A STUDY OF ION AND ELECTRON RESPONSES TO DC ELECTRIC FIELDS IN A PREMIXED METHANE-AIR FLAME

by

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A THESIS

Submitted in partial fulfillment of the requirements for the degree of Master of Science in Engineering in The Department of Mechanical and Aerospace Engineering to The School of Graduate Studies of The University of Alabama in Huntsville

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2015
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The effect of the ionic wind and of dissociative recombination on a premixed methane-air flame under a DC electric field is studied via mapping of the ion density with Langmuir probes. Additional metrics of study include the electron temperature and the brightness of chemiluminescence images. Ion densities were observed to increase near the burner with increasing electrode voltage up to 6 kV. Past this electrode supply voltage, ion densities ceased increasing and began to decline in some locations within the premixed flame. The increased ion density is caused by an increase in ionic wind force and cathode sheath thickness. The plateau in density is due to the cathode sheath fully encompassing the flame front which is the ion source, thereby collecting all ions in the flame. The spatial density data support the ionic wind hypothesis and provides further explanation of its limits based on the plasma sheath.
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<tr>
<td>$D$</td>
<td>Cathode sheath thickness</td>
</tr>
<tr>
<td>$d$</td>
<td>Electrode separation distance</td>
</tr>
<tr>
<td>$d_{i,n}$</td>
<td>Ion-electron intermolecular distance</td>
</tr>
<tr>
<td>$E$</td>
<td>Electric field</td>
</tr>
<tr>
<td>$E^*$</td>
<td>Fraction of energy transferred during particle collision</td>
</tr>
<tr>
<td>$e$</td>
<td>Elementary charge</td>
</tr>
<tr>
<td>$f$</td>
<td>Ionic wind force density</td>
</tr>
<tr>
<td>$g$</td>
<td>Electronic energy level degeneracy factor</td>
</tr>
<tr>
<td>$h$</td>
<td>Convection coefficient</td>
</tr>
<tr>
<td>$\hat{h}$</td>
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<tr>
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<tr>
<td>$k$</td>
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<tr>
<td>$k_p$</td>
<td>Reaction constant</td>
</tr>
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<td>$l_p$</td>
<td>Langmuir probe length</td>
</tr>
<tr>
<td>$m$</td>
<td>Particle mass</td>
</tr>
<tr>
<td>$m_i$</td>
<td>Ion mass</td>
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\( m_n \) Neutral mass

\( m_{i,n} \) Ion-neutral reduced mass

\( n_i \) Ion density

\( n_e \) Electron density

\( n_n \) Neutral density

\( n_{e,s} \) Electron density within the sheath

\( p \) Pressure

\( Q \) Partition function

\( Re_e \) Electric Reynolds number

\( r_p \) Langmuir probe radius

\( T \) Temperature

\( T_e \) Electron temperature

\( T_f \) Flame temperature

\( T_{TC} \) Temperature measured by a thermocouple

\( T_\infty \) Air temperature

\( U \) Plasma flow velocity

\( u_0 \) Bohm velocity

\( \bar{v} \) Ion-neutral thermal velocity

\( V_b \) Langmuir probe bias voltage
<table>
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<tr>
<td>$V_{breakdown}$</td>
<td>Breakdown voltage</td>
</tr>
<tr>
<td>$V_o$</td>
<td>Volume</td>
</tr>
<tr>
<td>$v_d$</td>
<td>Ion drift velocity</td>
</tr>
<tr>
<td>$X$</td>
<td>Mole fraction</td>
</tr>
<tr>
<td>$Z$</td>
<td>Bimolecular collision rate</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Ratio of Debye length to Langmuir probe radius</td>
</tr>
<tr>
<td>$\alpha_{DC}$</td>
<td>Dissociative recombination rate coefficient</td>
</tr>
<tr>
<td>$\alpha_c$</td>
<td>Plasma collision parameters</td>
</tr>
<tr>
<td>$\Delta H_0^f$</td>
<td>Change in heat of formation at STP</td>
</tr>
<tr>
<td>$\delta$</td>
<td>Bin size number of an image histogram</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Emissivity coefficient</td>
</tr>
<tr>
<td>$\epsilon_0$</td>
<td>Permittivity of freespace</td>
</tr>
<tr>
<td>$\lambda_D$</td>
<td>Debye length</td>
</tr>
<tr>
<td>$\lambda_{MFP}$</td>
<td>Mean free path</td>
</tr>
<tr>
<td>$\mu_i$</td>
<td>Ion mobility</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Stefan-Boltzmann constant</td>
</tr>
<tr>
<td>$\sigma_s$</td>
<td>Momentum transfer cross-section</td>
</tr>
<tr>
<td>$\sigma_y$</td>
<td>Symmetry factor for the rotational partition function</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Characteristic temperature</td>
</tr>
</tbody>
</table>
\( \chi \)  Dimensionless potential

\( \chi_s \)  Dimensionless potential within the sheath

\( \psi \)  Total brightness of an image

\( \omega_i \)  Stoichiometric coefficient
The scientific man does not aim at an immediate result. He does not expect that his advanced ideas will be readily taken up. His work is like that of the planter - for the future. His duty is to lay the foundation for those who are to come, and point the way. He lives and labors and hopes.

—Nikola Tesla
CHAPTER 1

INTRODUCTION

Equipped with his five senses, man explores the universe around him and calls the adventure Science.
—Edwin Hubble

A hydrocarbon flame is a self-sustaining propagation of a localized combustion zone. During combustion, the reactant species interact with internally excited molecules, creating ionized particles in a process known as chemical ionization, usually referred to as chemiionization for short. These ions and electrons are capable of exerting collective behavior, though their number density is low compared to that of the neutral particles. This combination of properties means a hydrocarbon flame fits the definition of a weak plasma.

As with all plasmas, hydrocarbon flames respond to external electrostatic fields and can be altered or controlled. This has been studied in the literature with various burner and electrode configurations and has produced wide range of results. Examples of observed responses include increased flame speed, reduction of CO$_2$ production in the flame’s exhaust stream, and improvements to flame stability via an increase in the flame’s blowoff velocity or a delay in the onset of thermoacoustic instabilities. Much of the research to date has covered the macroscopic effects of subjecting a flame
to an electric field; comparatively fewer papers have attempted to explain, from the perspective of plasma physics, why the flame behaves as it does when it is subjected to an electric field. Hypotheses do exist to explain the observed behaviors, and some are more accepted than others. However, no unified theory exists which can explain the macroscopic results or relate them to the weak plasma.

The present research aims to investigate the chemiionized plasmas response to electric fields over a range of operating conditions. This is done through the use of analysis methods and instruments commonly used with atmospheric plasmas, which reveal properties of the plasma such as the number and location of ions and the electron temperature. In doing so, this research develops an explanation grounded in plasma physics for why the flame behaves as it does, thereby helping to further the development of a comprehensive theory governing the response of flame plasmas to externally applied electromagnetic fields.
CHAPTER 2

BACKGROUND ON PLASMA ASSISTED COMBUSTION

Nothing in life is to be feared, it is only to be understood. Now is the time to understand more, so that we may fear less.

—Marie Curie

The underlying principles of plasma assisted combustion were first described by Chattock [1] in 1899. In these experiments, a non-conductive gas was ionized using a sharp point placed above a grounded metal plate. Using the electric field generated by this setup, Chattock was able to demonstrate that the ions generated by the electrically charged point were propelled by the electric field towards ground with a maximum pressure of 90 dyne/cm².

Research by Wilson [2] in 1931 attempted a similar experiment with the flue gases of several hydrocarbon flames. Wilson was able to catalog the electric properties of the flames and concluded that hydrocarbon flames contained a significant number density of ions, thus demonstrating that flames are a weakly ionized plasma. Building on this knowledge, Guenault and Wheeler produced a series of papers, one focused on the deformation of the flame surface [3], and one explaining the effects of transverse electric fields [4]. To explain why the flame behaved as they observed, Guenault and Wheeler proposed that a body forced was responsible. This mechanism was later
quantified by Lawton and Weinberg [5], who calculated the theoretical maxima for velocity, force, and pressure exerted on the flame by the body force, referred to as the ionic wind. In the time period following this early work, three mechanisms have been proposed to account for the responses of flames: ionic wind, ion-electron dissociative recombination, and thermal driven effects.

