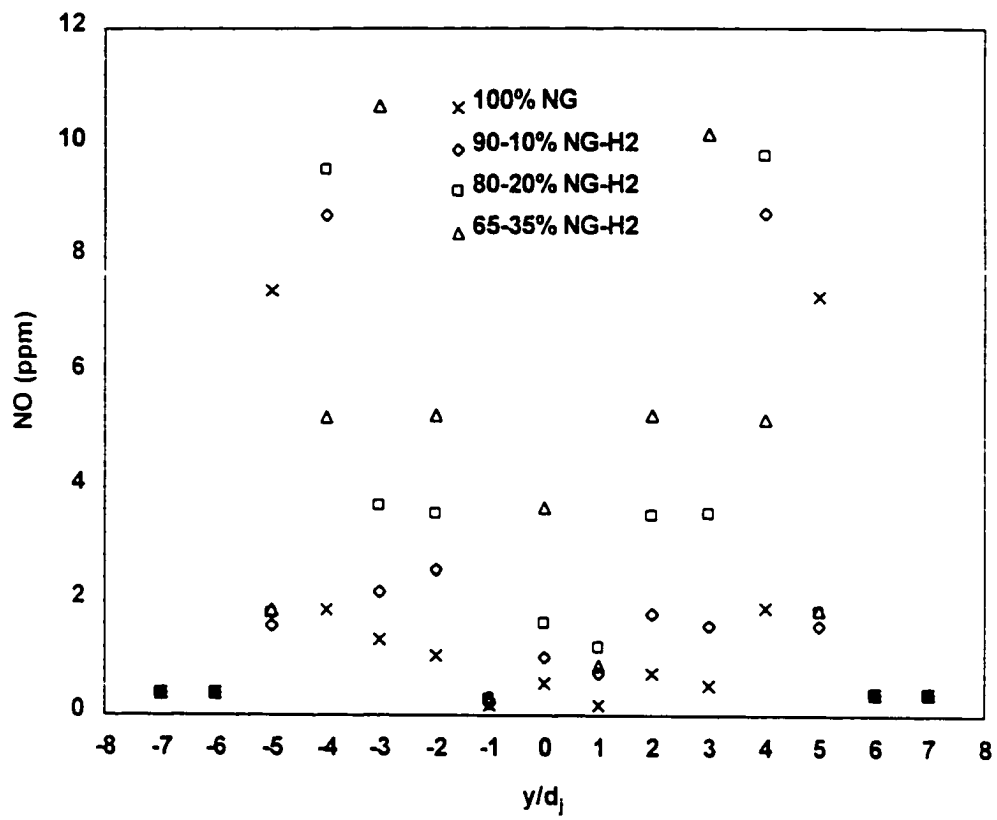


Fig. 6.10 Measured Radial NO Profiles at a Near-Burner ($0.25 L_f$) Location

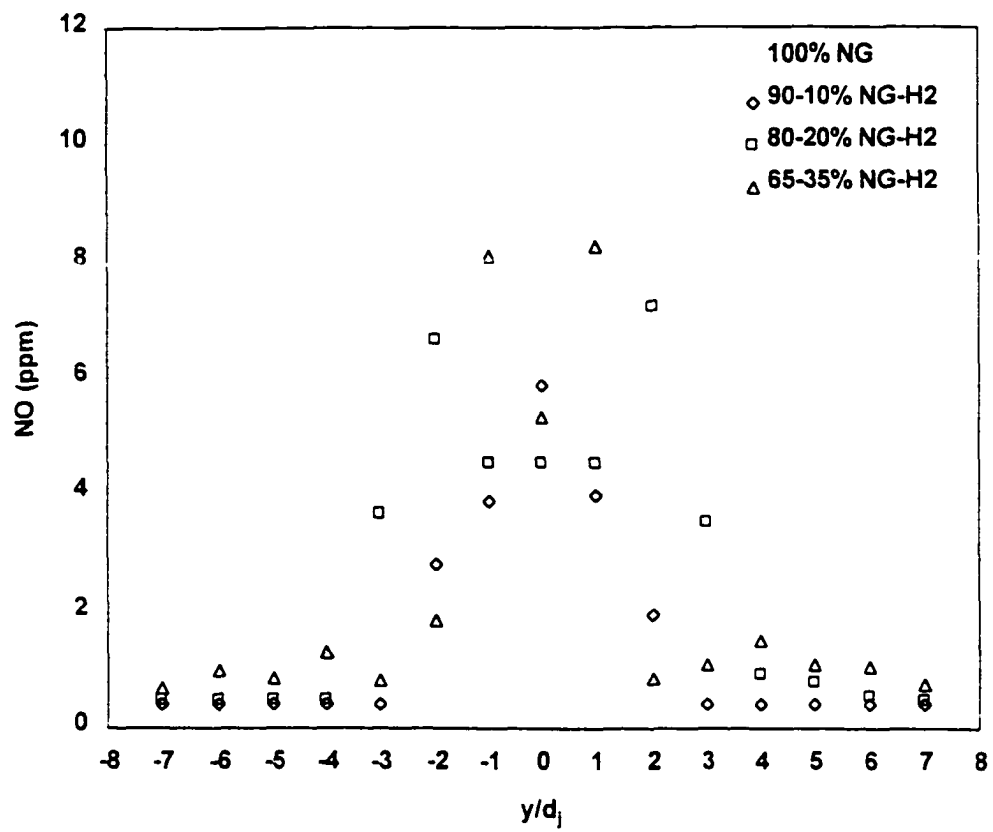


Fig. 6.11 Measured Radial NO Profiles at a Mid-Flame ($0.5 L_r$) Location

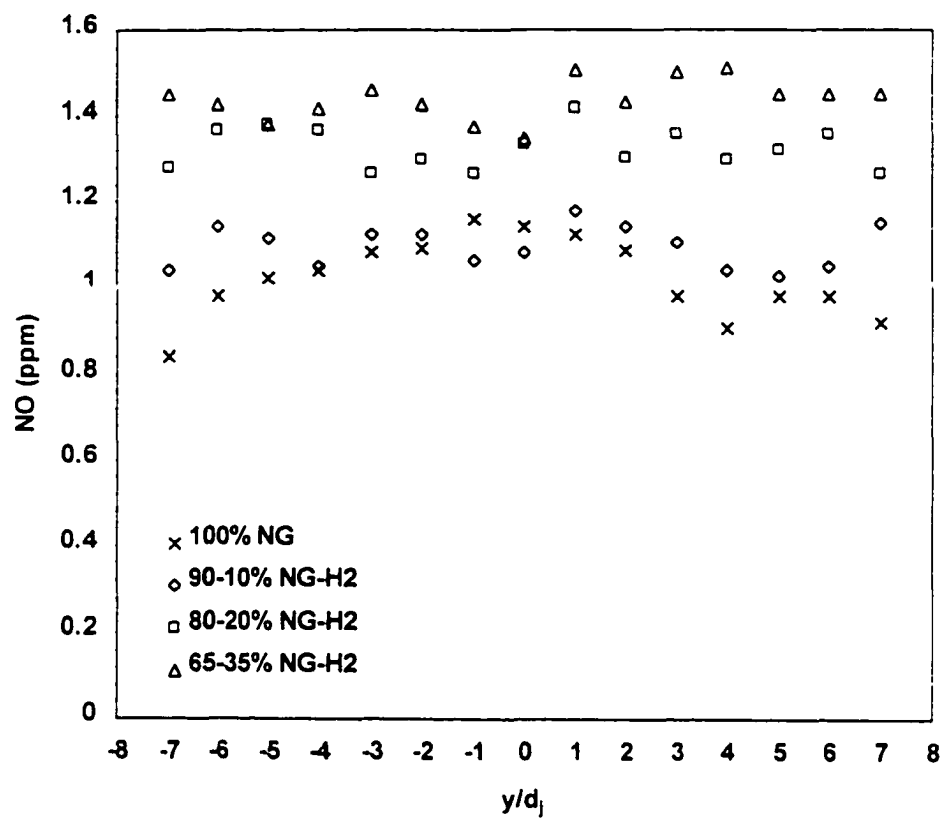


Fig. 6.12 Measured Radial NO Profiles at a Far-Burner ($0.75 L_r$) Location

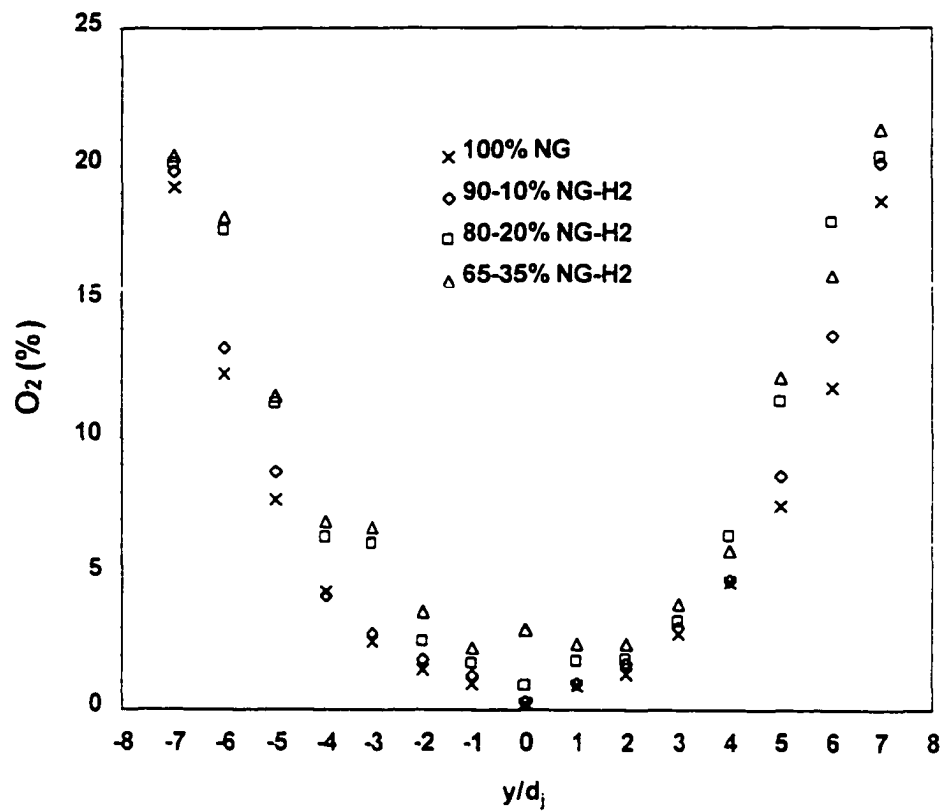


Fig. 6.13 Measured Radial O_2 Profiles at a Near-Burner ($0.25 L_f$) Location

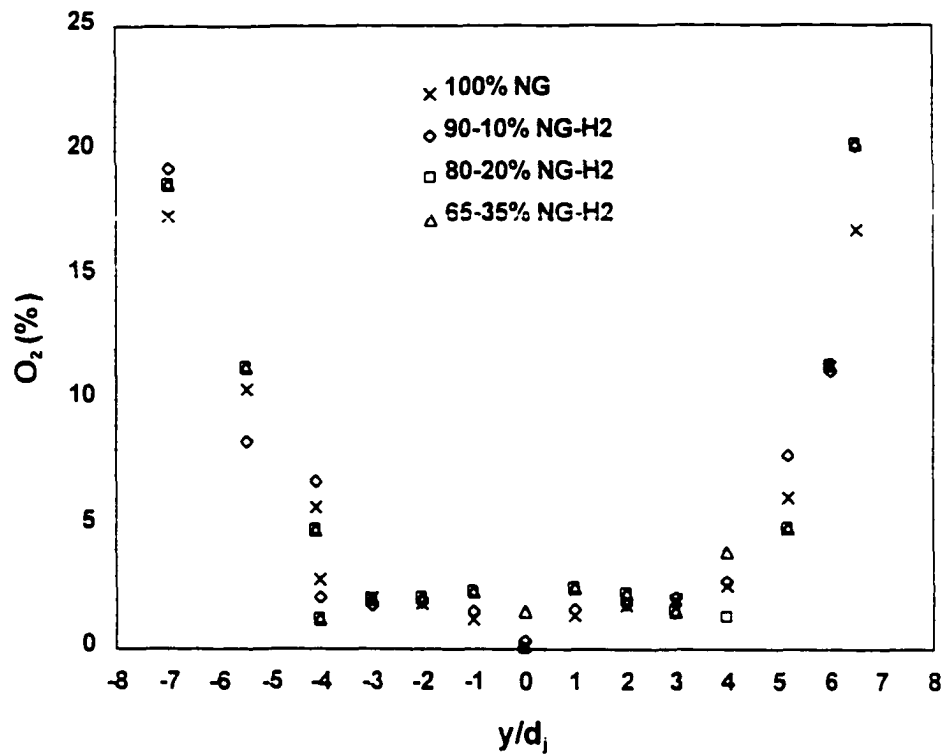


Fig. 6.14 Measured Radial O_2 Profiles at a Mid-Flame ($0.50 L_f$) Location

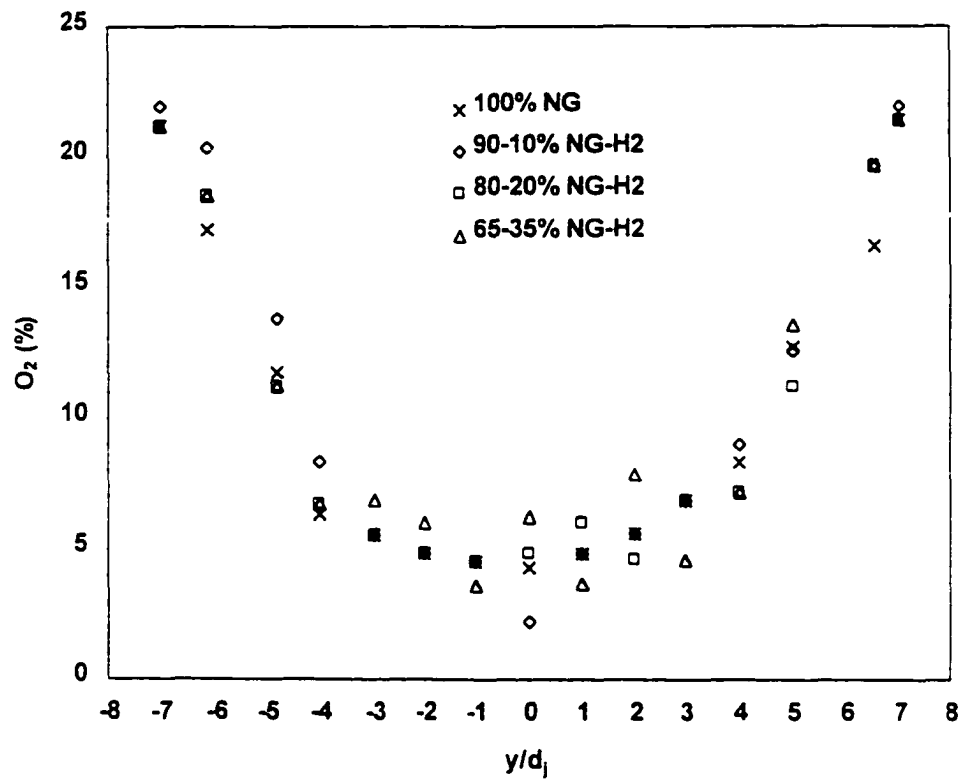


Fig. 6.15 Measured Radial O₂ Profiles at a Far-Burner (0.75 L_f) Location

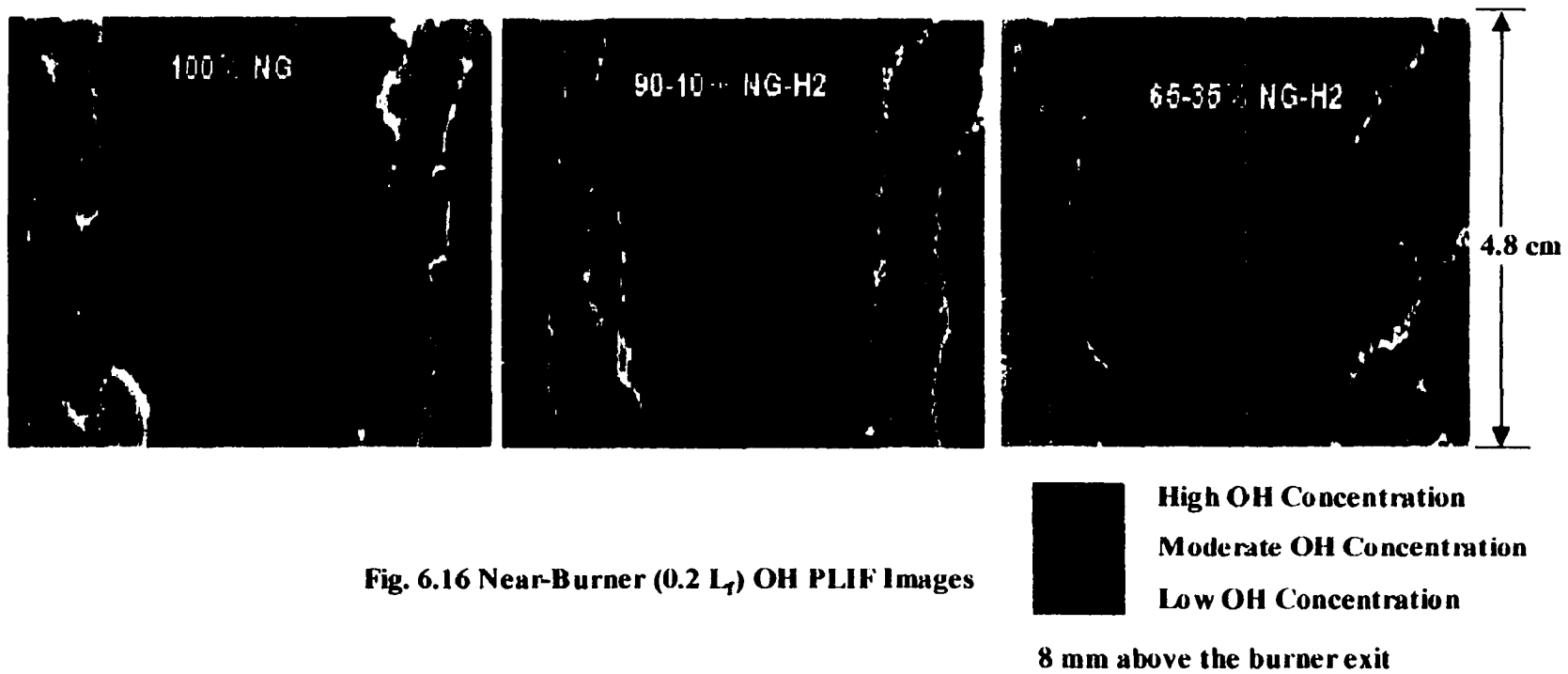


Fig. 6.16 Near-Burner ($0.2 L_f$) OH PLIF Images

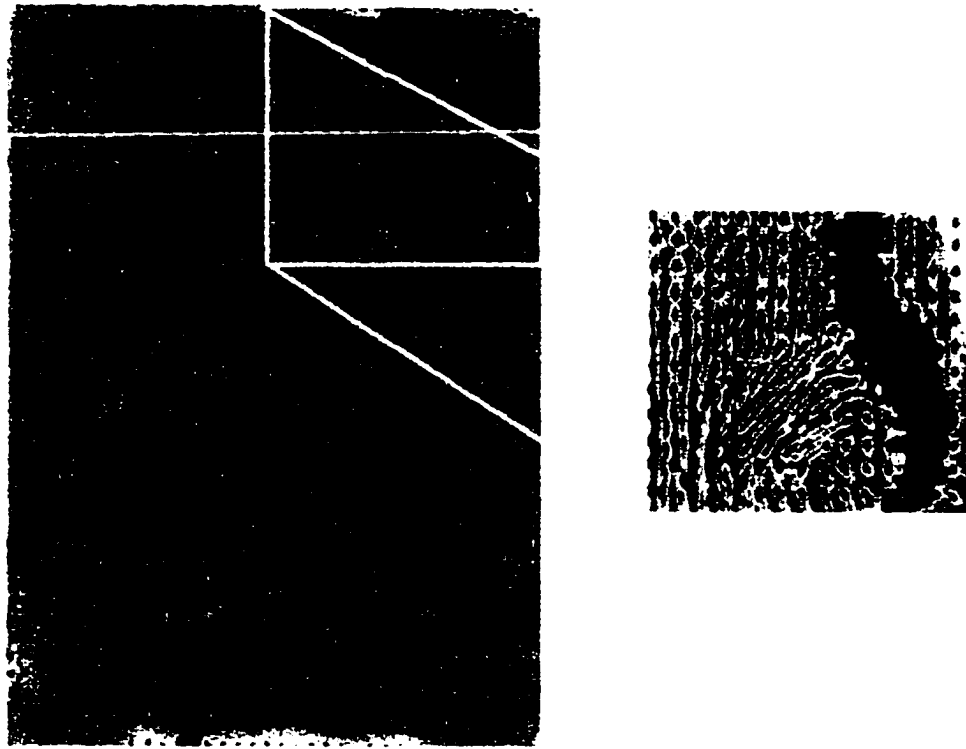


Fig 6.17 Superimposed Instantaneous OH PLIF Image on Averaged Computed Flow Field at a Mid-Flame ($0.25L_f - 0.75L_f$) location for 90-10% NG- H_2 Mixture ($Re_j=8500$)

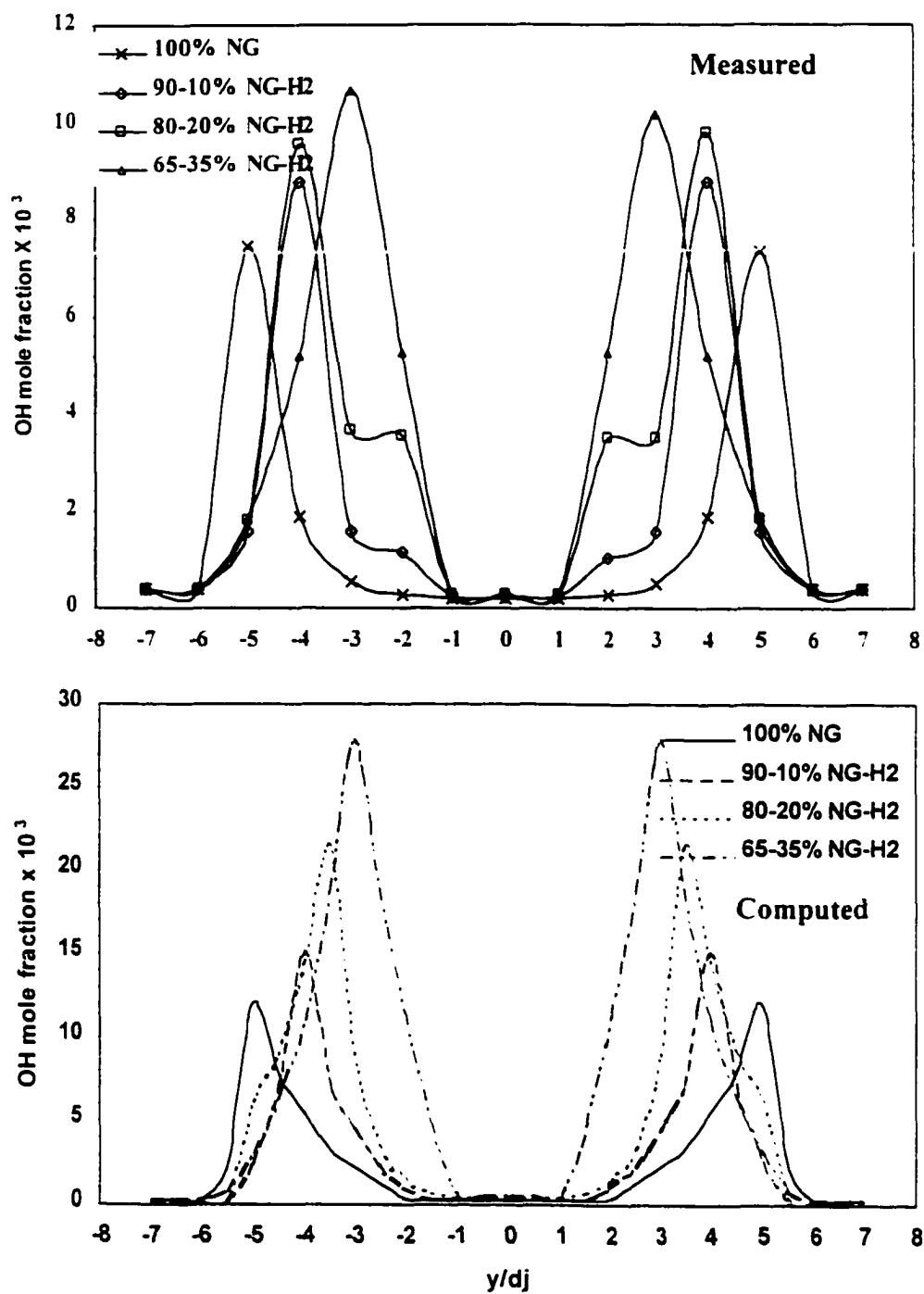


Fig. 6.18 Measured and Computed Radial OH Profiles at a Near-Burner ($0.25 L_f$) Location

