Quantitative Hydroxyl (OH) Concentration Calibration by Use of a Flat Flame Burner, Thermocouple and Planar Laser Induced Fluorescence (PLIF) System

A Thesis Presented

by

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to

The Department of Mechanical and Industrial Engineering

In partial fulfillment of the requirements
for the degree of

Master of Science

in

Mechanical Engineering

in the field of

Thermofluids

Northeastern University
Boston, Massachusetts

August, 2009
Abstract

A quantitative concentration calibration system has been constructed for a Planar Laser Induced Fluorescence (PLIF) system that was just recently established as being capable of accurately imaging the qualitative hydroxyl (OH) concentration in transient flames within a cylindrical constant volume vessel at desired radii. In conjunction with the PLIF system, this calibration system includes a flat flame burner, a thermocouple and positioning stand, gas flow meters, and control valves.

A calibration procedure was developed in order to apply absolute concentrations of OH to otherwise relative measurements obtained from the combustion vessel and to form a basis for future experiments. This procedure involved measuring the temperature in the hot gas region of a hydrogen-air flame, correcting for radiation losses, calculating the equilibrium OH concentrations at the measured temperature and applying these values to fluorescence images taken of the burner. Preliminary results are shown and discussed in relation to experimental uncertainties.

Modifications made to the existing system that significantly improved performance and enabled the capture of satisfactory fluorescence images are discussed.
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Acknowledgements

I would like to express my appreciation to my thesis advisor, Hameed Metghalchi. Thank you for giving me this opportunity to study under you in the BS/MS program here at Northeastern University. Thank you for your continued support and inspiring my enthusiasm in my work.

James Keck, thank you for always providing thoughtful discussions and advice to me and the rest of the laboratory. Your immense knowledge of combustion has helped me to learn a great deal.

I would like to extend a special thanks to my colleague, Kian Eisazadeh Far. You helped introduce me to this research and I have learned a lot from you. On a daily basis you answered any questions I had, helped me with experiments when needed and provided general guidance and encouragement.

John Doughty, thank you for the support that you provide to the laboratories in the basement of Forsyth and the rest of the MIE department to help them run smoothly. I appreciated the humor and insight you have offered me while helping me on various projects. Thank you to the MIE department and my combustion laboratory colleagues, especially Joyce Crane and Mimmo Elia for all your assistance. To the Young Scholars Cassandra Fach and Roger Cruz, thank you for helping me run experiments. I hope that you learned a lot during the time you spent here and that it has helped to inspire you.

I am especially grateful for the immense technical support that was provided by Spectra-Physics and LaVision. Chris Jaska and Javier of Spectra-Physics, you helped to provide an understanding of the Nd:YAG laser and answered all of my questions. Callum Gray and Stephan Wissel of LaVision, thank you for giving me clear guidance and insight as to how the system could be improved. Without the help from all of you, our PLIF system would not be where it is at today.

Last but certainly not least, I would like to say that I am extremely appreciative of my parents, friends and family for constantly giving me encouragement and support in all my endeavors.
Chapter 1 – Background

1.1 Laboratory Research

The combustion laboratory at Northeastern University located in the basement of Forsyth has been involved in many combustion studies involving laminar burning velocities of flames at high temperature and pressure, transient flame structure imaging in a cylindrical vessel and the effects of spark electrode geometry on flame kernels [1, 2]. More recently, flame structure and laminar burning speeds of JP8 in high temperatures and pressures have been studied because of the North Atlantic Treaty Organization’s interest in the “Single Fuel Concept” [3]. As an extension to this research, the combustion laboratory has acquired a laser spectroscopy system with the goal of studying flame structure by measuring intermediate species’ concentrations. An increased understanding of species concentrations will help in the development and validation of chemical kinetics computer models.

Much of the work done by a previous graduate student had been concentrated in validating the operation of the laser spectroscopy system, using it to capture the fluorescence of hydroxyl (OH) in a transient premixed methane-air flame in a cylindrical constant volume vessel. This work was successful in demonstrating the system’s basic functionality. The objective of this current research is to calibrate the data obtained in the cylindrical vessel to absolute concentrations of OH. This can be done by imaging the fluorescence of a well-defined, laminar flame, such as one achieved by a flat-flame burner. Flat flame burners have been used throughout combustion research because they produce a well-defined and controllable flame [4-6]. The properties of these flames are more readily calculated when compared with many other combustion events. When certain properties of these flames, namely the OH concentration, are combined with fluorescence pictures of the same flame, the colors of the image can be correlated to represent a specific concentration. Thus, the colors of the images captured using the cylindrical vessel represent absolute concentrations of OH and not just relative
fluorescence intensities. This process forms the basis of the concentration calibration, and when completed, the results can be extended to various other combustion processes.

The purpose of this thesis is to present preliminary results of the concentration calibration and explain its methodology and procedure to future users. First, the theory behind this work will be discussed, followed by an overview of the experimental design, including an in-depth description of each device, system improvements and the calibration procedure. Finally, preliminary results, conclusions and recommendations for future work will be discussed.

1.2 Theory

The planar laser induced fluorescence (PLIF) method is thoroughly explained in [1, 7], however a quick overview is presented here. When laser radiation is tuned to specific wavelengths, it will excite certain species (molecules) within a flame to a higher energy level. Fluorescence occurs when this excited state decays and emits radiation of a longer wavelength than the incident laser radiation. In the atmospheric pressure flame created by the burner, quenching is negligible and the fluorescence signal is proportional to the OH concentration [8]. If this OH concentration can be calculated, then a direct relationship between the fluorescence signal and OH can be found, forming the basis of a concentration calibration.
A very thorough overview of the PLIF system at Northeastern University was provided in Raymond Andrews’ Masters Thesis [1]. It is suggested that any operator review this as well as the manuals for each piece of equipment to gain a proficient understanding of the safety precautions, specifications and operation procedures.

2.1 Safety Precautions

The Nd:YAG is a class IV laser device and is a safety and fire hazard. Extreme caution should be exercised by the user when operating this and the dye laser. Direct and even indirect laser radiation can be harmful. The dye solution that is used as a lasing medium also presents a health risk and should be prepared over an absorbent medium, like BenchGuard. Chemical resistant gloves should be worn and other exposed skin covered. This section is in no way a full review of all safety precautions. The Nd:YAG and Dye laser user manuals should be consulted before beginning any work with the system [9, 10].

2.2 Laboratory Equipment

A representation of the experimental system is shown in Figure 1 and is identical to the one presented in Raymond Andrews’ Thesis with the exception of a few modifications. The main difference is the replacement of the cylindrical constant volume vessel with a flat flame burner.
The system works in much the same way but without the complication of a combustion timing system since fluorescence on the burner can be imaged continuously. The flat flame burner is supplied with fuel, oxidizer and an inert shield gas from tanks of compressed gas via a metering system. Needle valves enable the precise control of the mixture ratio. A laser pulse that is generated in the Nd:YAG pump and emitted from the Sirah dye laser passes through the flame and hot gas region over the burner as the Intensified Charge-Coupled Device (ICCD) camera images the fluorescence. A computer installed with software developed by LaVision is used to control the laser and camera, record and process images and control the electronic system timing.

2.2.1 Gas Metering System
The gas metering system consists of three tanks of compressed gas connected to a control board which houses the flow meters, needle valves and static mixer. Figure 2 presents a basic diagram of the current system. The fuel and oxidizer gas lines have shut-off valves
to isolate the flat flame burner when other experiments are run on different laboratory equipment.

![Gas Metering System schematic.](image)

The compressed gas is fed through three Omega Engineering, Inc. FMA-A2300 series mass flow meters. These meters use a “capillary type thermal technology to directly measure mass flow” which prevents the need for any temperature or pressure corrections. Although they are mass flow meters, the display is read in Standard Liters Per Minute (SLPM). SLPM are volumetric flow units that are referenced to specific standard conditions: Omega uses 21°C and 1 atmosphere (atm) to calibrate their meters [11]. So regardless of what the actual temperatures and pressures are, the meter reading always refers to these standard conditions. Due to this feature, it is very easy to calculate the mole flow rate based on the ideal gas law:

\[
\frac{n}{\text{min}} \left[ \text{mol} \right] = \frac{P}{R \cdot T} \left( \frac{Q}{K_{\text{cal}}} \right) \quad \text{Eq. (1)}
\]

where \( P \) is absolute pressure in kPa, \( Q \) is the volumetric flow rate displayed on the meter in SLPM, \( R \) is the universal gas constant and \( T \) is the temperature in Kelvin. \( K \) and \( K_{\text{cal}} \)
are conversion factors that are used when any gas other than the calibration gas is used. These factors are listed in the back of the manual for many different gases. The meters are calibrated to a standard using nitrogen, which is $K_{\text{cal}} = 1.00$; hydrogen is $K=1.01$ and nitrogen is $K=1.00$ [11]. The equivalence ratio,

$$\phi = \frac{n_{\text{fuel}}/n_{\text{oxidizer}}}{(n_{\text{fuel}}/n_{\text{oxidizer}})_{\text{st}}} \quad \text{Eq. (2)}$$

can easily be calculated once the mole flow rate is known for both air and hydrogen.

