

**DETERMINATION OF FLAME CHARACTERISTICS IN
A LOW SWIRL BURNER AT GAS TURBINE
CONDITIONS THROUGH REACTION ZONE IMAGING**

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CHAPTER 1

EXPERIMENTAL METHODS AND CONSIDERATIONS

The current chapter details the facilities and apparatus used to study the flame characteristics in a Low Swirl Burner. The selection and implementation of diagnostic techniques used in this study are explained, as are data analysis methods used to process the acquired data.

1.1 LSB configuration

Two LSB configurations, A and B are tested for this study. Each LSB configuration is built around a swirler with an outer diameter, d_s of 38 mm (1.5 in). Other key dimensions of the swirlers tested for this work are presented in Table 1.1.

Initial testing aimed at velocity field mapping and flame imaging is conducted on Configuration A, while Configuration B is used for a later series of tests aimed at visualizing the flame structure. The design of these two configurations is discussed in further detail in what follows.

1.1.1 Configuration A

In this configuration, the reactants reach the swirler through a converging nozzle that decreases linearly in diameter from the inlet diameter of 102 mm (4 in) to the outer diameter of the swirler, 38 mm (1.5 in). The swirler leads to a constant area nozzle, and is located one diameter upstream of an abrupt area change. At the area change, the reactants expand from the 38 mm (1.5 in) diameter nozzle into a 115 mm (4.5 in) diameter combustion zone. The expansion ratio is chosen so as to avoid confinement effects on the centerline flame flow field.[1]

The main combustion zone begins at the dump plane and is enclosed by a GE

Table 1.1: *The dimensions of the swirlers used and the respective perforated plates are presented. Each swirler is referred to by its vane angle (as in “ S_{37° ”).*

Geometric parameter	Swirler	
	S_{37°	S_{45°
Swirler data		
Outer diameter, d_s , mm	38	38
Diameter ratio, $\frac{d_i}{d_s}$	0.66	0.66
Vane angle, α	37°	45°
Theoretical Swirl Number, S	0.48	0.64
Perforated plate data		
Open area, mm^2	155.97	156.98
Blockage, %	71.54	71.36
Plate thickness, mm	1.27	1.27
Hole pattern	1 - 8 - 16	1 - 8 - 16
Hole location (dia), mm	0 - 10.2 - 19.1	0 - 10.2 - 19.1
Hole diameter, mm	2.79 - 2.79 - 2.84	2.82 - 2.82 - 2.83

214 quartz tube. The quartz tube is 300 mm (12 in) long and 115 mm (4.5 in) in 24
diameter. The thickness of the quartz tube is 2.5 mm (0.1 in). Configuration A is 25
illustrated in Figure FIXME. 26

1.1.2 Configuration B 27

In this configuration, the reactants approach the swirler through a smoothly con- 28
toured nozzle with a high contraction ratio designed to inhibit the formation of thick 29
boundary layers. The swirler again leads to a constant area nozzle which is FIXME 30
diameters in length. Following this, the reactants enter the combustion zone. 31

Unlike in Configuration A, there is no dump plane or quartz tube to provide 32
confinement to the combustion zone. Further, in this configuration, the annular flow 33
is separately controlled from the central flow, which allows one to control the mass 34
flow split directly, if needed. Finally, this configuration allows for adjusting the level of 35
turbulence present in the inlet flow by use of a turbulence generator located upstream. 36

The details of Configuration B are shown in Figure FIXME.

1.2 High Pressure Test Rig

Each of the two configurations is housed in a separate high pressure testing rig with optical access to study the flame. These rigs consist of an air and fuel supply system, a pressure vessel with adequate optical access and an exhaust system. The details of each rig are discussed in the following sub-sections.

1.2.1 Test Rig A

Preliminary experiments involving velocity field mapping and flame imaging are conducted in Test Rig A, shown in Figure FIXME. Preheated air at about 500 K is drawn from external tanks and metered through an orifice flow meter. The air enters the inlet nozzle of the LSB through a 1.8 m (6 ft) long, 102 mm (4 in) diameter straight pipe section. Fuel (natural gas) is metered using another orifice flow meter and injected at the head of the straight pipe section. The straight pipe section allows for the flow to be fully developed, and fully premixed before the reactants enter the burner. The combustor pressure and temperature are measured at the head of the inlet nozzle by a pressure transducer and a thermocouple respectively. In addition, the upstream pressure and the pressure differential are measured at the air and fuel orifice flow meters. For the preheated air stream, the upstream temperature is also measured. The measurements are used to calculate the four primary flow parameters (combustor pressure, preheat temperature, reference velocity and equivalence ratio) for the LSB in real time. All measurements are monitored and recorded during the course of the experiment by a LabView VI.