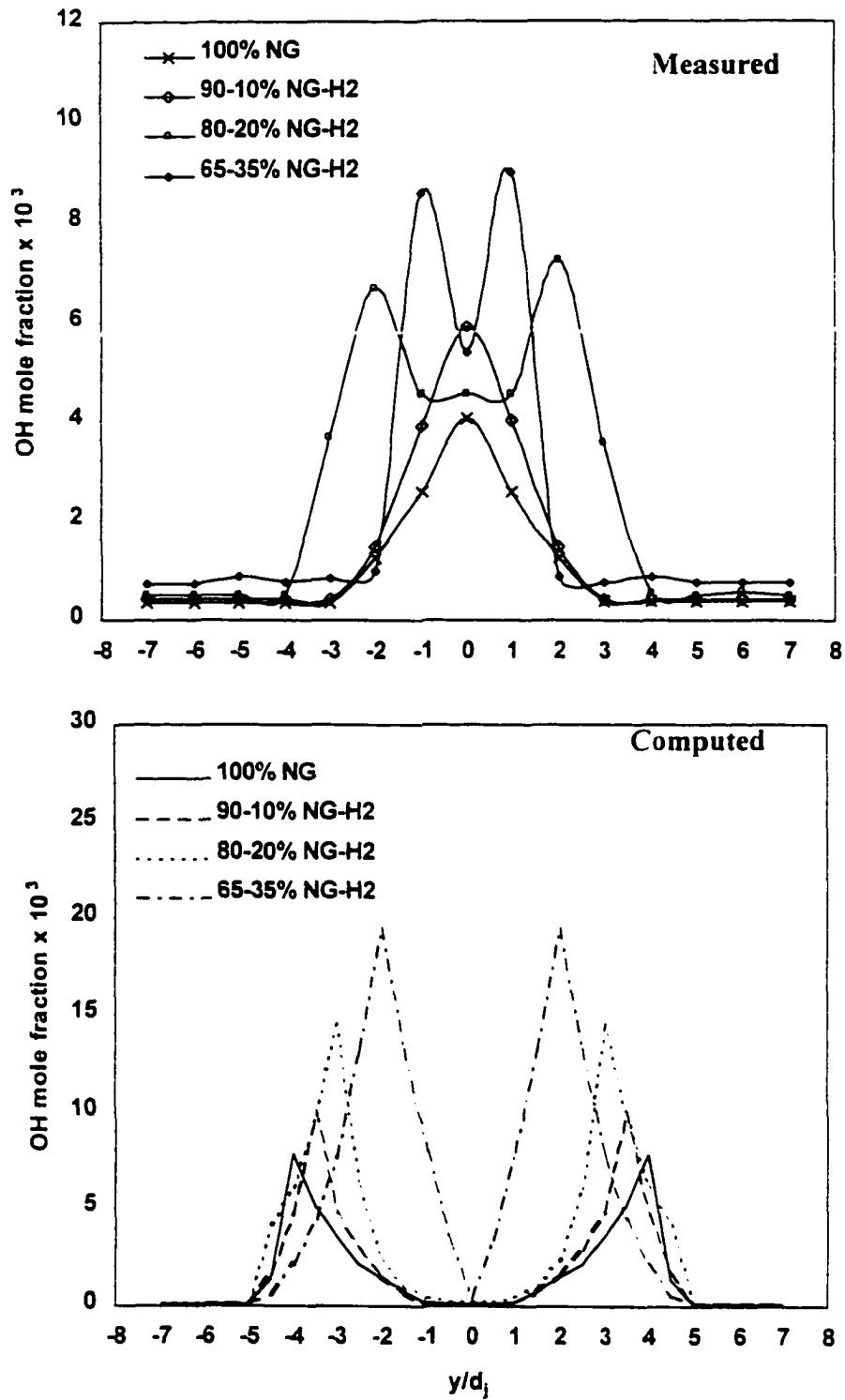


Fig. 6.19 Measured and Computed Radial OH Profiles at a Mid-Flame ($0.50 L_f$) Location

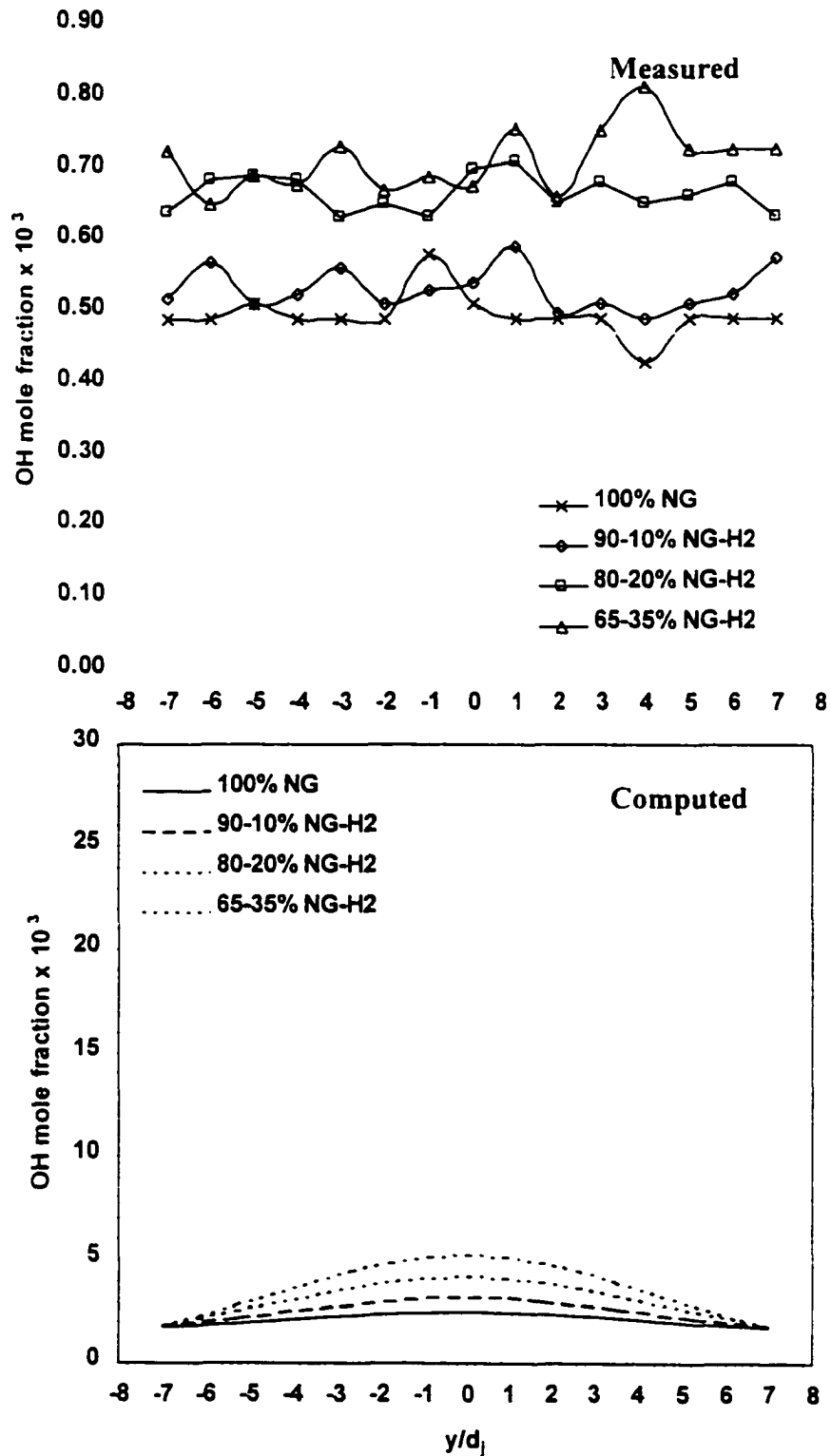


Fig. 6.20 Measured and Computed Radial OH Profiles at a Far-Burner ($0.75 L_f$) Location

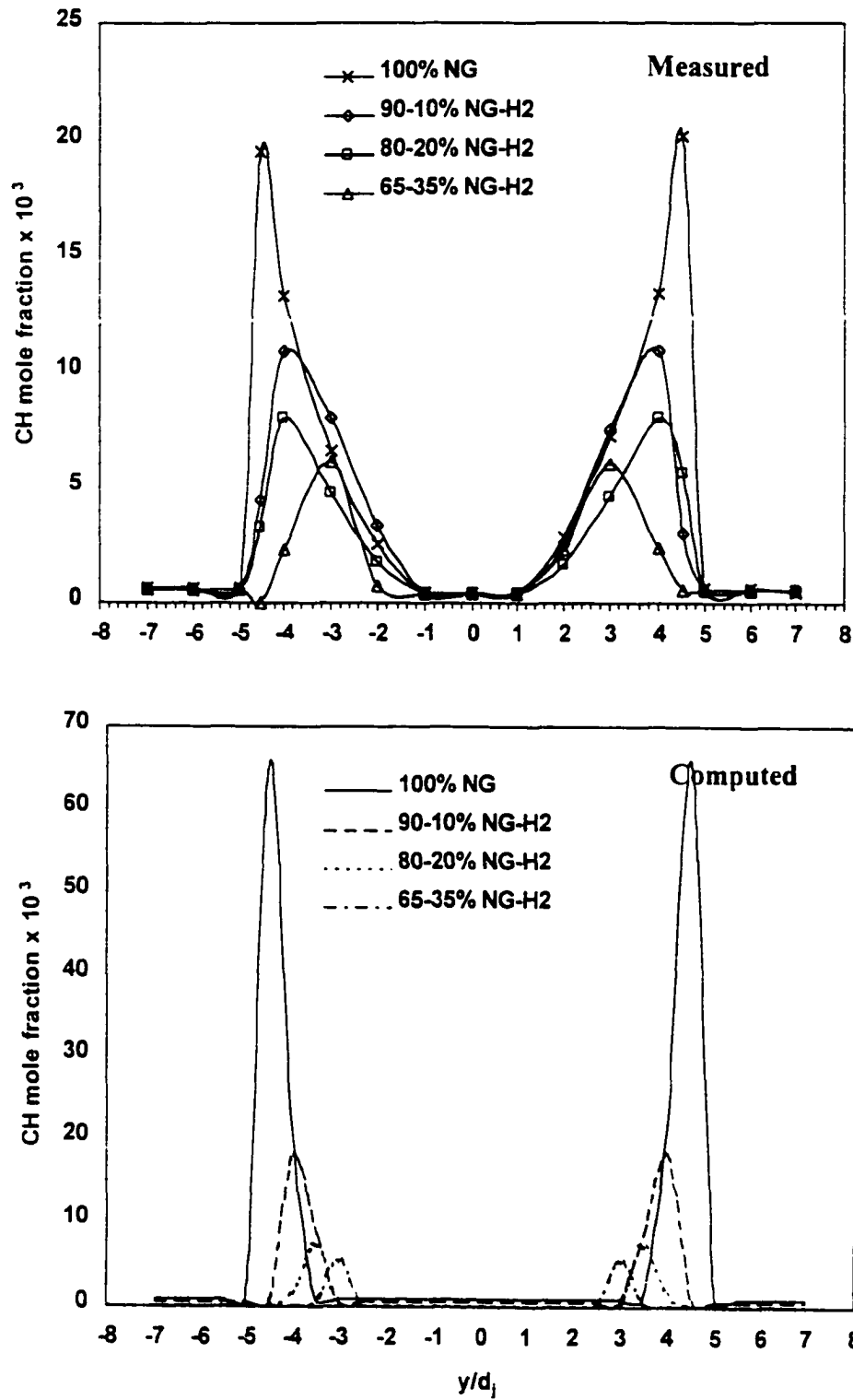


Fig. 6.21 Measured and Computed Radial CH Profiles at a Near-Burner ($0.25 L_f$) Location

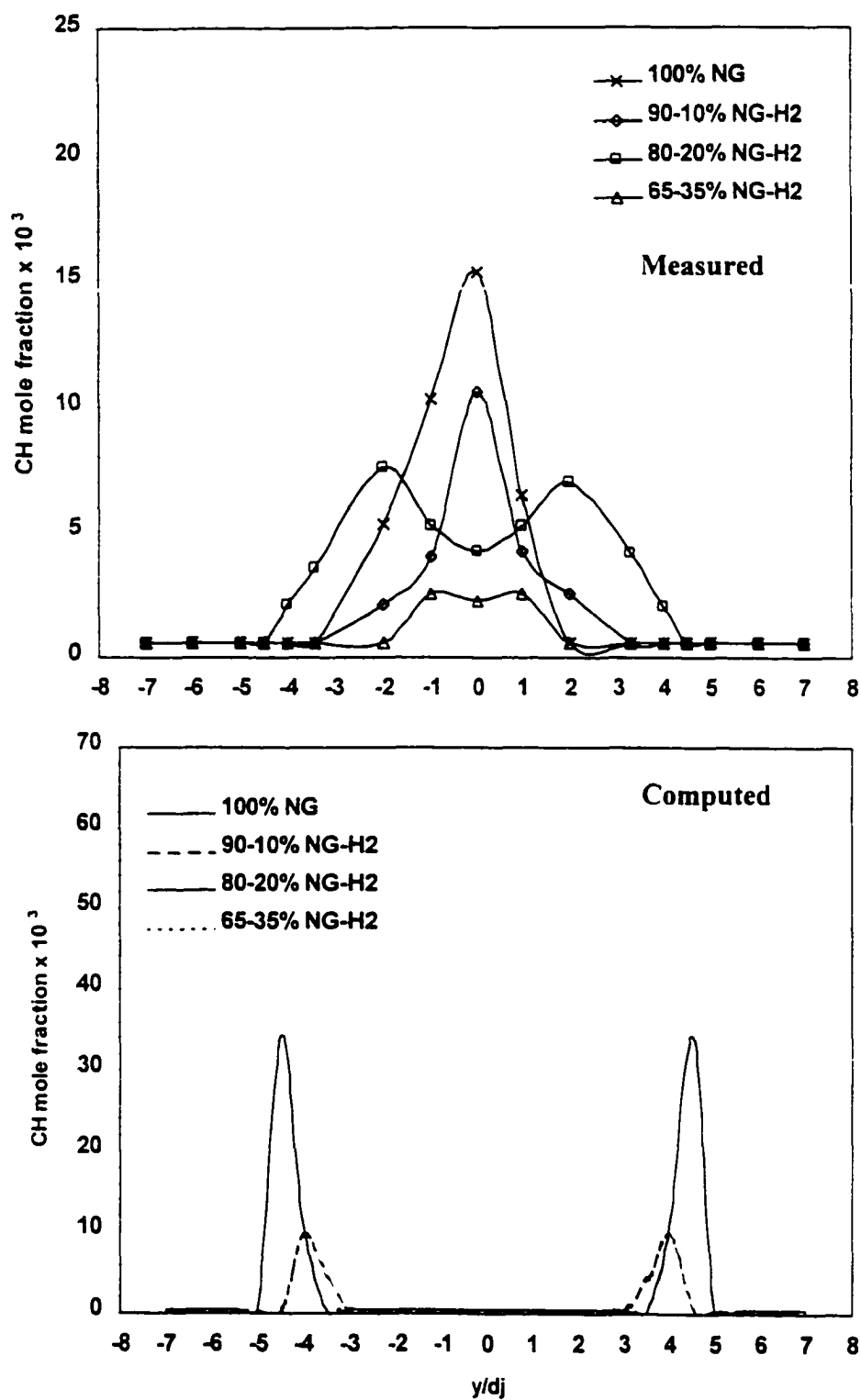


Fig. 6.22 Measured and Computed Radial CH Profiles at a Mid-Flame ($0.50L_f$) Location

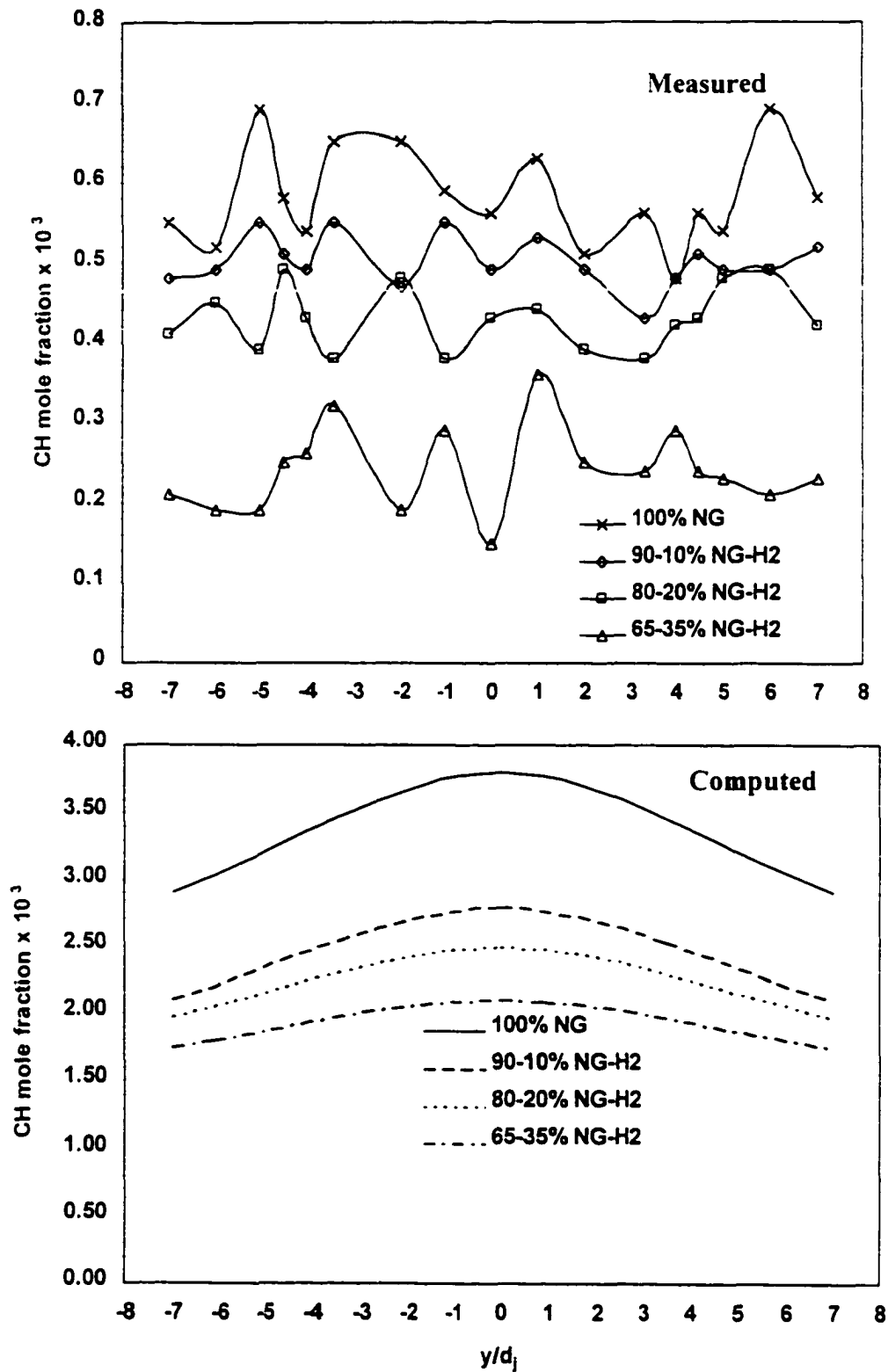


Fig. 6.23 Measured and Computed Radial CH Profiles at a Far-Burner ($0.75L_f$) Location

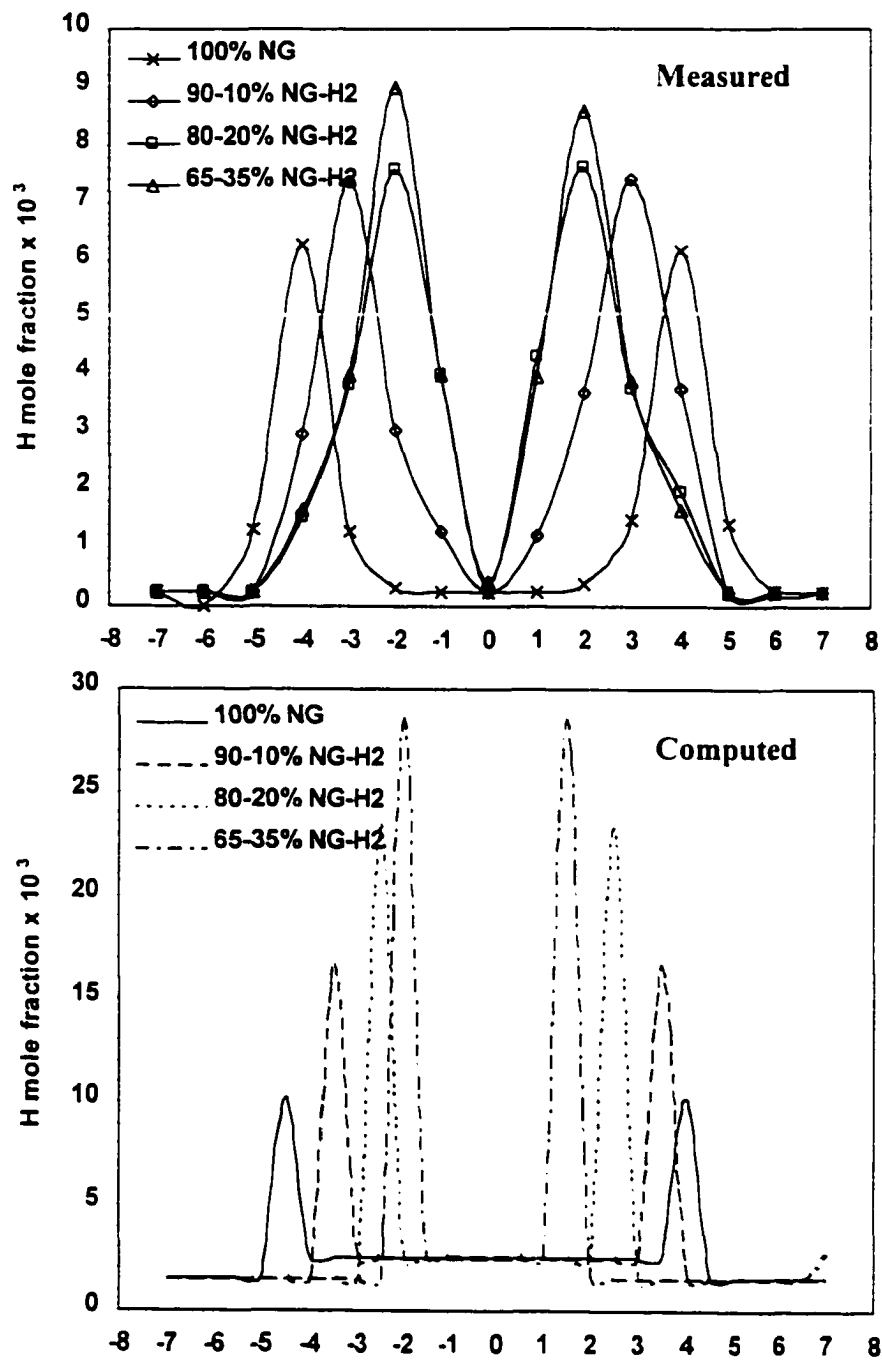


Fig. 6.24 Measured and Computed Radial H Profiles at a Near-Burner ($0.25L_f$) Location