2.1 Mechanisms Affecting the Flame

2.1.1 Ionic Wind

The most well-known and established mechanism is the ionic wind hypothesis, also known as the electro-hydrodynamic effect. Under this hypothesis, the electric field acts as a body force upon ions. Ions propelled by the ionic wind gain velocity, and the ion density downstream of the ionic wind increases, particularly in the vicinity of the cathode. The combined increase in ion velocity and ion density caused by the ionic wind has the effect of increasing both ion-ion and ion-neutral collision rates. Flame behavior is influenced by this change in collision rates, making the ionic wind a momentum driven process.

Most research into plasma assisted combustion today supports the ionic wind hypothesis [6] [7] [8] [9] [10] [11], either as the only mechanism acting on the flame or as the dominant one. Typically, the contributions of electron collisions are ignored; electrons are four orders of magnitude less massive than a single proton or neutron and the electrons present in a flame are very low energy for a plasma. The result is that ion-electron and neutral-electron collisions result in effectively no change in
the momentum of either ions or neutrals. Anion collisions are also typically ignored, since they are much less numerous than ions. An exception to this assumption comes from Kim et al., [6] who observed the blowoff velocity of a Bunsen type flame in an AC field. Kim et al. observed non-monotonic variation in the ratio of the flames blowoff velocity to its initial value. For AC fields with a frequency below 60 Hz, blowoff velocity decreased by as much as 30 %, with the greatest decrease occurring around 10 Hz. Above 60 Hz, the blowoff velocity increases exponentially, especially as the electric field strength increases. Kim et al. argue that this variation implies the contribution of anions is not negligible, and therefore must be accounted for when modifying a flame with an AC field.

2.1.2 Dissociative Recombination

A competing hypothesis to the ionic wind is a chemistry-driven process which involves dissociative recombination. This hypothesis, supported by Marcum, Ganguly, and Wisman [12] [13] [14] [15], as well as Boom et al. [16], states that Lawton and Weinbergs theoretical maximum pressure exerted by the ionic wind, which is 0.0004 atm, is insufficient to explain the magnitude and type of changes seen in hydrocarbon flames. Changes to flame behavior are instead brought about by a reaction known as dissociative recombination, wherein an ion and an electron recombine and split into two or more neutral particles. This process occurs naturally in hydrocarbon flames with several different ion species, with the majority of the reactions involving the hydronium ion. The typical reaction pathways for the hydronium ion are [17] [15]
Other examples of dissociative recombination reactions include

\[ H_3O^+ + e^- \rightleftharpoons H_2 + OH \] \hspace{1cm} (2.1)

\[ H_3O^+ + e^- \rightleftharpoons O + H + H_2 \] \hspace{1cm} (2.2)

\[ H_3O^+ + e^- \rightleftharpoons OH + 2H \] \hspace{1cm} (2.3)

\[ H_3O^+ + e^- \rightleftharpoons H_2O + H \] \hspace{1cm} (2.4)

Marcum, Ganguly, and Wisman suggest that dissociative recombination is enhanced by the electric field, and this drives the production of the light radicals. In their experiment, Marcum, Ganguly, and Wisman say that the increased presence of these radicals, which are highly diffusive species, reduces the global Lewis number of the flame, the ratio of the thermal diffusivity to the mass diffusivity of the flame, below unity. This reduction in Lewis number causes the premixed flame to adopt a wrinkled, unstable geometry. A slight variation of this hypothesis is provided by Boom et al., [16] who suggest that excitation of N\textsubscript{2} molecules due to collisions with
electrons may be responsible for additional chemical effects. In both cases, however, anion contribution to flame chemistry was ignored.

2.1.3 Thermal Effects

A third and much less known hypothesis is the thermal-driven process. Here, input electrical energy is converted to thermal energy. The conversion causes ionization of the neutral reactants, thus jumpstarting the appearance of a self-sustaining flame. Typically this is associated with exposure to an electrical arc, such as found in a spark plug engine, rather than the continuous combustion found in most laboratory burners. Thus, this hypothesis has yet to gain as much traction as the other two. However, a paper by Zhang et al. did report the appearance of thermally-driven processes in a coaxial burner, so it remains a possible contender. Furthermore, Zhang et al. observed all three mechanisms in the same flame, and using an electrode positioned 75 cm above the burner, identified three distinct voltage regimes corresponding to the different mechanisms. If the applied voltage was below 1 kV, ionic wind effects dominated. Between 1 kV and 3 kV, chemistry-driven effects predominated, and above 3 kV, all three mechanisms were coupled and observed affecting the flame.

2.2 Review of Experimental Setups

Experimental setups change sufficiently in the literature that observations can be highly variable. Therefore, a detailed discussion is required to adequately cover the entire range of possibilities. What follows is an overview of common burners,
electrodes, supply voltages, and instrumentation. A discussion of Computational Fluid Dynamics and its role in plasma assisted combustion research is also provided.

2.2.1 Burner Geometry

A number of different burners are used in the literature, the selection of which depends on factors such as desired geometry of the flame and whether it is premixed or a diffusion flame. Bunsen burners [2] [6] [7] [9] and flat flame burners [20] [21] [16] [22] [23] comprise the majority of burners used in the literature. Other geometries in use include Meker burners [24], nozzle-style burners [25] [6] [26], and various other burner designs which are not classified as Bunsen burners but which otherwise produce a cone-shaped flame [27] [19].

Bunsen burners are typically a rounded tube with a control valve near the burner inlet, although variants with a square tube exist. These burners produce a cone-shaped flame with two distinct layers, such as those seen in Figure 2.1. The inner layer of the flame is the premixed flame, which is brighter than the outer layer and has a very distinct boundary. The outer layer is the diffusion flame, consisting of fuel which did not react with the air in the premixed flame but is still energetic enough to react with air in the surroundings, and forms an amorphous sheet enclosing the premixed flame. Bunsen burners have been extensively studied in combustion research, so analytical models which describe Bunsen burner flames are easy to find.

Flat flame burners are designed such that the premixed flame rises uniformly above the burner surface, producing a flame which is entirely one-dimensional. This type of flame has a uniform velocity distribution and is not subject to flame stretch-
Figure 2.1: Example of a typical Bunsen flame. The flame consists of a bright, clearly defined premixed flame surrounded by an amorphous diffusion flame.

ing seen in Bunsen burners, which can alter the distribution of temperatures and molecular species. Thus, the flat flame burner produces a premixed flame which is as close to the ideal, planar flame as possible. In order to create a flat flame, the burner must have a surface with numerous outlet ports arranged in staggered formation and is small enough to prohibit flame propagation into the mixing chamber. The McKenna burner is an example of a commercially available flat flame burner, but many flat flame burners are custom built devices, such as the design used by
Hartung et al., [21] seen in Figure 2.2. Depending on how efficiently the fuel and air are mixed and the equivalence ratio of the flow, a flat flame burner may not produce a noticeable diffusion flame.

As these are open air burners, the flame burns at atmospheric pressure and is exclusively laminar. Previous work is usually not conducted at pressures above atmospheric or in the turbulent flow regime. Exceptions to this include research by Sakhrieh et al. [28], who used a high pressure chamber to study turbulent flames, and investigations of plasma discharges by Leonov et al. [29] and Starikovskii. [30]
2.2.2 Electrode Geometry

Electrode geometry varies heavily between experiments. Numerous geometries exist, but the electrodes typically seen in the literature are grid electrodes and ring electrodes. Grid electrodes exhibit a great diversity of forms, with some being custom made from wires or from perforated metal sheet. These are common because they are easy to construct and install. The electric field streamlines produced by grid electrodes tend to be approximately linear in shape. The field lines produced by ring electrodes, by comparison, are not truly linear. Occasionally, nozzle electrodes are used in experiments, wherein a charged nozzle is placed on top of the burner much like the setup used by Ryu et al. [10] Although this electrode geometry is the closest in form to that which would be used if the burner were part of a gas turbine, this geometry is not used very often due to its physical complexity and difficulty in modelling the equations, and thus its effects are not well quantified compared to other electrodes. A thorough understanding of how electrode geometry influences combustion is, at the moment, sparse because comparisons of electrode geometries in the same paper is rare.