Downstream from the flow meters are Swagelok Integral Bonnet Needle Valves, which are used to accurately adjust the flow rate in each gas line. These valves were ultimately chosen because they provided the smoothest operation and greatest control over several other needle valves that were tested. The fuel and oxidizer are then passed through a static mixer and poppet valve before entering the flat-flame burner. The static mixer consists of a tube with various spokes that intrude upon the flow to create turbulence that ensures sufficient mixing. The poppet valve is used as a deterrent for flashback.

### 2.2.2 Flat Flame Burner

The burner is a McKenna Flat Flame Burner made by Holthuis & Associates and is depicted in Figure 3. It is constructed with a steel housing, sintered bronze burner plug and shroud ring and a copper cooling coil. The overall height and diameter, excluding the inlet and outlet tubes, are 60 mm and 120 mm, respectively. The sintered bronze burner plug is roughly 60 mm in diameter and the shroud ring has a 75 mm outer diameter. The center chamber is used for the oxidizer-fuel mixture and the outer ring is for the shield gas. The sintered bronze plug and ring help distribute an even flow of the mixture and shroud gas as well as minimizing the chance of flashback. The cooling circuit is designed to minimize any radial temperature gradient and provide additional assurance against flashback [12].
The shield gas is important because it stabilizes the flame above the plug and minimizes the entrainment of air from the surrounding environment. It is possible to run experiments without the shroud gas; however all of the results presented in this paper included it.

### 2.2.3 Thermocouple and Stand

A bare wire Type B thermocouple from Omega was used for all the temperature measurements and has a bead and wire diameter of 0.36 mm and 0.20 mm, respectively. Type B thermocouples (Pt 6% Rh/Pt 30% Rh) have one of the highest limit-of-operation temperatures within an oxidizing atmosphere, making them useful in flame temperature measurements. One disadvantage however, is that Omega doesn’t have a Type B thermocouple with a smaller bead size. The thermocouple leads were isolated using a two-hole alumina ceramic insulator and held in place by a clamp attached to a high precision “Y-Z Axis Metric Stage” from Edmund Optics. A picture of this set-up can be seen in Figure 4. This translation stage can be positioned up to an accuracy of 0.1 mm and allows the thermocouple to be moved back and forth across the burner in one direction or axially to and from the burner’s surface.
2.2.4 Pump Laser

A Spectra-Physics Quanta-Ray Lab-190 Laser is used to produce the initial pump laser beam. A generalized diagram is shown in Figure 5.

Figure 4 – Thermocouple Stand and Burner in the experimental set up.

Figure 5 – Spectra-Physics Nd:YAG (pump) laser schematic, adapted from [1].
The Lab-190 uses a flash lamp to optically stimulate neodymium-doped yttrium aluminum garnet (Nd:YAG) in the crystal’s red and near infrared absorption bands. This absorption causes a population inversion, depicted in Figure 6. Atoms in the crystal are excited from energy level $E_1$ to $E_4$ by the flash lamp; however the atoms are unstable in this elevated level.

![Figure 6 – Depiction of population inversion, adapted from [9].](image)

Since there is a greater transition probability to $E_3$ than back to $E_1$ the atoms quickly decay to $E_3$. This third level is metastable, so the atoms remain at this level for a relatively long amount of time, roughly $230 \mu$s. From $E_3$, there are several other energy levels (not pictured) that the atoms could decay to, the most probable being $E_2$. The transition from $E_3$ to $E_2$ is called the lasing transition because this is where the fundamental wavelength of 1064 nm is generated. The $E_2$ state is also unstable, so once atoms decay from $E_3$, they quickly transition down from $E_2$ back to $E_1$. This behavior causes the population of atoms in the third energy level to grow larger than that of the second level, hence the population inversion. This inversion is what allows light passing through the crystal to be amplified, with a larger inversion producing a larger amplification [9].
The pump laser also uses a Q-switch to produce short bursts of high intensity beams. Without it, the length of the laser pulse would be similar to the length of the flash lamp and have a low maximum power. The Q-switch allows the Nd:YAG crystal to reach the maximum population inversion at which point the switch quickly opens causing a quick release of energy, forming a beam that is less than 10 ns long and a peak optical power of tens of megawatts. This high powered beam is then sent through several non-linear conversion processes. Two such processes, frequency doubling and frequency mixing, occur in the Nd:YAG’s Harmonic Generator (HG) which houses potassium dideuterium phosphate (KD*P) crystals. Depending on the configuration of these crystals, the HG can be used to either polarize the fundamental wavelength (1064 nm) or generate the second harmonic (532 nm), third harmonic (355 nm) or fourth harmonic (266 nm). The second and third harmonics are often used with dye laser systems because they have high dye conversion efficiency. Finally, just before the beam exits the Nd:YAG, it is reflected by two Dichroic mirrors which are designed to reflect only specific wavelengths of light and allow everything else to pass through it [9].
2.2.5 Dye Laser

The dye laser is a Sirah Precision Scan SL Dye Laser. A simplified diagram of the dye laser is shown in Figure 7.

![Figure 7 – Sirah Dye Laser schematic, adapted from [1].](image)

The dye laser works on similar principles as the pump laser except that the dye laser cannot generate its own beam and thus is dependant on an external source. However, dye lasers are very important in fluorescence studies because of their ability to selectively emit a specific wavelength. This output wavelength needs to be tunable in order to excite certain quantum transitions of the intermediate species under study, causing them to fluoresce. Dye lasers use liquids as a lasing medium because they generate a much broader range of wavelengths when compared to solid mediums like an Nd:YAG [10].

When the pump beam exits the Nd:YAG it is reflected 180° upward by two mirror boxes and into the dye laser. The pump beam passes through the first beam splitter, $M_1$, where a small portion of the beam’s power is diverted toward the 20 mm dye quartz cell. This
portion of the beam stimulates the dye and causes it to fluoresce. Take for example Coumarin 153, shown in Figure 8. When this dye solution is stimulated by a pump beam at 355 nm, it fluoresces within the range and efficiencies shown.

![Tuning curve Coumarin 153](image)

**Figure 8 – Excitation spectrum of Coumarin 153 using a 355 nm pump [10].**

A fundamental wavelength can be selected out of this broad spectrum by the use of the prism expander (PE) and the mirror/gravings G₁ and G₂, see Figure 7. The section of the dye laser (the 20 mm dye quartz cell, PE, G₁ and G₂) makes up the Resonator. Once the beam is generated in the Resonator, it passes through a series of polarizing plates called Brewster Plates, a turning prism and then reflected back into the same 20 mm dye quartz cell. As this happens, the remainder of the beam passes through M₁, is reflected by 2 turning prisms and passes through a second beam splitter, M₄. Another small portion of the beam is reflected onto the 20 mm dye quartz cell as the large majority of the original pump beam passes through M₄. The secondary split beam intersects the beam from the Resonator (that was reflected back into the dye quartz cell) causing stimulated emission. This action amplifies the beam by a small amount and is called the Preamplifier [10].
The preamplified beam that is emitted from the 20 mm dye quartz cell is expanded through a telescope and enters the 40 mm dye quartz cell where the beam is intersected yet again. However, the remaining 75% of the pump beam’s original power is incident on the 40 mm dye quartz cell and results in the final amplification of the dye laser beam. This dye quartz cell is the Main Amplifier. The final beam passes through three more devices before finally exiting the laser housing. The first device is the Frequency Conversion Unit (FCU), which doubles the frequency and halves the wavelength of the fundamental beam. The compensator steers the beam back to its original path after it has passed through the FCU. Lastly, a wavelength separation unit, comprised of four Pellin-Broca prisms, diverts the residual fundamental beam onto a beam dump to ensure that only the second harmonic passes through and exits the dye laser [10].