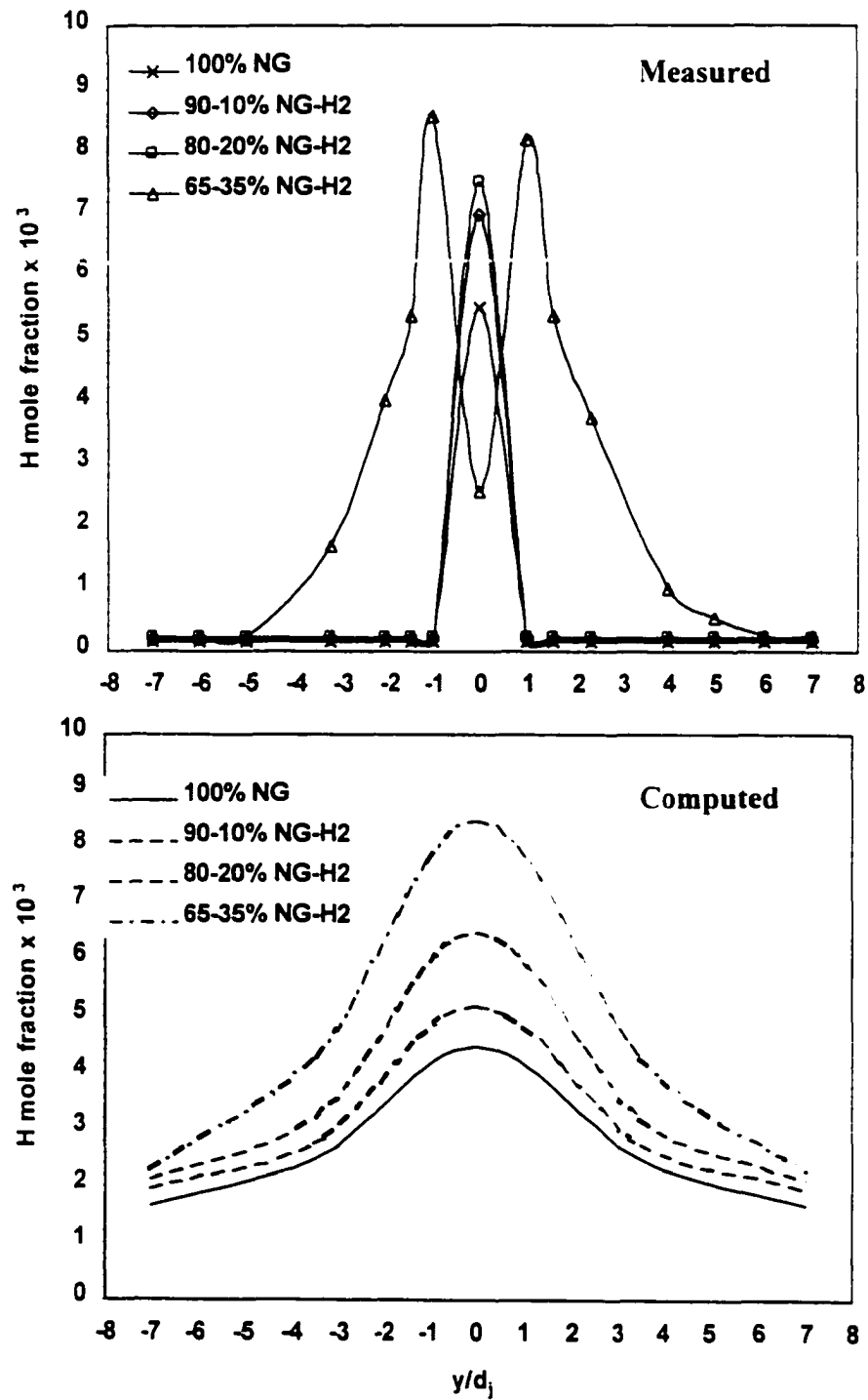


Fig. 6.25 Measured and Computed Radial H Profiles at a Mid-Flame ($0.50L_f$) Location

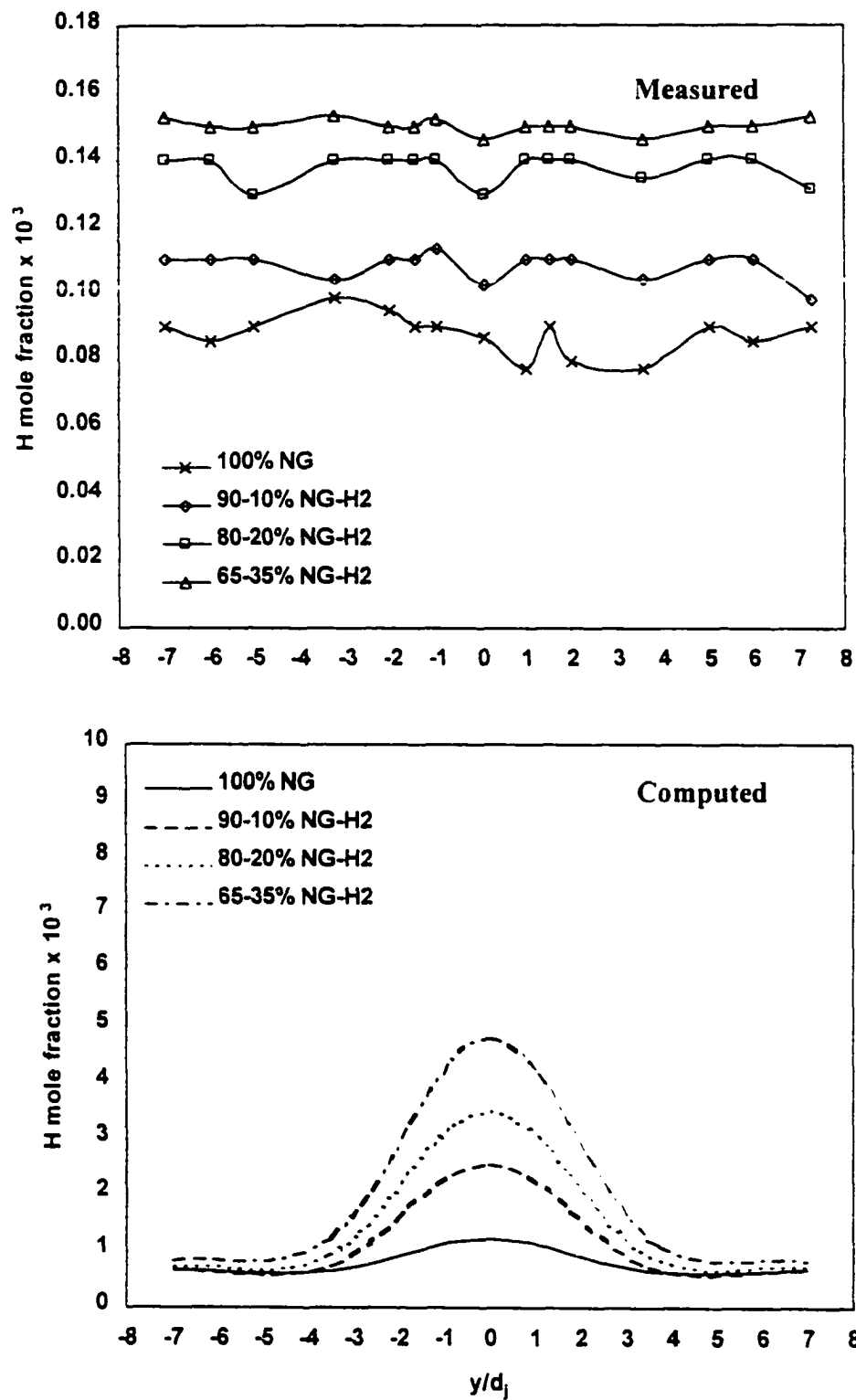


Fig. 6.26 Measured and Computed Radial H Profiles at a Far-Burner ($0.75L_f$) Location

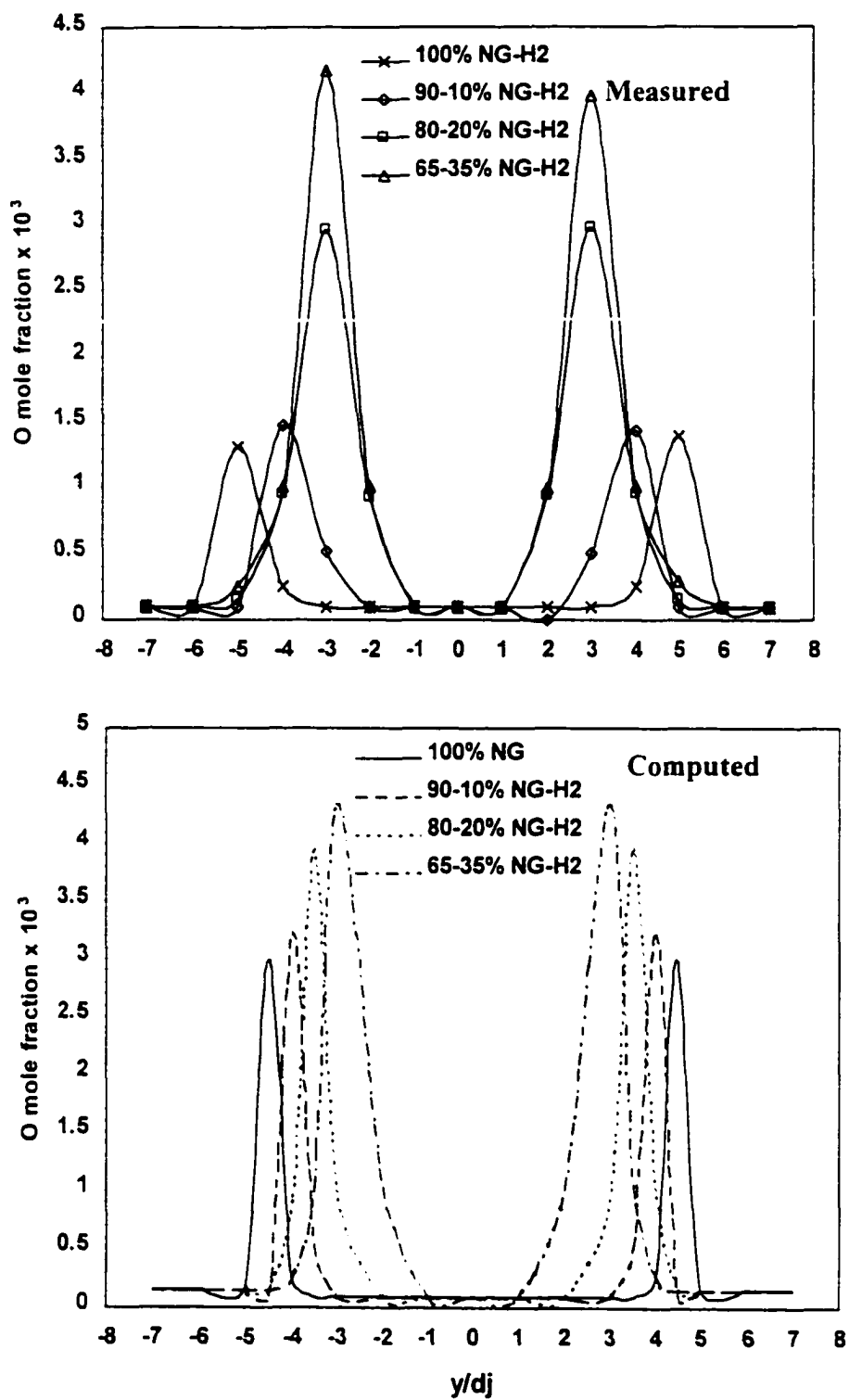


Fig. 6.27 Measured and Computed Radial O Profiles at a Near-Burner ($0.25L_f$) Location

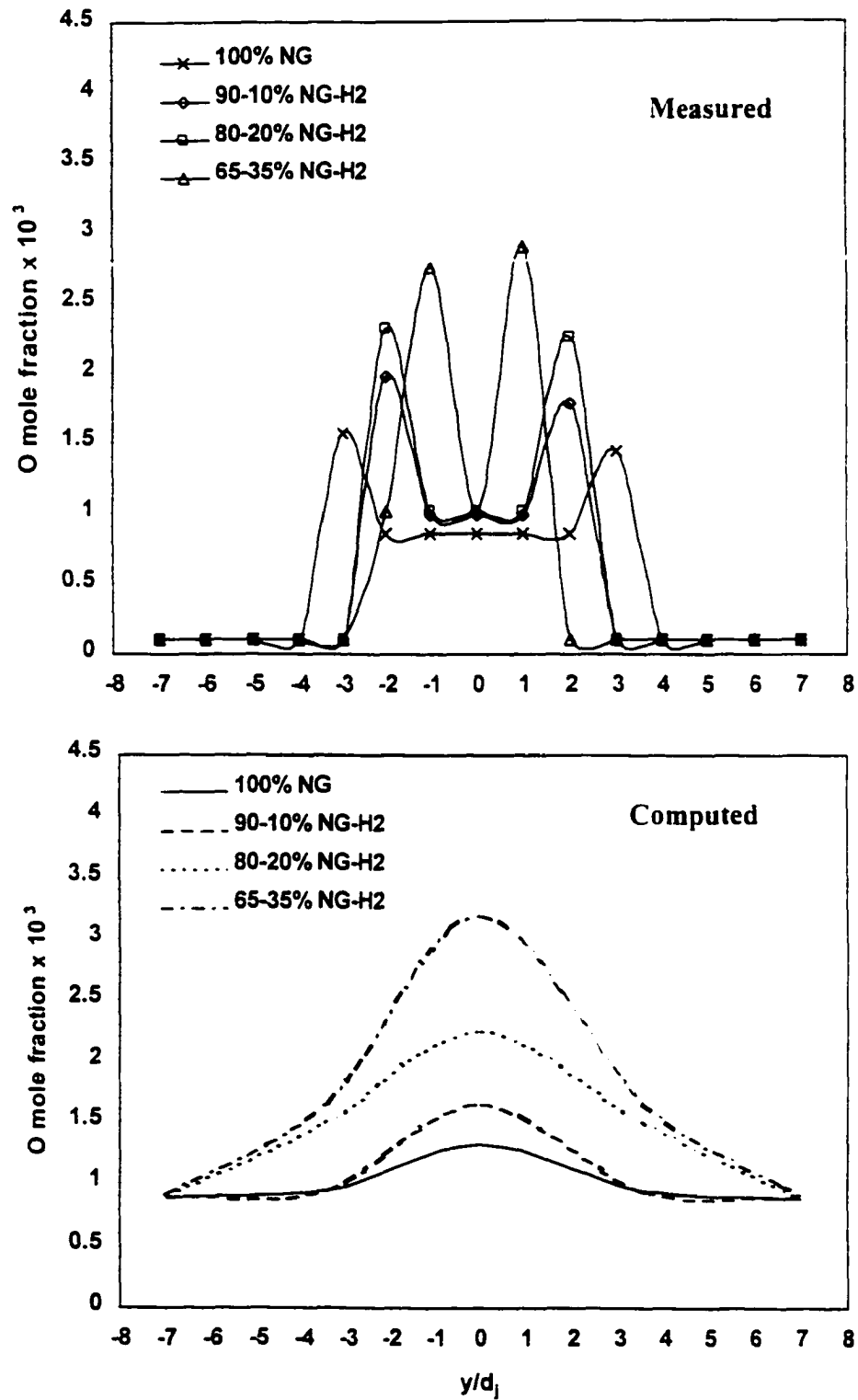


Fig. 6.28 Measured and Computed Radial O Profiles at a Mid-Flame ($0.50L_f$) Location

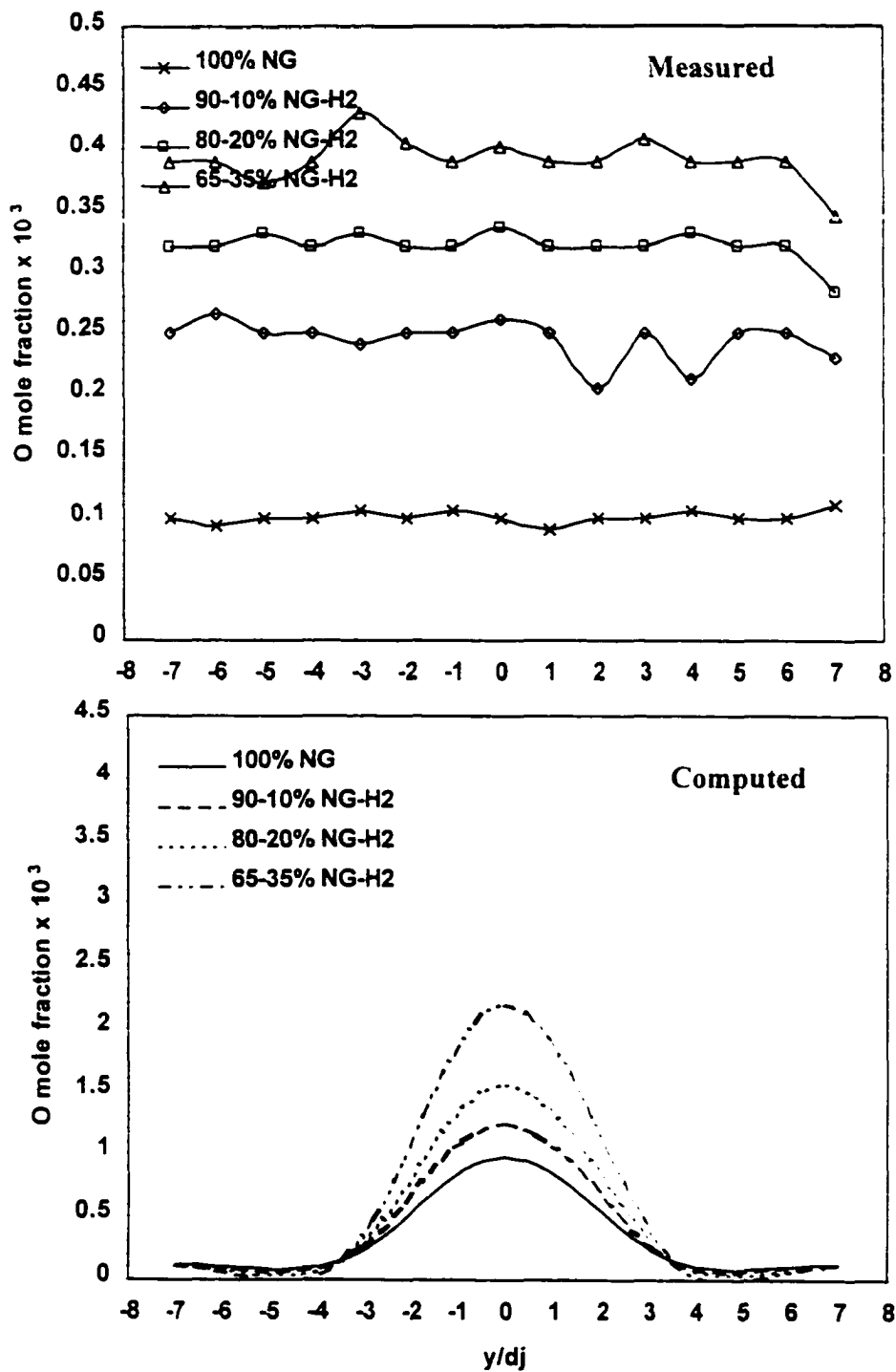


Fig. 6.29 Measured and Computed Radial O Profiles at a Far-Burner ($0.75L_f$) Location

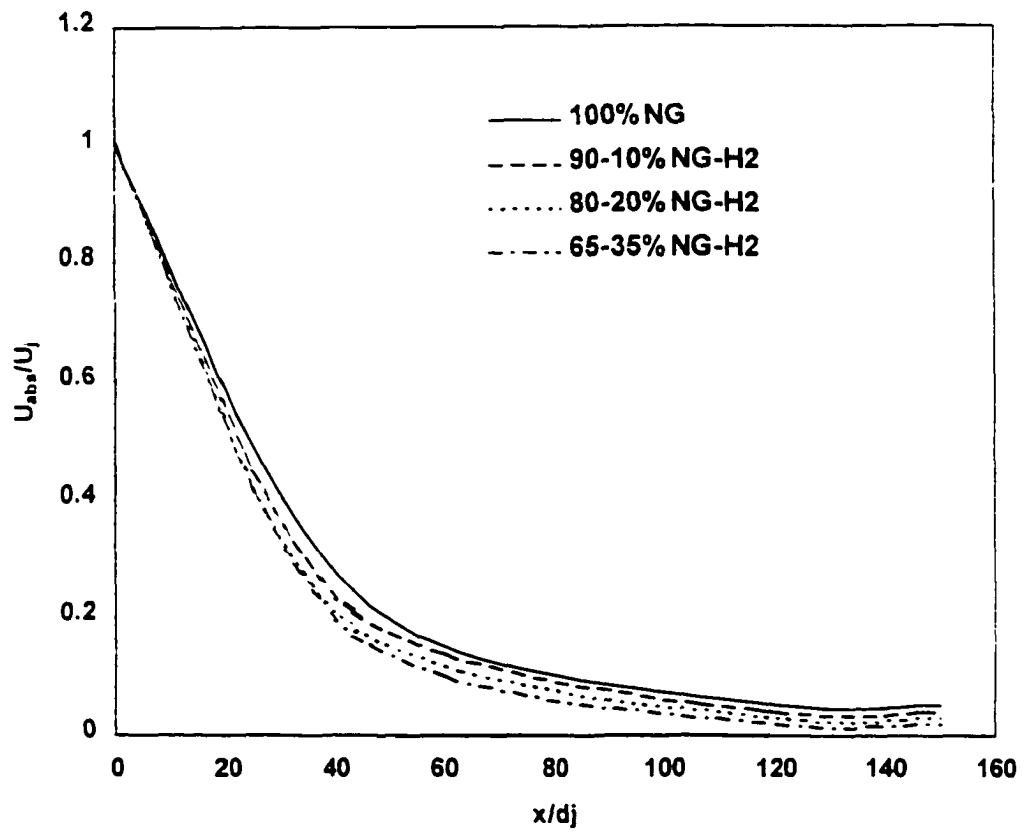


Fig. 6.30 Axial Profiles of Computed Absolute Velocity ($Re_j = 8500$)

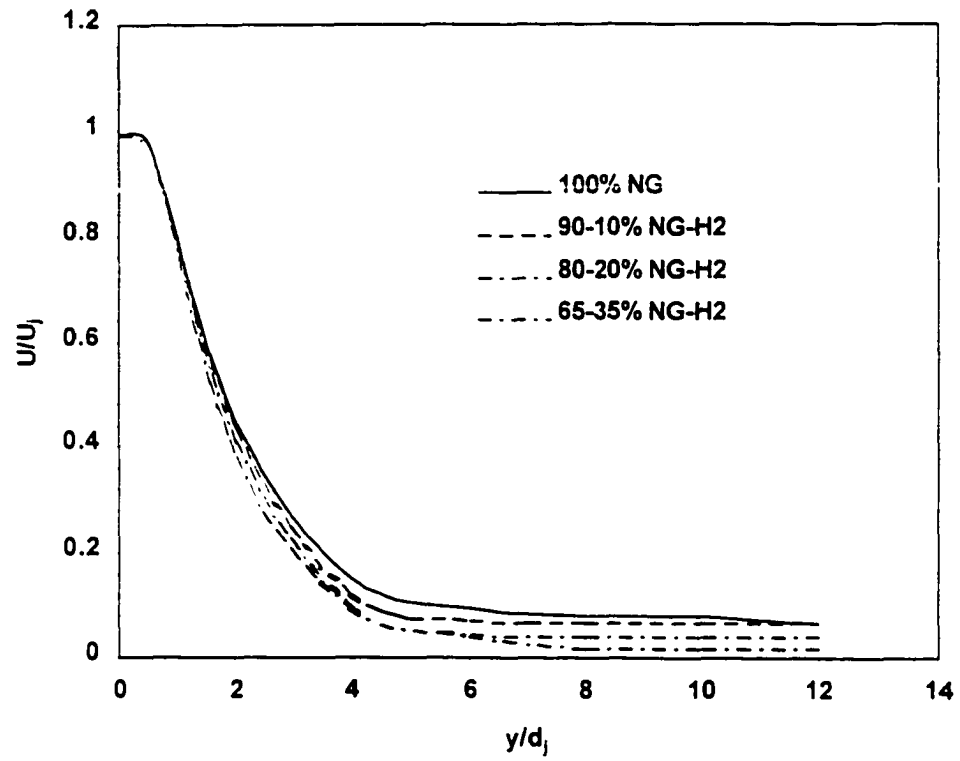


Fig. 6.31 Radial Profiles of Computed Axial Velocity at $x/d_j = 25d_j$ ($Re_j = 8500$)

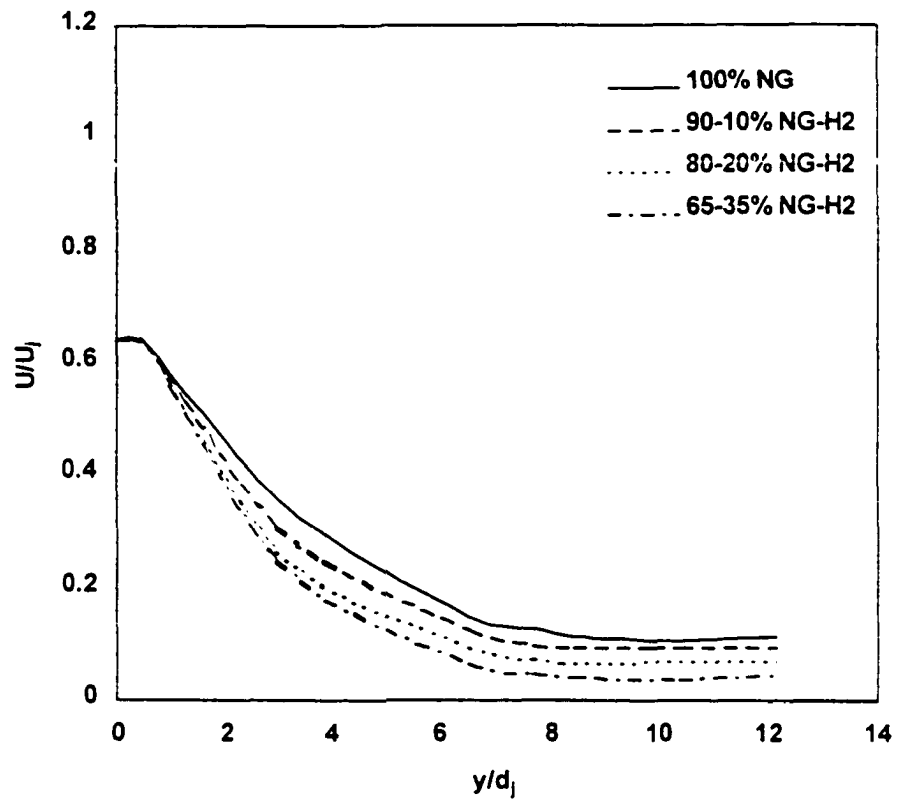


Fig. 6.32 Radial Profiles of Computed Axial Velocity at $x/d_j = 50d_j$ ($Re_j = 8500$)