Regardless of electrode geometry, almost all in the literature are positively biased. A few workers have used at negatively biased electrodes, and the results dont always agree. Marcum and Ganguly [12] stated that they did attempt observations with a negatively biased electrode, but could not detect any flame modification. This is disputed by Kim et al. [6] and Hu et al. [31], both of whom demonstrate an increase in blowoff velocity using a DC voltage with negative polarity.
2.2.3 Electrode Voltage

As with electrode geometry, the supply voltage for the electrode shows a diversity of forms. Since many effects become more obvious as the electric field grows stronger, a high potential is frequently needed and it is common to see maximum values of up to 10 kV. The global electric field produced by this is highly variable and depends on the separation distance of the electrodes in the experimental setup. Notable exceptions include research by Zhang et al. [19] and by Marcum and Ganguly [12], who tend to run at a maximum of 3 kV, producing a global electric field of 60 and 75 kV/m respectively. The type of voltage applied to the electrode, on the other hand, is highly varied in the literature, and tends to either be DC [7] [32] [16] [28] [20] or nanosecond-duration pulsed DC [12] [30] [27] [33] with AC sources being less common. [6] [10] The effect of both DC and pulsed DC voltage is noticeably different compared with AC voltage, as these modes can promote the collapse of the flame front into a wrinkled geometry under certain conditions. [12] [13] [14] [33] AC voltages do not do this, though in lifted flames, the flame front was observed to move up and down in proportion to the frequency, [6] [10] an effect which is believed to be tied to the collision time of the ions. AC voltages were also observed by Kim et al. [34] to affect flame spread rates, with low frequency AC suppressing spread rates and high frequencies (those above 200 Hz) increasing spread rates. Surprisingly, Kim et al. also observed that excessive voltage caused flame extinction, and that increasing the frequency up to 400 Hz resulted in an approximately linear decline in the voltage required to achieve this. All three voltage modes draw very little power regardless of
the applied voltage. At most, the electrodes in the literature dissipate on the order of 1 W electrical power, exhibiting control over flames with a thermal power on the order of 1 kW, [12] [13] [16] [30] making this a very energy-efficient way of influencing the flame.

2.2.4 Instrumentation

2.2.4.1 Imaging Techniques

Choice of instrumentation varies widely depending on the experiment. Observing the macroscopic effects of the electric field typically calls for a high speed camera, and information about the flame can be obtained through recording the emissions from certain molecules within the flame via specially made band-pass filters. This process, called chemiluminescence, is one of the simplest optical diagnostics on flames, and is a very common method used in the literature. [13] [14] [19] [20] [35] A similar method is laser diagnostics, such as Planar Laser-Induced Fluorescence (PLIF) imaging, which uses a laser to excite molecules within the flame in addition to the bandpass filter. Like conventional chemiluminescence setups, PLIF provides a non-intrusive way of observing the flame. However, it provides a more detailed results than can be achieved with a passive setup and demonstrates a strongly linear relationship between emission luminescence and laser power, thus it has gained popularity and has appeared in a number of recent publications. [7] [9] [27] [36] Other methods used for observing macroscopic changes to the flame include Particle Image Velocimetry (PIV), which uses lasers to demonstrate how the flow field in a 2-dimensional flame
changes in response to an electric field, [7] [9] [33] and temperature observations with pyrometers, which were used by Marcum and Ganguly [12] as well as Saito et al. [32] to record changes in the temperature profile in response to electric fields.

2.2.4.2 Non-Imaging Techniques

Imaging methods are useful for looking at the entire flame at a single point in time and looking at radical production, but are not capable of observing plasma-related properties. Mass spectrometers have been successfully used by Goodings et al. [37] [38] [39] and Hayhurst et al. [40] to gain information about ions within the flame. However, the most common instrument used to do this is a Langmuir probe, an electrostatic probe which is inserted into the plasma, and in its simplest form is a thin wire. Irving Langmuir’s probe theory was originally developed for use with probes measuring the properties of collisionless plasmas (see Chapter 3 for a full definition), found in in plasmas with a high energy or which exist under low pressure. Flames are low energy plasmas with a high density, making them highly collisional. Although Langmuir’s theory cannot be used with these types of plasmas, a corrected probe theory was developed by Su and Lam [41] which applies to collisional plasmas, allowing the use of Langmuir probes in flames without a significant reworking of either the Langmuir probe or the instruments associated with it. Extensive commentary on this modified probe theory, including how to identify different measurement regimes with a Langmuir probe, is found in the work of Smy. [42]

A variety of Langmuir probes exist in different configurations. Depending on
the needs of the measurement, Langmuir probes can be found in planar, cylindrical, and spherical shapes, [43] but Langmuir probes in the literature are generally cylindrical probes, a discussion of which is given by Parrot et al. [44] Cylindrical probes can be further classified based on how many wires are electrically connected to the data acquisition system. Single probes, such as the one shown in Figure 2.3, contain only one wire, while double probes have a second wire present. Both single probes and double probes are used in taking measurements of flames. Triple probes have also appeared in the literature, [45] but this is a very uncommon configuration.

2.2.5 CFD

Thus far, determination of flame properties in this area of research has exclusively been done via physical measurements. Computational Fluid Dynamics simulations of flames, to date, comprise only a relative handful of the published literature on Plasma Assisted Combustion, though this is slowly changing. An early attempt at numeric simulations of electrically modified flames in multiple dimensions came from Hu et al., [31] who used the PHOENICS and CHEMKIN computer codes to solve
the mass, momentum, and energy equations for both co-flowing and a candle-type flames. Using these codes, Hu et al. generated plots of the flame temperature, radical mass fractions, and flow fields as a function of position, both for an unmodified flame as well as with a positive voltage applied. Both a 20X20 and a 40X40 grid were used in this experiment with the majority of the nodes clustered near the reaction zone, though the authors did determine that a 20X20 grid was sufficient to generate an accurate solution. As a result of this success, more journal articles which either include sections of CFD simulations or which focus on simulation entirely have been forthcoming, such as Boom et al., [16] Belhi et al., [11] and Prager et al. [46] Belhi et al. completed an in-depth analysis of the flame, taking an approach similar to what Hu et al. took, but noted that the 31 elementary reactions found in Hu’s work were insufficient to accurately model ion production, and opted to include 375 elementary reactions. In doing so, they were able to claim more accurate results than reported by earlier simulations, though their predictions for ion densities fell short of experimental results obtained by Goodings et al. [39] Given the increasing success and accuracy of these simulations, it can be expected that CFD will play an increasing role in this field.

2.3 Review of Literature Results

Several different types of observations have been recorded in the literature up to this point. Based on the common types of instrumentation previously reviewed, these observations include changes in flame speed, temperature, stability, and the pro-
duction of pollutants, both gaseous and solid. These effects positively correlate with increasing potential, and are typically associated with positively biased electrodes.

2.3.1 Flame Speed and Temperature

Both flame speed and temperature have been observed to change upon exposure to an electric field, and the effect is more pronounced with more powerful electric fields. This was observed by Marcum and Ganguly, [12] who measured the radial temperature profile of the flame at different locations using color pyrometers. In its normal state, the flames temperature profile tended to take on a bell curve shape, with the peak corresponding to the central axis of the flame. Applying an electric field, however, caused this distribution to flatten out, resulting in a temperature profile with a greatly reduced difference between the colder edges of the flame and the center. The same observations were recorded by Saito et al., [32] who observed that the effects were most strongly pronounced along the outer edge of the flame.