### 2.2.6 Camera

The camera used in this research is specially designed to image photons emitted from fluoresced species. The LaVision NanoStar camera is an intensified charge-coupled device (ICCD) camera of which the basic operation is explained in [1]. The lens currently installed on the camera does not have any direct magnification capability and can only be focused between roughly 2.5 and 15 feet. To “zoom,” the whole camera must be physically moved. The lens’ focus range therefore limits the distance between the camera and the combustion event, which directly affects the camera’s zoom and thus the size of the imaged object.

### 2.2.7 Energy Meter

The power of both the Nd:YAG and dye lasers is measured using a thermopile detector sensor and digital meter display made by Newport Corporation (model numbers 818P-015-18HP and 842-PE, respectively). The thermopile detector functions by absorbing the incident laser radiation on the white disk in the center of the head, pictured in Figure 9.
A series of thermocouples, also known as a thermopile, are used to measure the temperature increase of the absorber. The voltage generated by the thermopile is interpreted by the calibrated digital meter as a unit of power, most commonly in watts. This model of detector can be used for wavelengths of 190-600nm and 925-2,500nm. It can withstand a maximum continuous average power of 15 watts and 4 J/cm² using a pulsed laser at a wavelength of 532 nm, pulse length of 7 ns and pulse frequency of 10 Hz [13]. Both of these operational limits well exceed the current operating conditions of the laser system.

The energy meter is operated by first connecting the detector to the meter and pressing the power button located on the lower right of the digital meter as shown in Figure 10.
The initial display of the meter will depend on the saved settings, but most likely will show the power in real time. This display can be changed by pressing the “Menu” button which will give the user access to the tool bar displayed at the top of the screen. Under the “Display” menu, the user can set the meter to show the real time or average power, a line plot or even a tuning needle. The second tool bar item sets the display range from as little as 0 to 100 mW or as high as 30 W. The “Settings” menu is where new or changed settings can be saved or loaded. The last menu, “Ctrl,” is where the meter’s display can be toggled between watts and joules. The meter is turned off by pressing and holding the power button [14].

There are several important steps to ensure proper operation of the energy meter. The wavelength setting must be changed to correspond with the wavelength of beam that is to be measured. This setting is activated by pressing the right button on the directional pad. Pressing “Enter” will bring up a ‘numpad’ where the desired wavelength can be entered. Once the wavelength is selected, the detector should be set up so that the beam is directed close to the center of the white disk. Once it is set in place, the detector should be left alone for a while to allow it come to equilibrium with the surrounding environment. The manual also suggests that the digital meter be allowed to warm up for an hour before being used for measurements, however this was rarely done and did not noticeably affect power measurements. After the power ceases to drift and only fluctuates, the meter can
be zeroed by pressing the left button on the directional pad. This eliminates any zero offset error that may have been present when the meter was initially turned on [14].

The manual also suggests useful maintenance suggestions. It states that a fully charged battery will take approximately 15 months before it is discharged, which can cause battery breakdown or significantly reduce battery life. Therefore, the meter should be charged often or removed completely if unused for longer periods of time in order to prevent damage. The meter should also be turned off when not in use to prevent battery leakage. Each thermopile detector is calibrated for a specific digital meter, and this calibration is warranted for a period of one year. After this time has expired, the meter and detector should be sent back to Newport Corporation for re-certification [14]. The energy meter was received close to the beginning of June 2009. It should be recalibrated in June 2010.

2.3 System Improvements

The initial attempts to complete the calibration process were hampered by several system inefficiencies. Resolving these issues became one of the primary focuses of this present research. As detailed in previous sections, much time was devoted to study these systems in greater detail so as to learn how the experimental design could be improved. This work is presented in the current section.

2.3.1 Fluorescence

The main issue with the calibration process was that the fluorescence images captured by the camera had a very low intensity. The count gradient was very low because the maximum counts rarely exceeded 10 or 15. This made it more difficult to distinguish between high and low concentrations of OH and would have proved to be an imprecise calibration. A large count gradient would give more certainty to the concentration distribution. The best way to improve this is to increase the power of the experimental beam.
**Changing the Dye Solution**

The first improvement made to the dye laser system was changing the lasing medium. The system was originally installed using a Coumarin 153 dye solution. The efficiency and fluorescence spectrum of this particular dye can be seen in Figure 8, in section 2.2.5 *Dye Laser*. The peak efficiency of this dye is 15% and occurs at a fundamental wavelength of 535 nm. OH excitation in the (1,0) transition is commonly targeted by tunable dye lasers and requires a beam wavelength near 285 nm, corresponding to a fundamental of 570 nm [1]. The chart for Coumarin 153 shows that a maximum efficiency of 5-7% can be expected for this beam output.

After discussions with a laser specialist at LaVision, it was learned that Rhodamine 6G dye is the standard used for OH-PLIF, and not Coumarin 153. Rhodamine 6G dye has several advantages over Coumarin 153: an increased dye stability of several weeks instead of several days; a significantly higher dye efficiency, as shown in Figure 11; and an increased pump beam power. The peak efficiency of Rhodamine 6G is roughly 28%, and drops to around 25% near 570 nm.

![Tuning curve Rhodamine 6G](image)

*Figure 11 – Excitation spectrum of Rhodamine 6G using a 532 nm pump [10].*
The trade-off however, is that this dye has a much smaller tuning range, and therefore a less effective range than the Coumarin dye; only 17 nm as opposed to 57 nm. One very important consideration to make when changing dyes is whether or not the emissions from the new dye overlap with the absorption from the old dye. If it does, it will decrease the dye laser output or in some cases prevent lasing altogether. Changing dyes within a “family,” such as Coumarins or Rhodamines, usually won’t have a severe impact; however this is not the case when changing from Coumarin 153 to Rhodamine 6G so it is important to have two sets of dye circulators, one for each type, to prevent contamination [10].

The dye solution of Rhodamine 6G is mixed the same way that Coumarin 153 is mixed because the relative dye solution concentrations are dependant on the Sirah Dye Laser configuration and not on the particular dye. Rhodamine 6G should be mixed in a concentration of 0.09g/liter for the Resonator/Preamplifier dye circulator and one-third this concentration (0.03g/liter) for the Main Amplifier dye circulator. The Sirah service manual, section M.3.4, offers more information and examples on dye solution preparation. It is suggested that this section be read as well for full comprehension of this process [10].

**Pump Beam Alignment**

After changing the type of dye solution, it may be necessary to retune the mirrors and lenses within the dye laser, but if the dye solution is simply replaced by a fresh mixture of the same dye or the Nd:YAG has not been altered, retuning may not be necessary. Retuning the dye mirrors and lenses is best done after the Nd:YAG has been tuned using the steps explained in section *Nd:YAG Modifications and Tuning*. After tuning the Nd:YAG, the pump beam may need to be redirected into the dye laser correctly. This is done primarily by adjusting the three knobs located on the mirror boxes (see Figure 1), although the reversing prisms may also be adjusted if *absolutely* necessary (see Figure 7). Do *not* use the beam splitter adjustment screws to steer the pump beam. Ideally the pump beam should enter and exit through the center of each opening on each mirror box; however this is hardly realistic to achieve since it is much more important to align the
beam within the dye laser. There is also a “Beamtool1” that can be placed in the beam path at the locations shown in Figure 12 within the dye laser. The blue vertical line should be aligned with either of the lines drawn on the base plate [10].

Ideally the beam should land on the numbered cross-hairs, but this might not be the optimum case, and it is best to use the tool only as a guide. The most important thing is to make sure that the beam does not get truncated or cut-off as it passes through any of the mirrors or prisms. This can be checked by interrupting the beam with a business card at each location. Ultimately the whole beam profile should pass through the center of the last 90° prism (M5). This adjustment process can be an iterative and time-consuming one but once it is accomplished the beam is positioned correctly [10]. While adjusting the pump beam’s alignment, the Nd:YAG should be turned on in the Long Pulse (LP) mode via the laser remote in order to avoid unnecessary dangers caused by a full power beam.