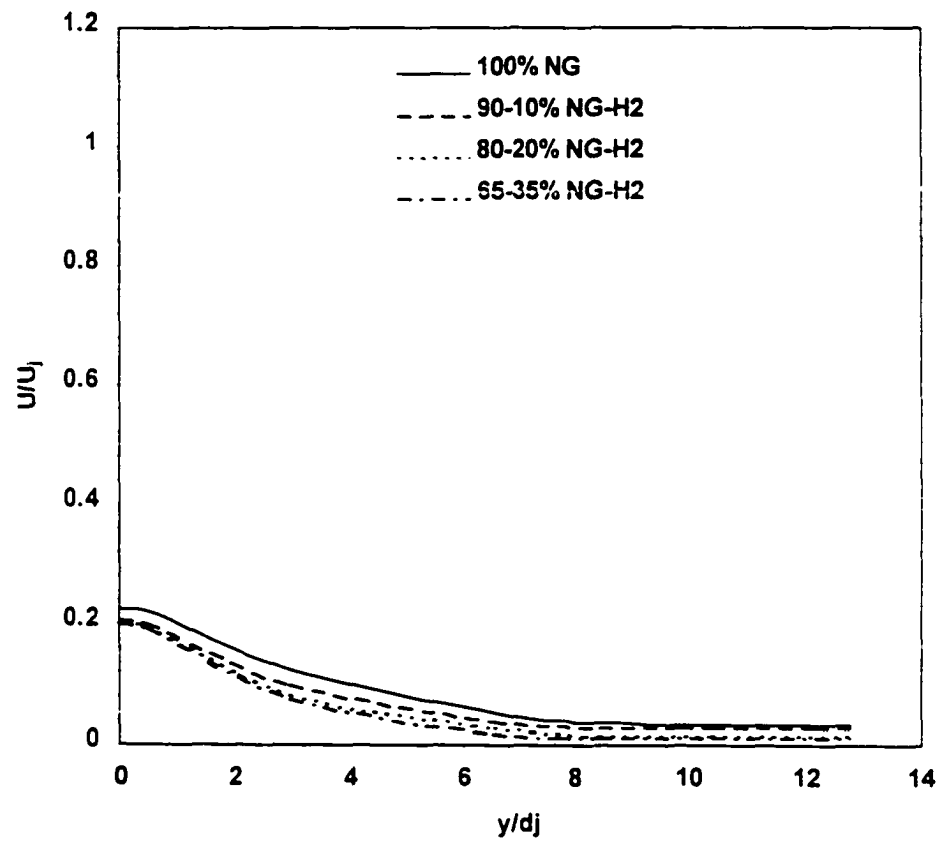


Fig. 6.33 Radial Profiles of Computed Axial Velocity at $x/d_j = 75d_j$ ($Re_j = 8500$)

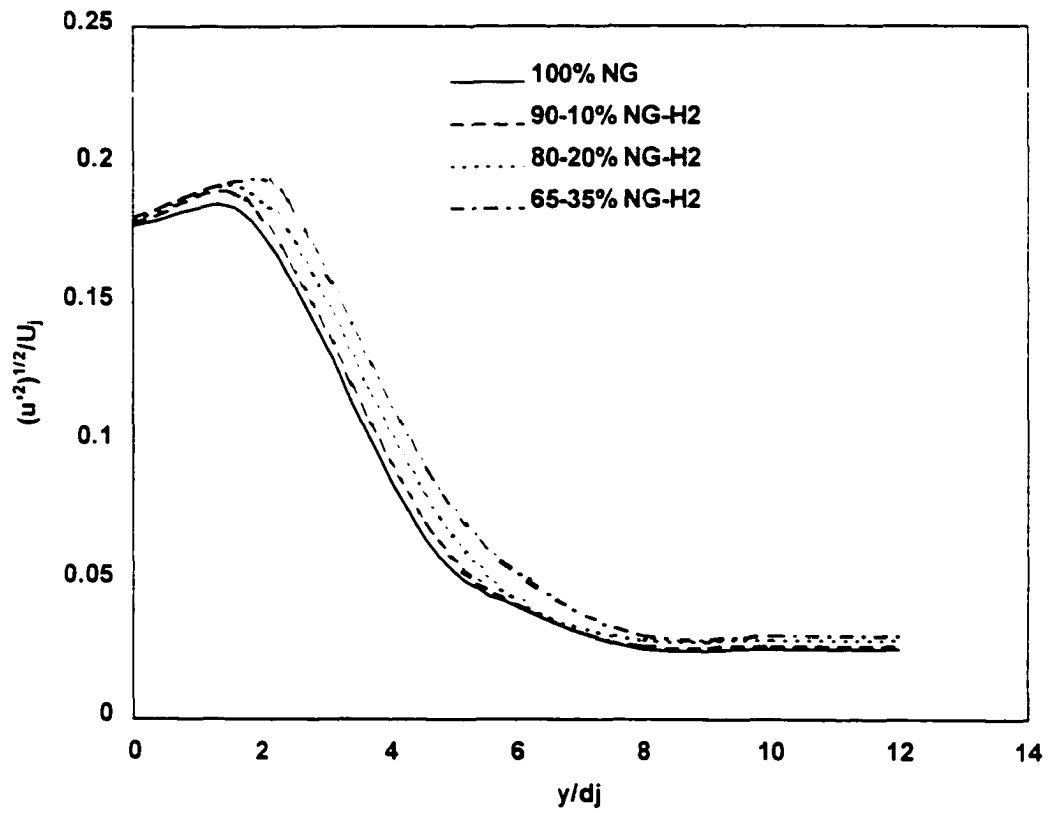
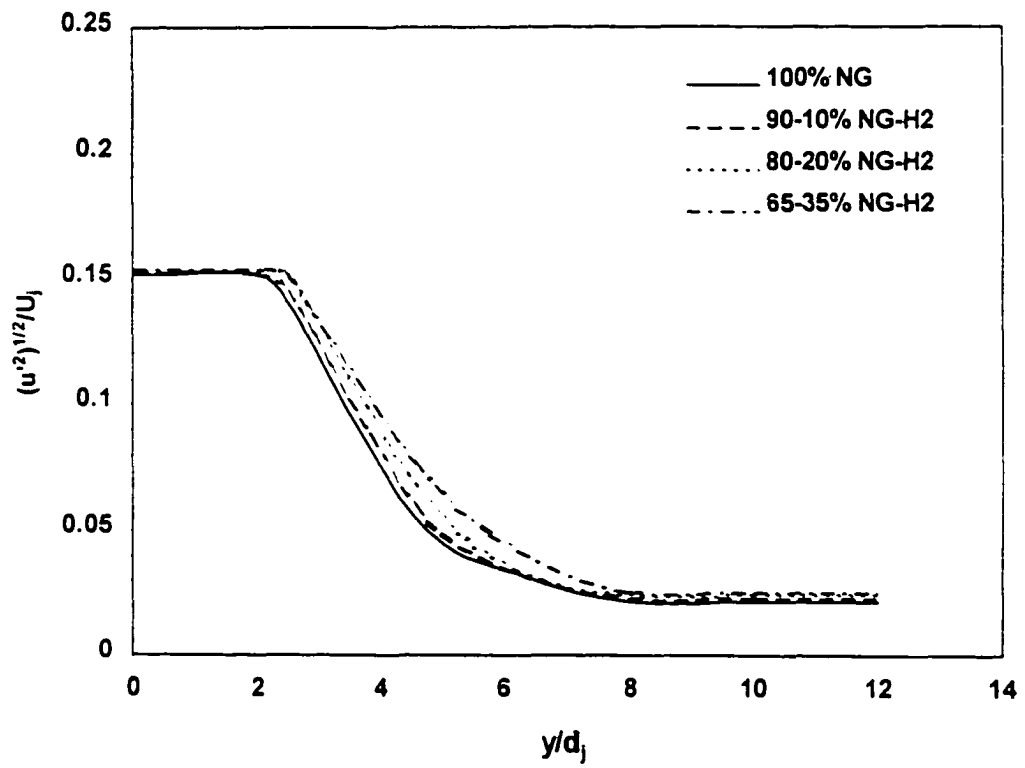
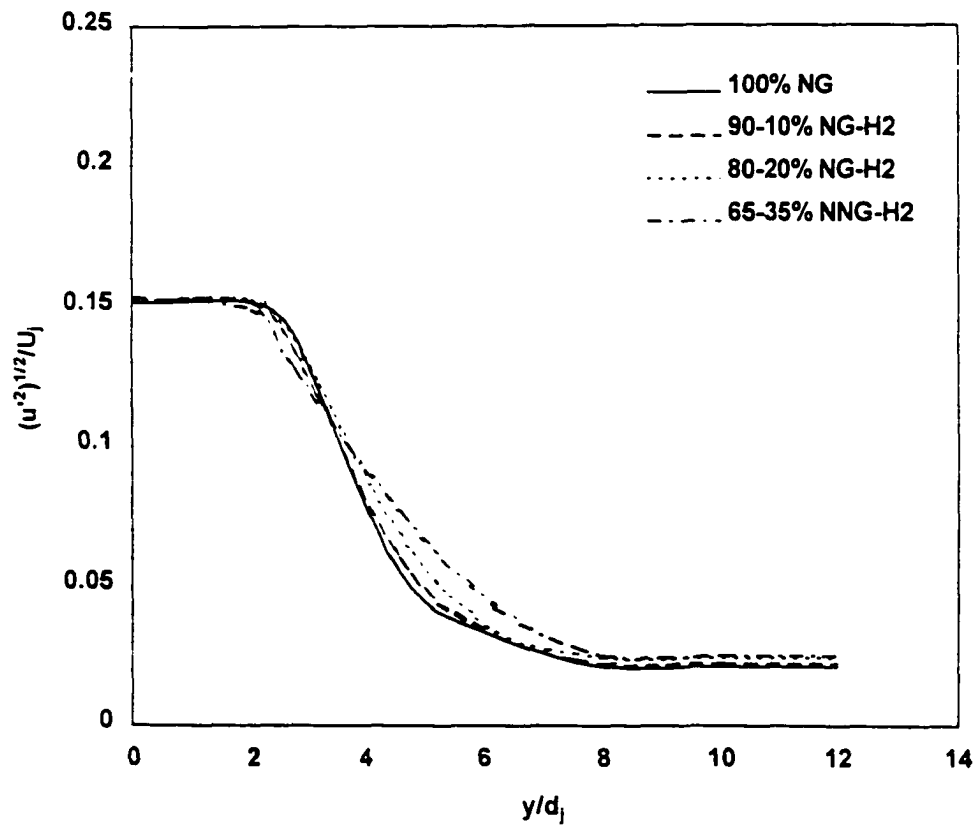


Fig. 6.34 Radial Profiles of Computed Axial Turbulent Fluctuation at $x/d_j = 25d_j$ ($Re_j = 8500$)



**Fig. 6.35 Radial Profiles of Computed Axial Turbulent Fluctuation at $x/d_j = 50d_j$
($Re_j = 8500$)**



**Fig. 6.36 Radial Profiles of Computed Axial Turbulent Fluctuation at $x/d_j = 75d_j$
($Re_j = 8500$)**

Chapter Seven

Global Flame Characteristics

Flame Radiation and Pollutant Emissions

In order to use the hydrogen-hydrocarbon composite fuel effectively in a practical combustion system, the global characteristics such as flame length, flame radiation etc. of the composite fuel flame need to be properly determined. On the other hand, in many cases pollution emissions of the flame is one of the limiting factors for combustion chamber design. In this chapter, the effect of hydrogen addition on flame length, flame radiation and pollutant emissions will be presented. Also, the scaling issues of these global characteristics with jet exit Reynolds number and Froude numbers will be discussed.

As mentioned earlier, information regarding flame length, flame radiation and pollutant emission are highly important for combustion chamber design. Estimation of flame length is required for combustion chamber sizing procedure. Depending upon the fuel used, turbulent diffusion flames can be highly radiating. For certain practical applications such as industrial furnaces, radiant superheaters etc., flame radiation is a desired attribute to contribute in heating loads. On the other hand, in some other applications, radiation losses can contribute to loss efficiency (diesel engine) or safety hazards (e.g. industrial flaring operations). Also, in aero propulsion and terrestrial power generation gas turbine engines excessive flame radiation losses impose a difficult challenge on combustion chamber linings and turbine blade inlet vanes. And, on top of everything, due to present stringent environmental regulations, emission of pollutants such as NO_x and CO is one of the most important considerations that have to be

evaluated prior to combustion system design. This chapter will describe the relation of flame length, radiation heat loss, and NO_x and CO emission indices with hydrogen content in the mixture. A scaling technique will also be presented for general representation of the data.

7.1 TURBULENT FLAME LENGTH

The primary factors which affect the turbulent flame lengths are (i) jet exit momentum force and active buoyant force on flame, Fr_f (ii) flame stoichiometry, $Y_s = [1/(\phi_s + 1)]$ (iii) ratio of air and fuel density, Fr_d , and obviously (iv) exit diameter of the burner, d_j . A global flame Froude number can be defined based on the analysis presented by Delichatsios, 1993 and Bahador, 1993

$$Fr_f = \frac{U_j Y_s^{1.5}}{\left(\frac{T_{ad} - T_x}{T_x} g d_j \right)^{0.5} \xi^{0.25}} \quad [7.1]$$

where, ξ is the density ratio of fuel to air. A condition $Fr_f > 5$ represents the regime of pure momentum-dominated flame. Delichatsios, 1993 proposed a general expression for flame length

$$L^* = \frac{L_f Y_s}{d_j \xi^{0.5}} \quad [7.2]$$

Also, it was shown that for $Fr_f < 5$ i.e., for buoyancy-dominated region the relation of equivalent flame length and Flame Froude number is

$$L^* = \frac{13.5 Fr_f^{\frac{1}{2}}}{(1 + 0.07 Fr_f^2)^{\frac{1}{4}}} \quad [7.3]$$

and for momentum dominated regime i.e. $Fr_f > 5$ the equivalent flame length L^* is constant and has value of 23. With the visible flame length, a global flame residence time (T_G) can be defined based on the argument presented in Turns and Myhr, 1991.

$$T_G = \frac{\rho_f W_f^2 L_f Y_s}{\rho_F d_j^2 U_j} \quad [7.4]$$

where ρ_f is the density of the flame gases and it can be approximated as the density at flame equilibrium temperature. Sample calculations associated with Froude Numbers are presented in Appendix VI and VII.

7.2 SCALING PARAMETERS FOR RADIATION HEAT LOSSES

Radiant heat losses from hydrocarbon flame primarily occur through two sources: First due to the molecular sources i.e., infrared spectrum broadband radiation from CO_2 and H_2O , and second, continuum radiation from soot particles at short wavelengths. For a methane diffusion flame the wavelength ranges associated with molecular radiation and continuum radiation are 2.5-4.5 μm and 1.2-1.5 μm respectively. So, the total radiation from the flame can be written as the summation of radiation from soot particles and radiation from gas molecules, i.e.,

$$Q_R = q_s + q_g$$

where q_s and q_g are radiation from soot particles and gas molecules respectively. Although, the radiation from soot particles can be modeled in a more complex, for

scaling purposes a simple model can be conceived by considering the whole flame as a total source.

$$Q_R = \epsilon \sigma V_f (T_f^4 - T_x^4) \quad [7.5]$$

where V_f is flame volume (approximated as $0.0027 L_f^3$). T_f is the apparent flame temperature based on flame radiation. ϵ is the flame emissivity, and σ is the Boltzman constant ($5.67051 \times 10^{-8} \text{ W/m}^2\text{-K}^4$). The primary problem of the simplified analysis is to assign a proper value of ϵ . For the present investigation a constant value of 0.55 was used. Although this approach neglects the wavelength and temperature dependence of ϵ , the approximation is fairly reasonable because it is an average value determined from an actual flame measurement. Now if a wide-angle radiometer located at distance R (half of the visible flame length) from the flame axis and measuring a radiant heat flux of q_R is measured then the radiant heat loss can be alternatively modeled as

$$\dot{Q}_R = q_R^* (4\pi R^2) \quad [7.6]$$

The total heat release by combustion of fuel can be expressed as

$$\dot{Q}_T = \dot{m}_F \Delta H_c \quad [7.7]$$

where $\dot{m}_F$ is the fuel mass flow rate and ΔH_c is the enthalpy of combustion of this fuel.

For hydrogen-hydrocarbon composite fuel the above expression can be rewritten as

$$\dot{Q}_T = (\dot{m}_{HC} \Delta H_{cHC}) + (\dot{m}_{H_2} \Delta H_{cH_2}) \quad [7.8]$$

Hence, the radiative heat release factor can be defined as the ratio of radiative heat loss rate to the total heat released rate by the fuels.

$$F = \frac{\dot{Q}_R}{\dot{Q}_T} \quad [7.9]$$

If the flame volume is assumed to be proportional to the cube of the burner diameter i.e., d_b^3 (Turns and Myhr, 1991) and the fuel flow rate to $d_j^3 U_j$, then the radiative heat loss fraction can be scaled as

$$F \approx \epsilon T_f^4 \frac{d_j}{U_j} \quad [7.10]$$

Hence, the scaling analysis shows that the radiation heat loss fraction decreases and mean flame temperature increases as the convective timescale d_j/U_j decreases. Furthermore, the radiation heat loss fraction can be related to the jet exit Reynolds number ($Re_j = \rho U_j d_j / \mu$) and the jet exit Froude Number ($Fr_j = U_j^2 g / d_j$) in the following fashion

$$F \approx \epsilon \sigma T_f^4 (\nu Re_j)^{0.33} (g Fr_j)^{-0.75} \quad [7.11]$$

This expression can be further written as a function of densimetric Froude number

$$F \approx \epsilon \sigma T_f^4 (\nu Re_j)^{0.33} (\xi g Fr_d - g Fr_d)^{-0.75} \quad [7.12]$$

where ξ is the ratio of fuel and air density.

7.3 EMISSION INDICES

The emission index (EI) of pollutant species, defined as the mass of the species emitted per unit mass of the fuel burned, was calculated by using the expression from Gollahalli (1994) and Turns (1996). This index is the most important parameter to determine the emission potential of any specific fuel. The *emission index* for a species 'i' is the ratio of the mass of species 'i' to the mass of fuel burned by the combustion process (Turns, 1996).

$$EI_i = \frac{m_{i, emitted}}{m_{F, burned}} \quad [7.13]$$

In principle, the emission index is a dimensionless quantity. However, units such as g/kg are used to avoid working with very small numbers. For the combustion of hydrocarbon fuels in air, the emission index can be determined from concentration (mole fraction) measurements of the species of interest, together with those of all the C-containing species. Assuming all of the fuel carbon appears either as CO₂ or CO, the emission index is expressed

$$EI_i = \left(\frac{\chi_i}{\chi_{CO} + \chi_{CO_2}} \right) \left(\frac{x M_i}{M_F} \right) \quad [7.14]$$

Physically, the first bracketed term in the above equation represents the number of moles of *i* per mole of carbon originating in the fuel, while the other term provides the necessary conversion to mass units. It is obvious that the measurement of emission index should be independent of any dilution by air. Since all of the measured concentrations appear as ratios, the effect of the diluent is canceled. Nitric oxide is the primary oxide of nitrogen produced and emitted from most practical combustors. In combustion systems, nitric oxide is formed by three chemical mechanisms (Bowman, 1992)

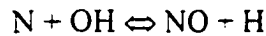
- (i) Oxidation of atmospheric (molecular) nitrogen via the *Thermal-Zeldovich* mechanism.
- (ii) *Prompt NO* mechanism.
- (iii) Oxidation of nitrogen containing organic compounds in fossil fuels via the *Fuel NO* mechanism.

The *Thermal-Zeldovich* mechanism consists of two chain reactions

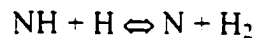
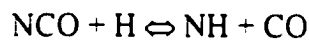
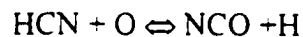
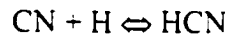




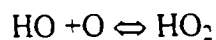
This reaction mechanism can be extended by adding the reaction



This three-reaction set is referred to as the *Extended Zeldovich* mechanism. The calculated rate parameters for these reactions clearly reveal that NO formation through this route is strongly dependent on burned gas temperature and somewhat weakly dependent on the O₂ concentration (Hanson, 1994). *Prompt NO* is defined as the NO formed at a rate faster than that formed by the thermal mechanism. With this definition, three sources of Prompt NO in hydrocarbon fuel can be identified: (a) Super-equilibrium O and OH concentrations in the reaction zone and post flame processes, which greatly accelerate the thermal mechanism (b) Fenimore mechanism, in which a reaction sequence is initiated by hydrocarbon radicals present close to the reaction zone



(c) reaction of O atoms with N₂O via the three body recombination reaction (N₂O intermediate mechanism). Organic compounds bound chemically in the fuel are the principal source of *Fuel NO* formation. The extent of conversion of this organic nitrogen to NO is dependent on the fuel-air ratio and the combustion temperature. The dominant reactions in the mechanism of NO₂ formation are



It is also removed by the equation



The HO_2 radicals are formed in relatively low temperature regions; hence, NO_2 formation occurs when NO molecules from high temperature region diffuse or are transported by fluid mixing into the HO_2 rich region. The NO_2 destruction reactions are active at a temperature more than 1750 K with the presence of sufficient H . Although at high temperatures the concentration of OH is also high, the NO_2 removal reactions do not usually reverse due to the large forward reaction rate coefficient of this reaction. Hydrocarbon flames have lower NO_2 production than that of hydrogen flames due to lower OH radical concentrations (Barlow and Carter, 1996). Besides, in hydrocarbon flames CH and CN radicals mainly dominate the NO_2 removal process if the local temperature is less than 1750 K (Heard, et. al., 1992). These two factors, i.e. lower formation and higher destruction rates of NO_2 , are responsible for the lower NO_2 emission from hydrocarbon flames than from hydrogen flames.

Carbon monoxide (CO) considered as another air pollutant besides NO_x , is formed due to incomplete oxidation of carbon and dissociation of CO_2 . In the primary reaction zone of the flame where the temperature is within 10% of the adiabatic flame temperature, free radicals like H , O and OH are formed which initiate the branching chain reaction of CO formation (Westenberg, 1971). On the other hand, Turns and Bandaru (1993) predicted that due to radical recombination in the secondary reaction zone at sufficient concentration of oxygen, most of the CO is converted into CO_2 .



The amount of CO present in flames or post flame gases generally depends upon the factors like stoichiometry, chemical kinetics and its oxidation to form CO₂. This is why the presence of CO in the exhaust gases usually indicates incomplete combustion. Turns and Bandaru (1993) have shown that in lean mixture flames, the CO formation is due to insufficient residence time. They also predicted that CO formation has a strong correlation with *in-flame soot formation*. Since both of them compete for oxygen, usually either CO or NO_x increases at the expense of the other. However, there exist some combustion situations where both pollutants may increase or decrease.

7.4 RESULTS AND DISCUSSIONS

7.4.1 Turbulent Flame Length

The variation of normalized visible flame length (L_f/d_j) with hydrogen content in the mixture is shown in Fig. 7.1. It can be seen that flame length decreases as hydrogen content increases in the mixture. With the increase of hydrogen in the mixture, the chain carrying radicals/atom pool such as of H, O and OH increases, which enhances the combustion rate of natural gas (Chapter Six). As a global effect of this, the composite fuels with higher hydrogen content burn faster and thereby reduce the overall convective time scale. Hence, the fuel travels a short length before combusting, which ultimately shortens the visible flame length. The global convective time scale, (T_G), defined by Eq. 7.4 also reveals the same. The variation of global convective time scale with hydrogen content in the mixture is shown in Fig 7.2. The convective time scale, which is a representation of global flame residence time, decreases as hydrogen concentration

increases in the mixture. This also supports the argument mentioned earlier i.e. the increase in hydrogen content increases the overall combustion performance. Also, on a global scale, the percent hydrogen content in the mixture shows a linear correlation [$-1.4356(\%H_2) + 142.51$; $R^2=0.99$] with the convective time scale. The variation of nondimensional flame lengths (L^* ; defined by Eq. 7.2) with hydrogen content in the fuel mixture is shown in Fig. 7.3. The L^* , which primarily shows the effect of flame stoichiometry and density, increases as the hydrogen concentration increases in the mixture. This trend indicates that the decrease in visible flame length is slower than the decrease in flame stoichiometry (f_s) and density as the hydrogen content increases in the mixture. The nondimensional flame length (L^*) correlates fairly well ($R^2=0.94$) with hydrogen content in the mixture and yield a linear relation [$0.0233(\%H_2)-16.755$]. The effects of Flame Froude number on flame length are shown in Fig. 7.4. Clearly, the effect of buoyancy on the composite fuel flames are still important for the range of Flame Froude Number encountered in the present investigation. Although the trend predicted by Eq. 7.3 is similar to that measured, the values are by a factor of approximately 1.4 higher. The non-dimensional flame length increases with Flame Froude number and follows a power correlation of $L^* = 17.16Fr_f^{0.16}$ with a R^2 value of 0.93.