2.3.2 Stability

Changes to the stability of the flame are one of the key reasons for this research. Due to demand for reduced emissions from combustion, burners are increasingly being made to run leaner mixtures, thus reducing the formation of NOx. Lean combustion is, however, very susceptible to instabilities. Overall, stability increases with exposure to electric fields. This is most noticeable in lean and ultra-lean flames approaching the fuels Lower Flammability Limit (LFL). Ganguly [14] demonstrated this with a lean
Bunsen burner flame. The flame was first exposed to an electric field, then conditions were changed such that quenching would occur. While exposed to the electric field, however, the flame continued to burn. Removing the electric field inevitably resulted in the flame quenching. Similar results were found by Bak et al., [47] [26] while testing a premixed methane/air flame in a combustion tube. Combustion was tested at equivalence ratios both slightly under and slightly above the Lower Flammability Limit. Although complete combustion did not occur while the equivalence ratio was below the LFL, a flame was able to be sustained. Furthermore, combustion at equivalence ratios above the LFL occurred with a greater intensity, thus confirming the results found by Wisman et al. [13] as well as Marcum and Ganguly. [12] Other stability related findings include results found by Kim et al. [6] while testing the blowoff velocity of Bunsen burner flames. Electric fields were observed to increase the blowoff velocity in the experiment by a factor of 7, making the flame more resistant to flameouts while requiring electrical power on the order of only a few watts. This stabilizing effect was confirmed by Belhi et al. [11] via numeric simulation, as well as Sakrieh et al., [28] who determined that pressure enhanced electric field effects on the blowoff velocity. Of particular interest, however, is the control of more dangerous combustion instabilities. One such example of this is the thermoacoustic instability, wherein the heat release by combustion and acoustic perturbations in the equipment respond to each other in a positive feedback loop. This is not only noisy, it also contributes to mechanical wear on the burner. In more extreme cases, instabilities like this can damage or destroy more delicate components, such as turbine blades. Research published by Volkov et al. [20] suggest, however, that electric fields are an
effective way of dealing with thermoacoustic instabilities in flat flames by increasing their resonance frequency. They further demonstrated that this is most effective on lean flames, since lean combustion tends towards a lower resonance frequency.

2.3.3 Pollution Reduction and Control

Experiments by Saito et al. [32] measured the production of soot in an acetylene diffusion flame as a function of applied voltage and electrode position above the burner. Soot suppression was most effective when the electrode was placed higher above the burner, and at this electrode height, the soot mass ratio relative to an unmodified flame fell below 10% of the initial value when the supply voltage exceeded 5 kV. Almost all soot production ceased when the supply voltage reached 8 - 9 kV. Since Saito et al. observed increased temperatures, and higher combustion temperatures tend to promote more complete combustion, it is reasonable to conclude that the electrode voltage is coupled with the combustion temperature, and thus drives soot suppression this way. Research has also been done on the suppression of noxious gases as well, as demonstrated by Zhang et al. [19] Molar concentrations of CO and NO were tracked in the flames exhaust; both exhibited variability in concentration, but overall NO concentration was shown to decrease in the long run. CO concentration was also observed to fall, but was seen to sharply increase when the electrode had a potential above 3 kV. These patterns were tested over a range of flame speeds, and the patterns tended to hold no matter what the speed was. Although Sakhrieh et al. [28] were able to identify a reduction in CO, their results with other pollutants were mixed. N\textsubscript{2}O, a pollutant responsible for smog formation, was reduced in
concentration, but production of the compound NO increased. Between these two compounds, Sakrieh et al. concluded that the total NOx production increased by as much as 25% during the experiment.

2.3.4 Ion Densities

Differing results can be found for many of the flame properties investigated in the literature. However, ion densities appear to be the most consistent regardless of how the measurements were taken or what the experimental configuration was. Measurements are typically taken with Langmuir probes and the results are typically reported in units of m\(^{-3}\) or cm\(^{-3}\), although mass spectroscopy is a technique which has been used by some authors. [38] [39] Ion density measurements by Calcote, [48] Wortberg, [23] Goodings et al., [38] and MacLatchy [49] have all used physical instruments to measure the ion density of flames. Different peak ion densities were identified by the authors, but with the exception of MacLatchy, whose peak ion density was 10\(^{17}\) m\(^{-3}\), all of the authors measured a peak ion density on the order of 10\(^{16}\) m\(^{-3}\). Ion densities have also been the subject of CFD simulations by Belhi et al. [11] and Prager et al. [46]; both authors also reported a peak ion density on the order of 10\(^{16}\) m\(^{-3}\). The range of these results extend from 1.3 X 10\(^{16}\) m\(^{-3}\) for Belhi’s results all the way up to 4 X 10\(^{16}\) m\(^{-3}\) for Goodings et al.’s results. A more detailed comparison of these results and how they compare with the present work is provided in Table 5.1, found in Chapter 5, Section 5.1.1.

If desired, it is possible to alter the number density of ions, and can be accomplished through a couple of methods. Increasing the equivalence ratio can increase
ion density, but past a certain point, increasing the equivalence ratio reduces the number density of ions, as demonstrated by MacLatchy [49]. Changing the equivalence ratio from 1.2 to 1.7 caused a drop in number density within the premixed flame by as much as a factor of 4. More frequently, however, the flame is doped with something which is known to increase ionization. Different doping agents have different effects on the number density, which was demonstrated by Goodings et al. [39] using a hydrogen flame. Doping with salts tended to show a large increase in the ion saturation current, while doping with hydrocarbons tended to show a large increase in the electron saturation current. An interesting effect of doping is that it tends to shift the location of peak ionization. [49] Furthermore, very low concentrations of doping agents are required to accomplish this, implying that the chemiionized plasma is very sensitive to alterations.

2.3.5 Electron Temperature

Unlike ion density, the electron temperature in flames is not consistent across experiments, even though experiments exclusively use Langmuir probes, most them being double probes. Even within experiments, electron temperatures are typically reported as a range of values because results can vary by as much as 2,000 K or more depending on where the measurements are taken.

A good illustration of how results vary significantly in a single experiment is taken from Silla and Dougherty’s comparison of electron temperature measurements from single and double probes. [50] Double probe measurements were more consistent,
but their results ranged from 1,170 K - 2,200 K. Single probe results showed greater variability; depending on where the measurements were taken, electron temperatures as low as 1,120 K and as high as 7,720 K were observed. Very high electron temperatures are also reported by Engel and Cozens, who measured an electron temperature of 8,700 K, and Bradley and Matthews, who measured an electron temperature of 30,000 K. [51]

However, these examples are part of the extreme high end of temperature measurements, and other authors do not report such high electron temperatures. Porter reports a temperature range of 1,430 K - 2,000 K, [52] Clements and Smy report a temperature range of 1,500 K - 2,400 K, [24] and Wild et al. report a temperature range of 2,400 K - 4,000 K. [22] Although the range of values presented here is very wide, prior research focusing only on electron temperature is far less common than research which covers other properties. A more detailed comparison of these results and how they compare with the present work is provided in Table 5.2, found in Chapter 5, Section 5.1.2.
CHAPTER 3

METHODOLOGY

It’s easy to sleep floating around it’s very comfortable. But you have to be careful that you don’t float into somebody or something!

—Sally Ride

3.1 Experimental Setup

Two primary configurations are used in the present work. When measuring ion density, the configuration in Figure 3.1 is used. When measuring electron temperature, the configuration in Figure 3.2 is used. For measurements where imaging is needed, the only instrument present is an ICCD camera, and the only optics necessary are a single bandpass filter. The experimental setup used in this case is shown in Figure 3.3.

3.1.1 Burner

The present work makes use of a custom-built flat flame burner based a design used by Hartung et al. [21] A grid made from perforated brass sheet is used to prevent combustion inside the mixing chamber and defines the surface of the burner. Further flashback suppression is provided by steel wool placed in the mixing chamber near the
Figure 3.1: Experimental setup for measuring ion density. A single Langmuir probe is used in this configuration.

Figure 3.2: Experimental setup for measuring electron temperature. A double Langmuir probe is used in this configuration.
Figure 3.3: Experimental setup for measuring the relative concentration of combustion radicals using chemiluminescence. The camera pictured here uses a single bandpass filter.

chamber inlet. This also has the effect of promoting mixing of the fuel and oxidizer prior to combustion. A premixed flame is studied instead of a diffusion flame. This was chosen because most real combustors operate with premixed flames.