Figure 12 – Beamtool 1 placement locations within dye laser.
Dye Beam Optimization

The next step is to adjust the dye beam components within the dye laser. Although the following descriptions will provide a thorough overview of the adjustments made during this research, it is highly suggested that the sections on optimization in the dye laser manual be read as well [10]. On a day-to-day basis, the adjustment of the Resonator, Preamplifier and Main Amplifier dye cells most likely will not need tuning. For every day tuning, adjusting the M1, M4 and M5 knobs are generally sufficient. The Sirah Dye Laser Service Manual also states that changing the laser wavelength less than 200 nm most likely will not warrant any dye cell adjustment, but when the Nd:YAG or mirror boxes are adjusted or the dye solution is changed to a new type it may be beneficial to check the dye cells’ alignment. Start the Resonator adjustment procedure by shutting both shutters S1 and S2, removing the protective metal covering to the left of the 20 mm dye quartz cell. Place the “Beamtool 4” upside down in front of the Brewster Plates, as shown in Figure 13, so that the beam is reflected onto the wall of the laboratory [10].

![Figure 13 – Picture of the Brewster Plates with and without Beamtool 4.](image)

Extra care must be taken to avoid excess laser radiation, especially with the protective covers removed and while the laser beam is diverted outside the manufacturer’s intended path. The pump beam power should be turned down as low as possible while still maintaining the dye laser beam. Figure 14 depicts the dye cell as the beam from the
gratings passes through it. The ideal position of this beam is to run parallel, as close as possible, along the inside wall of the quartz cell.

If the dye cell is positioned incorrectly, there will most likely be a reflection off the inside wall of the quartz cell, depicted by Figure 14. If this happens, there will be two separate laser spots on the laboratory wall from the reflection and main resonator beam. Make adjustments to the dye cell by varying the linear displacement (the lower left screw) first until the two laser spots have obtained their maximum luminosity. Next, adjust the rotation of the dye cell (the upper right screw) by a $\frac{1}{4}$ turn in the direction that lessens the distance between the laser spots. Alternately repeat these two steps until the spots converge and are at maximum luminosity; when this occurs, the Resonator dye cell is optimized. The protective metal covering can now be reinstalled. There are other, more advanced, tuning procedures that are covered in Section RO of the dye manual but they were not attempted during the course of this research and will not be covered here [10].

The second step is to do a simple adjustment of the Preamplifier. Like the Resonator, there are more complicated procedures in the dye manual, however they were not performed. The goal of this adjustment is much the same as the Resonator in that the beam should be positioned as close as possible to the inside wall of the quartz cell, as shown in Figure 15. Close shutters $S_1$ and $S_2$ and turn the laser power down as low as
possible, while maintaining visibility of the whole beam profile from the Resonator. As the resonator beam is reflected from the Resonator back into the Preamplifier (see Figure 7) it is rotated 90°, so that a vertical displacement of the resonator beam is changed to a horizontal one of the preamplifier beam as shown in Figure 15.

Place a thick piece of paper in the beam path just before the main amplifier so the beam profile can be visually monitored. Make adjustments by turning the left screw of the beam splitter, M₁, to move the resonator beam up and down until the beam looks similar to Figure PO-7 of [10]. A visual inspection serves as a good first estimate but fine tuning should be done by using the power meter. This way the affects of minute changes can be easily monitored. Open shutter S₁ and start adjusting the left screw of M₁ until a maximum power is observed. Next adjust the height of the second split beam by turning the left screw of beam splitter M₄ until the power peaks again. Repeat this procedure for the right screws of M₁ and M₂ to tune the horizontal position of the pump beam on the dye cell. A final fine adjustment of the resonator’s pump beam (right screw of M₁) will ensure the optimal horizontal position of the preamplifier beam [10].
With the preamplifier beam optimized, it is now possible to tune the Main Amplifier. Just like the resonator and preamplifier, the dye laser beam should pass through the main amplifier close to the inner wall of the quartz cell. The rotation of the 40 mm dye quartz cell is made using a similar looking knob as the other dye cell, however linear adjustments have to be made manually by loosening two M5 socket cap screws located behind the cell. This dye cell was not tuned while research was being done but if adjustments need to be made the manual should be consulted before making any changes. The 90° turning prism, M5, should be tuned however, and works the same way as the beam splitters. Use the left screw to move the pump beam’s location on the dye cell up and down and the right screw to move it left and right. Use the energy meter to monitor the beam’s power while making changes.

The frequency conversion unit (FCU) is the last item that needs to be tuned. This requires using the “Sirah Control 2.6” software installed on the computer. With the software open, the tuning interface is activated by selecting Motor > First Conversion > Tune. The FCU’s motor rotates the unit by user defined increments each time a button on the interface is clicked or by pressing the F5-F8 keys. The incremental adjustment value can be changed by pressing the F2 key. It is useful to display the motor counts by selecting Motor > First Conversion > Display while tuning with the energy meter so that when the optimal position is reached, the FCU’s motor position can be recorded. The full tuning range of the FCU is very large in comparison to the useful tuning range of the experimental beam. Care must be taken when making adjustments because the beam can be lost if the FCU is rotated too much and may be difficult to find the correct motor position again.

After all of the previously described adjustments are made, the power of the final experimental beam should be measured with the energy meter. If the power is satisfactory then no other adjustments should be made. However, if the power is still low, minor adjustments can be made to the Resonator/Preamplifier dye cell, beam...
splitters and prism, M₁, M₄ and M₅ (all in this order) while the experimental beam’s power is being monitored.

As mentioned previously on a day-to-day basis, or if just the dye solution has been refreshed, retuning the pump beams location within the dye laser or adjusting the dye cells’ alignments is not usually necessary. However, if this is the case and the beam power tends to fluctuate, the beam splitters and 90° turning prism (M₁, M₄ and M₅) can be tweaked in order to peak the beam power again. It is advantageous to tune M₁ before M₄ because the vertical displacement of the resonator pump beam affects the horizontal location of the preamplifier beam within the dye cell.

**Nd:YAG Modifications and Tuning**

The decision to change the laser dye from Coumarin 153 to Rhodamine 6G also meant that the Nd:YAG’s beam wavelength needed to be changed to the second harmonic (532 nm) in order to allow the dye to generate the desired range of wavelengths. As described in section 2.2.4 *Pump Laser*, the HG creates different harmonics of the fundamental wavelength by using different combinations of special crystals. The desired output can be looked up in Table 1 and Table 2 to find the correct setting of the HG.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Arm Position</th>
<th>Crystal Position</th>
</tr>
</thead>
<tbody>
<tr>
<td>1ˢᵗ</td>
<td>0</td>
<td>First stage crystals out of beam path</td>
</tr>
<tr>
<td></td>
<td>I</td>
<td>Type I SHG crystal in beam path</td>
</tr>
<tr>
<td></td>
<td>II</td>
<td>Type II SHG crystal in beam path</td>
</tr>
<tr>
<td>2ⁿᵈ</td>
<td>O</td>
<td>Second stage crystals out of beam path</td>
</tr>
<tr>
<td></td>
<td>T</td>
<td>THG crystal in beam path</td>
</tr>
<tr>
<td></td>
<td>F</td>
<td>FHG crystal in beam path</td>
</tr>
<tr>
<td>Output of Interest</td>
<td>HG Settings</td>
<td></td>
</tr>
<tr>
<td>-------------------</td>
<td>-------------</td>
<td>---</td>
</tr>
<tr>
<td>( \lambda ) (nm)</td>
<td>Polarization Crystal</td>
<td>Main Stage Position</td>
</tr>
<tr>
<td>1064</td>
<td>Horizontal</td>
<td>0</td>
</tr>
<tr>
<td>Vertical</td>
<td>Horizontal</td>
<td>I</td>
</tr>
<tr>
<td>Horizontal</td>
<td>Vertical</td>
<td>I</td>
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<tr>
<td>Vertical</td>
<td>Horizontal</td>
<td>II</td>
</tr>
<tr>
<td>532</td>
<td>Horizontal</td>
<td>Vertical</td>
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<td>Vertical</td>
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</tr>
<tr>
<td>Horizontal</td>
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<td>I</td>
</tr>
<tr>
<td>Vertical</td>
<td>Vertical</td>
<td>II</td>
</tr>
<tr>
<td>355</td>
<td>Horizontal</td>
<td>Horizontal</td>
</tr>
<tr>
<td>Vertical</td>
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<td>II</td>
</tr>
<tr>
<td>Horizontal</td>
<td>Horizontal</td>
<td>II</td>
</tr>
</tbody>
</table>

The original settings of the HG that were used to pump Coumarin 153 are highlighted in blue and the current settings used to pump Rhodamine 6G are highlighted in yellow in Table 2. The dye laser optics allows it to be pumped with an s-polarized (vertical) 532 nm or 355 nm beam without any modifications. The Nd:YAG’s Dichroic mirrors also have been optimized for vertical polarization, so it is always best to make sure that the HG produces such a beam. The HG also has two types of second harmonic generation (SHG) crystals, as shown in Table 2 that can be used to generate the 532 nm beam; however the Type II has a slightly higher conversion efficiency than Type I and is typically used for dye laser applications [9].