7.4.2 Radiative Heat Loss Fraction

Fig. 7.5 shows the variation of heat release factor with the increase of hydrogen content in the mixture. Radiative heat loss decreases as hydrogen content increases in the mixture. Since an increase in hydrogen content decreases soot and carbon dioxide

formation, the radiative heat losses from both continuum and molecular sources decrease. Initially, the rate of decrease is high, however the rate decreases as more hydrogen is added to the mixture. Most likely, comparative increase in radiation from H_2O near $2.5\mu\text{m}$ at higher hydrogen contents slows down the decrease rate of radiant heat release factor at higher hydrogen concentration. The variation of apparent flame temperature, T_f , derived from radiation measurement is shown in Fig. 7.6. The apparent flame temperature shows an indication of how hot or cold the flame was felt to the surroundings. This information is essential to design the combustion chamber linings. The variation of computed adiabatic equilibrium flame temperature with hydrogen content in the mixture is also shown in the same figure. The apparent flame temperature decreases with hydrogen content in the mixture despite the slight increase in adiabatic flame temperature. The adiabatic flame temperature increases since hydrogen has a higher adiabatic flame temperature (2382 K, Choudhuri, 1997) than that of natural gas¹ (2225K, Choudhuri, 1997), and it indicates possible maximum flame temperature. In fact, in chapter six it was shown that flame temperature increases with the increase of hydrogen content in the mixture. However, the apparent flame temperature decreases since radiation from both molecular and continuum band decreases with the increase of hydrogen content in the mixture. Hence, despite an increase in flame temperature the surroundings feel a much cooler flame as the hydrogen content is increased in the fuel mixture. The relation between convective time scale, T_G , and radiative heat loss fraction is shown in Fig. 7.7. Although the data are not perfectly correlated [$T_G=9.71F+3.64$; $R^2=0.84$], in general an increase in convective time scale increases flame radiation.

¹ Considering only CH_4

7.4.3 Emission Indices

The variation of NO emission indices with hydrogen content in the mixture is shown in Fig. 7.8. The NO emission increases as hydrogen content increases in the mixture. As discussed in Chapter Six the overall flame temperature increases rapidly as hydrogen is added to the system because of the higher energy input and lower flame radiation. In a methane diffusion flame *thermal* or *Zeldovich Mechanism* is a dominant route of NO production. An increase in flame temperature further enhances the NO formation through this route. On the other hand, the lower CH formation (Chapter Six) at higher hydrogen concentration decreases the NO destruction through *Fenimore mechanism*. As a combined effect the NO emission increases as hydrogen concentration increases in the fuel mixture. For the 100% NG the EI_{NO} is 0.23 g/kg. of fuel which increases by 8.6% as 5% hydrogen is added to the mixture. The variation of NO_x emission with hydrogen content in the mixture is shown in Fig. 7.9. The NO_x emission also increases with the increase of hydrogen concentration in the mixture. This indicates that similar to NO, NO_2 also increases as the hydrogen concentration increases in the mixture. Despite an increase in H concentration, the N_2O destruction through N_2O intermediate mechanism is not dominant in the flames investigated in the present study. For the 100% NG the EI_{NO_x} is 0.58 g/kg of fuel which increases by 3% as 5% hydrogen is added to the fuel mixture.

The variation of CO emission index with hydrogen concentration in the mixture is shown in Fig. 7.10. As expected, the CO emission index decreases with the increase of hydrogen concentration in the mixture. Primarily there are two reasons behind it: First, for the same Reynolds number, an increase in hydrogen concentration results in a lower

carbon input to the flame, and thereby reduces CO formation: Second, an increase in hydrogen concentration prompts more OH formation, and since OH is a dominant radical for CO oxidation, the overall CO formation in the flame drops. This phenomenon is evident in the decreasing trend of CO emission index.

The scaling of Emission indices with Flame Froude Number is shown in Figs. 7.11, 7.12 and 7.13. It can be seen that an increase of Flame Froude Number results in a higher NO and NO_x formation. The NO and NO_x emission indices exhibits exponential correlations with the Flame Froude number, whereas the CO emission index yields power law relation with the Flame Froude Number. The correlations are shown in the figures with the respective R² values. The scaling of emission indices with the heat release rate is shown in Figs. 7.14-16. With the increase of heat input to the flame, the emission indices of NO and NO_x increase. Since a higher heat input in a low radiating flame results in a higher flame temperature, this trend is justifiable. The exponential correlations of NO and NO_x with the heat release rates are $[EI_{NO}=1.19e^{0.35Q_T}]$ and $[EI_{NO_x}=0.42e^{0.15Q_T}]$ with correlation coefficients of 0.9 and 0.97. The CO emission index decreases with the flame heat release rate. The higher flame temperature with lower carbon input and higher availability of OH radicals is responsible for this trend. The CO emission index shows a power relation $[EI_{CO}=2.67Q_T^{-0.48}; R^2=0.9]$ with total heat release rate.

7.4.4 Volumetric Soot Concentration

The axial distribution of volumetric soot concentration (Appendix VII), as a line-of-sight average normal to the flame axis, with the axial distance from the burner is shown in

Fig. 7.17. For all mixture conditions soot concentration increases from the near-burner to mid-flame regions attains a peak value in the mid-flame regions and decreases in the far-burner region. This trend indicates a three-regime soot history inside the flame. The region with increasing concentration is the *soot inception* region followed by the *soot growth* and *burning region* in the mid-flame and far-burner regions. With the increase in hydrogen content in the mixture, the soot concentration decreases. This is expected since a lower carbon input at higher hydrogen concentration results in lower PAH (Polycyclic Aromatic Hydrocarbons) formation, which ultimately decrease the soot inception process. For the 100% NG flame the maximum measured volumetric soot concentration is 16.9×10^{-7} g/cc which decreases to 10.7×10^{-7} g/cc as 10% hydrogen is added to fuel mixture. The maximum soot concentrations in the 80-20% and 65-35% NG-H₂ mixtures are 7.9×10^{-7} g/cc and 4.9×10^{-7} g/cc respectively.

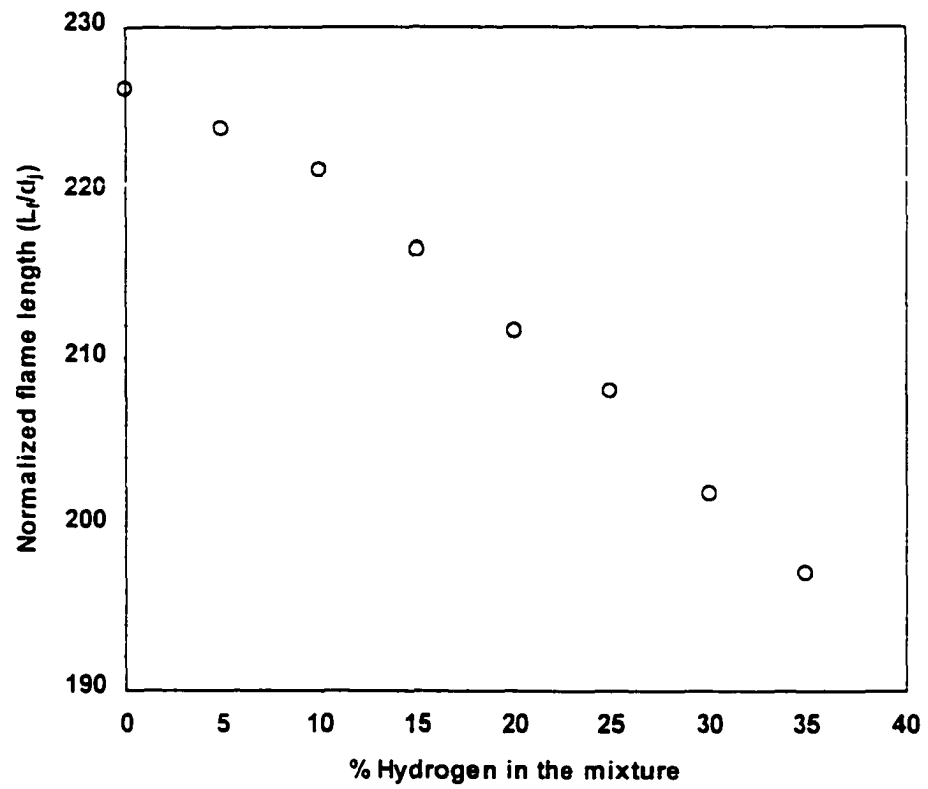


Fig. 7.1 Variation of Normalized Flame Length with % Hydrogen in the Mixture

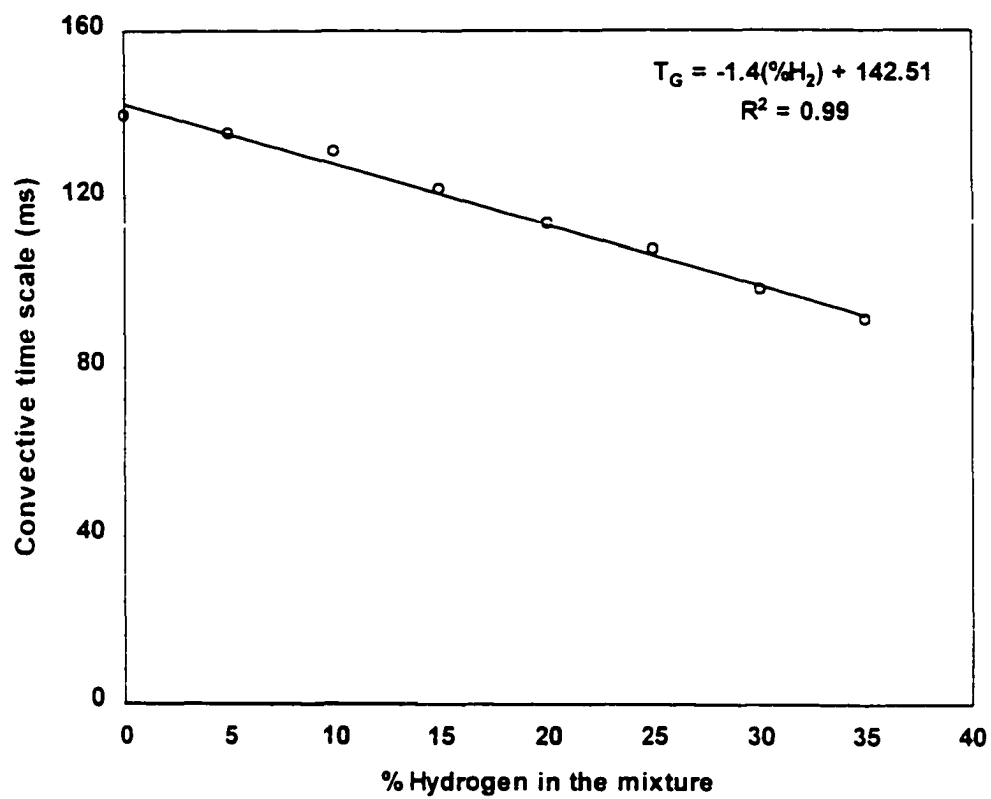


Fig. 7.2 Variation of Convective Time Scale with % Hydrogen in the Mixture

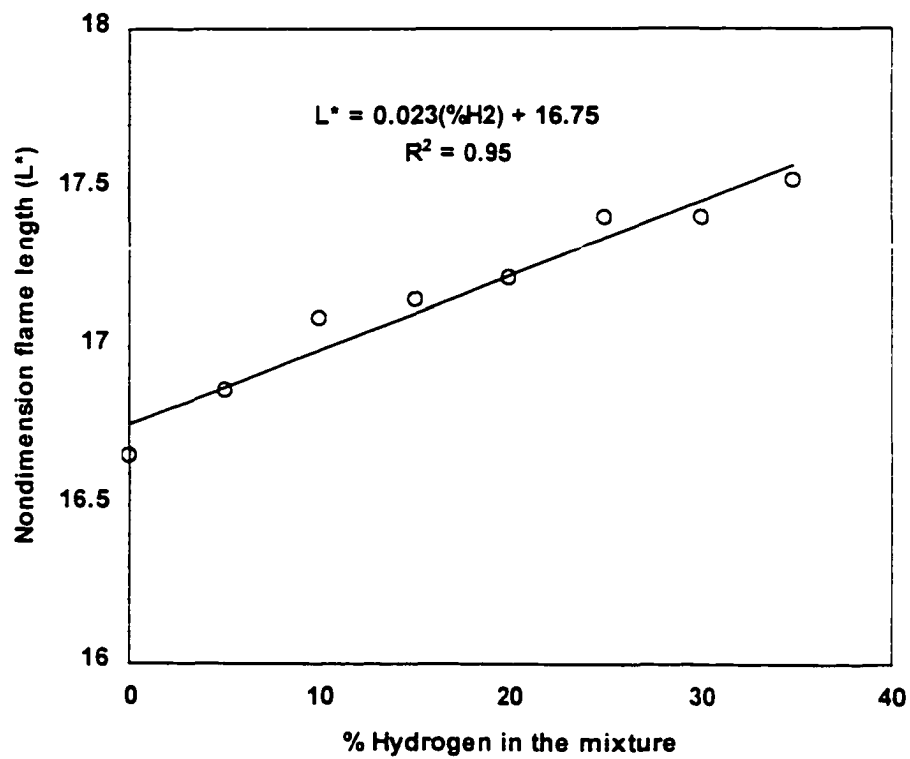


Fig. 7.3 Variation of Nondimensional Flame Length with % Hydrogen in the Mixture

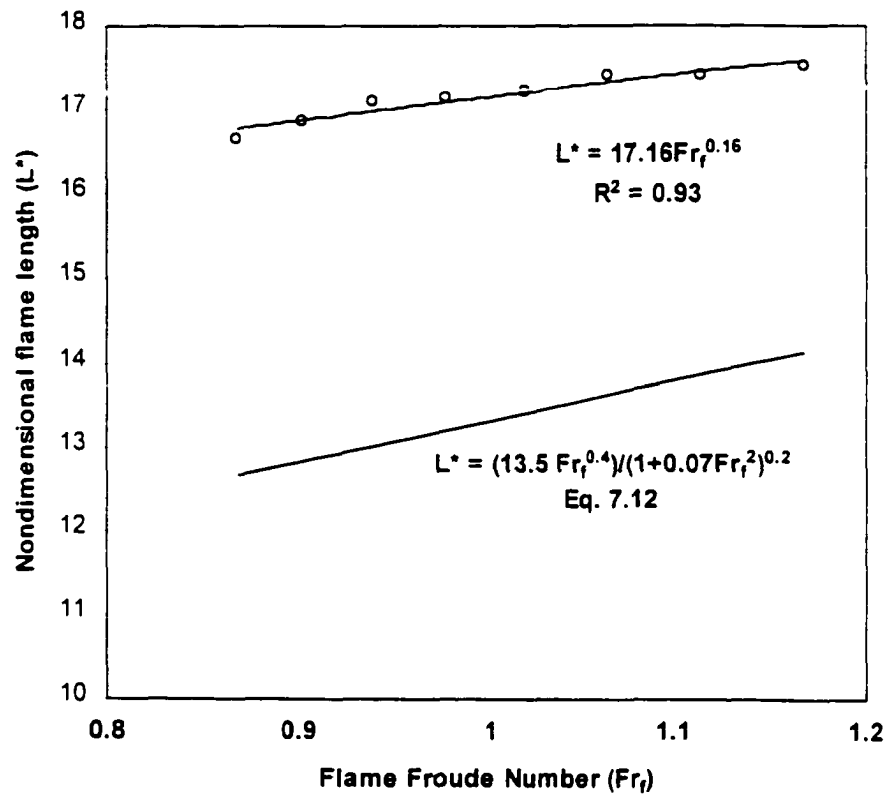


Fig. 7.4 Variation of Nondimensional Flame Length with Flame Froude Number

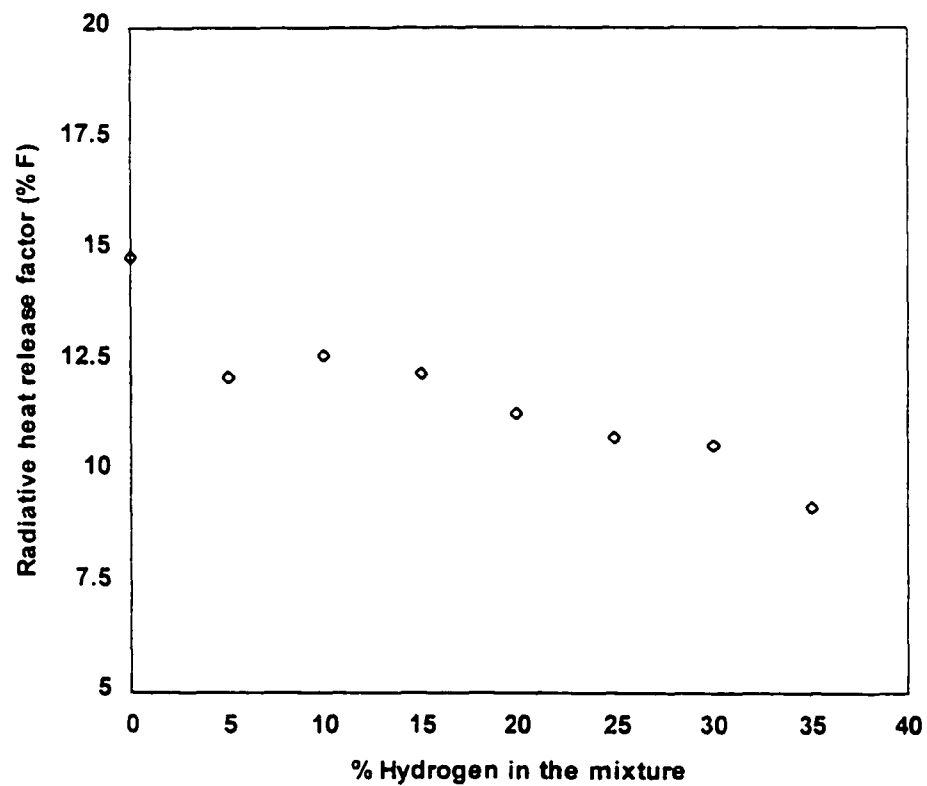


Fig. 7.5 Variation of Radiative Heat Release Factor with % Hydrogen in the Mixture

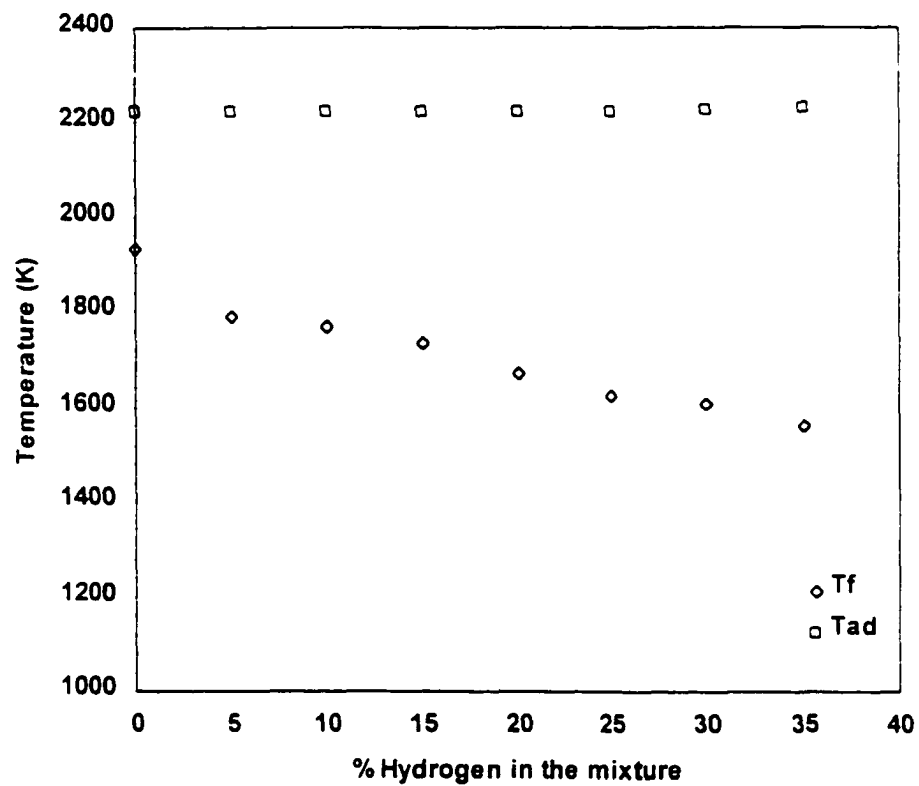


Fig. 7.6 Variation of Apparent (T_f) and Adiabatic Flame Temperature (T_{ad}) with % Hydrogen in the Mixture

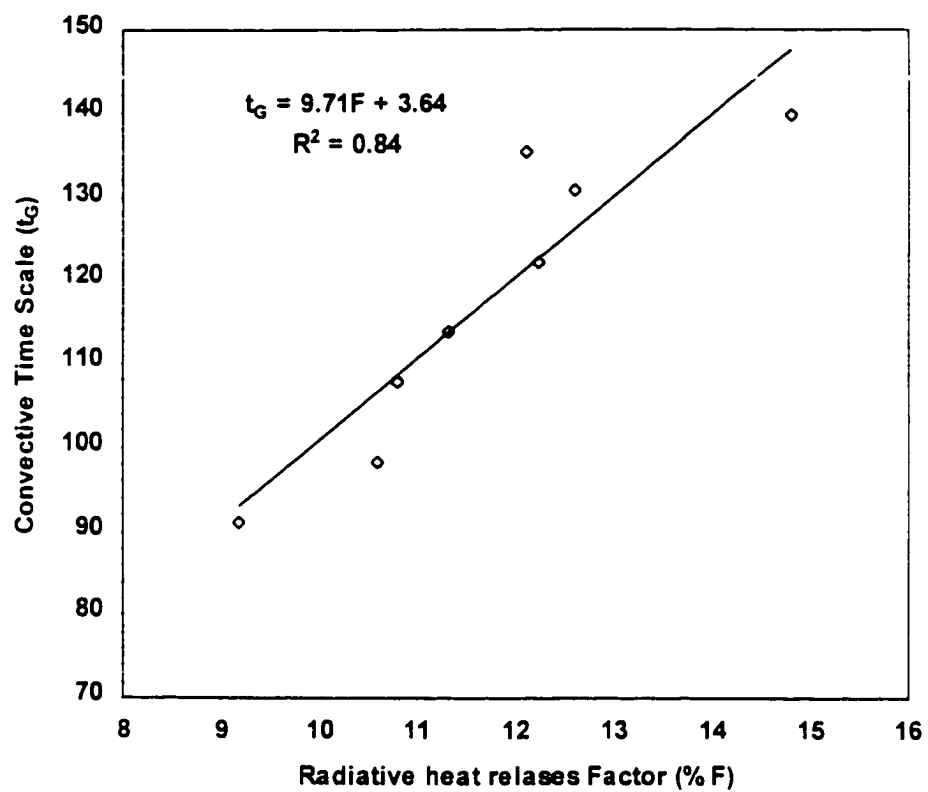


Fig. 7.7 Variation of Convective Time Scale (t_G) with Radiative Heat Loss Factor