Methane is used as the oxidizer for this flame and compressed air is used as the oxidizer. The equivalence ratio, the fuel-air ratio of the flame compared against the fuel-air ratio of a stoichiometric reaction, is fixed at 1.0. The fuel volumetric flow rate is set at 0.72 SLM and the oxidizer volumetric flow rate being set at 6.81 SLM. This results in a total volumetric flow rate of 7.53 SLM.
3.1.2 Electrode

A flat grid electrode made from perforated steel sheet is used to generate the electric field and is suspended above the burner from a non-conductive unistrut bar. This electrode design is very straightforward to implement and generates straight electric field lines from the electrode to the burner surface, which is connected to an electrical ground to create a complete circuit. The electrode is placed 7.62 cm above the burner. A Matsusada AU-10P60 series high voltage power supply provides up to 10 kV of DC power to the electrode. In this configuration, the electrode supplies a global electric field up to 131.2 kV/m. This is equivalent to a reduced electric field, the ratio of electric field to the density of neutral particles, of 38 Td. To compare, the breakdown voltage for a situation like the one in the present research can be calculated using Paschen’s law

\[
V_{breakdown} = \frac{4.36 \times 10^7 \times pd}{Ln(pd) + 12.8} \quad (3.1)
\]

This yields a breakdown voltage of 324 kV, or a global electric field of 4260 kV/m. The experiment therefore occurs well below the threshold for Townsend discharge, which is where arcing occurs due to collisional ionization of molecules. Current is limited to a maximum of 60 mA.
3.1.3 Langmuir Probe

A Langmuir probe is used to collect the majority of data. The probe filament is made from 0.127 mm diameter tungsten wire, which is held in place and protected using a 2 mm diameter alumina tube. 22 AWG copper wire is inserted into the opposite end of the tube to electrically connect the filament to a power source and to prevent the filament from sliding out of the tube. The exposed length of tungsten wire ranges between 2.54 mm for the double probe and 4.70 mm for the single probe, with the wire length being accounted for in calculations. The double probe used in this research is arranged in an asymmetric geometry, with the two probe cores offset by 90°. Both probes are connected to a Keithley 2410 Sourcemeter, which is used to bias the probes as deemed appropriate.

Both the single and double core probes are positioned in the flame using a Velmex two axis linear stage. After recording data, the probe is retracted and allowed to cool before attempting measurements again so as to extend the lifetime of the filaments and reduce the risk of erroneous measurements. For the single probe, the maximum acceptable residence time was deemed to be 7 seconds. The basis for this determination is shown in Figure 3.4.

3.2 Sheaths

Central to the present work is the concept of sheaths. In plasma physics, the sheath is region which exists between a plasma and a solid surface. Here, because electrons move faster than ions, the electron flux to the surface is greater as well.
Initially, a negative charge builds up on the surface. The result is that the surface begins to repel electrons and attract ions. This process quickly reaches equilibrium, wherein a transition region between the plasma and the surface exists which is predominantly occupied by ions. This means sheath violate the common assumption of quasi-neutrality, where it is assumed that an equal number of ions and electrons exist in the plasma and are distributed such that the plasma as a whole can be assumed to have no net charge. While it is possible for a sheath to be ion-repelling, the majority of sheaths found outside of laboratory conditions are electron-repelling.

Sheaths can also be thought of as a voltage boundary condition between the plasma and the surface. Both have a potential, but the surface is frequently a ground. As shown in Figure 3.5, when a plasma comes into contact with a surface that ex-
Figure 3.5: Example of the potential within a plasma in contact with a surface with an arbitrary potential. The sheath is shown along the domain from $x = 0$ to $x = D$, and shows the steep decline in potential when transitioning from the plasma to the surface. Figure is adapted from Xu [53]

Ions must achieve sufficient energy to enter the sheath, otherwise they will be shielded from the surface potential along with the rest of the plasma. As they approach the sheath, ions are accelerated until they either enter the sheath or fail to do so. The region over which this occurs is called the pre-sheath. As shown in Figure 3.5, the pre-sheath lacks the sharp potential gradient of the sheath and changes potential slowly. Furthermore, the pre-sheath is both much larger than the sheath.
and can be assumed to be quasi-neutral like the plasma is. In general, the pre-sheath can be ignored, as it is not as critical to understanding topics in plasma physics as the sheath is.

### 3.3 Langmuir Probe Theory

The original application for Langmuir probes is in a fully ionized, stationary plasma under a vacuum. In this environment, the plasma is said to be collisionless, which means the relation of the ion-neutral mean free path to the Debye length is

\[ \lambda_{MFP} >> \lambda_D \]  

(3.2)

where the Debye length is defined as

\[ \lambda_D = \sqrt{\frac{\epsilon_0 k T_e}{n_e e^2}} \]  

(3.3)

Plasmas are only collisionless when the electron temperature is high, such as in a Tokomak reactor, or the electron density is low, as it is in a vacuum chamber. In a typical flame, the particle density is at least three orders of magnitude greater than that of a vacuum plasma, but the electron temperature is close to that of the flame. This makes the flame a collisional plasma.

In the case of a flowing plasma, the important quantities governing the probe,
as described by Smy [42], are the electric Reynolds number

\[
Re_e = \frac{2r_p U_e}{\mu_i kT_e} \quad (3.4)
\]

the ratio of the Debye length to the probe radius

\[
\alpha = \frac{\lambda_D}{r_p} \quad (3.5)
\]

and the dimensionless potential

\[
\chi = \frac{eV_b}{kT_e} \quad (3.6)
\]

The results obtained with a Langmuir probe depends on its configuration. When inserted into a plasma, if the probe does not receive a bias, a single probe will read the floating voltage of the plasma. This is where no net current is collected by the probe, since the plasma is quasi-neutral. A double probe will read no voltage until it is biased, since the voltage on both probe cores will be equal. Applying a bias voltage to any Langmuir probe produces a current-voltage response which depends mostly on the properties of the plasma. The typical response is seen in Figure 3.6, which produces an S-curve shape. Double probes always produce a current-voltage response like this; single probes do produce S-curves like this, but they tend to be very asymmetric, such as the one shown in Figure 3.6, and are more likely to do this for collisionless plasmas. Collisional plasmas often do not produce S-curves, instead
Figure 3.6: Typical current-voltage behavior of a double probe as the bias voltage is increasingly positive. Plot is adapted from Wild et al. [22]

producing a current-voltage response appearing more similar to a J-curve, such the one shown in Figure 3.8.

The S-curve response of a Langmuir probe can generally be broken up into three separate regions. The first two are the saturation regions, where the probe has been sufficiently biased such that it has collected all ions or electrons in its immediate vicinity. These regions correspond to the regions on the ends of the curves seen in fjdjks, which contain both ion saturation (left side) and electron saturation (right side). Because the probe has collected all of the opposite charges in these regions, it is possible to calculate ion density from the current reaching the probe. The third region, located in the middle of the curve, is typically referred to as the linear region. The slope of the linear region is proportional to the inverse of the electron temperature, so a shallower current-voltage response is associated with more energetic
electrons. Details on how to exactly calculate the electron temperature are described later in Section 3.3.2.

### 3.3.1 Ion Density

Smy [42] describes three different flow regimes associated with Langmuir probes in flowing collisional plasmas: Diffusion-convection, sheath-convection, and E field-convection. In the diffusion-convection regime, the boundary layer which forms on the probe due to the plasma flow is thicker than the probe sheath, while the opposite is true of the sheath-convection regime. The probe operates in the diffusion-convection regime if \( R_e \alpha^2 \lambda^2 \ll 1 \) and is operating in the sheath-convection regime.
if $Re_e \alpha^2 \chi^2 >> 1$. In the E field-convection regime, the sheath extends so far out from the probe surface that ions may enter the sheath but wind up being swept downstream without ever reaching the probe. In this regime, $Re_e \alpha^2 > 1$. Equations describing the ion currents to a negatively biased probe exist for all three regimes. Although Smy does identify research which has been conducted in all three regimes, ion density measurements in the literature are typically associated with the sheath-convection regime.

To ensure continuous operation in the sheath-convection regime, a probe bias
of -5 V was selected. For this type of analysis, the electron temperature is frequently assumed to be equal to the flame temperature. The operating characteristics of the single probe at the specified bias voltage are shown in Table 3.1.