The pressure vessel enclosing the combustor is designed to withstand pressures of up to 30 atm and is insulated from the combustor by a ceramic liner. Cooling for the pressure vessel and the quartz tube is provided by a flow of cold air introduced at

the head of the pressure vessel. Optical access to the combustor is provided through
four 150 mm (6 in) \times 75 mm (3 in) quartz windows located 90° apart azimuthally.
The view ports allow the combustor to be imaged from the dump plane to an axial
distance of 150 mm (6 in) downstream.

The exhaust from the combustor is cooled by circulating cold water through a
water jacket enclosing the exhaust pipe section. The length of the exhaust pipe
section is about FIXME. The exhaust pipe section terminates in an orifice plug to
provide the back pressure to the combustion chamber. Different diameter orifices
are used for each reference velocity condition to be tested. The exiting products are
finally released to the building exhaust system.

1.2.2 Test Rig B

FIXME

1.3 Diagnostics

1.3.1 Laser Doppler Velocimetry

The velocity field of the LSB is mapped using a TSI 3-component LDV system.
Three wavelengths (514 nm, 488 nm and 476 nm) are separated from the output of a
5 W Argon ion laser by an FBL-3 multicolor beam generator. The individual beams
are split into two coherent beams which are then focused to intersect and produce
interference fringes within an ellipsoidal measurement volume with dimensions of the
order of 100 μ m. For this purpose, two transceiver probes are mounted 90° apart
about the axis of the LSB. One transceiver probe focuses the 514 nm and 488 nm
beams in planes perpendicular to each other, while the second probe focuses the 476
nm beams orthogonal to the other two beams. Particles in the flow field crossing the
interference fringes scatter the laser light elastically and produce a sinusoidal signal
whose frequency is proportional to the velocity of the particle. The transceiver probes

collect this scattered light and each wavelength is detected separately by a PDM- 87
1000-3 three-channel photodetector module. The output from the photodetector is 88
processed by an FSA-3500-3 signal processor. The resulting three components of the 89
particle/flow velocity are recorded by the FlowSizer software. 90

Since the airflow is very sparsely populated by particles, the flow needs to be 91
artificially seeded to facilitate LDV measurements in a reasonable amount of time. 92
The seeding particles to be used and their mean diameter are decided by the char- 93
acteristics of the flow to be imaged. Since the LSB flow field is a reacting one, the 94
particles need to have high melting points. Further, the particles need to be small 95
enough to follow the flow closely and large enough or reflective enough to scatter light 96
efficiently in the measurement volume. Based on these requirements, commercially 97
available alumina particles with a mean particle diameter of $5\text{ }\mu\text{m}$ were chosen for this 98
study. In order to uniformly seed the flow, a novel seeding generator was designed as 99
described in Appendix A. The seeding particles were introduced slightly upstream of 100
the 1.8 m (6 ft) long straight pipe section in Test Rig A. 101

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CHAPTER 2

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CH PLIF SIGNAL MODELING AND VALIDATION

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2.1 Fluorescence Signal Intensity

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As described in Chapter FIXME 2, the excitation scheme used in this study produces 111
fluorescence through a three-step process. First, the CH radicals in the ground state 112
 $X^2\Pi, v = 0$ are excited by the incident radiation to the second electronically excited 113
state $B^2\Sigma^-, v = 0$. This excitation occurs near the R-bandhead and targets the 114
ground state CH radicals present in the rotational energy levels, $N = 5$ through 9. 115
The upper electronic state $B^2\Sigma^-, v = 0$ is nearly degenerate with the $A^2\Delta, v = 1$ 116
energy level. This leads to the population of the $A^2\Delta, v = 0, 1$ energy levels due to 117
collisional energy transfer. The resulting fluorescence collected is primarily the result 118
of three spontaneous transitions — $A \rightarrow X(1, 1)$, $A \rightarrow X(0, 0)$ and $B \rightarrow X(0, 1)$. 119
These transitions are shown in Figure FIXME. 120

The primary goal of this exercise of modeling the CH fluorescence signal intensity 121
is to gage the feasibility of using CH PLIF to study various premixed flames, rather 122
than to quantitatively calculate the amount of CH present in the flames. As such, 123
we are more interested in the order of magnitude of the PLIF signal, rather than the 124
absolute value of it. 125

The intensity of the CH fluorescence signal may be written as a function of the 126
amount of CH radicals present in the excited state and the probability of spontaneous 127
emission from said state. Symbolically, this may be written as shown in Equation 128
B.1. 129

$$S = nVA \quad (2.1)$$

In Equation B.1, S is the total number of photons emitted per unit time, n is the number of excited CH radicals in a unit volume, V is the volume from which the signal is observed. The Einstein coefficient for spontaneous emission, A represents the probability of spontaneous emission between the two involved energy states. The predicted signal intensity represents the total number of photons emitted in all directions. In reality, only a fraction of these emitted photons will be recorded by the collection system. This fraction is a function of the experimental setup and depends on the collection angle, the efficiency of the optics and the detector used to record the signal. This fraction is left out because our objective is only to predict the relative variation in the signal between various premixed flames.