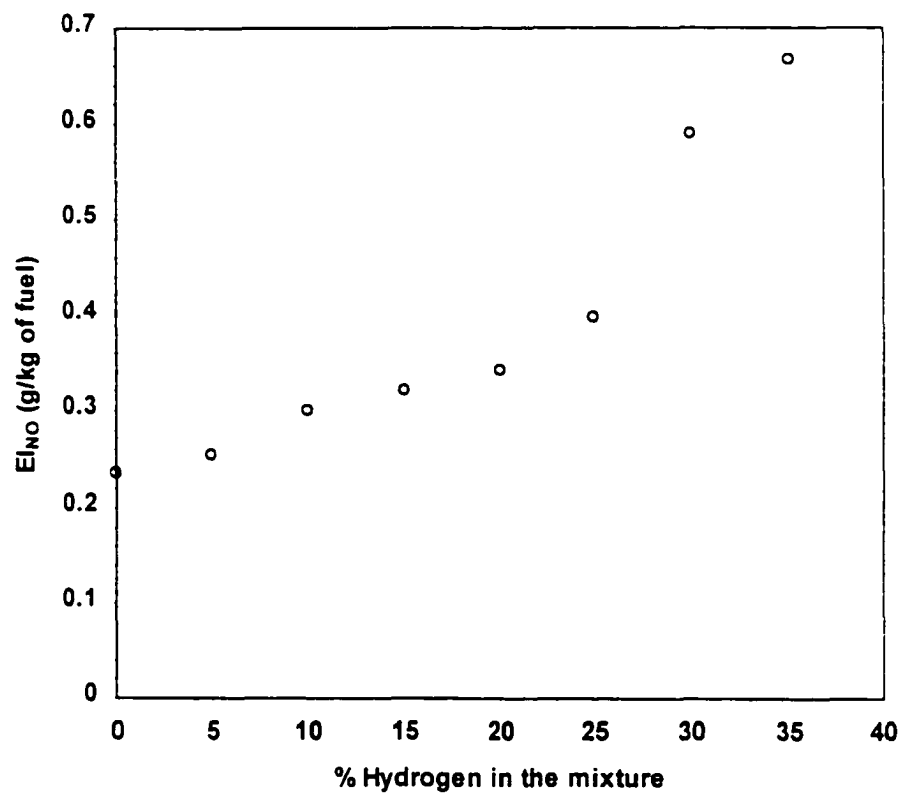


Fig. 7.8 Variation of NO Emission Index % Hydrogen in the Mixture

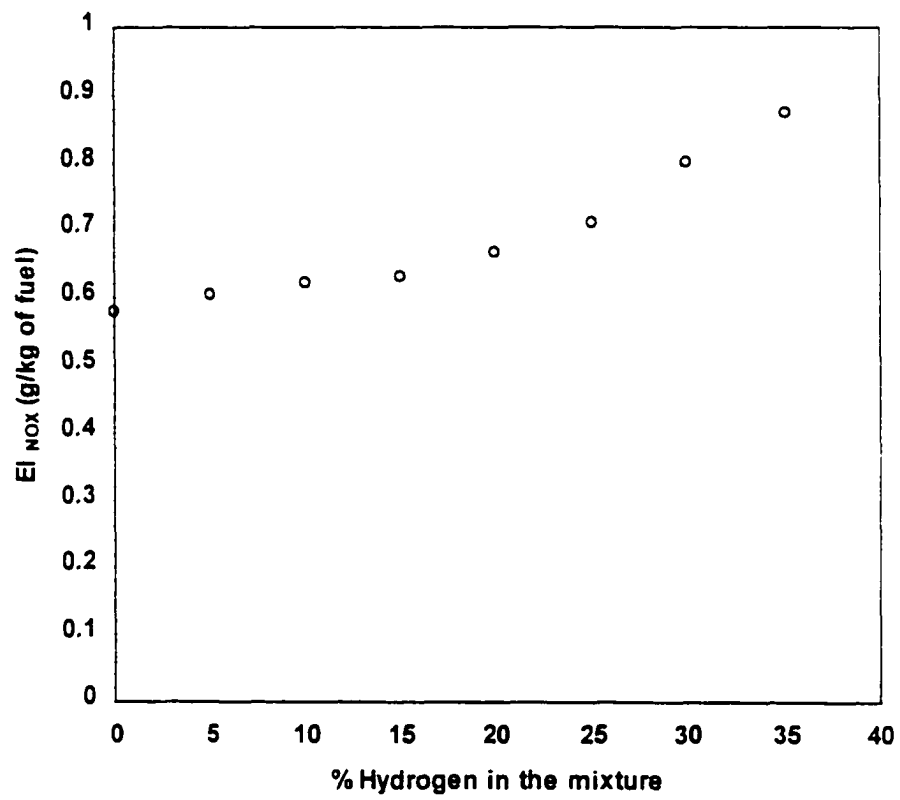


Fig. 7.9 Variation of NO_x Emission Index % Hydrogen in the Mixture

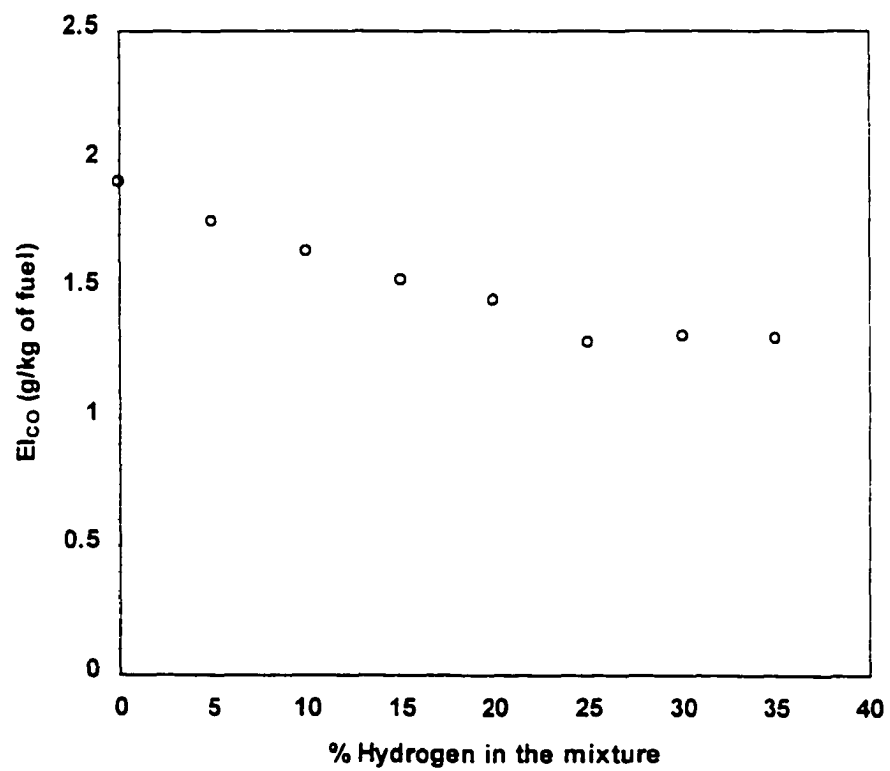


Fig. 7.10 Variation of CO Emission Index % Hydrogen in the Mixture

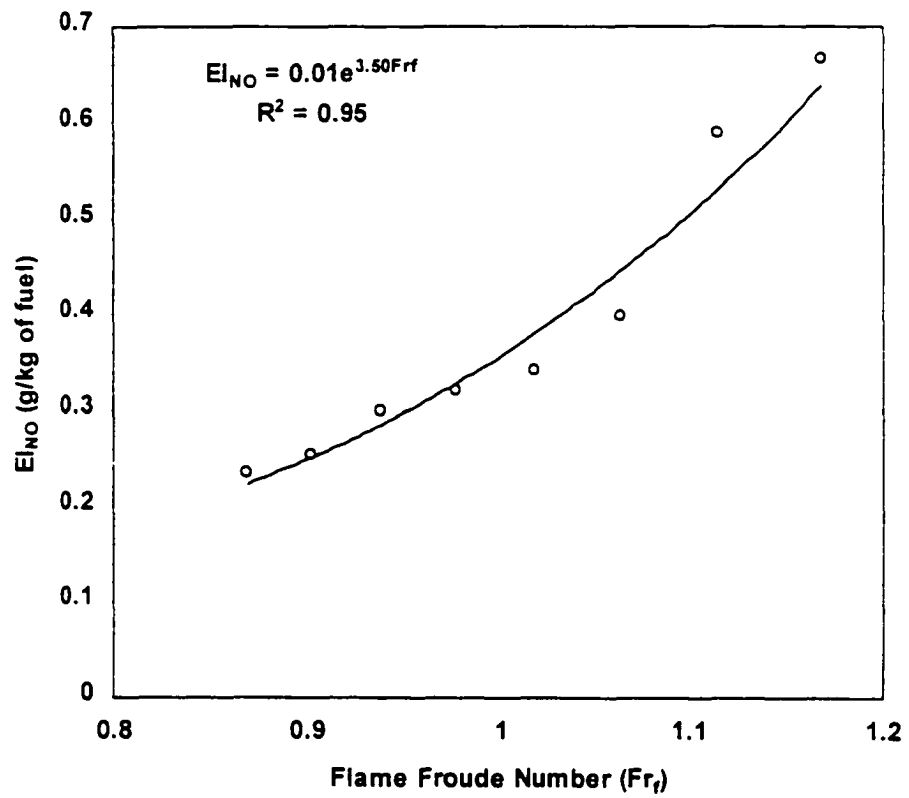


Fig. 7.11 Variation of NO Emission Index with Flame Froude Number

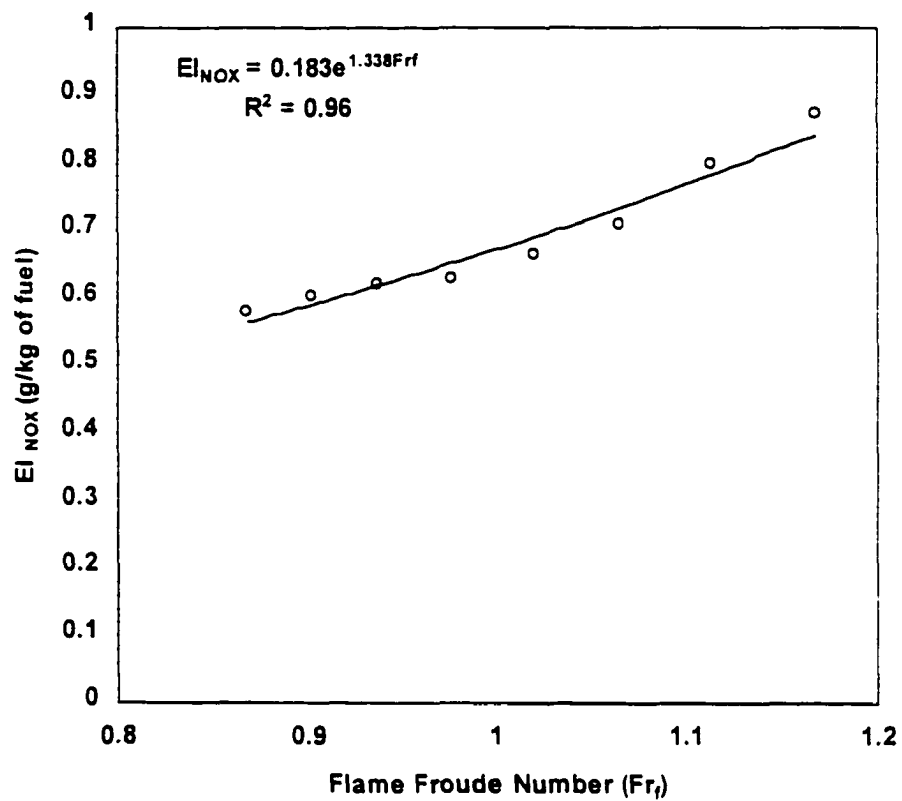


Fig. 7.12 Variation of NO_x Emission Index with Flame Froude Number

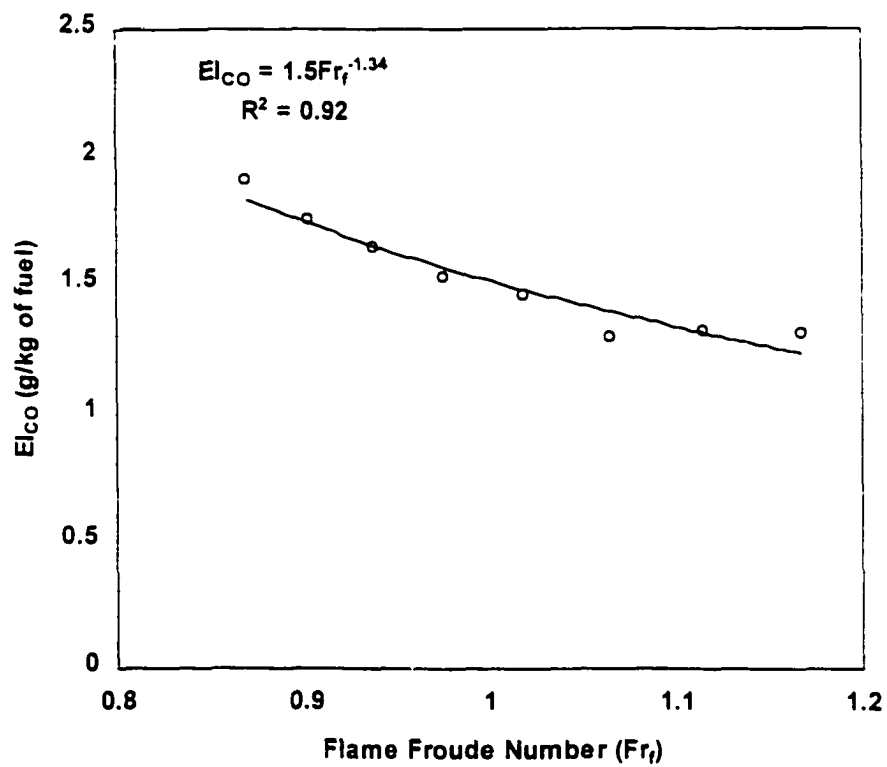


Fig. 7.13 Variation of CO Emission Index with Flame Froude Number

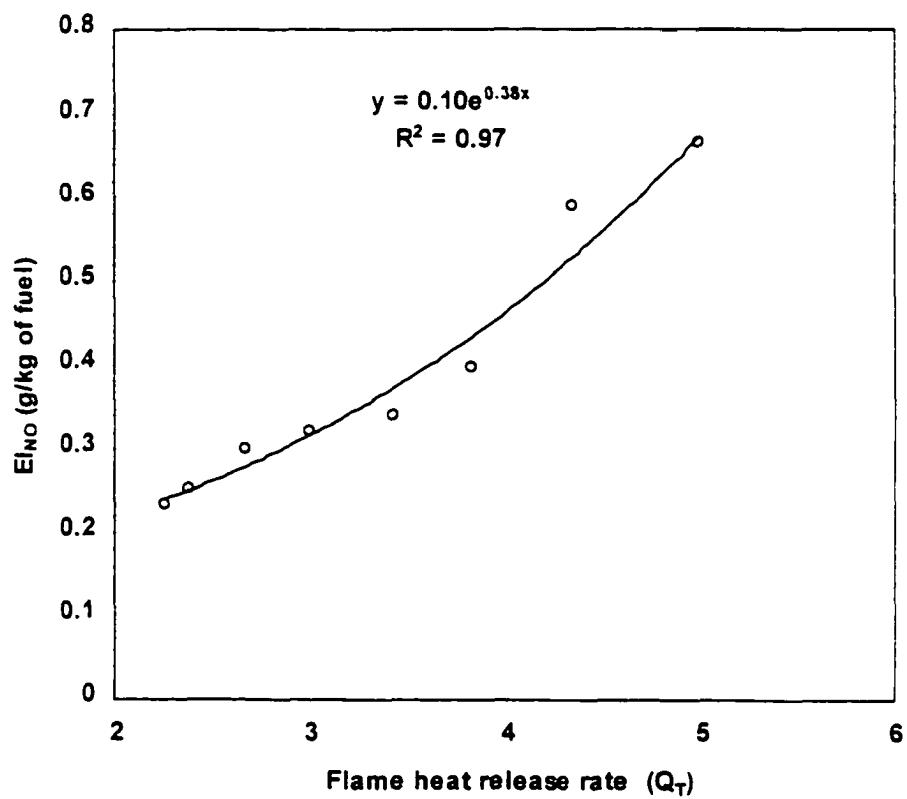


Fig. 7.14 Variation of NO Emission Index with Total Heat Release Rate

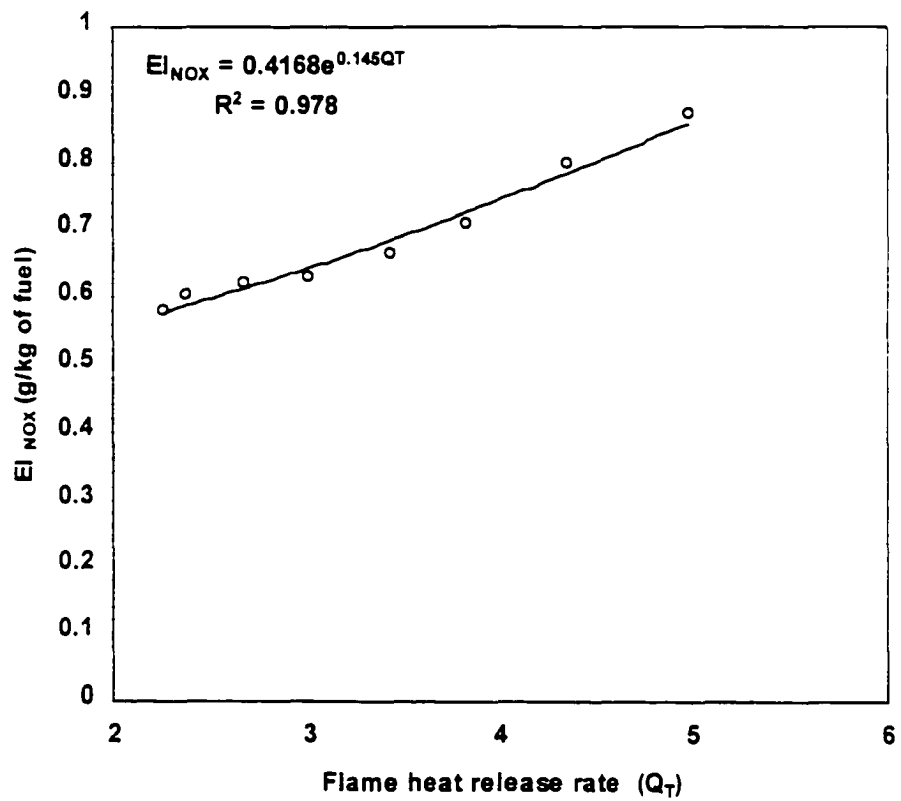


Fig. 7.15 Variation of NO_x Emission Index with Total Heat Release Rate

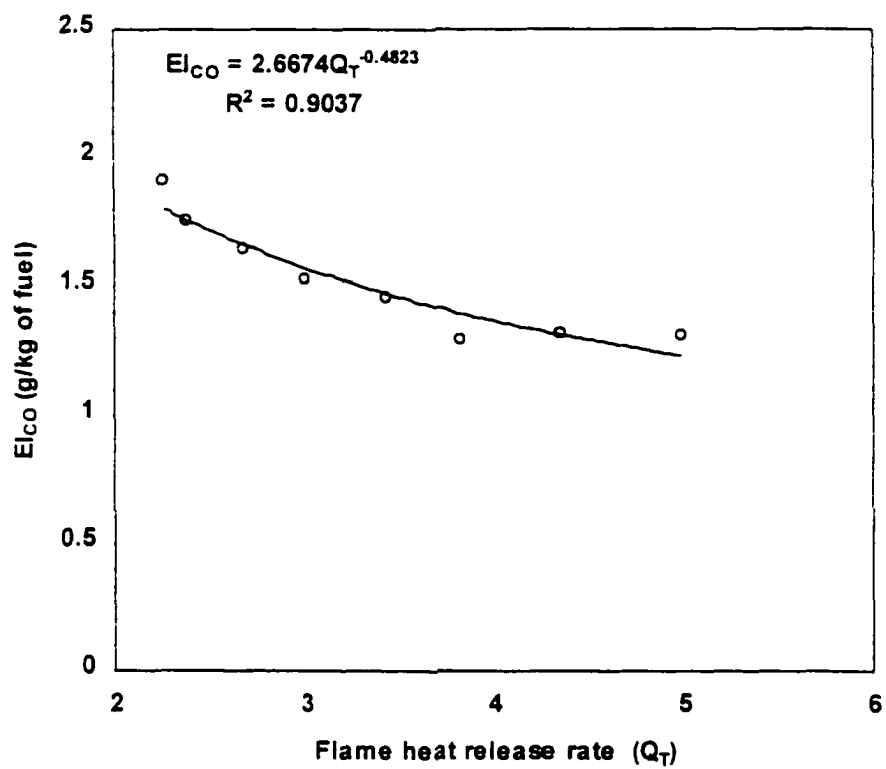


Fig. 7.16 Variation of CO Emission Index with Total Heat Release Rate

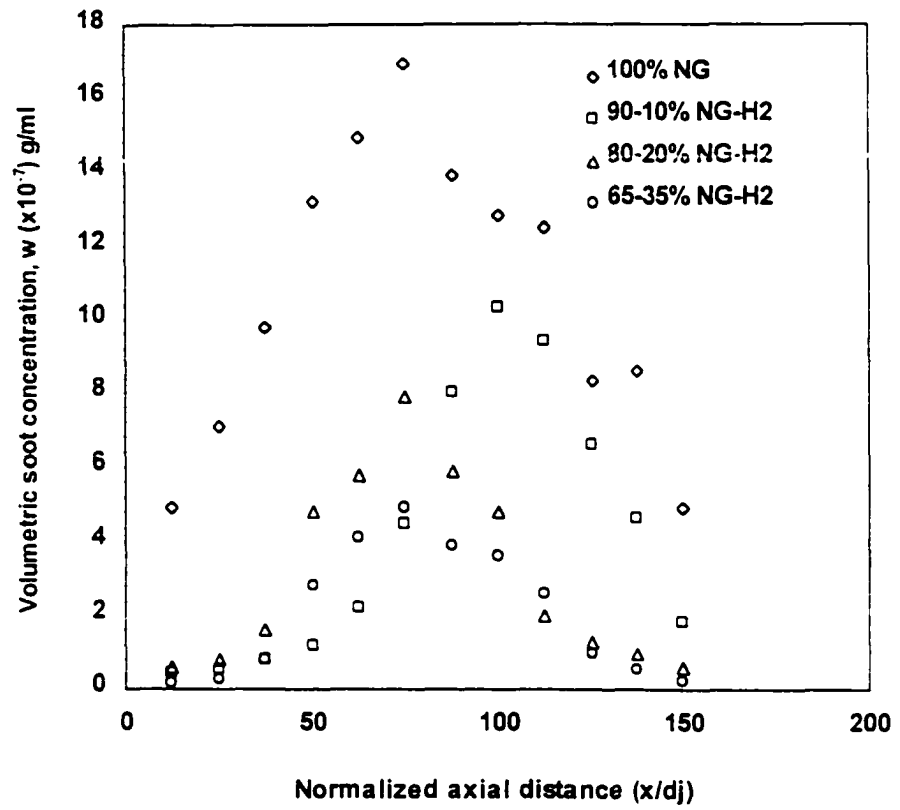


Fig. 7.17 Axial Distribution of Volumetric Soot Concentration

Chapter Eight

Conclusions and Recommendations

8.1 General Remarks

The characteristics of turbulent diffusion flames of three composite mixtures of hydrogen and natural gas (90-10%, 80-20% and 65-35%) by volume are presented in this dissertation. The combustion behavior of these composite fuels is evaluated against the characteristics of a 100% natural gas flame, which is considered as the base-line condition of the present investigation. The fuel jet exit Reynolds number is maintained constant at 8700, which ensured fully turbulent flames. The range of corresponding Flame Froude number depending on the fuel mixture is 0.85-1.18 and hence both buoyancy and momentum are significant.

As mentioned in Chapter One, this study is a broader extension of the earlier studies conducted in the Combustion and Flame Dynamics Laboratory of the University of Oklahoma by the author. Chronologically, the first study evaluated the feasibility of the hydrogen-hydrocarbon composite fuels and investigated the global combustion behavior of these composite fuels. (Choudhuri and Gollahalli, 1998a, 2000a, Choudhuri, 1997). In that study, laminar diffusion flame configuration was used for simplicity. Subsequently, intermediate radical concentration measurements were carried out to extend the understanding of flame chemistry of composite fuels (Choudhuri and Gollahalli, 2000c). Also, a numerical computation of the flow-field was done using detailed chemical kinetics to understand the interaction of flow and chemistry (Choudhuri and Gollahalli, 2000b). Similar to other two studies, a laminar flame configuration was also used in this

study to avoid the complexities of the flow-field. All these studies demonstrated superior combustion characteristics of hydrogen-hydrocarbon composite mixtures. However, before planning to implement the use of composite fuels in practical combustors, turbulent combustion of these fuels needs to be thoroughly investigated. Hence, in the present study, the detailed of combustion characteristics of the natural gas-hydrogen composite fuels are studied using both experimental and numerical approaches.

One of the primary rationales of using hydrogen-hydrocarbon composite fuels is their increased flame stability compared to hydrocarbon fuels. The stability behavior of hydrogen-natural gas composite fuels for different burner sizes is presented in Chapter Five. Since fuel jet velocity is the key determining factor of diffusion flame stability, all measurements and computation in that chapter are presented in terms of jet exit velocity rather than jet exit Reynolds number. Also, predictions of the flame stability (Lift-off and Blowout Velocity) of the hydrogen-natural gas composite mixtures using currently available theories are presented. The primary difficulty in using those theories is the evaluation of laminar flame speed of hydrogen-hydrocarbon mixtures and the equivalent stoichiometric mass fraction for the composite fuel. Linear interpolation of these parameters between pure hydrogen fuels and hydrocarbon fuels yields erroneous predictions. However, a theory proposed by Tanford and Pease (1947), along with the current computational technique produces a good agreement of predictions and measurements of blowout velocities. The analysis has emphasized that the flame stability of any fuel primarily depends on the concentrations of OH, H and O radicals, and their binary diffusion coefficients. In the present study, the equilibrium

concentrations of these radicals are computed, and the laminar flame speed of the composite fuels is calculated. In Chapter Six, the flame structure measurements show that OH, H and O concentrations increase with the increase of hydrogen content in the mixture but the increase is not linear. Cold jet mixing (isothermal mixing) of the fuel and air in the lifted part of the flame is often considered to be one of the key factors, which determine the stability. This aspect of flame stability is also investigated in the present investigation. The acetone PLIF images of cold jet reveal that the addition of hydrogen enhances the overall mixing process of fuel and air. It is found that turbulence intensity increases with the increase of hydrogen concentration in the mixture, which ultimately increases the local turbulent flame speed. On the other hand, the axial flow velocity decreases faster with the increase in hydrogen concentration in the mixture. Therefore, the combined effect of higher turbulent flame speed and lower axial flow velocity in the lifted part increases the flame stability of natural gas-hydrogen mixtures. Also, a local equivalent equivalence ratio (EFI) is defined to demonstrate the effect of local stoichiometry on flame blowout mechanism.