Table 3.1: Electric characteristics of the single probe

<table>
<thead>
<tr>
<th>(Re_e\alpha^2\chi^2)</th>
<th>14.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Re_e\alpha^2)</td>
<td>0.016</td>
</tr>
<tr>
<td>(\alpha\chi)</td>
<td>6.45</td>
</tr>
</tbody>
</table>

If ion densities are to be measured, the quantity \(\alpha\chi\) is important. When \(\alpha\chi \ll 1\), the sheath can be assumed thin. In this case, the sheath is not much wider than the Langmuir probe. In the case where \(\alpha\chi > 1\), the sheath is much wider than the Langmuir probe, and so must be treated as thick. At the specified bias voltage, the quantity \(Re_e\alpha^2\chi^2\) is much greater than unity, so the probe is for certain not operating in the diffusion-convection regime. Furthermore, \(Re_e\alpha^2 < 1\), so the probe is operating in the sheath-convection regime. The value of \(\alpha\chi\) means that the sheath is not thin compared to the probe diameter, so a thick sheath ion density equation applies. For cylindrical probes operating in the sheath-convection regime, Clements et al. [43] provide a means of calculating the ion density within a thick sheath

\[
n_i = \left( \frac{I_{i,0}}{5.3(\epsilon_0\mu_i)^{0.25}V_b^{0.5}\rho_p^{0.25}l_p} \right)^{1/0.75} * \frac{1}{eU} \quad (3.7)
\]

3.3.2 Electron Temperature

For electron temperature measurements, probe cooling of the plasma is an issue, since this produces unreliable electron temperature measurements. If probe
cooling is to be avoided, one of the two conditions must be satisfied [42]

\[
\frac{m_e \bar{v}_e r_p}{m_i \lambda_{MFP} U} < 1 \quad (3.8)
\]

\[
Re_e \alpha^2 \chi^2 \sim 1 \quad (3.9)
\]

This can be addressed by recording probe currents over a wide range of voltages, a step which is often a necessary part of techniques for measuring electron temperature in the first place.

One drawback of using single probes when trying to measure electron temperature is they do not reach electron saturation easily. A typical single probe’s current-voltage plot appears like a J-curve the more positively biased it becomes, such as the curve shown in Figure 3.8. This is why it is necessary to calculate the natural logarithm of the probe current when calculating the electron temperature from measurements produced by a single probe. Additionally, as was determined by the work of Silla and Dougherty [50] electron temperatures measured by a single probe can produce a wide range of results depending on how close to ion saturation the temperature calculation is made and single probes tend to produce very high temperature measurements. These issues mean that a single probe is often not an accurate means of obtaining electron temperatures in a high pressure plasma. An alternative device for measuring electron temperatures is the use of a double probe, which is seen more often in the literature and has a variety of well tested approaches to calculating the electron temperature from current measurements. A method described by Saito et
al. [8] shows how to calculate the electron temperature in a collisional plasma where the probes are the same length

\[ T_e = \frac{eI_{i,0}}{6.16 k} \frac{dI}{dV_b} \]  

(3.10)

3.4 Chemiluminescence

A complete schematic showing the relationship between different ion reactions in Prager et al. [46] demonstrates the importance of the molecule CH to ion production. A relevant ionizing reaction involving CH is

\[ CH + O \rightleftharpoons CHO^+ + e^- \]  

(3.11)

This reaction enables the following reactions to take place

\[ CHO^+ + H_2O \rightleftharpoons H_3O^+ + CO \]  

(3.12)

\[ CHO^+ + e^- \rightleftharpoons CO + H \]  

(3.13)

\[ CHO^+ + C_2H_5OH \rightleftharpoons H_3O^+ + CO + C_2H_4 \]  

(3.14)

\[ CHO^+ + CH_2CO \rightleftharpoons C_2H_5O^+ + CO \]  

(3.15)

\[ CHO^+ + CH_3 \rightleftharpoons C_2H_5O^+ + H \]  

(3.16)

\[ CHO^+ + CH_3OH \rightleftharpoons CH_5O^+ + CO \]  

(3.17)
Additionally, CH is a reactant in many non-ionizing reactions, such as the following [54]

\[ CH + CH_4 \rightleftharpoons C_2H_4 + H \]  \hspace{1cm} (3.18)
\[ CH + O \rightleftharpoons CO + H \]  \hspace{1cm} (3.19)
\[ CH + O_2 \rightleftharpoons CHO + O \]  \hspace{1cm} (3.20)
\[ CH + CO \rightleftharpoons HC_2O \]  \hspace{1cm} (3.21)
\[ CH + CO_2 \rightleftharpoons CHO + CO \]  \hspace{1cm} (3.22)
\[ CH + H_2O \rightleftharpoons CH_2O + H \]  \hspace{1cm} (3.23)

CH is therefore an important radical when investigation hydrocarbon flames.

The same is true of OH, which is also produced in significant quantities in a hydrocarbon flame. An example of OH reactions in a flame are as follows [54]

\[ OH + CH_4 \rightleftharpoons H_2O + CH_3 \]  \hspace{1cm} (3.24)
\[ OH + OH \rightleftharpoons H_2O + O \]  \hspace{1cm} (3.25)
\[ OH + H + M(2) \rightleftharpoons H_2O + M(2) \]  \hspace{1cm} (3.26)
\[ OH + HO_2 \rightleftharpoons H_2O + O_2 \]  \hspace{1cm} (3.27)
\[ OH + CO \rightleftharpoons CO_2 + H \]  \hspace{1cm} (3.28)
\[ OH + CH \rightleftharpoons CHO + H \]  \hspace{1cm} (3.29)
Both CH and OH are not easy to detect. However, it is possible to detect the excited forms of these radicals, usually represented as CH* and OH*, using a specially designated bandpass filter. In the present work, 50 images of the flame are collected at each applied voltage for both CH* and OH* using a Quantel ICCD camera at a sampling rate of 5 Hz. Each image is a 6,000 µs exposure, and the image intensifier is applied to each image for 2,500 µs. The CH* filter works at a wavelength of 431 nm and the OH* filter works at a wavelength of 308 nm.

Images are post-processed using the program ImageJ. Each image is passed through a 50 pixel sliding paraboloid background subtraction algorithm to remove any remaining noise and then then cropped down to focus entirely on the premixed flame. An average image is created from this set of images.

Trends in ion production are assessed via the total brightness of the average image, which is assessed using the image histogram. The approach used here to determine the total brightness weighs each pixel against its intensity. This has the effect of exaggerating changes in the image brightness, but also helps to downplay the contribution of noise and dim pixels not associated with the flame to the total brightness. An equation for the total image brightness used in this analysis is as follows

$$\psi = \sum_{i=1}^{n} \delta_{i}x_{i}$$  \hspace{1cm} (3.30)
3.5 Limitations

3.5.1 Probe Limitations

One of the key limitations of the current research involves the Langmuir probe itself. The Langmuir probe used is constructed with a thin tungsten wire being utilized for the filament. Tungsten's high melting point makes it the preferred metal for making the filament. Although cooling is used in some Langmuir probes, the small size prevents active cooling from being effectively utilized with this probe. Experience has shown that the probe response when heated over long periods of time can change. Additionally, the oxidizing environment inside the flame is destructive to the probe filament, and over time, the flame will consume the filament. Both issues are solved by retracting the probe after every measurement and allowing it to cool.

Even under ideal circumstances, measurements with Langmuir probes can be noisy. The flame presents an even more challenging environment because it is so weakly ionized and exists at a much higher pressure compared to the plasmas Langmuir probes are most commonly used to observe, such as astrophysical plasmas. Noise can be dealt with in several ways, depending on the type of measurement needed. For ion density measurements, a sample of at least 100 current measurements can be collected and averaged, providing the mean and standard deviation of the ion density values. A similar approach can be taken when finding electron temperature with the double probes, where five sets of measurements are taken to determine the average current-voltage response of the plasma. 11 point Savitsky-Golay smoothing is then applied to the averaged data in order to remove any additional noise present.
3.5.2 Chemiluminescence Limitations

Although less prone to noise than a Langmuir probe, the flame images are not without unique problems. The small size of the premixed flame tends to result in very dim images. With the addition of an optical filter to capture the CH* images, it becomes almost impossible to visualize the flame, which risks the loss of image detail. Capture rates with the ICCD camera are also very slow compared with a high speed camera, thus the flame may shift slightly over time. To avoid a loss of detail in the images, each image is recorded and stored as a 16 bit Tiff. This does not solve problems with the difficulties visualizing the flame, however. To make the flame visible, a set of 50 images are captured and merged into an average image, and each average image is expressed on a nonstandard intensity scale. This is permissible because it changes how the pixels are represented visually but does not change the actual pixel intensity. Averaging the images also has the effect of preventing any motion of the premixed flame from becoming an issue; the premixed flame moves very little to begin with, thus any motion in the premixed flame will be erased by the averaging process.