The steps to changing the HG is as follows: Decide what type of beam needs to be generated and which type of SHG crystal is best. In this case, a vertically polarized beam of 532 nm is required, and a Type II crystal is most efficient. Move the crystal translation arms to the positions shown on the chart, so that the peg of the tuning knob sits in the notch with “II” next to it. Push the second translation arm all the way into the HG; this removes it from the beam path. Do not move the crystals in and out of the beam path while the laser is running. Depending on the current orientation, the whole HG housing will need to be rotated to the position shown in Figure 16.
The easiest way to determine the polarization of the Nd:YAG beam is to look at the direction of the long translation arm associated with the last harmonic being generated. The polarization is parallel to this arm (or perpendicular to the crystal tuning axis). In this case, the second arm is removed from the beam path so the first arm is the one that determines the polarization. Once the steps described are completed, the beam pump will pulse vertically polarized 532 nm light [9].

The next crucial step is to change the Dichroic mirrors. Remove the cover of the pump laser by unscrewing the six screws on the top. Unscrew the knurled ring holding the mirror in place, as shown in Figure 17.
Remove the mirrors by pushing them out and replace them with the mirrors corresponding to the pumped wavelength. In this case, the 355 mirrors were replaced by the 532 mirrors. Screw the knurled ring snugly back onto the mirror stand. Be sure to install the mirrors so that the arrow drawn on the rim of the mirror points towards the side that the beam will reflect from (i.e. when both mirrors are installed, the arrows should point in at each other) as shown in Figure 5. Changes to the HG and the Dichroic mirrors usually cause the beam to displace from the original path (hence, the adjustments made in sections Pump Beam Alignment and Dye Beam Optimization). If the beam displacement is small, as it probably will be, then it is best to tune the mirror boxes instead. If need be, horizontal adjustments are made by turning the lower brass hex nut on the back of the mirror stands. Vertical adjustments made using the upper nut, shown in Figure 17. When finished, replace the cover on the pump laser [9].

The last step is to peak the laser power. Set up the energy meter at the output of the Nd:YAG and set it up as per the instructions in section 2.2.7 Energy Meter and turn on the laser in Q-Switch mode at full power. Safety glasses should be worn whenever the
laser is on, especially at full power. Slowly adjust each crystal by turning each ‘angle turning knob,’ shown in Figure 16, one at a time until the beam power peaks. Only a small portion of the full tuning range of the knob will actually produce any output beam so large adjustments to find the new optimum position are common after changing the HG settings. Next, slowly turn the ‘input polarization rotator,’ located at the front of the HG shown in Figure 16, until the power peaks again. This rotates the polarization of the 1064 nm (fundamental) beam to optimize it with the harmonic generation crystals for maximum conversion efficiency. This rotator is very sensitive so a very steady hand is needed to get sufficient results. As a last check, carefully tune the knobs and rotator again to ensure the beam is optimized.

2.3.2 Imaging

Improvements were also made to the LaVision NanoStar camera in order to improve image quality. As discussed previously in section 2.2.6 Camera, the distance between the flat flame burner and the camera was limited by the focal range of the lens. Discussions with a LaVision system specialist revealed that there were lens spacers that could be inserted in between the lens and photocathode. The lens spacers, shown in Figure 18, act as a zoom for the camera.
Figure 18 – Different Sized Lens Spacers. No. 1 and 2 are placed on the camera.

Figure 19 illustrates this concept using a thin lens approximation. The gray bar on the left represents the photocathode where the object is imaged and the three lines emanating from the object represent three rays of light reflecting from a single point.

Figure 19 – Illustration of the effect of adding lens spacers.
For simplicity only these three lines are shown, but naturally light would be reflected in all directions and from every “point”. The “plus” signs are the two focal points and center of each lens. As spacers are added to increase the distance between the lens and the photocathode, the object must be brought closer to the camera (or vice versa) in order for the image to remain in focus, which is indicated by the intersection of the rays of light on the photocathode. As a result, the image of the object on the photocathode becomes larger.

2.3.3 Results
After making all of the aforementioned changes to the laser and imaging systems, the improvements were tested and documented by measuring the power of the new experimental beam and imaging OH excitation on the flat flame burner. The experimental beam’s energy increased from approximately 6 mJ/pulse (60 mW) to 24 mJ/pulse (240 mW) as a result of changing the Nd:YAG’s beam to 532 nm and the dye laser’s lasing medium to Rhodamine 6G. This four-fold increase of energy is contributed to the fact that the pump beam’s power increases as the number of non-linear harmonic generation processes is decreased as well as to the fact that the Rhodamine 6G dye is roughly 3.5 times more efficient at 570 nm than Coumarin 153. Theoretically, the Nd:YAG’s power should double, from 2.5 W at 355 nm to 5 W at 532 nm [1], however an increase to only 4.1 W was seen. The overall effect of the system improvements are clearly shown in Figure 20. The picture of the OH fluorescence takes up the full frame lending to a much higher detailed image and the peak number of counts has increase from about 10 to over 75 which produces a higher “resolution”.
What is not shown in Figure 20 was the camera’s gain, a sort of multiplier of the raw electronic signal that serves to increase the picture’s intensity. This value was also reduced from 90 to 60 for the before and after shots, respectively. Lowering the gain is advantageous because it increases the signal-to-noise ratio, improving image quality. Overall the system changes made significant improvements to the OH fluorescence images which will be useful in helping to increase the accuracy of the concentration calibration. There are some other possible improvements that might increase the performance slightly that are discussed in Chapter 4 – Conclusions and Recommendations.

2.4 Experimental Procedure

Research for the calibration concentration began with the purchase and assembly of the flat flame burner, temperature measurement and gas metering system. Preliminary tests were conducted in order to verify that the system mechanics worked properly and matched general theoretical performances as well as to get the operator familiar with the system operation. A bare wire, uncoated type B thermocouple, with a bead and wire size of 0.36 mm and 0.20 mm, respectively, was used for all temperature measurements. A methane-air mixture had been initially used for experiments for the flat flame calibration because it had been used in previous work related to this research, but was traded for hydrogen after the preliminary temperature measurements gave seemingly questionable
data. Hydrogen temperature measurement trends tended to better match theoretical predictions but was not without problems, as will be discussed in the following sections.

2.4.1 Thermocouple Temperature Correction

In all thermocouple measurements made in high temperature, non-isothermal environments such as those found in combustion studies, the temperature that the thermocouple reads is not the true temperature of the gas. Therefore, there needs to be some correction applied to account for this discrepancy. Three heat transfer phenomena affect the temperature measurement of a thermocouple inserted into a flame: radiation, conduction and convection. Radiation transfers energy between objects in the form of electromagnetic waves and is emitted by any object with a finite temperature. Conduction occurs when there is a temperature gradient within a stationary medium. Convection refers to the heat transfer that occurs when there is a temperature difference between an object and a fluid that flows past it. Generally speaking, all three phenomena can contribute to the heating or cooling of an object, depending upon the conditions of it and its environment [15]. When measuring temperature within a flame or hot gas region, convection to the thermocouple is most often the main contributor to its heating and conduction and radiation usually account for its cooling. Under less extreme conditions, radiation and conduction losses can be considered negligible. However as temperature increases, the radiation loss increases rapidly as a fourth order equation. In regions of steep temperature gradients, conduction will significantly affect the thermocouple readings as well.