This formulation of the signal intensity implicitly makes the following assumptions.

1. The fluorescence emission is predicted at steady state.
2. The collection volume is optically thin and an emitted photon is not reabsorbed within the flame itself. This is a reasonable assumption to make, since the flame thickness and the thickness of the laser sheet are both typically quite small.

As described earlier, an accurate model of the CH system should involve five energy levels — $X(0)$, $B(0)$, $A(1)$, $A(0)$, and $X(1)$ ¹. Such a model would also have to account for collisional transfers between each of these levels, in addition to spontaneous and stimulated transitions. The mathematical solution quickly becomes complicated and tedious. Further, it would involve several rate coefficients that have not been measured in experiments done so far.

¹In this notation, the letter represents the electronic energy level and the number in the parentheses represents the vibrational quantum number of the energy level

CHAPTER 3

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LSB FLAME CHARACTERISTICS

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In Chapter 2, we introduced the salient features of the Low Swirl Burner (LSB) flow field and discussed the mechanisms by which the LSB flame is stabilized. Further, various characteristics of the LSB flame that can be measured from flame images were outlined. To recapitulate, these are the flame location, flame shape and the flame structure. The first two are quantified by the flame standoff distance, X_f , and the flame angle, θ_f , respectively.

In the same chapter, we introduced the four flow parameters that describe an operating condition for the LSB — the combustor pressure, p , the preheat temperature, T , the mass-averaged inlet velocity (also called the reference velocity, U_0 , and the equivalence ratio of the premixed reactants, ϕ . We further introduced a geometric parameter — the angle of the vanes of the swirler, α , which affects the amount of swirl present in the flow field.

The LSB flame is imaged over a range of operating conditions and the effect of flow and geometric parameters on the reacting flow field is investigated. This results of the investigation are presented in this chapter.

3.1 Effect of reference velocity

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In typical gas turbine applications, varying the loading on the engine does not affect the reference velocity. However, since the reference velocity is a design parameter, the effect it has on the flame characteristics has implications for the design of future LSB-based gas turbine engines.

One of the key objectives of this thesis is to investigate how the LSB flame stabilization operates at high pressure conditions. The simple model described earlier

predicts a self-similar flow field for the LSB at all reference velocities. This implies
that the reference velocity will have no discernible impact on the flame standoff dis-
tance. This result is very desirable for gas turbine designers, since the flame location
and shape can be assumed to be constant. Limited testing conducted in earlier works
confirms this behavior at atmospheric pressure conditions with no preheat.

In order to verify the validity of this model at high pressure conditions in the
presence of substantial preheat, the LSB was operated at a pressure of 6 atm over a
range of reference velocities from 10 m/s to 40 m/s. For these tests, the S_{37° swirler
was used. In a parallel series of tests, the S_{45° swirler was tested at a pressure of 3 atm
at a reference velocities of 40 and 80 m/s. The location of the flame was measured
from CH* chemiluminescence images and the results are presented in Figure FIXME.

There is essentially no systematic variation in the flame standoff distance or the
flame angle for the low velocity, S_{37° tests. The increase in reference velocity continues
to produces a concomitant increase in the turbulent flame speed at the flame stabi-
lization location, negating any change in the flame's location. In other words, the flow
field appears to retain its self-similarity, even at elevated pressures and temperatures.

However, when the S_{45° swirler was tested at higher reference velocities, the flame
location shifted downstream sharply. This indicates potential limitations to the simple
flame stabilization model that may not predict the behavior of the LSB flame at
elevated pressures and temperatures, particularly at high reference velocities.

To examine the probable cause of this limitation more closely, consider the effect of
increasing the reference velocity on the turbulent combustion regime where the LSB
combustor operates. Previous studies have primarily operated the LSB in the flamelet
regime where the modified Damköhler model predicts the behavior of the turbulent
flame speed with reasonable fidelity. At elevated pressures, both the laminar flame
speed of the reactants, S_L and the flame thickness, δ_f are diminished. This places
the operating regime higher and more to the right on a Borghi diagram, as shown

in Figure FIXME. While previously, increasing the reference velocity did not affect the turbulent combustion regime, at elevated pressures, the flame is more likely to transition into the thin reaction zone. This transition causes a drop-off in the S_T/S_L plot and the turbulent flame speed no longer increases in step with the increased levels of turbulence. This results in the observed downstream shift of the high pressure LSB flame at high reference velocities.