Because chemiluminescence is an image-based approach to analyzing the flame, absolute measurements of species concentrations cannot be made without a reference image for which a quantity is known. For this experiment, a reference image describing how dense the population of CH* is at a given location in the flame would be necessary to determine exactly how much CH* is present. This could not be obtained, thus absolute measurements with the chemiluminescence system is impossible. Relative
changes to the CH* can still be measured, however. This is accomplished using the total brightness of the average image.

3.6 Assumptions

The following simplifying assumptions are made in the present work

- The plasma is axisymmetric
- The diffusion flame produced by the burner does not affect the premixed flame
- Electric field streamlines are one-dimensional along the vertical axis
- Magnetic fields can be neglected

Assuming that the plasma is axisymmetric can be justified due to the shape of the flame, which is laminar and therefore axisymmetric for this burner type. Other workers who have observed these types of laminar flames have also reported axisymmetric flame properties, an unsurprising result given that laminar flames tend not to have the kind of localized perturbations that turbulent flames do. The plasma is therefore treated as axisymmetric about the center axis of the burner. Using this assumption, it is necessary to obtain measurements for only half the flame.

Treating the electric field streamlines as one dimensional is possible due to the shape of the electrode, a flat metal plate with no large cavities in the center. In order for the electric field streamlines to be perfectly straight, the electrode would need to be significantly larger than the burner. The electrode has a finite size that is only slightly larger than the burner, so some curved streamlines will appear, particularly
near the edge of the flame. A simulation of the electric field, shown in Figure 3.9, shows that the curving is minor near the burner surface and almost nonexistent in the middle of the flame.

Magnetic fields require current flow to be generated. Current flow is present in the electrode and in the burner going to ground, but the current in both cases is on the order of $10^{-9}$ Amperes. The magnetic field resulting from this is vanishingly small, thus magnetic field effects on the flame can be neglected.
Figure 3.9: Plot of potential produced by the experimental setup with streamlines present. The white space in the middle represents the burner. The streamlines become nonlinear only near the edge of the burner, justifying the assumption that the streamlines are straight lines. Image produced using FEMM.
CHAPTER 4

RESULTS

With insufficient data it is easy to go wrong.

—Carl Sagan

4.1 Flame Temperature

The flame temperature was measured with 1/16 in. diameter, ungrounded, type K thermocouple with an inconel sheath placed in the center of the premixed flame. Thermocouples used in measuring the temperatures of hot gases have very high radiative heat losses which need to be compensated for. To do this, the heat transfer to the thermocouple by convection was assumed equal to the heat transfer away from the thermocouple by radiation and that conduction within the thermocouple was not important. This approach, as described by Brundage et al., [55] results in the following equation

\[ h(T_f - T_{TC}) = \varepsilon \sigma (T_{TC}^4 - T_{\infty}^4) \] (4.1)

The thermocouple recorded a temperature of 1123 K in steady state at the stoichiometric equivalence ratio, which corresponds to a film temperature of 710 K. Using the properties of air at the film temperature and a flow velocity of 0.369 m/s,
this results in a corrected flame temperature of 2153 K.

4.2 Langmuir Probe Analysis

For ion collection to occur, the Langmuir probe must be biased sufficiently such that all of the ions near the probe can reach it, but it must not be so excessively biased that the probe begins collecting ions far downstream of it. Additionally, the flow velocity of the flame must not be so high that the ions are swept downstream before being collected by the probe. Tests with the single probe, wherein the bias was swept through multiple values, were used to determine what bias should be used for ion collection. The results show that any value above -2 V will result in ion saturation.

4.3 Ion Density

Ion densities show a distinct distribution. Densities are very low near the radial edge of the burner and increase the closer the probe is moved to the burner centerline, with peak densities being coincident with the center axis of the burner and 5 - 10 mm from the burner edge. Ion densities are also highest near the burner surface, with densities falling off rapidly as the probe becomes axially more distant from the burner surface.

Applying a DC electric field was observed to increase the ion density in the premixed flame. Near the burner, ion densities increased by as much as an order of magnitude. As seen in Figure 4.1 and Figure 4.2, the maximum gain in ion densities occurs when the electrode is biased to 6 kV. The unmodified flame has a maximum
density of $2 \times 10^{16}$ m$^{-3}$, but this increases to $1 \times 10^{17}$ m$^{-3}$ once the bias voltage reaches 6 kV. At higher voltages, however, the ion densities start falling. A similar pattern, as shown in Figure 4.3 and Figure 4.4, can be seen 5 mm downstream of the burner, although here the ion densities peak at 4 kV instead of 6 kV.

![Graph](image.png)

**Figure 4.1**: Ion densities measured at 0 mm above the burner surface for an electrode voltage of 0 - 4 kV.
Figure 4.2: Ion densities measured at 0 mm above the burner surface for an electrode voltage of 6 - 10 kV.

An overall picture of ion responses to the electric field can be seen in the ion density gradient maps, shown in Figure 4.5, Figure 4.6, and Figure 4.7. Initially, ions have a fairly even distribution, ranging from $1 \times 10^{16} \text{ m}^{-3}$ to $3 \times 10^{16} \text{ m}^{-3}$. The distribution changes significantly once electric fields have been applied. The significant concentration of ions near the burner is particularly visible, with the ion density growing to $1 \times 10^{17} \text{ m}^{-3}$. Notably, Figure 4.6 and Figure 4.7 show that this concentration comes at the expense of ion densities far away from the burner, with measurement locations 4 mm and 5 mm from the burner showing particularly strong depletion.
Figure 4.3: Ion densities measured at 5 mm above the burner surface for an electrode voltage of 0 - 4 kV.

Figure 4.4: Ion densities measured at 5 mm above the burner surface for an electrode voltage of 6 - 10 kV.
Figure 4.5: Gradient map of ion concentrations at an electrode voltage of 0 kV. The solid black line represents the premixed flame.

Figure 4.6: Gradient map of ion concentrations at an electrode voltage of 6 kV. The solid black line represents the premixed flame.

Figure 4.7: Gradient map of ion concentrations at an electrode voltage of 10 kV. The solid black line represents the premixed flame.
4.4 Electron Temperature

The approach used by Saito et al. [8] produces the results shown in Table 4.1. At the center of the flame, electron temperature has a general tendency to increase as the electric field strength is increased as well. The effect is particularly strong at 0 mm above the burner; although electron temperature is lowest near this location, the electron temperature increases very quickly and becomes the highest out of all electron temperature measurements. The electron temperature at this point climbs significantly when subjected to a low strength electric field and stays elevated until the electrode voltage reaches 4 kV. Some electron temperature measurements were unable to be determined due to probe errors; these measurements are marked as “n/a”.

In the absence of an electric field, the electron temperature ranges from 3000 K to 6600 K, with lower electron temperatures tending to occur close to 0 mm above the burner and 5 mm above the burner.

Table 4.1: Electron temperatures measured in the center of the flame as a function of electrode voltage

<table>
<thead>
<tr>
<th>Voltage (kV)</th>
<th>Distance from burner (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>3276</td>
</tr>
<tr>
<td>2</td>
<td>14417</td>
</tr>
<tr>
<td>4</td>
<td>14863</td>
</tr>
<tr>
<td>6</td>
<td>11401</td>
</tr>
<tr>
<td>8</td>
<td>4160</td>
</tr>
<tr>
<td>10</td>
<td>4737</td>
</tr>
</tbody>
</table>
The same general trend towards greater electron temperatures as the electrode voltage increases can be seen at the edge of the flame, as shown in Table 4.2. Additionally, the range of electron temperatures near the edge of the flame is the same as it is near the center of the flame, and when no electric field is applied, the distribution of temperatures shows the same pattern wherein the electron temperatures tend to be higher as the probe is moved farther away from the burner.

**Table 4.2:** Electron temperatures measured at the edge of the flame as a function of electrode voltage

<table>
<thead>
<tr>
<th>Voltage (kV)</th>
<th>Distance from burner (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0 3734 3072 4040 5276 6497 4845</td>
</tr>
<tr>
<td>2</td>
<td>5098 5759 6813 10730 6335 4829</td>
</tr>
<tr>
<td>4</td>
<td>5568 5342 n/a 7837 3968 n/a</td>
</tr>
<tr>
<td>6</td>
<td>6969 3639 n/a n/a n/a n/a</td>
</tr>
<tr>
<td>8</td>
<td>8225 3186 n/a n/a n/a n/a</td>
</tr>
<tr>
<td>10</td>
<td>n/a 1758 n/a n/a n/a n/a</td>
</tr>
</tbody>
</table>

Although electron temperature does increase, it either sharply drops once the electrode voltage exceeds 4 kV or the probe measurements are unable to extract meaningful electron temperatures. This occurs at multiple locations within the flame. For electrode voltages past 4 kV, it is therefore reasonable to conclude that the probe measurements are unreliable.