In Chapter Six, the details of the inflame measurements of flame temperature, stable species concentration (CO_2 , CO, NO, and O_2), and intermediate radicals/atoms concentration (OH, CH, H and O) are presented. The measured values are also compared with the computed values using a simplified reaction mechanism. As mentioned earlier, the OH, H and O concentrations play a key role in determining the local flame velocity. On the other hand, the local flame temperature dictates the formation and destruction of these key radicals. Laser Induced Fluorescence technique is used to measure radical

concentrations. Although relative concentrations of these radicals give an overall picture of the changing flame chemistry of composite mixtures, absolute concentrations are necessary for deeper understanding of the reaction kinetics. However, quantification of LIF signals require the modeling of collisional quenching, which is often impossible because of the lack of information regarding collider molecules. In the present investigation, combined measurements of LIF and Raman Spectra are done to tackle this problem. The Raman signals are used to measure the collider species concentration. Also, a comparison of existing collision models is done to identify their validity in diffusion flame. It is found that the method of estimation yields poor results in LIF signal quantification process. The chemistry of diffusion flames is largely controlled by the local flow-field behavior. As it is seen from the velocity-species concentration composite picture, a strong recalculation zone exists in the vicinity of the primary reaction zone and fuel moves quickly towards this location. The CO concentration decreases in most locations of the flame with the increase in hydrogen concentration. Lower soot formation and higher OH concentration are responsible for this. The inflame NO production increases as the flame temperature increases with the increase of hydrogen concentration in the mixture. Although, in most cases (especially in near-burner region) the computed velocity field agrees well with the predicted trends, the magnitude of velocities is different from measured values. The two-step simplified reaction kinetic mechanism employed in prediction is most likely responsible for the discrepancy. However, current computational capability and numerical schemes are limited in using detailed chemical kinetics in turbulent reacting mixtures.

The global flame characteristics i.e. flame length, pollutant emission, radiative heat loss fraction, and volumetric soot concentrations are presented in Chapter Seven. The correlation of flame length and hydrogen concentration in the fuel mixture is shown. It is found that the flame length decreases with the increase of hydrogen concentration in the mixture. As seen in flame structure measurements the reactivity of fuel mixtures increase with the increase of hydrogen concentration in the mixture, i.e., higher OH, H, O and flame temperature, which ultimately shorten the time required to burn the fuel and thereby reduces the overall flame length. By the same argument convective time scale decreases with the increase of hydrogen content in the mixture. The measured flame length is also compared with theoretical calculations in Chapter Seven. Although the trends are similar, the predicted values are 1.4 times higher than the measured values. The radiation heat losses decrease because of the reduction of emission from both CO₂ and soot bands. As seen in the flame structure measurements that CO₂ decreases with the increase of hydrogen concentration in the mixture. Similarly, axial soot concentration decreases with the increase of hydrogen content in the fuel mixture because of lower carbon input and higher soot oxidation due to higher OH concentration. As expected from flame structure measurements, the CO emission index decreases with the increase of hydrogen concentration in the mixture. On the other hand, NO and NO_x emission indices increase because of the higher flame temperature at higher hydrogen concentration in the mixture.

In general, the computational model predicts the flame temperature and species concentrations reasonably well in the near-field region. However, the agreement

between measurement and prediction is poor in the mid-flame and far-burner regions. It is apparent that the growth of solid soot particles and their heterogeneous kinetics controlled combustion in the far-burner region make the flow-field exceedingly complicated. Since the present mathematical formulation and computational capability are limited and do not account for these complex physical phenomena the computed values are significantly different from those of experimental measurements. Also, the critical assumption of neglecting flame radiation, which is considerable in the mid-flame and far-burner regions, seems to have a significant impact on the computed values. Although the computational predictions of mid-flame and far-burner flame structure are not accurate quantitatively, the qualitative agreement of computation and measurement certainly justifies the use of numerical technique in conjunction with a calibrating technique in many practical applications where time and resource limit the use of detailed experimental investigation.

8.2 Conclusions

In the present study the measurements of global flame characteristics lead to the following results. With the increase of hydrogen concentration in the mixture:

- i. the maximum laminar flame speed increases and exhibits an exponential relation. $S_{L,max} = 34.2 e^{0.02(\%H_2)}$; ($R^2=0.95$, $0 \leq \% H_2 \leq 100$), with the volumetric concentration of hydrogen in the composite fuel.
- ii. The blowout velocity increases. A single relation, $\frac{U_b}{S_{L,max}} \left(\frac{\rho_F}{\rho_A} \right)^{1.5} = 0.017 \frac{HS_{L,max} \rho_F}{\mu_F}$; ($R^2=0.99$, $0 \leq \%H_2 \leq 35$), correlating burner diameter, blowout velocity and hydrogen content in the mixture is proposed. The characteristics flame height is

defined as, $H = 8 \left[\frac{Y}{Y_s} \left(\frac{\rho_F}{\rho_A} \right)^{0.5} - 5.8 \right] r_j$; ($1\text{mm} \leq r_j \leq 4\text{mm}$). The correlation is

critically sensitive to the value of the laminar flame-speed of the mixed fuel.

iii. the maximum lift-off height at blowout condition decreases. A general

correlation, $\frac{U_b}{S_{L,max}} \left(\frac{\rho_F}{\rho_A} \right)^{1.5} = 50.54 \frac{L_b S_{L,max} \rho_F}{\mu_F}$; ($R^2=0.99$, $0 \leq \%H_2 \leq 35$), between lift-

off height at blowout conditions and the percent of volumetric hydrogen concentration in the mixture is also determined.

iv. visible flame length decreases. A correlation [$t_G = -1.4(\%H_2) + 142.51$; $R^2=0.99$,

$0 \leq \%H_2 \leq 35$] between convective time scale (derived from visible flame length) and hydrogen concentration is formulated. The general correlation between the nondimensional flame length, L^* , and the volumetric percentage of hydrogen in the mixture is $L^* = 0.023(\%H_2) + 16.75$; $R^2=0.95$; $0 \leq \%H_2 \leq 35$.

v. radiative heat loss fraction decreases. The convective time scale (t_G) exhibits a linear relation [$t_G = 9.71(\%F) + 3.64$; $R^2=0.84$; $0 \leq \%H_2 \leq 35$] with the radiative heat release factor of the flame.

vi. the CO emission index decreases. The CO emission index shows a power relation [$El_{CO} = 1.5 Fr_f^{-1.34}$, $R^2=0.92$, $0 \leq \%H_2 \leq 35$] with the flame Froude number.

The correlation between CO emission index and flame heat release rate (Q_T) is $El_{CO} = 2.667 Q_T^{-0.4823}$, $R^2=0.90$, $0 \leq \%H_2 \leq 35$.

vii. the NO_x emission index increases and shows an exponential relation [$El_{NO_x} = 0.183 e^{1.338 Fr_f}$, $R^2=0.96$, $0 \leq \%H_2 \leq 35$] with the flame Froude number. The

correlation between NO_x emission index and flame heat release rate (Q_T) is

$$\text{EI}_{\text{NO}_x} = 0.4168e^{0.145Q_T}, R^2 = 0.978, 0 \leq \%H_2 \leq 35.$$

Where, %F= percent radiative heat loss fraction, %H₂= volumetric percentage of hydrogen in the mixture, H= characteristic flame height, EI =emission indices, Fr_f= flame Froude number, L^{*}= nondimensional flame length, L_b= maximum lift-off height at blowout condition, Q_T= Total heat release rate by the fuel, R²= correlation coefficient, r_f= burner radius, S_{L, max}= maximum laminar flame speed, t_G= convective time scale, U_b= blowout velocity, Y = fuel Mass Fraction, Y_s= stoichiometric fuel mass fraction, ρ_F= fuel density at burner exit condition, ρ_A=ambient air density, μ_F = kinematic viscosity of the fuel at burner exit condition.

The flow and flame structure measurements and predictions reveal that with the increase of hydrogen concentration in the mixture

- i. the mixing rate of fuel and air increases. The acetone PLIF images of NG-H₂ cold jet show that as the hydrogen concentration increases the fuel mixes rapidly with the incoming air.
- ii. the axial velocity in the lift-off region of the flame decays faster.
- iii. the local axial turbulence intensity and turbulent flame speed increase.
- iv. flame temperature increases. In near burner location, the 100% NG flame has a peak temperature of 1620 K, which increases to 1850 K as 10% hydrogen is added to the mixture. Also, both measured and predicted radial profiles of flame temperature show the inward movement of the stoichiometric contour.

- v. the inflame concentration of CO₂ and CO decreases while NO concentration increases.
- vi. the inflame OH, H and O radical concentrations increase while CH concentration decreases. In near burner location, the peak OH, H and O concentration increases 15.2%, 12.6% and 11.8% respectively with the addition of 10% hydrogen in the pure NG flame. On the other hand, in the same location the CH concentration reduced sharply (77%) as 10% hydrogen is added into the pure fuel.
- vii. the axial volumetric soot concentration decreases. The peak soot concentration decreases from a value of 17×10^{-7} g/ml to 10.2×10^{-7} g/ml as 10% hydrogen is added to the 100% NG flame.

8.3 Practical Implications

The present study provides previously non-existent information about the combustion behavior of hydrogen-hydrocarbon composite fuel, which can be effectively used for designing or improving various practical combustors such as lean burning gas turbine combustors for terrestrial and aerospace applications. The generalized correlations of blowout velocity, lift-off height and hydrogen concentration in the fuel mixture can be used to determine the amount of hydrogen required to achieve the limits of flame stability in the combustion chamber. Similarly, the generalized relations of flame lengths, hydrogen concentration and flame radiation are crucial for combustion chamber dimensioning. Also, the quantification of pollution characteristics of the hydrogen-hydrocarbon composite

fuel will be beneficial to improve flame stability without violating the environmental regulations.

8.4 Recommendations for Future Work

A further study on hydrogen-hydrocarbon composite fuels using premixed or partially premixed configuration will benefit the use and applicability of the composite fuels in some practical combustors. Also, a further investigation is required to develop a simplified reaction mechanism, which will better represent the chemistry of the mixed fuel. In most of the mid-flame locations, where multi-step reaction kinetics dominate the combustion chemistry, the numerical scheme fails to predict the proper trend. Instead of using two global hydrocarbon reactions, a better approach might be to use two global reactions and two elementary reactions (Penko, 2000).

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Appendix I

Methane Reaction Mechanism

Peter's (1993) mechanism for methane combustion

Reaction	A kg-mol/m ² s	n	E KJ/kg-mol
<i>H₂O₂ chain reaction</i>			
R1 H-O ₂ ⇒OH+O	2.00×10 ¹⁷	0.0	70560
R2 OH+O⇒H+O ₂	1.57×10 ¹⁶	0.0	3528
R3 O-H ₂ ⇒OH+H	5.06×10 ⁰⁷	2.67	26376
R4 OH+H⇒O+H ₂	2.22×10 ⁰⁷	2.67	18354
R5 H ₂ +OH⇒H ₂ O+H	1.00×10 ¹¹	1.60	13860
R6 H ₂ O+H⇒H ₂ +OH	4.31×10 ¹¹	1.60	76734
R7 OH+OH⇒O+H ₂ O	1.50×10 ¹²	1.14	420
R8 O+H ₂ O⇒OH+OH	1.47×10 ¹³	1.14	71400
<i>H₂O formation and consumption</i>			
R9 O ₂ +H+M⇒HO ₂ +M	2.30×10 ²¹	-0.8	0
R10 HO ₂ +M⇒O ₂ +H+M	3.19×10 ²¹	-0.8	196056
R11 HO ₂ +H⇒OH+OH	1.50×10 ¹⁷	0.0	4200
R12 HO ₂ +H⇒H ₂ +O ₂	2.50×10 ¹⁶	0.0	29064
R13 HO ₂ +OH⇒H ₂ O+O ₂	6.00×10 ¹⁶	0.0	0
R14 HO ₂ +H⇒H ₂ O+O	3.00×10 ¹⁶	0.0	7224
R15 HO ₂ +O⇒OH+O ₂	1.80×10 ¹⁶	0.0	-1705.2
<i>H₂O₂ formation and consumption</i>			
R16 HO ₂ +HO ₂ ⇒H ₂ O ₂ +O ₂	2.50×10 ¹⁴	0.0	-5280
R17 OH+OH+M⇒H ₂ O ₂ +M	3.25×10 ²⁵	-2.0	0
R18 H ₂ O ₂ +M⇒OH+OH+M	1.69×10 ²⁷	-2.0	202986
R19 H ₂ O ₂ +H⇒H ₂ O+OH	1.00×10 ¹⁶	0.0	15036
R20 H ₂ O ₂ +OH⇒H ₂ O+HO ₂	5.40×10 ¹⁵	0.0	4200
R21 H ₂ O+HO ₂ ⇒H ₂ O ₂ +OH	1.80×10 ¹⁶	0.0	135198
<i>Recombination reaction</i>			
R22 H+H+M⇒H ₂ +M	1.80×10 ¹⁶	-1.0	0
R23 H+OH+M⇒H ₂ O+M	2.20×10 ²⁵	-2.0	0
R24 O+O+M⇒O ₂ +M	2.90×10 ²⁰	-1.0	0
<i>CO/CO₂ mechanism</i>			
R25 CO+OH⇒CO ₂ +H	4.40×10 ⁰⁹	1.5	-3108
R26 CO ₂ +H⇒CO+OH	4.96×10 ¹¹	1.5	90048
<i>CH consumption reactions</i>			
R27 CO ₂ +CH⇒CHO+CO	3.40×10 ¹⁵	0.0	29064
R28 CH+O ₂ ⇒CHO+O	3.00×10 ¹⁶	0.0	0
<i>CHO consumption reactions</i>			
R29 CHO+H⇒CO+H ₂	2.00×10 ¹⁷	0.0	0
R30 CHO+OH⇒CO+H ₂ O	1.00×10 ¹⁷	0.0	0
R31 CHO+O ₂ ⇒CO+HO ₂	3.00×10 ¹⁵	0.0	0
R32 CHO+N⇒CO+H+M	7.10×10 ¹⁷	0.0	70560
R33 H+CO+M⇒CHO+M	1.14×10 ¹⁸	0.0	9996
<i>CH₂ consumption reactions</i>			
R34 CH ₂ +H⇒CH+H ₂	8.40×10 ¹²	1.5	1407
R35 CH+H ₂ ⇒CH ₂ +H	5.83×10 ¹²	1.5	13125
R36 CH ₂ +O⇒CO+H+H	8.00×10 ¹⁶	0.0	0
R37 CH ₂ +O ₂ ⇒CO+OH+H	6.50×10 ¹⁵	0.0	6300
R38 CH ₂ +O ₂ ⇒CO ₂ +H+H	6.50×10 ¹⁵	0.0	6300
<i>CH₂O consumption reactions</i>			
R39 CH ₂ O+H⇒CHO+H ₂	2.50×10 ¹⁶	0.0	16758

R40	$\text{CH}_2\text{O}+\text{O}\Rightarrow\text{CHO}+\text{OH}$	3.50×10^{16}	0.0	146832
R41	$\text{CH}_2\text{O}+\text{OH}\Rightarrow\text{CHO}+\text{H}_2\text{O}$	3.00×10^{16}	0.0	5040
R42	$\text{CH}_2\text{O}+\text{M}\Rightarrow\text{CHO}+\text{H}+\text{M}$	1.40×10^{20}	0.0	321300
<i>CH₃ consumption reactions</i>				
R43	$\text{CH}_3+\text{H}\Rightarrow\text{CH}_2+\text{H}_2$	1.80×10^{17}	0.0	63210
R44	$\text{CH}_3+\text{H}_2\Rightarrow\text{CH}_4+\text{H}$	3.68×10^{16}	0.0	44436
R45	$\text{CH}_3+\text{H}\Rightarrow\text{CH}_4$	6.00×10^{19}	-1.0	0
R46	$\text{CH}_3+\text{O}\Rightarrow\text{CH}_2\text{O}+\text{H}$	7.00×10^{16}	0.0	0
R47	$\text{CH}_3+\text{O}_2\Rightarrow\text{CH}_2\text{O}+\text{OH}$	3.40×10^{14}	0.0	37548
R48	$\text{CH}_3+\text{H}_2\Rightarrow\text{CH}_4+\text{H}$	8.39×10^{04}	3.0	34692
R49	$\text{CH}_3+\text{H}_2\text{O}\Rightarrow\text{CH}_4+\text{OH}$	2.63×10^{08}	2.10	71190
<i>CH₄ consumption equation</i>				
R50	$\text{CH}_4+\text{H}\Rightarrow\text{CH}_3+\text{H}_2$	2.20×10^{22}	3.0	36708
R51	$\text{CH}_4+\text{O}\Rightarrow\text{CH}_3+\text{OH}$	1.20×10^{10}	2.1	32004
R52	$\text{CH}_4+\text{OH}\Rightarrow\text{CH}_3+\text{H}_2\text{O}$	1.60×10^{09}	2.1	10332

Appendix II

Estimated Uncertainties

The uncertainties related to measurements can be classified into two categories: *Precision or Random error* and *Bias or Fixed error*. Random error, which is dependent on sample size, can be estimated statistically, whereas, estimation of biased error needs detailed idea about measurement techniques and flowfield properties. Usually probe measurement of any reacting flow experience some common biased error like gradient biasing, calibration error, error associated with the least count of the instrument etc. Therefore, it is possible to estimate the biased error by carefully determining its effect on a specific measurement. Overall uncertainties of a measurement can be expressed as

$$\omega = \sqrt{(P_r^2 + B^2)}$$

where

P_r = Precision error of mean = $t_{\alpha/2} \cdot S_r$

S_r = Standard deviation

$$S_r = \frac{S_r}{\sqrt{n}} = \frac{\sqrt{\left(\sum (x_i - \bar{x})^2\right)}}{\sqrt{n(n-1)}}$$

$t_{\alpha/2} = 2.262$ (for $\nu = 9$ or $n=10$)

In this present study, the random errors have been estimated by repeating some of the experiment 10 to 12 times. All instruments were calibrated prior to each day's measurements so it can be expected that measurements are free from calibration errors.. Since we are mostly interested in a comparative study, we neglected the effect of other bias errors (gradient biasing, least count etc.) assuming that their effects are constant in all cases.

Relative OH-atom measurement for 65-35% NG-H₂ flame at mid-flame location (25% of the visible flame length)

Observation	OH-concentration Relative unit x 10 ⁻²
1	15.96
2	15.02
3	14.32
4	15.83
5	16.52
6	15.10
7	15.92
8	15.95
9	16.6
10	16.78
Mean	15.8
S _x	0.78
S _x	0.25
P _x	0.56
% error of mean	3.53

Maximum estimated uncertainty of relative OH concentration at 95% confidence interval
 $\approx 4\%$ of the mean value

Appendix III

Thermo-Physical Properties of the Composite Fuel

Density and Viscosity

$$\text{Density: } \rho_{mix} = \sum_{i=1}^n X_i \rho_i$$

$$\text{Viscosity } \mu_{mix} = \frac{\sum_{i=1}^n X_i \mu_i}{\sum_{i=1}^n X_i \phi_i}$$

where

$$\phi_i = \frac{1}{\sqrt{8}} \left[1 + \frac{M_i}{M_j} \right]^{-1.2} \left[1 + \left(\frac{\mu_i}{\mu_j} \right)^{1.2} \left(\frac{M_i}{M_j} \right)^{1.4} \right]$$

where

Q_F = Fuel flow rate (m^3/sec)

μ_{mix} = Viscosity of the mixed fuel (N-s/m^2)

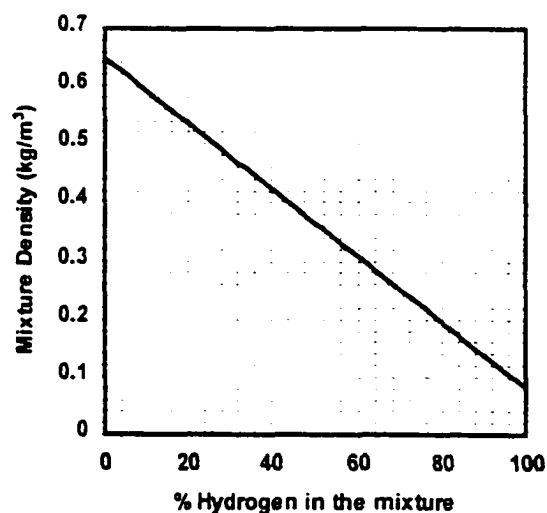
ρ_{mix} = Density of the mixed fuel (Kg/m^3)

M = Molecular weight of the fuels

χ_i = Species mole fraction

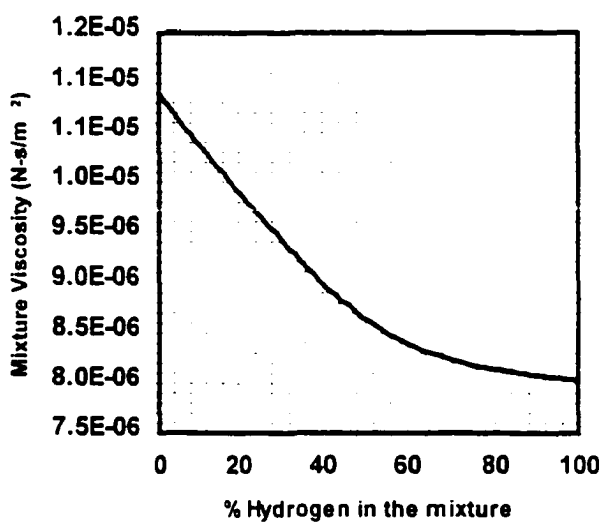
Mixture Density (at 16°C)

Mixture		Density
Methane	Hydrogen	
%	%	Kg/m^3
100	0	0.648
95	5	0.619665
90	10	0.59133
85	15	0.562995
80	20	0.53466
75	25	0.506325
70	30	0.47799
65	35	0.449655
50	50	0.36465
40	60	0.30798
20	80	0.19464
10	90	0.13797
0	100	0.0813



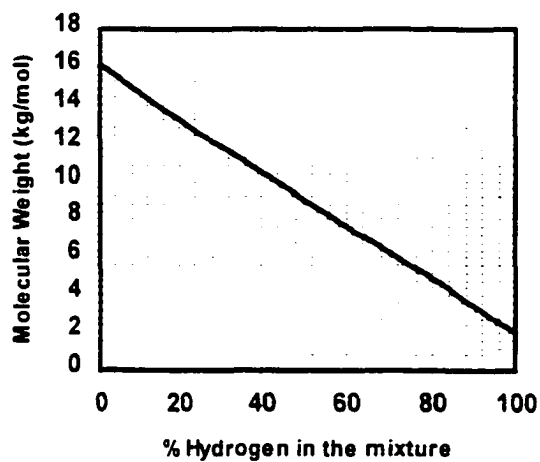
Mixture Viscosity (at 16°C)

Mixture		
Methane	Hydrogen	Viscosity
%	%	N-s/m ²
100	0	1.09E-05
95	5	1.06E-05
90	10	1.04E-05
85	15	1.01E-05
80	20	9.89E-06
75	25	9.65E-06
70	30	9.43E-06
65	35	9.21E-06
50	50	8.65E-06
40	60	8.37E-06
20	80	8.15E-06
10	90	8.07E-06
0	100	8.03E-06



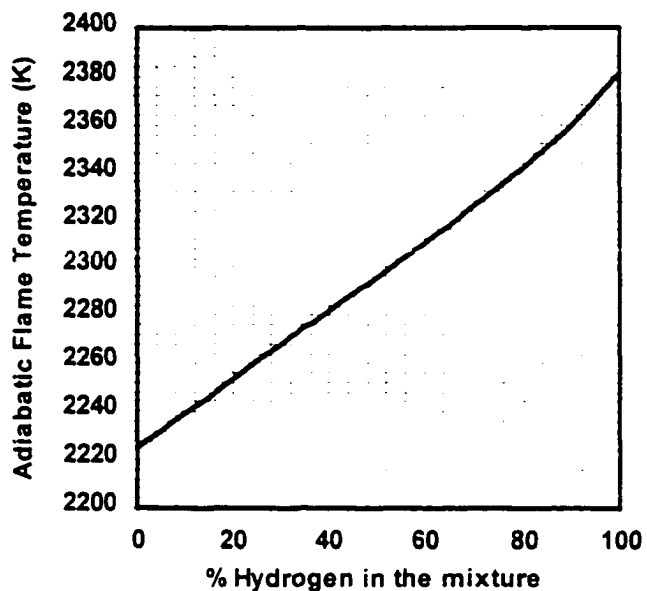
Molecular Weight

Methane	Hydrogen	Mol. Weight
%	%	Kg/mol
100	0	16.043
95	5	15.34165
90	10	14.6403
80	20	13.2376
65	35	11.13355
50	50	9.0295
40	60	7.6268
20	80	4.8214
10	90	3.4187
0	100	2.016