3.2 Effect of preheat temperature

The preheat temperature of the reactants is a key flow parameter for the LSB due to two reasons. First, The temperature of the incoming flow directly affects its viscosity and consequently, the velocity field. Additionally, the rates of most chemical reactions in the flame zone are acutely sensitive to the temperature of the reactants. Thus, studying the effect of the preheat temperature on the LSB flame and flow field is important.

In order to explore this in greater detail, the velocity field of the combustor was mapped using Laser Doppler Velocimetry (LDV). The conditions were chosen to study the effect of increasing the preheat temperature on both reacting and non-reacting LSB flow fields. Further, the study includes both low and high reference velocity cases. The relevant flow parameters relating to these tests are presented in Table FIXME. All LDV tests were limited to atmospheric pressure conditions. Implementing the LDV technique at elevated pressures proved difficult due to beam steering issues, coupled with impractical turn-around times between successive runs.

The normalized centerline mean and rms axial velocity profiles for the three cases are presented in Figure FIXME. The abscissa represents the distance from a point called the virtual origin, X_0 . The virtual origin is defined as the imaginary location where the extrapolated linear axial velocity profile reaches the reference velocity in magnitude. The extrapolation is indicated in Figure FIXME by a dashed line.

As noted in Chapter FIXME 2, previous studies[2] reported that mean axial stretch — the normalized slope of the linear decay of axial velocity — at the inlet of the combustor was self-similar, regardless of the Reynolds number, Re of the operating condition. Further, it was reported that the velocity decay was steeper for reacting cases compared to non-reacting cases.

The results presented in Figure FIXME however, show that even though Cases 1 and 2 have similar Re , their mean velocity profiles have very different slopes. Further, the reacting and non-reacting cases (both at preheated conditions) have similarly steep slopes. This indicates that the mean axial stretch in the near field of the LSB flow field is a stronger function of the preheat temperature than Re . The presence of preheat results in increased viscosity that enhances the momentum transport in the radial direction. This causes the velocity decay to be steep for preheated cases, compared to cases without preheat.

These results suggest that holding S_T constant, at higher preheat temperatures, the flame would stabilize closer to the dump plane because of the faster velocity decay and reduced local flow velocities. In reality, a faster velocity decay would produce greater u' values and increase S_T , further causing the flame location to shift upstream. Furthermore, in view of the steep velocity profile, it may be anticipated that any changes in the stabilization location caused by perturbations in the local flow field (and hence or otherwise, the local turbulent flame propagation velocity) are likely to be of diminished magnitude in the presence of preheat. All of this leads to an intuitive result — the LSB flame behaves more stably at high preheat conditions.

3.3 Effect of swirler vane angle

As described in Chapter FIXME 3, the LSB swirlers tested for this study are designed to have the same mass flow splits. The S_{45° swirler has a higher vane angle, resulting in greater blockage to the flow passing through the annular section. In order to

compensate for this, the perforated plate covering the central section has slightly smaller holes. The net effect retains the same mass flow split as in the S_{37° swirler.

Earlier, in Chapter FIXME 2, we discussed how the swirler vane angle relates to the amount of swirl imparted to the incoming flow. According to Equation FIXME, a swirler with a higher vane angle will produce greater swirl in the reactants. Previous work in swirl combustion[3, 4] has pointed out that increased swirl shortens the flame by enhancing the swirl-induced radial pressure gradients. The data acquired in the present investigation is in agreement with this observation. Operated at identical inlet conditions, the S_{45° swirler stabilizes a flame closer to the dump plane and with a larger flame angle compared to the S_{37° swirler.

This result highlights an interesting trade-off for the designers of LSB-based gas turbine engines. The S_{45° flame is located further upstream and has a more concentrated region of heat release. This enhances the strength of the toroidal recirculation zone near the dump plane, which may be powerful enough under certain conditions (as we shall see in the Section 3.4) to even cause the flame to attach itself to the lip of the inlet. All of this means that the S_{45° flame is more stable and will resist perturbations in the incoming flow better than the S_{37° flame. However, the presence of a strong recirculation zone in the flow field of the S_{45° swirler will entrain more hot products and retain them longer near the zone of heat release. This is a recipe for the production of thermal NO_x . While no emission measurements were made as part of this study, it may be reasonably anticipated that the NO_x performance of the S_{45° swirler is worse than the S_{37° swirler. The trade-off for gas turbine engine designers is thus between flame stability and emissions performance.

3.4 Effect of equivalence ratio

The LSB is primarily intended for fuel-lean operation in order to utilize its low NO_x emission performance. As a result, most of the testing was done as close to the target

ϕ of 0.56 as possible. However, limited testing was done at 12 atm at both a slightly rich ($\phi \approx 0.58$) and a slightly lean ($\phi \approx 0.53$) condition to explore the sensitivity of the LSB flame to limited changes in equivalence ratio. The S_{45° swirler was used for these tests. The corresponding averaged and Abel-deconvoluted flame images are presented in Figure FIXME.