Temperature corrections were made by assuming that radiation from the invisible flame to the thermocouple and heat generation from thermocouple surface reactions were negligible. The thermocouple bead was also assumed to be in thermal equilibrium with its environment after a period of about 30-60 seconds. Starting from a basic heat transfer balance where there is convection to the bead, radiation loss from the bead and conduction loss down the lead wires, solving for the burned gas temperature gives:
\[ T_b = \frac{\varepsilon \sigma D}{\text{Nu}_D k_b} \left( T_i^4 - T_R^4 \right) + T_i + \frac{k_i D}{\text{Nu}_D k_b} A_l \frac{dT}{dn} \]  
Eq. (3)

where \( \text{Nu}_D \) is the Nusselt number for the bead, \( k_b \) is the thermal conductivity of the burned gas, \( \varepsilon \) is the emissivity of the thermocouple, \( \sigma \) is the Stefan-Boltzmann constant, \( D \) is the diameter of the bead, \( T_i \) is the recorded thermocouple temperature, \( T_R \) is room temperature, \( k_i \) is the thermal conductivity of the thermocouple leads, \( A_l \) is the cross-sectional area of the leads, \( A_b \) is the surface area of the bead and finally \( dT/dn \) is the temperature gradient of the bead-lead junction. The Nusselt number was approximated by the flow around a sphere [15-17] given by

\[ \text{Nu}_D = 2 + \left( 0.4 \text{Re}_D^{1/2} + 0.06 \text{Re}_D^{2/3} \right) \text{Pr}^{0.4} \left( \frac{\mu}{\mu_s} \right)^{1/4} \]  
Eq. (4)

where \( \text{Re}_D \) is the Reynolds number for a sphere, \( \text{Pr} \) is the Prandtl number of the burned gas, and \( \mu \) and \( \mu_s \) is the dynamic viscosity of the burned gases at \( T_b \) and roughly \( T_t \), respectively. Emissivity was calculated using a temperature dependant equation for the emissivity of platinum [17]:

\[ \varepsilon = 9.6 \times 10^{-5} \times T_i + 0.056 \]  
Eq. (5)

The most accurate temperature corrections include conduction effects in the calculation, however this is a challenging and iterative process. Conduction effects could easily be determined if the temperature along the thermocouple leads was known, however only the temperature of the bead is measurable. An estimated temperature profile can be obtained by translating the thermocouple in the direction of the leads but this measures the bead temperature, and not that of the leads. In order to estimate the real temperature profile, an iterative calculation could be used [17].

In another work, the effects of conduction losses were considered by measuring the temperature across a co-flowing burner using three thermocouples with different bead
and wire thickness. They corrected the measurements for radiation loss (and excluded conduction) using a similar approach as Equation 3 and found that with increasing lead thickness, the effects of conduction losses increased. For their experimental set-up they determined that conduction losses were negligible with a lead thickness of roughly less than 75 μm and that only a radiation correction for the bead was sufficient [18]. The thermocouple used for experiments in this research had a wire thickness of 200 μm, well above this cut-off. In the two previous works just mentioned, the thermocouples had been suspended in the flame with the leads extending horizontally outwards in opposite directions. In this configuration the leads traverse through short semi-isothermal regions and large temperature gradients near the edges of the hot gas regions. In this current work, the thermocouple was placed in a vertical position with the leads extending downstream, away from the burner. In this configuration the leads extend through a semi-isothermal region for a longer distance than the previous two works (see Figure 22, section 2.4.3 Flame Front Location). The thermocouple bead is also the leading section in the flow of gas which possibly reduces the disturbances caused by the horizontal leads, leading to a more ideal flow around the bead. Using these rationales, the conduction term of Equation 3 was neglected, to obtain:

\[
T_b = \frac{\varepsilon \sigma D}{NuDk_b} \left( T_i^4 - T_R^4 \right) + T_i
\]  

Eq. (6)

The properties of the burned gas were estimated using the properties of air [15, 16, 18]. An excel chart and macro were set up so that the temperature correction was calculated using temperature dependant properties of air instead of estimating an average value for the whole range of temperatures. Using this methodology, radiation corrections were approximately 45 to 150 K for temperature measurements of 1200 to 1700 K, respectively.
2.4.2 Flow Meter Calibration

The equations used to calculate the equivalence ratio based on the volumetric flow meters were discussed in section 2.2.1 Gas Metering System and involve a correction factor for gases other than the calibration gas, nitrogen. Since the correction factor for both nitrogen and air are \( K=1 \), it should only be necessary to correct the reading from the hydrogen meter. The calibrated correction factor, determined by Omega, for hydrogen is \( K_{H_2}=1.01 \) which is presumably used for the full range of the flow meter [11]. Figure 21 shows the results of the temperature measurements made in various equivalence ratios and hydrogen flow rates, using \( K_{H_2}=1.01 \). Measurements were made at a height of 8 mm above the burner and corrected for radiation losses using the equations in the previous section.

![Comparison of Corrected Thermocouple Measurements at 8 mm Above the Burner in Different Flow Rates of Hydrogen; \( K_{H_2}=1.01 \)](image)

Figure 21 – Temperature versus equivalence ratio. In order of increasing temperature, \( H_2 \) is 2, 3, 4, 5, 8, 10, 12 and 15 SLPM.
The factory correction factor was used for each flow rate and it can be seen that the overall temperature increases and the peak temperature shifts as the hydrogen flow rate was increased. The overall temperature increase with respect to flow rate is expected. The equilibrium position of the flame front, the difference between the laminar flame speed and gas velocity, increases in height as the total gas flow rate is increased [19]. This presumably reduces the heat transfer to the burner and as a result the flame temperature approaches the adiabatic temperature. The shift in the temperature peak with respect to flow rate was not expected however, because the temperature-equivalence ratio profile should be independent of flow rate, which in this case it clearly is not.

After investigating several possible mechanical reasons for this error, it was eventually presumed that the hydrogen meter correction factor, \( K_{\text{H}_2} = 1.01 \), provided by Omega was incorrect. Since the flow meters are calibrated to a standard pressure and temperature, Equation 1 can be substituted into Equation 2 and the resultant equation simplified significantly:

\[
\phi = C \frac{\dot{Q}_{\text{fuel}}}{\dot{Q}_{\text{oxidizer}}} K_{\text{H}_2} \quad \text{Eq. (7)}
\]

where \( C \) is a constant, the stoichiometric mole fraction, and is close to 2.39 for hydrogen-air. Equation 7 shows that for a given fuel-to-air flow ratio, the equivalence ratio can be adjusted by simply changing the hydrogen correction factor, \( K_{\text{H}_2} \). This acts as a shifting mechanism for the data in Figure 21 and therefore is only a matter of determining a calibration curve that takes into account a flow dependant correction factor. Figuring out how the data in Figure 21 should be shifted by the flow dependant correction factor was unclear, as it will be shown in Chapter 3 – Preliminary Results.

### 2.4.3 Flame Front Location

The next step in the experimental procedure was to measure the temperature profile above the burner in the direction of the gas flow. The thermocouple bead was placed at the center of the burner and positioned as accurately as possible to 0.2 mm above the
burner then moved incrementally along the axis of the burner until the bead reached a height of 25 mm. Measurements were taken at three different equivalence ratios and a hydrogen flow rate reading of 8 SLPM. The radiation corrected temperature results are shown in Figure 22.

![Axial Temperature Profile Above the Burner: H₂ at 8 SLPM](image.png)

**Figure 22 – Axial temperature profile of the burner with H₂ reading at 8 SLPM.**

The small peak close to 1 mm indicates that the flame front is within that region and several millimeters beyond this can be considered to be in chemical equilibrium. The temperature profile is also fairly level in the post flame region [20]. Future temperature readings were made in this region above the burner because of the shallower temperature gradients and equilibrium conditions, which were necessary for the concentration calibration.
2.4.4 Fluorescence Imaging

OH fluorescence imaging forms the core of the experimental data gathered for calibration. This process is begun by turning on the following systems: Nd:YAG and dye lasers, dye circulators, pump laser and flat flame burner coolant, ICCD camera, flow meters and the data acquisition computer. Refer to [1] for an in depth description of the system operation and a guide to the experimental process. While some of the equipment warms up, open the LaVision software and begin a new project. Check the settings of various equipment parameters such as the camera’s delay, gate and gain and the dye laser’s fundamental wavelength. Experiments were conducted with the following values: delay – 0.14 μs, gate – 0.10 μs, gain – 50 to 70 and a fundamental wavelength of 565.344 nm. When the laser is fully operational, set up the energy meter to measure the power of the beam. Record this value both as a way to track the history of the power level and to determine whether or not the dye laser needs tuning or a fresh batch of dye solution.

Once the camera has warmed up (the LED on the back is a steady green light) it must be calibrated. This is part of the pre-processing discussed in [1] and is necessary in order to account for any image distortion that is caused by the optics of the camera, experimental apparatus and the camera’s oblique viewing angle to the laser sheet. Place the calibration plate in the intended path of the laser, take off the OH filter from the lens and follow the image distortion calibration procedure. Finish the rest of the pre-processing calibration by recording a sheet and background image as per [1]. Secure the OH filter back on the camera. The laser and data acquisition system should now be ready to take experimental images.