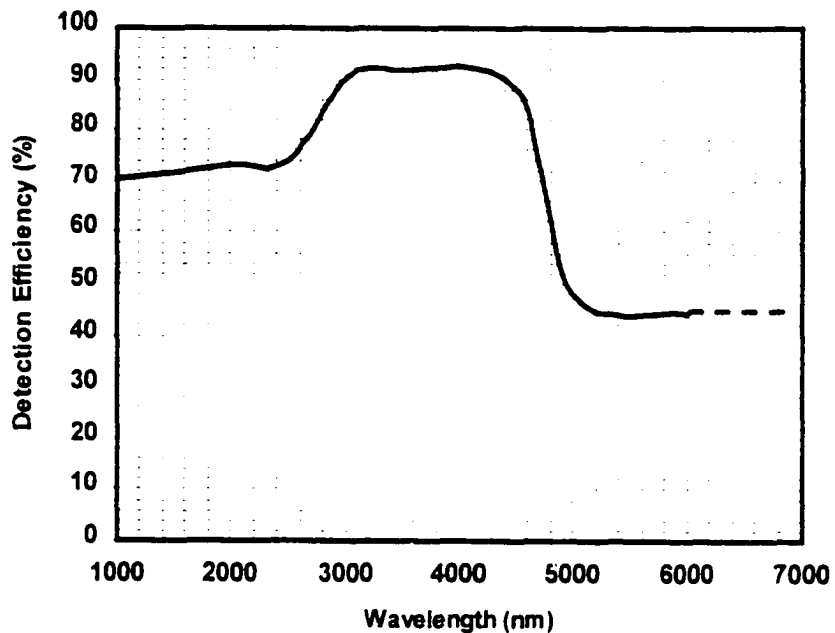


Adiabatic Flame Temperature

Hydrogen	Adiabatic Flame Temperature
0	2225
5	2232.135
10	2239.27
15	2246.405
20	2253.54
25	2260.675
30	2267.81
35	2274.945
50	2296.35
60	2310.62
80	2342.3
91.3	2362.3
100	2382



Radiometer Detection Efficiency*



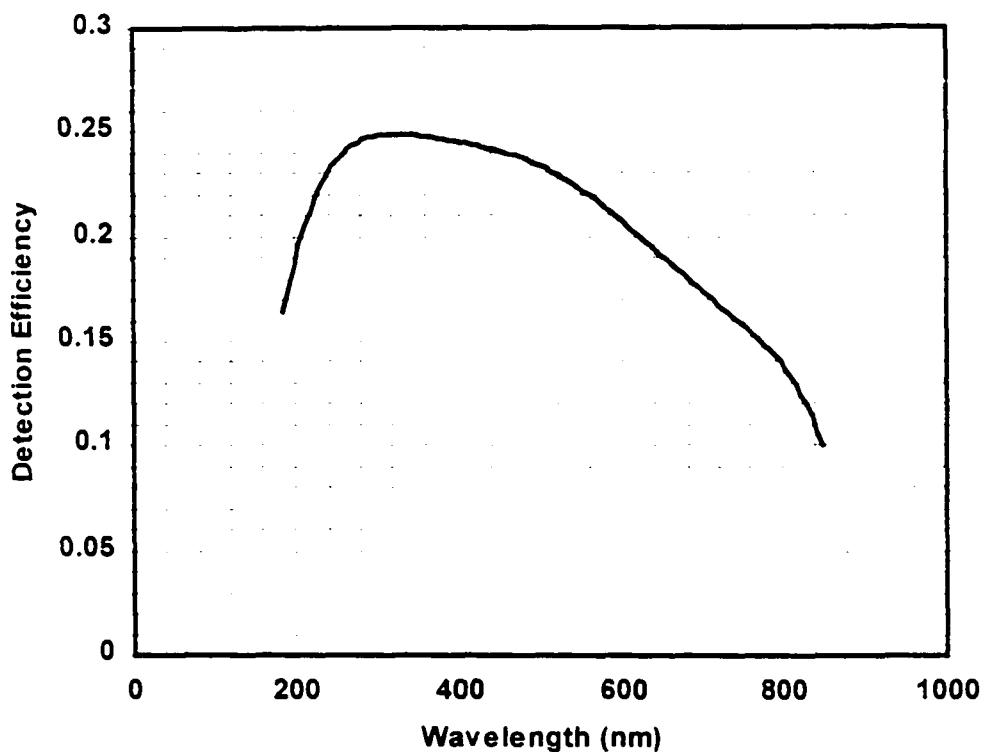
* Source : Hy-Cal Engineering Inc., Santa Fe Springs, CA (<http://www.specdata.com>)

Appendix IV

Calculations Related to Spectroscopic Modeling

Photomultiplier Tube Characteristics

Detection Efficiency*

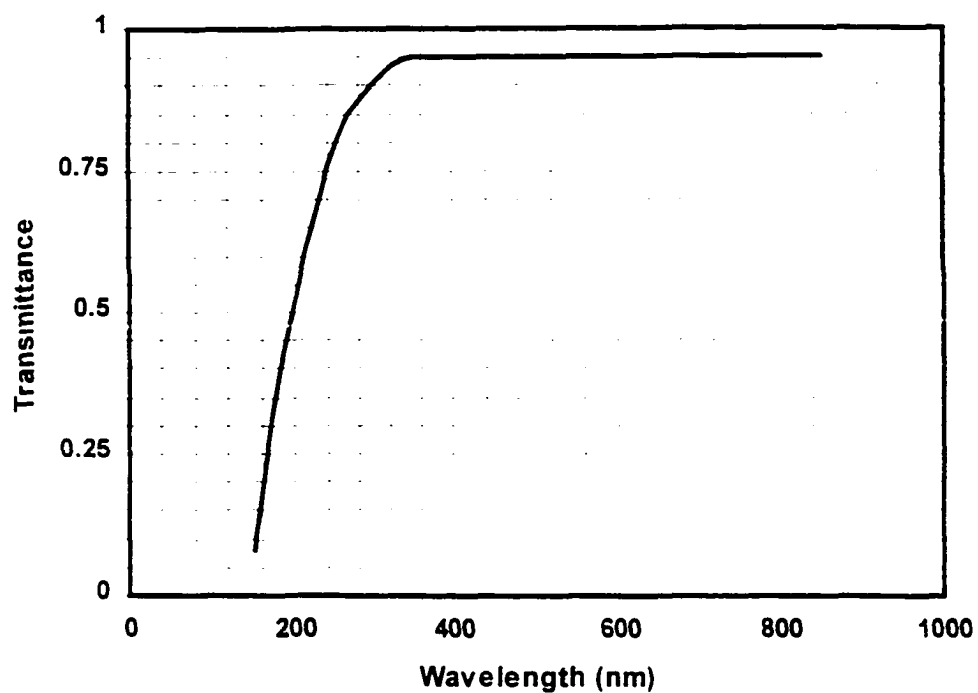


General Features*

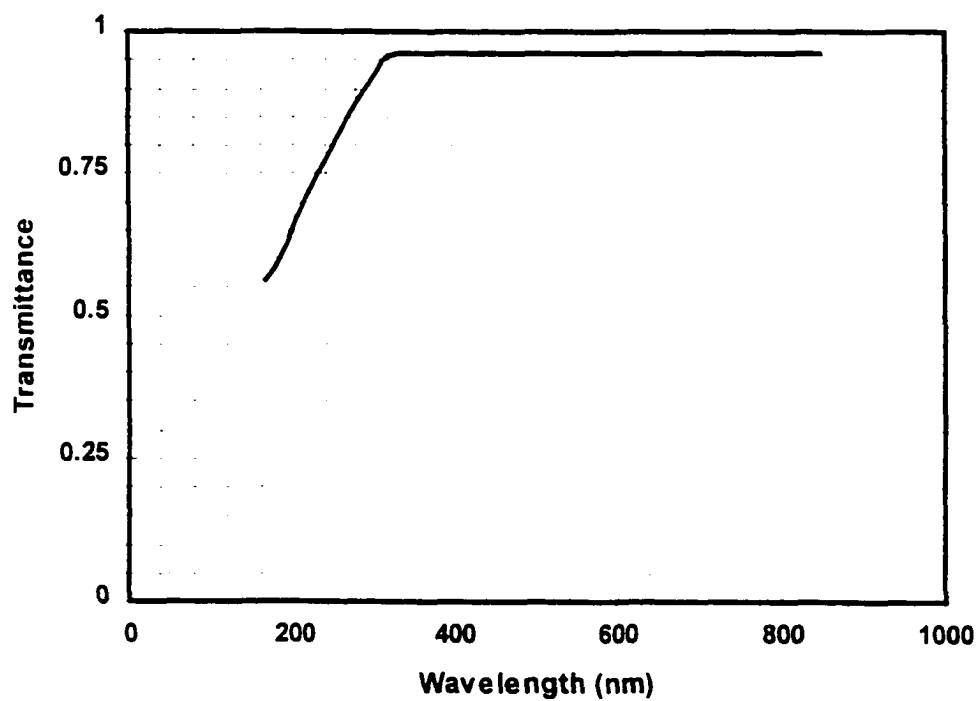
Type	End-on Photomultiplier Tube
Photocathode Material	Bialkali
Window Material	UV Fused Silica
Wavelength Range	185 nm- 850 nm
Peak Wavelength	365 nm
Maximum Rated Voltage	1500 V
Operating Voltage	1000 V
Cathode Responsivity	85 mA/W
Anode Responsivity	1.8×10^5 A/W
Anode Dark Current	2nA
Rise Time	12 ns

* Technical Specifications Handbook, Hamamatsu Photonics K K, Shizuoka-ken, 438-01, Japan

Transmission Efficiency of the Window Material*



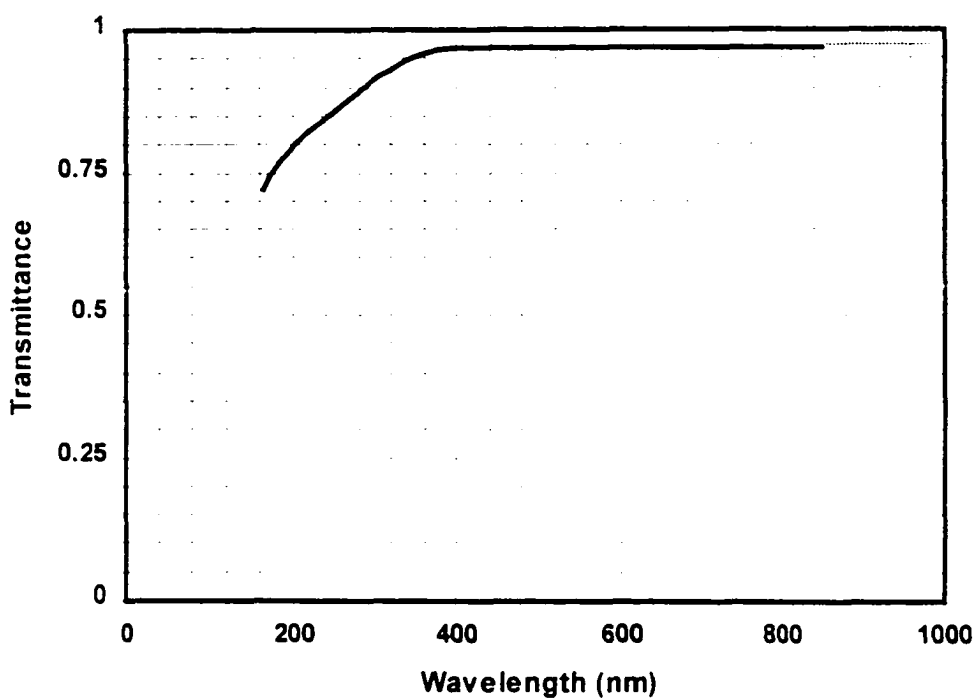
Transmission Efficiency of the Focusing Lenses*



* Technical Specifications Handbook, Hamamatsu Photonics K K, Shizuoka-ken, 438-01, Japan

* Photonics Handbook, Oriel Instruments, 250 Long Beach Boulevard, Stratford, CT 064972, USA

Transmission Efficiency of the Monochromator*



For OH Detection Scheme

Experimental Constants:

At 315 nm

$$\begin{aligned}
 \text{Transmission Efficiency, } \eta_T &= \text{Transmittance of the window material} \times \text{Transmittance of} \\
 &\quad \text{focusing lenses} \times \text{Transmittance of the monochromator} \\
 &= 0.955 \times 0.96 \times 0.87 \\
 &= 0.797
 \end{aligned}$$

$$\begin{aligned}
 \text{Detection Efficiency, } \eta_D &= \text{Detection efficiency of the photomultiplier tube} \\
 &= 0.25
 \end{aligned}$$

$$\text{The collection solid angle} = \Omega/4\pi = 10^{-2}$$

$$\text{The experimental constant, } C = \Omega/4\pi \times \eta_T \times \eta_D = 10^{-2} \times 0.797 \times 0.25 = 0.002$$

* Photonics Handbook, Oriel Instruments, 250 Long Beach Boulevard, Stratford, CT 064972, USA

Stern-Volmer Coefficient:

Equation 3.5

$$SV = \frac{A_{12}}{A_{12} + P + Q + S}$$

For this particular Transition S is negligible and

$$A_{12} = 3.24 \times 10^5 \text{ s}^{-1} \text{ and } P = 0.57 \times 10^{10} \text{ s}^{-1}$$

Again

$$Q = \sum_i \chi_i q_{ix}$$

$$q_{ix} = a_i \sigma_q T^{0.5}$$

$$\sigma_q = \sigma_{qx} e^{\frac{E}{kT}}$$

For OH at 1200 K using Table and measured concentration of the collider species

$$Q = 0.14 \text{ s}^{-1}$$

$$\therefore SV = \frac{3.24 \times 10^5}{(3.32 \times 10^5) + 0.57 \times 10^{10} + (0.14)} = 5.68 \times 10^{-5}$$

Mole fraction Calculation:

$$n_A = C \frac{I_L}{h\nu_{ge}} \chi \beta_{ge} \sigma_{ge} \Delta l$$

$$\sigma_{ge} = \frac{B_{ge} h\nu_{ge}}{\Delta\nu c}$$

Now,

$$B_{ge} = 3.461 \times 10^{15} \text{ m}^3 \cdot \text{s}^{-2} \text{ J}^{-1}$$

$$h = 6.626 \times 10^{-34} \text{ Js}$$

$$\nu_{ge} = c/\lambda = (2.997 \times 10^8)/(285.265 \times 10^{-9}) = 1.050 \times 10^{15} \text{ Hz}$$

$$\Delta\nu = 18 \times 10^9 \text{ Hz}$$

$$\therefore \sigma_{ge} = 4.466 \times 10^{-22} \text{ m}^2$$

Again

$$I_L = 100 \times 10^{-6} \text{ J}$$

$$\beta_{ge} = 0.023$$

$$\Delta l = 0.042 \text{ m}$$

$$\text{and } n_A = 856 \text{ photoelectron}$$

Then

$$856 = 0.002 \frac{100 \times 10^{-6}}{6.626 \times 10^{-34} \times 1.050 \times 10^{15}} \times \chi_{OH} \times 0.023 \times 4.466 \times 10^{-22} \times 0.042$$

$$\therefore \text{the mole fraction of OH, } \chi_{OH} = 6.9 \times 10^{-3}$$

Appendix V

Equivalent Fuel Formula and Equivalence Ratio

Equivalent Fuel Formula

80-20% NG-H₂ mixture

Mass fraction of NG (CH₄) in the mixture = 0.969

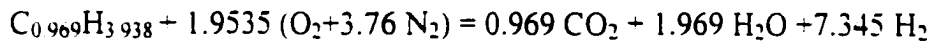
Mass fraction of H₂ in the mixture = 0.031

Equivalent Fuel Formula: C_xH_y

$$x = (0.969 \times 1) = 0.969$$

$$y = (0.969 \times 4) + (0.031 \times 2) = 3.938$$

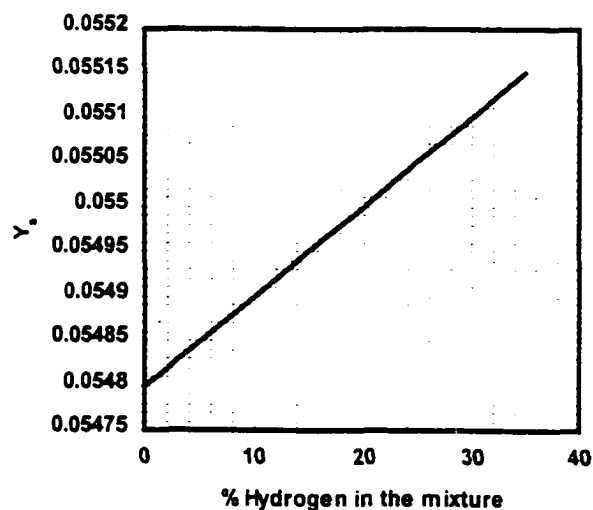
Hence



Stoichiometric

$$\text{Air/Fuel ratio } (A/F)_s = (4.76 \times 1.9535 \times 28.97) / [(12 \times 0.969) + (2 \times 3.938)] = 17.26$$

$$Y_s = \frac{1}{(A/F)_s + 1} = 1 / (17.26 + 1) = 0.0547$$



Equivalence Ratio

For a mixture $Y_{CH_4} = 0.25$, $Y_{air} = 0.6$, $Y_{H_2} = 0.15$

Eq. 5.7 (a)

$$EFI = \frac{\frac{C_{CH_4}}{C_A - \left(\frac{C_{H_2}}{C_A} \right)_s}}{\left(\frac{C_{CH_4}}{C_A} \right)_s}$$

Eq. 5.7 (b)

$$CFI = \frac{\frac{C_{CH_4}}{C_A}}{\left(\frac{C_{CH_4}}{C_A} \right)_s}$$

Eq. 5.7 (b)

$$HFI = \frac{\frac{C_{H_2}}{C_A}}{\left(\frac{C_{H_2}}{C_A} \right)_s}$$

Molecular Weights:

$$M_{CH_4} = 16$$

$$M_{H_2} = 2$$

$$M_A = 28.97$$

$$C_{H_2}/C_A = 0.029 \quad [2H_2 + (O_2 + 3.76N_2) = 2H_2O + 3.76 N_2]$$

$$C_{CH_4}/C_A = 0.058 \quad [CH_4 + 2(O_2 + 3.76N_2) = CO_2 + 2H_2O + 7.52 N_2]$$

$$EFI = \frac{1}{0.058} \left[\frac{0.25 \times 16}{0.6 \times 28.97 - \frac{0.15 \times 2}{0.029}} \right] = 9.79; HFI = \frac{0.15 \times 2}{\frac{0.6 \times 28.97}{0.029}} = 0.6$$

$$CFI = \frac{\frac{0.25 \times 16}{0.6 \times 28.97}}{0.058} = 3.96$$

Appendix VI

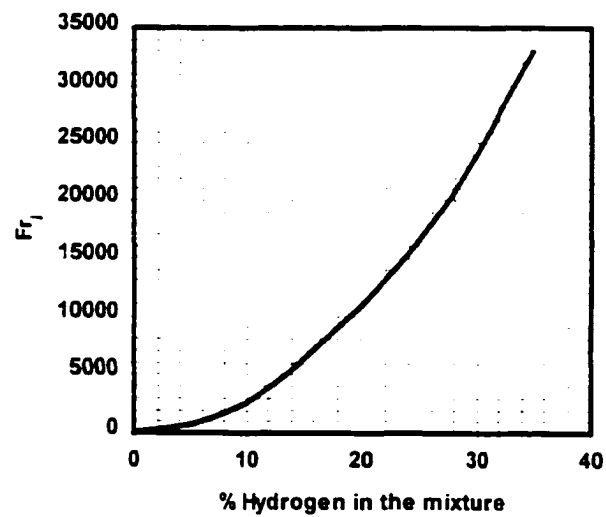
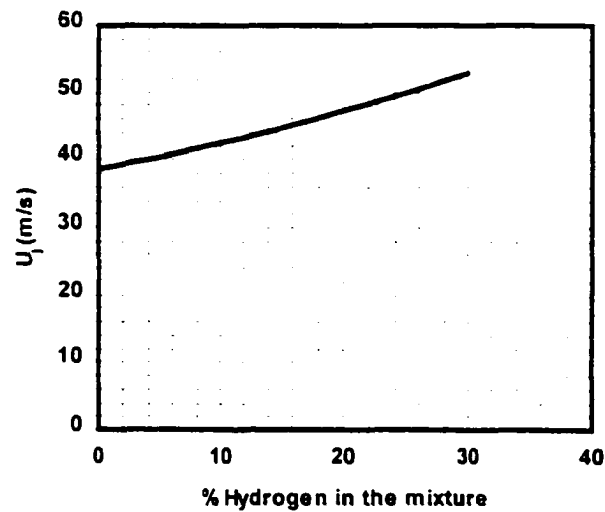
Froude Number Calculation

Jet Exit Froude Number

$$Fr_j = \frac{U_j^2}{gd_j}$$

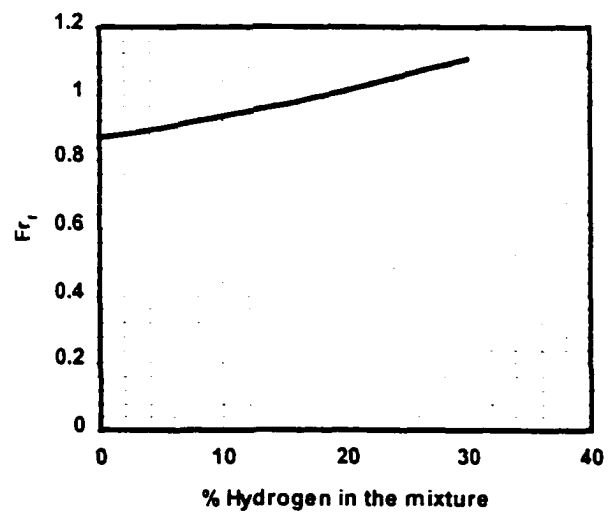
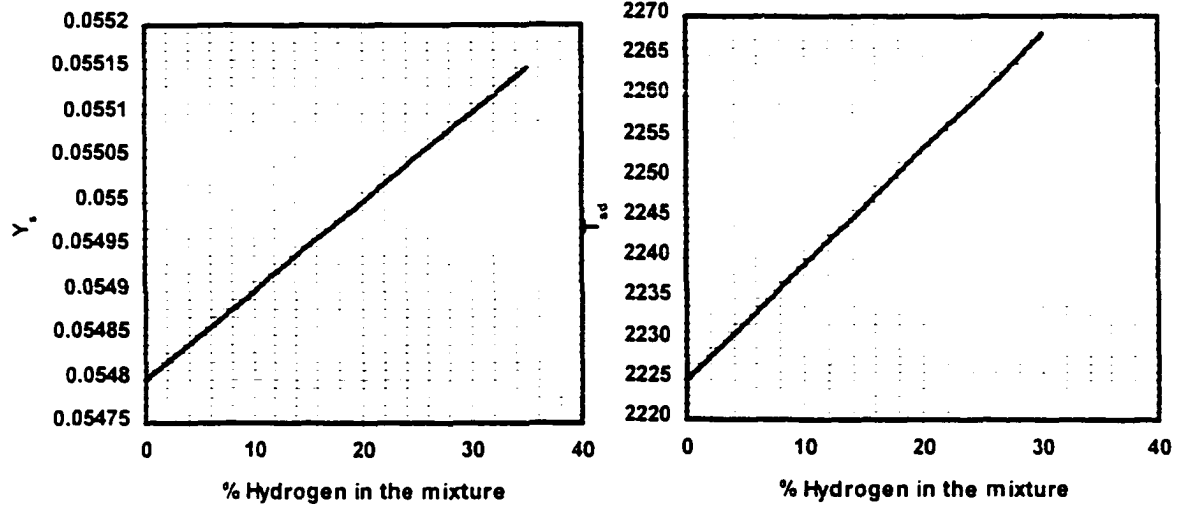
For $Re=8700$, 80-20% NG- H_2 , and $d_j=3.8$ mm

$$Fr_j = (47.15)^2 / (9.81 * 0.0038) = 59638$$



Flame Froude Number

$$Fr_f = \frac{U_j Y_s^{1.5}}{\left(\frac{T_{ad} - T_\infty}{T_\infty} g d_j \right)^{0.5} \xi^{0.25}}$$



Non-dimensional Flame Length

$$L^* = \frac{L_f Y_s}{d_j \xi^{0.5}}$$

For Re=8700, 80-20% NG-H₂, and d_j=3.8 mm

L_f = 0.804 m: Y_s=0.055 d_j= 0.0038m: ξ=0.45736

L* = 17.30

Appendix VII

Soot Concentration

Volumetric Soot Concentration (w_{soot})

The volumetric soot concentration of soot is calculated using the method described by Yagi and Iino (1960)

The spectral emissivity of a soot particle can be expressed as

$$w_{soot} = 1 - e^{-\frac{4.14wt}{\lambda}}$$

where

t = flame thickness

w = volumetric soot fraction (ml/ml)

λ = wavelength of the laser source

According to the Kirchoff's law, emissivity is equal to absorptivity

Applying this assumption

Absorptivity α = Emissivity ϵ of the soot particle

$$\text{Since } \alpha = \frac{I_i - I_e}{I_e}$$

where I_i = Intensity of the incident laser beam

I_e = Intensity of the emitted laser beam

Assuming that the soot material density is 2g/cc (Kamal, 1995) the soot concentration can be expressed as

$$w_{soot}(\text{g/cc}) = -2 \ln \left(\frac{I_o}{I_i} \right) \times \frac{\lambda}{4.14t}$$

Using equation the above for the experiment 80-20% NG-H₂ at Re 8700($x/d = 75$)

$$I_i = 65.82 \text{ mW}$$

$$I_e = 63.70 \text{ mW}$$

$$t = 0.0127 \text{ m}$$

$$\lambda = 635 \text{ nm}$$

$$w = -2 \times \ln\left(\frac{63.70}{65.82}\right) \times \frac{635 \times 10^{-9}}{0.0127} = 7.9 \times 10^{-7} \text{ g cc}$$