Two characteristics of the flame are immediately obvious from these images.

First, the zone of heat release, marked by the region from which CH^* chemiluminescence is observed, is increasingly compact at fuel-rich conditions. Virtually all other flame images acquired at a leaner condition show a long flame, with the heat release distributed over the entire visible area of the combustor. The compactness of the heat release zone indicates potentially poor NO_x performance at these conditions.

Second, the fuel-rich flame brush can be observed to wrap around and anchor itself on the dump plane. This is particularly observable in the Abel-deconvoluted image. The attached region is not as bright as the rest of the flame brush, indicating that the flame may be attaching itself intermittently. This intermittent behavior can be confirmed from the instantaneous images where it is visible on some of the acquired images, but not others. This behavior was alluded to in Section 3.3 as being the result of the enhanced toroidal recirculation zone produced by this swirler. Thus, the intermittent attachment of the flame to the inlet indicates the increased importance of the toroidal recirculation zone in stabilizing the flame.

It should be noted that the reliance on a toroidal recirculation zone to anchor the flame to the inlet is one of the primary flame stabilization mechanisms used by traditional swirl combustors. Thus, LSB swirlers with high vane angles tend to behave like traditional swirl combustors at fuel-rich conditions.

3.5 Effect of combustor pressure

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In a typical gas turbine application, the combustor pressure is expected to vary directly with the loading of the engine. Like the preheat temperature, the combustor pressure affects the LSB flame both through the fluid mechanics of the flow and the kinetics of the chemical reactions in the flame. The effect of the combustor pressure on the fluid mechanics of the LSB flow field can be captured by its effect on the Reynolds number of the flow. However, as noted in Section 3.2, the Reynolds number may not be an important parameter for the LSB, particularly in the near field where the flame stabilization occurs. On the other hand, the effect of the combustor pressure on the reactions occurring in the flame are more dominant. Increasing the combustor pressure results in a lower laminar flame speed and reduced flame thickness for methane-air flames. According to the modified Damköhler model discussed earlier, the reduced laminar flame speed should have little or no effect on the flow field, since the contribution from S_L in Equation FIXME is vanishingly small, even at the lowest reference velocities of our test conditions. However, as suggested by our discussion in Section 3.1, the validity of the simple model at elevated pressure conditions is questionable.

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In order to resolve the uncertainties regarding how the LSB flame responds to combustor pressure, the flame was imaged over a range of operating conditions from 3 atm to 12 atm. For these tests, the reference velocity and the equivalence ratio were held constant. However, the temperature of the reactants continues to increase with pressure. The reason for this was discussed in Chapter FIXME 3 and is attributable to the reduced heat losses in the connecting pipes at the high flow rates required to pressurize the LSB. The flame location and shape inferred from the flame images are presented in Figure FIXME.

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At low to moderate pressures, the flame location is nearly invariant for S_{37° , but

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moves upstream for the S_{45° cases. This observation is explained as follows. The flame stabilization location for the S_{45° swirler is closer to the dump plane compared to the S_{37° swirler. This results in enhanced heat transfer to the dump plane and consequently to the incoming reactants. This feedback is even more effective as the temperature of the incoming reactants increases. This causes the upstream shift of the S_{45° flame, while the S_{37° flame is less affected by these processes.

At high pressures, however, both flames are observed to move downstream, despite the increasing preheat temperatures. The apparent decrease in the turbulent flame speed at these conditions is an unexpected result and the modified Damköhler model is insufficient in accounting for this observation. Figure FIXME also shows that the flame angle for both cases decreases slightly with pressure. This suggests that the turbulent flame speed was consistently decreasing with pressure. In light of this, the nearly constant location of the S_{37° flame could be attributed to the effects of increasing combustor pressure and preheat temperature nearly canceling each other out at the lower pressures.

3.6 Flame structure

APPENDIX A

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SEEDER DESIGN

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A new seeder was designed for use in high pressure implementations of diagnostic techniques like Laser Doppler Velocimetry (LDV), Particle Image Velocimetry (PIV), etc.

The previous design, as shown in Figure FIXME, was a fluidized bed seeding generator. Seeding particles in a cylindrical vessel are fluidized by an air-turbine vibrator. Air is introduced into the vessel in the form of two opposing jets directed tangentially to produce a small amount of swirl in the flow field. Particles are picked up by the air flow and the swirl aids in separating the heavy/coagulated clumps of seeding particles by centrifugal acceleration.

This design had several shortcomings. First, it is observed that the seeding density of the seeded flow generally decreases over time, even if the seeding particles have not been depleted. The seeding particles tend to coagulate over time, due to the buildup of moisture, static charge, etc. In such cases, the vibrator can no longer effectively fluidize the particles. Further, the tangential introduction of the air flow preferentially depletes particles near the walls of the container, leaving the center relatively undisturbed. The cumulative effect of these phenomena diminishes the effectiveness of the seeder.