Set up the thermocouple stand next to the flat flame burner as shown in Figure 4 and position the bead to a desired position. This position is where the temperature will be measured, the average fluorescent image intensity and the OH concentration will be calculated. A ruler is useful in placing the thermocouple bead precisely at the desired height and to center the bead over the burner. Use the translational stage to move the thermocouple out of the way for now.
The flow meters will display an equilibrium value once they have had a sufficient amount of time to warm up. If this reading is not zero, it can be adjusted by tuning a potentiometer. There is a small access hole on the left side of each meter where a small flat-headed screw driver can be used to tune the potentiometer via a screw. The flat flame burner and gas metering system is now set up. Use Equations 1 and 2 to calculate the flow rates for a desired equivalence ratio or vice versa. Open the shut-off valves, needle valves and adjust the pressure on the tank regulators. Ignite the burner either by a striker or cigarette lighter before adjusting the flow meters to the desired value so that a build up of hydrogen doesn’t form above the burner. Once the fuel, oxidizer and co-flow have been adjusted to the appropriate values, fluorescence images can be taken.

Dialing the laser to maximum power will turn on the laser if the remote is set to ‘fixed,’ otherwise ‘on’ must be selected in the software, under Laser Control. Turn the camera gain down to 30 or so and take a single image by clicking ‘take’. Verify that the signal intensity does not exceed the camera’s maximum 4096 count intensity. If not, take pictures at each increase of gain until the desired value is reached, in this case 50-70. Switch from ‘take’ to ‘grab’ to image the flame continuously at 5 frames per second. If necessary, adjust the co-flow until the fluorescence image of the flame becomes as steady as possible. When this is achieved, record images of the flame. Generally speaking, 10 images would be sufficient, however taking roughly 50 images produced smoother results overall, which is only more beneficial for the concentration calibration process. When recording is finished, stop the laser from pulsing and position the thermocouple back to the original position over the burner’s surface. Wait 30 to 60 seconds for it to reach steady state and record the temperature. Reposition the stand so the thermocouple is off to the side again, out of the laser’s line-of-sight. Repeat this recording process for each desired equivalence ratio and/or flow rate.

When recording is finished, turn off the valves to all three compressed gas tanks and cease the Nd:YAG laser pulsing. The rest of the system may remain on especially if other tests are planned. Using the software provided by LaVision, post process the fluorescence images by following the guidelines detailed in [1, 21]. This processing.
includes the image distortion correction, averaging, energy correction, background subtraction, sheet processing and sheet image correction. When the OH concentration is known at a specific location, the final concentration calibration can occur.

2.4.5 OH Calibration

The images taken using the previous section’s process only contain the qualitative concentration of OH based on the relative intensities of the fluorescence photon emission. These pictures are not very useful because they do not portray a quantitative concentration; data that is valuable when studying combustion processes. One way to correlate the image intensity (i.e. color) to a specific concentration is to measure the flame temperature at a given equivalence ratio and use this to calculate OH concentrations. To do this, a chemical kinetics program called Stanjan was used to calculate the theoretical equilibrium OH mole fraction at temperatures measured in the hot gas region. A version of this software is available for free online [22]. Since the flat flame burner produces an atmospheric and well-defined flame, it was possible to measure the axial temperature profile (see Figure 22) to get an approximation of the flame front, or reaction zone. It appeared that the flame front occurred at a height close to 1 mm above the burner [20]. Well downstream of this zone, the products of combustion are considered to be in chemical equilibrium. It is this assumption of equilibrium conditions that makes the use of Stanjan possible.

After applying a radiation correction, these temperatures were entered into Stanjan as the “starting temperature” (equilibrium temperature) along with a starting pressure of 1 atmospheres. N, O and H were entered as “Elements” and H, H₂, O, O₂, N, N₂, NO, NO₂, H₂O, H₂O₂, OH and HO₂ were entered as “Additional Species.” The values of the mole fraction for each reactant were entered corresponding to the equivalence ratio at that given temperature measurement. Lastly, the calculation constraints were constant pressure and temperature to match the given experimental conditions.

Once all of the OH mole fractions were calculated and recorded for each experimental condition, the results were applied to the images via the software. Before doing so
however, a mask needs to be created around the area where the thermocouple was. The best method of doing this is to take the OH filter off the camera and image the thermocouple in its original location. Using this image, create a mask around the immediate area of the bead. The concentration calibration screen can be accessed by clicking the “Concentration” button on the top toolbar in the LaVision software. Using the mask that was just created, the software will average the intensity of the pixels within the masked area. This averaged intensity corresponds to the temperature that the thermocouple measured within the masked region and therefore, the OH mole fraction calculated by Stanjan is related to this averaged intensity value. Entering this OH concentration value into the LaVision software will generate a calibration curve, which relates intensity (in units of counts) to units of ‘OH mole fraction.’ Entering data points for other experimental conditions should help populate a trendline. Ultimately this curve is used to apply a quantitative concentration to other pictures, such as the ones captured in [1] and other future experiments.
Chapter 3 – Preliminary Results

This section covers the preliminary results obtained using the experimental procedure and assumptions made in the preceding sections. The results are described as preliminary because of some uncertainties and the possibility that the data could be improved upon.

3.1 Temperature Measurements

In section 2.4.2 *Flow Meter Calibration*, the issue of a flow-dependant hydrogen correction factor was discussed. Using the original factory correction factor, $K_{H2}=1.01$, developed results that were not in agreement with theory. It was therefore necessary to determine a calibration curve for the meter itself. It was originally assumed that because the adiabatic temperature peaks at $\phi=1.07$ that the measured temperature should as well. The calibration curve shown in Figure 23 shifts the temperature data so that the peak is the same equivalence ratio as the adiabatic peak.

![Hydrogen Flow Meter Correction Factor Calibration Curve](image)

**Figure 23 – Calibration curve for $H_2$ meter correction factor for a temperature peak at $\phi=1.07$.**

The results of applying the correction factors in Figure 23 to the data in Figure 21 are show in Figure 24.
Comparison of Corrected Thermocouple Measurements at 8 mm Above the Burner in Different Flow Rates of Hydrogen

![Graph showing temperature versus equivalence ratio using a variable $K_{H2}$ factor. In order of increasing temperature, H2 is 2, 3, 4, 5, 8, 10, 12 and 15 SLPM.](image)

The correction factor calibration curve in Figure 23 was assumed to be correct until an investigative experiment was conducted: In this test, temperature was measured at a height of 10 mm above the burner and a hydrogen reading of 8 SLPM through an extremely fine equivalence ratio sweep. The sweep had an average of 0.01 between each equivalence ratio point. Following the OH concentration calibration procedure described earlier, the corrected temperatures were entered into Stanjan. The equivalence ratio corresponding to each temperature measurement were calculated using $K_{H2}=1.32$, obtained from Figure 23, which corresponds to 8 SLPM of hydrogen. Fluorescence images were recorded at every temperature measurement and then corrected using the post-processing procedure. The average intensity of the masked region (near the thermocouple bead) from each picture is plotted versus equivalence ratio, along with the theoretical OH Mole Fraction calculated by Stanjan. This comparison is shown in Figure
25 as a percentage of the peak value, so that a comparison between the trends could be made. While they have fairly similar shapes, it is obvious that these two curves don’t align: there is a disparity between the experimental and theoretical results. One possible rationale is that the hydrogen correction factor is still wrong, meaning that the measured temperature doesn’t peak at $\varphi=1.07$, as it was originally assumed.

![Comparison of Theoretical OH Mole Fraction and Average Fluorescence Intensity, $K_{H_2}=1.32$](image)

**Figure 25 – Comparison of the Theoretical OH Mole Fraction and Experimental Fluorescence.**

To test this theory, a simple investigation was done to see how the theoretical OH profile was affected by variations in $K_{H_2}$ values. The independent variables in this study are the temperature measurements and the volumetric flow rate *readings*. For example, a temperature measurement will always be the same for a given set of flow rate *readings* of air and hydrogen, independent of the value of $K_{H_2}$. However, since the equivalence ratio is a calculated unit (see Equation 7), neither temperature or equivalence ratio are dependant of each other because the latter is dependant of $K_{H_2}$. Due to this temperature independence, the change in the theoretical OH mole fraction can be studied when a single set of temperature data is paired with several sets of equivalence ratios. Figure 26
shows the theoretical results from Stanjan using various values of $K_{\text{H}_2}$, thus different sets of equivalence ratios.