Second, the fluidized bed requires a minimum amount of seeding particles to function effectively. This requires the seeder to be refilled even before all the seeding particles are consumed.

Third, when designed for high pressure applications, the seeder will become quite heavy due to flanges and other fittings. Such a setup cannot be easily fluidized using a reasonable-sized air-turbine vibrator.

The new seeder design is shown in Figure FIXME, and resembles a funnel with a 371
swirler located halfway up the stem. A perforated base plate holds the swirler and the 372
seeding particles in the conical section of the swirler. Due to the steep angle of the 373
sides of the conical section, the seeding particles continuously collapse into the central 374
section. This negates any need for vibrating the system. Air is introduced from the 375
bottom of the seeder and enters the vessel by passing through the swirler. Since all 376
the air enters this way, there is a considerable amount of swirl in the resulting flow 377
field, Heavy/coagulated seeding particles are flung outward, while lighter particles are 378
carried with the air. After a sufficient distance to allow for the cyclonic separation 379
to be effective, the seeded air passes through another perforated plate which further 380
limits the presence of large clumps of particles. The exiting air is now spatially and 381
temporally uniformly seeded. 382

APPENDIX B

CH PLIF QUENCHING MODEL

In order to calculate the intensity of the quenched CH PLIF signal in a flame, an improved model of the CH system was constructed and analyzed. According to this new model, CH radicals from the X ground state are excited to the $B(0)$ upper state. This is followed by collisional transfer to the $A(1)$ and $A(0)$ states. The transfer between the nearly degenerate $A(1)$ and $B(0)$ states is partially reversible. The transfer between $B(0)$ and $A(0)$ is not reversible. This is followed by spontaneous emission as CH radicals transition from the A states to the X state. This results in a pseudo-three-level model as shown in Figure FIXME.

Figure FIXME indicates the rates of the various processes discussed. The subscripts 0, 1 and 2 represent the electronic energy levels X , A and B respectively. Processes involving the $A(0)$ state are differentiated from those involving the $A(1)$ state by a prime ($'$). With the exception of the nearly degenerate $A(1)$ and $B(0)$ states, most collisional excitation steps are neglected due to their low probability.

In this formulation, the signal intensity of the CH PLIF emission is given by Equation B.1.

$$S = (n_1 A_{10} + n'_1 A'_{10} + n_2 A_{20})V \quad (\text{B.1})$$

The spontaneous emission coefficients, A_{10} , A'_{10} and A_{20} are obtained from various published papers[5, 6, 7]. The values used for this analysis are presented in Table B.1.

Equations B.2–B.4 describe the time variation of the number density of CH radicals in each excited state.

Table B.1: *The coefficients of spontaneous emission for transitions in the CH system are provided.*

Transition	Symbol	A, s ⁻¹
$B \rightarrow X(0, 0)$	A_{20}	2.963×10^6
$A \rightarrow X(1, 1)$	A_{10}	1.676×10^6
$A \rightarrow X(0, 0)$	A'_{10}	1.832×10^6

$$\frac{dn_1}{dt} = -(A_{10} + Q_{10} + R_{12})n_1 + R_{21}n_2 \quad (\text{B.2})$$

$$\frac{dn'_1}{dt} = -(A'_{10} + Q'_{10})n'_1 + R'_{21}n_2 \quad (\text{B.3})$$

$$\frac{dn_2}{dt} = W_{02}n_0 + R_{12}n_1 - (A_{20} + Q_{20} + R_{21} + R'_{21})n_2 \quad (\text{B.4})$$

At steady state, the rate of change of the number density is minimal. Under this assumption, the LHS of Equations B.2–B.4 can be set to zero. This results in a closed set of linear equations in terms of the populations of the upper states. This set of equations is presented in Equation B.5.

$$\begin{bmatrix} A_{10} + Q_{10} + R_{12} & 0 & -R_{21} \\ 0 & A'_{10} + Q'_{10} & -R'_{21} \\ -R_{12} & 0 & A_{20} + Q_{20} + R_{21} + R'_{21} \end{bmatrix} \begin{bmatrix} n_1 \\ n'_1 \\ n_2 \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ W_{02}n_0 \end{bmatrix} \quad (\text{B.5})$$

The solution to Equation B.5 is shown in Equations B.6–B.8.

$$n_1 = \frac{R_{21}}{(A_{10} + Q_{10} + R_{12})(A_{20} + Q_{20} + R_{21} + R'_{21}) - R_{12}R_{21}} W_{02}n_0 \quad (\text{B.6})$$

$$n'_1 = \frac{(A_{10} + Q_{10} + R_{12})R'_{21}}{(A'_{10} + Q'_{10})((A_{10} + Q_{10} + R_{12})(A_{20} + Q_{20} + R_{21} + R'_{21}) - R_{12}R_{21})} W_{02}n_0 \quad (\text{B.7})$$

$$n_2 = \frac{(A_{10} + Q_{10} + R_{12})}{(A_{10} + Q_{10} + R_{12})(A_{20} + Q_{20} + R_{21} + R'_{21}) - R_{12}R_{21}} W_{02}n_0 \quad (\text{B.8})$$

These expressions can be further simplified by noting various observations made 410
in studies of the CH system. For instance, previous work[8, 9] has reported that the 411
B state is slightly (about 1.3 times) more prone to quenching compared to the *A* 412
state. We can thus make the following assumptions. 413

$$Q_{10} = Q'_{10} = Q \quad (\text{B.9})$$

$$Q_{20} = 1.3Q \quad (\text{B.10})$$

Next, it has been reported[10] that the electronic energy transfer rate from *B* to 414
A state accounts for 0.24 times the total collisional removal from the *B* state. 415

$$\frac{R_{21} + R'_{21} - R_{12}}{Q_{20} + R_{21} + R'_{21} - R_{12}} = 0.24 \quad (\text{B.11})$$

$$\therefore \frac{R_{21} + R'_{21} - R_{12}}{Q} = 0.4105 \quad (\text{B.12})$$

We further know[9, 10] that the collisional transfer from the *B*(0) energy level 416
populates the nearly degenerate *A*(1) level about four times faster than the *A*(0) 417

level.

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$$\frac{R_{21} - R_{12}}{R'_{21}} = 4 \quad (\text{B.13})$$

Finally, it was observed[9] that the rate of forward transfer from $B(0)$ to $A(1)$ is about 1.6 times the reverse process.

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$$\frac{R_{21}}{R_{12}} = 1.6 \quad (\text{B.14})$$

Collating Equations B.12–B.14, we obtain a closed set of linear equations. This can be solved to eliminate R_{21} , R_{12} and R'_{21} in terms of Q as shown in Equation B.15.

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$$\begin{bmatrix} R_{21} \\ R'_{21} \\ R_{12} \end{bmatrix} = \begin{bmatrix} 5.1966 \\ 0.4872 \\ 3.2479 \end{bmatrix} Q \quad (\text{B.15})$$

Substituting Equations B.9, B.10 and B.15 into Equations B.6–B.7 leads to simplified expressions for the populations of the upper electronic states purely as a function of the respective Einstein coefficients and the collisional quenching rate. These are presented in the following Equations B.16–B.18.

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$$n_1 = \frac{5.1966Q}{(A_{10} + 4.2479Q)(A_{20} + 6.9838Q) - 16.8780Q} W_{02} n_0 \quad (\text{B.16})$$

$$n'_1 = \frac{0.4872Q(A_{10} + 4.2479Q)}{(A'_{10} + Q)((A_{10} + 4.2479Q)(A_{20} + 6.9838Q) - 16.8780Q)} W_{02} n_0 \quad (\text{B.17})$$

$$n_2 = \frac{(A_{10} + 4.2479Q)}{(A_{10} + 4.2479Q)(A_{20} + 6.9838Q) - 16.8780Q} W_{02} n_0 \quad (\text{B.18})$$

The quenching rate, Q of excited CH radicals is calculated by using the quenching cross-sections of various species. The quenching cross-sections are measures of the effectiveness of each collision between a given species and an excited CH radical. The

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effectiveness of the collision also depends on the velocity of collision between the two species, g_j and the abundance of the species, n_j . This relationship is formalized in Equation B.19.

$$Q = \sum_j g_j \sigma_j n_j$$

$$Q = \sum_j \sqrt{\frac{8kT}{\pi\mu_j}} \sigma_j \frac{pN_A}{RT} X_j \quad (\text{B.19})$$

In Equation B.19, μ_j represents the reduced mass of the colliding CH- j molecules, p is the pressure, N_A is Avogadro's Number, R is the Universal Gas Constant, T is the temperature, and X_j is the mole fraction of species j . The mole fractions of the various species in the flame, as well as the temperature across the flame are obtained from Chemkin simulations. The expression for the reduced mass is given in Equation B.20.

$$\mu_j = \frac{m_j m_{CH}}{m_j + m_{CH}} \quad (\text{B.20})$$

The quenching cross-sections of various species are obtained from various published papers[11, 12, 13] and are functions of temperature. The functional forms used in this study are presented in Table B.2.