![Comparison of Theoretical OH Mole Fraction for Different Values of $K_{\text{H}_2}$](image)

Figure 26 – OH Mole Fraction as a dependence on $K_{\text{H}_2}$

It is seen that regardless of what the $K_{\text{H}_2}$ value is, there is a sharp decline in OH concentration at $\phi \approx 1.0$. Also, as $K_{\text{H}_2}$ changes, so does the equivalence ratio at which the measured temperature peaks (section 2.4.2 Flow Meter Calibration). Since OH is a function of both temperature and equivalence ratio, this indicates that the location of the OH profile trend is dependant more on the equivalence ratio than on where the temperature peaks. If the opposite were true and the OH profile was heavily dependant on temperature, it would have shifted around too, corresponding to different values of $K_{\text{H}_2}$. Further more, this leads to the same assumption that the calibration of $K_{\text{H}_2}$ is incorrect because the theoretical OH profile is stationary with regards to a change of $K_{\text{H}_2}$, whereas the experimental profile is not. The reason the experimental intensity profile (which is, or should be, proportional to the OH profile) shifts around, is for the same reason the temperature peak shifts. The intensity profile is plotted versus a calculated equivalence ratio, like the measured temperature, which changes with different values of
The next step, then, is to find a value of $K_{H2}$ where the theoretical OH mole fraction profile matches the experimental intensity profile.

When $K_{H2}$ is chosen to be 1.14, the OH profile better coincides with the intensity profile, seen in Figure 27, but the temperature peak shifts in the lean direction to $\phi \approx 0.94$.

![Comparison of Theoretical OH Mole Fraction and Average Fluorescence Intensity, $K_{H2}=1.14$](image)

**Figure 27 – Comparison of the Theoretical OH Mole Fraction and Experimental Fluorescence.**

This discrepancy between the expectation that the temperature-equivalence profile should peak at $\phi = 1.07$ and the OH-equivalence profile indicating that temperature peaks at $\phi \approx 0.94$ causes some uncertainty in the data and experimental equipment. For the purpose of the OH concentration calibration however, values of $K_{H2}$ (shown in Figure 28) were chosen with the intent of matching the theoretical and experimental OH profiles. The values were chosen by shifting the temperature data in Figure 24 to that shown in Figure 29. Using the hydrogen flow meter correction factor calibration curve shown in Figure 28, temperature measurements were made at several different equivalence ratios and flow rates at 10 mm above the burner. The corrected experimental data is shown in Table 3.
Figure 28 – Calibration curve for H₂ meter correction factor for a temperature peak at \( \phi = 0.94 \).

Figure 29 – Temperature versus equivalence ratio using a variable \( K_{H2} \) factor. In order of increasing temperature, H2 is 2, 3, 4, 5, 8, 10, 12 and 15 SLPM.
Table 3: Preliminary Experimental Temperature Measurements.

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Table 4: Preliminary Average Intensity Values.

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3.2 Fluorescence Pictures

For each temperature measurement shown in Table 3, images of OH fluorescence were captured using the ICCD Camera and are shown in Appendix: Experimental Fluorescence Images. Table 4 lists the average intensity values calculated within the masked area created around the thermocouple bead for each image.
3.3 OH Concentration

The corrected temperature data and calculated equivalence ratios presented in section 3.1 Temperature Measurements were entered into Stanjan to calculate the theoretical OH concentrations. This data is listed in Table 5.

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3.4 Final Concentration Calibration

The OH concentration calibration performed by the LaVision software based on the data in Table 4 and Table 5 is depicted in Figure 30. This OH concentration calibration curve was obtained by using the H₂ flow meter corrective factor calibration curve for K,H₂ in Figure 28 and a radiation correction for temperature measurements using Eqs. 4-6. Although the correlation in Figure 30 between counts and mole fraction has a good deal of scatter, it serves well as preliminary data, demonstrating the possible capability of the system.
Figure 30 – Fluorescence concentration calibration of OH Mole Fraction.
Chapter 4 – Conclusions and Recommendations

The feasibility of an OH concentration calibration using the PLIF system, gas control system, thermocouple temperature measurements and flat flame burner has been demonstrated. This research has been the extension of a previous work that demonstrated the capability of a PLIF system for imaging transient flames in a constant volume combustion chamber [1]. Combining the success of the previous effort with that of the current one will enable the combustion research laboratory at Northeastern University to study the hydroxyl concentration in many different fuels. Using the information and procedures described in this work, a user should also be able to easily modify the system with confidence to measure the concentration of other intermediate species, such as NO and CH, for example. Such work would provide the laboratory with valuable information to create or add to a database of results useful to chemical kinetics modelers.

Although the general feasibility of the laser and flat flame burner system has been shown, there are several recommendations for how to improve the accuracy of the results. The uncertainty of the flow meters was discussed at length, and the core problem is due to the fact that it was unclear whether or not they were calibrated correctly to begin with. These meters should be calibrated accurately so that the results obtained with them can be done so with confidence. Probably the best way to do so would be to compare the digital read out to that of a calibrated Venturi tube in order to develop a calibration curve. Another possible method would be to sample the products of combustion using a gas analyzer. The Northeastern combustion laboratory has an analyzer that is capable of measuring oxygen content and could be used to calibrate the meters in fuel-lean conditions. This method may be an easier alternative, albeit a less accurate one.

Another recommendation is to verify the validity of the assumptions made when applying the thermocouple temperature corrections. Although the assumptions seemed to be appropriate, the lead diameter was above a critical diameter that one group found to be the limit of negligible conduction effects [18]. The other issue is that a true isothermal region of large enough importance is very rarely found when working with burners of this
scale, therefore conduction affects all measurements to varying degrees. However, only further analysis will determine how accurate these assumptions are.

Lastly, both the Nd:YAG and Dye lasers would benefit from tuning by a technical specialist. Although this thesis and the laser service manuals describe the tuning process, the best possible option would be to have a manufacturer trained expert peak the laser. Users of the laser system should take this opportunity to learn as much as possible about this process from the specialist.
References


Appendix: Experimental Fluorescence Images

Figure 31 – Hydrogen (SLPM): 2, Air (SLPM): 5.4, $K_{H_2}=0.91$, $\phi=0.80$

Figure 32 – Hydrogen (SLPM): 2, Air (SLPM): 4.8, $K_{H_2}=0.91$, $\phi=0.90$
Figure 33 – Hydrogen (SLPM): 3, Air (SLPM): 8.9, $K_{H_2}=0.99$, $\varphi=0.80$

Figure 34 – Hydrogen (SLPM): 3, Air (SLPM): 7.9, $K_{H_2}=0.99$, $\varphi=0.90$
Figure 35 – Hydrogen (SLPM): 4, Air (SLPM): 12.4, $K_{H_2}=1.04$, $\varphi=0.80$

Figure 36 – Hydrogen (SLPM): 4, Air (SLPM): 11, $K_{H_2}=1.04$, $\varphi=0.90$
Figure 37 – Hydrogen (SLPM): 5, Air (SLPM): 16.7, $K_{H_2}=1.12$, $\phi=0.80$

Figure 38 – Hydrogen (SLPM): 5, Air (SLPM): 14.8, $K_{H_2}=1.12$, $\phi=0.90$
Figure 39 – Hydrogen (SLPM): 8, Air (SLPM): 27.2, $K_{H_2}=1.14$, $\phi=0.80$

Figure 40 – Hydrogen (SLPM): 8, Air (SLPM): 24.2, $K_{H_2}=1.14$, $\phi=0.90$
Figure 41 – Hydrogen (SLPM): 10, Air (SLPM): 35.6, $K_{H_2}=1.195$, $\varphi=0.80$

Figure 42 – Hydrogen (SLPM): 10, Air (SLPM): 31.7, $K_{H_2}=1.195$, $\varphi=0.90$
Figure 43 – Hydrogen (SLPM): 12, Air (SLPM): 42.9, $K_{H_2}=1.20$, $\varphi=0.80$

Figure 44 – Hydrogen (SLPM): 12, Air (SLPM): 38.2, $K_{H_2}=1.20$, $\varphi=0.90$
Figure 45 – Hydrogen (SLPM): 15, Air (SLPM): 55.3, $K_{H2}=1.237$, $\phi=0.80$

Figure 46 – Hydrogen (SLPM): 15, Air (SLPM): 49.2, $K_{H2}=1.237$, $\phi=0.90$