The term $W_{02}n_0$ in Equations B.16–B.18 represents the rate of pumping of the ground state CH radicals. The current excitation scheme targets multiple transitions in the R-bandhead. The pumping rate for each transition is the product of the number of CH radicals present in the appropriate level, the Einstein absorption coefficient for that energy level, B_i and the amount of laser energy available at the appropriate frequency, E_i . As a result, the term is actually a summation over the individual energy levels. Equation B.21 presents this symbolically.

Table B.2: *The functional form of the quenching cross-sections of various species with CH are provided.*

Species	$\sigma, \text{\AA}^2$
H ₂	$6.1 \exp(-686/T)$
H	$221T^{-0.5} \exp(-686/T)$
O ₂	$8.61 \times 10^{-6} T^{1.64} \exp(867/T)$
OH	$221T^{-0.5} \exp(-686/T)$
H ₂ O	9.6
CH ₄	$52.8T^{-0.5} \exp(-84/T)$
CO	8.31
CO ₂	$8.67 \times 10^{-13} T^{3.8} \exp(854/T)$
C ₂ H ₆	13.4
N ₂	$1.53 \times 10^{-4} T^{1.23} \exp(-522.1/T)$
C ₃ H ₈	22

$$\begin{aligned}
 W_{02}n_0 &= \sum_i B_i I_i n_i \\
 W_{02}n_0 &= \sum_i B_i \frac{E_i}{A_c} \frac{p N_A X_{CH}}{RT} f_i
 \end{aligned} \tag{B.21}$$

Table B.3 presents the values of B_i for the transitions targeted by the current 449
excitation scheme.[14] Assuming a Gaussian line shape for the laser, and using the line 450
strengths from LIFBASE, the relative amount of energy absorbed by each transition 451
can be calculated. These values are also presented in Table B.3. 452

In Equation B.21, A_c is the area of cross-section of the laser beam and f_i is the 453
Boltzmann fraction of the population at the energy level i . The expression for the 454
Boltzmann fraction at the energy level corresponding to the vibrational quantum 455
number v and rotational quantum number J is given in Equation B.22. 456

$$f(v, J) = \frac{\exp\left(\frac{-hcE_v(v)}{kT}\right) (2J+1) \exp\left(\frac{-hcE_r(v, J)}{kT}\right)}{Q_{rv}} \tag{B.22}$$

Table B.3: *The coefficients of absorption for selected transitions in the CH X($v = 0$) system are provided.*

N''	λ , nm	B , $\text{m}^2\text{J}^{-1}\text{s}^{-1}$	E (normalized)
R1			
5	387.2698	7.677×10^9	0.0568
6	387.1899	7.665×10^9	0.1706
7	387.1677	7.610×10^9	0.1483
8	387.206	7.519×10^9	0.1479
9	387.308	7.397×10^9	0.0126
R2			
5	387.2289	7.539×10^9	0.1080
6	387.1549	7.569×10^9	0.1128
7	387.1371	7.539×10^9	0.0841
8	387.1786	7.464×10^9	0.1311
9	387.283	7.354×10^9	0.0279

The vibrational energy, $E_v(v)$ of a level is calculated according to Equation B.23, 457
while the rotational energy, $E_r(v, J)$ is calculated according to Equation B.24. 458

$$E_v(v) = \omega_e \left(v + \frac{1}{2}\right) - \omega_e x_e \left(v + \frac{1}{2}\right)^2 + \omega_e y_e \left(v + \frac{1}{2}\right)^3 - \omega_e z_e \left(v + \frac{1}{2}\right)^4 \quad (\text{B.23})$$

$$E_r(v, J) = \left\{ B_e - \alpha_e \left(v + \frac{1}{2}\right) \right\} J(J+1) - \left\{ D_e + \beta_e \left(v + \frac{1}{2}\right) \right\} J^2(J+1)^2 \quad (\text{B.24})$$

The spectroscopic constants in Equations B.23 and B.24 are found in literature[15] 459
and are provided here in Table B.4. 460

The rovibrational partition function, Q_{rv} is a summation over all available vibra- 461
tional and rotational levels in the particular electronic state. For the ground state 462
of the CH molecule, there are five available vibrational quantum numbers, $v = 0$ to 463
 $v = 4$. The CH system falls under Hund's Case b and hence, the appropriate rota- 464
tional quantum number to use is N . Each vibrational level has twenty-two possible 465
values for N from $N = 1$ to $N = 22$. For each rotational quantum number N , there 466
are two possible values of J given by $N \pm \frac{1}{2}$. 467

Table B.4: *Spectroscopic constants for the CH X²Π level are presented.*

Constant	Value, cm ⁻¹
ω_e	2860.7508
$\omega_e x_e$	64.4387
$\omega_e y_e$	0.36345
$\omega_e z_e$	-1.5378×10^{-2}
B_e	14.459883
α_e	0.536541
D_e	1.47436×10^{-3}
β_e	-2.530×10^{-5}

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