### 4.5 Chemiluminescence

Prior to chemiluminescence imaging, measurements of the flame height were taken using an image of the flame with no electric field applied. Measurements indi-
cated that the premixed flame had a height on the order of 5 mm above the burner surface. Images of the CH* and OH* concentrations within the premixed flame are shown in the false color images seen in Figure 4.8 and Figure 4.9. The brightest regions of the images, corresponding to the locations of maximum CH* concentration, appear in the middle of the premixed flame. The general shape of this bright region does not shift in response to the electric field, implying that the premixed flame does not change shape in response to the electric field. Additionally, CH* production falls off very rapidly downstream of the premixed flame, with only token amounts of CH* appearing in the diffusion flame. The same can be seen in the image of OH*.

![Image of a CH* chemiluminescence image for the unmodified flame](image)

**Figure 4.8:** Example of a CH* chemiluminescence image for the unmodified flame
The total brightness ratio, the ratio of the total brightness of a image for any given electrode voltage to the total brightness when no voltage is applied, is plotted in Figure 4.10, which presents the results for both CH* and OH*. A clear downward trend in image brightness can be seen as the electric field strength increases, with the image brightness stabilizing at an electrode voltage between 4 kV and 6 kV. This is similar to the results of the ion density measurements, which peak at the same applied electrode voltage. Since the total brightness is a function of the brightness of each pixel, a decrease in brightness points to a reduced radial concentration overall. Although both radicals show a decrease in image brightness as more voltage is applied to the electrode, CH* shows a slightly greater downward trend.
Figure 4.10: Ratio of total brightness at a given electrode voltage to the total brightness of the unmodified flame for both CH* and OH*

4.6 Error Analysis

Uncertainties in the physical aspects of the system were either determined by measurements or by manufacturer specifications. The tungsten filament had an uncertainty in diameter of ±0.0019 mm, while the probe length had an uncertainty of 0.025 mm. Uncertainty in the Velmex position was determined by the minimum step size of each axis. The horizontal minimum step size was 0.025 mm while the vertical minimum step size was 0.0064 mm.

Uncertainties in ion density are shown in Table 4.3. The uncertainties calculated here show both a spatial dependence and a voltage dependence similar to the mean ion densities in both locations. At 0 mm, uncertainties tend to be very similar, regardless of electrode supply voltage, although some variability is present. This is
not the case at 5 mm, where the uncertainty of the probe sometimes exceeds 100%. This can be attributed to the probe registering positive currents during operation when it should be registering only negative currents. Close to the boundary of the premixed flame, therefore, the Langmuir probe may have accuracy problems in a strong electric field.

**Table 4.3:** Uncertainties in ion density measurements

<table>
<thead>
<tr>
<th>Voltage (kV)</th>
<th>Uncertainty percentage of mean at 0 mm</th>
<th>Uncertainty percentage of mean at 5 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>17.9%</td>
<td>18.7%</td>
</tr>
<tr>
<td>2</td>
<td>35.8%</td>
<td>39.9%</td>
</tr>
<tr>
<td>4</td>
<td>24.4%</td>
<td>19.2%</td>
</tr>
<tr>
<td>6</td>
<td>22.3%</td>
<td>67.7%</td>
</tr>
<tr>
<td>8</td>
<td>20.3%</td>
<td>148%</td>
</tr>
<tr>
<td>10</td>
<td>22.4%</td>
<td>251%</td>
</tr>
</tbody>
</table>

Electron temperature presents a difficult situation when trying to determine uncertainty. Normal methods of determining uncertainty, such as taking the standard deviation of multiple measurements, are unavailable due to the lack of a large number of measurements. Uncertainty in electron temperature is therefore estimated at 30% of the values presented in Table 4.1. These values are shown below in Table 4.4.

Uncertainties in the image data are shown in Table 4.5. A MATLAB algorithm analyzed all 50 images captured by the camera found the uncertainty to be consistently 5% of the meant total brightness. This pattern held for both the CH* and OH* images across all electrode voltages.
Table 4.4: Uncertainties in electron temperature

<table>
<thead>
<tr>
<th>Voltage (kV)</th>
<th>Temperature uncertainty at 0 mm (K)</th>
<th>Temperature uncertainty at 5 mm (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>983</td>
<td>1216</td>
</tr>
<tr>
<td>2</td>
<td>4325</td>
<td>1818</td>
</tr>
<tr>
<td>4</td>
<td>4459</td>
<td>n/a</td>
</tr>
<tr>
<td>6</td>
<td>3420</td>
<td>n/a</td>
</tr>
<tr>
<td>8</td>
<td>1248</td>
<td>n/a</td>
</tr>
<tr>
<td>10</td>
<td>1421</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Table 4.5: Uncertainties in chemiluminescence measurements

<table>
<thead>
<tr>
<th>Voltage (kV)</th>
<th>Uncertainty percentage of mean for CH*</th>
<th>Uncertainty percentage of mean for OH*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>7.5%</td>
<td>5.0%</td>
</tr>
<tr>
<td>2</td>
<td>5.2%</td>
<td>5.4%</td>
</tr>
<tr>
<td>4</td>
<td>5.5%</td>
<td>5.5%</td>
</tr>
<tr>
<td>6</td>
<td>5.2%</td>
<td>5.1%</td>
</tr>
<tr>
<td>8</td>
<td>4.6%</td>
<td>6.1%</td>
</tr>
<tr>
<td>10</td>
<td>4.3%</td>
<td>4.9%</td>
</tr>
</tbody>
</table>
CHAPTER 5

DISCUSSION

If you want to have good ideas you must have many ideas. Most of them will be wrong, and what you have to learn is which ones to throw away.

—Linus Pauling

5.1 Comparison with Literature

5.1.1 Ion Densities

Table 5.1 compares the ion densities of flames measured by prior authors. Of all ion density measurements, MacLatchy’s results are the highest for an unmodified flame, exceeding all other measurements by a full order of magnitude. Peak ion density in the present work approaches MacLatchy’s numbers, but requires an electric field to do so. Results presented here are very close to other results found in the literature. Differences in the ion densities presented by the previous authors listed can be attributed to differences in experimental configurations, most notably the burners used. Furthermore, both Belhi et al. and Prager et al. obtained results from simulations based on GRI 3.0 rather than experimental measurements, and the simulations relied on a very lean flame. In general, however, the ion densities of
Table 5.1: Ion densities of selected flames with no electric field present

<table>
<thead>
<tr>
<th>Author</th>
<th>Fuel</th>
<th>Oxidizer</th>
<th>Peak ion density (m$^{-3}$)</th>
<th>Pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present work</td>
<td>$CH_4$</td>
<td>Air</td>
<td>$2 \times 10^{16}$</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Wortberg [23]</td>
<td>$CH_4$</td>
<td>air</td>
<td>$1.8 \times 10^{16}$</td>
<td>101 kPa</td>
</tr>
<tr>
<td>MacLatchy [49]</td>
<td>$C_3H_8$</td>
<td>air</td>
<td>$4 \times 10^{17}$</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Belhi et al. [11]</td>
<td>$CH_4$</td>
<td>air</td>
<td>$1.3 \times 10^{16}$</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Goodings et al. [39]</td>
<td>$CH_4$</td>
<td>$O_2$</td>
<td>$1.3 \times 10^{16}$</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Prager et al. [46]</td>
<td>$CH_4$</td>
<td>air</td>
<td>$1.7 \times 10^{16}$</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Calcote [48]</td>
<td>$C_3H_8$</td>
<td>air</td>
<td>$1.4 \times 10^{16}$</td>
<td>101 kPa</td>
</tr>
</tbody>
</table>

Previous work are comparable to the results of the present work, so it is reasonable to conclude that peak ion density is independent of burner geometry.

5.1.2 Electron Temperatures

The present work is among the few comprehensive attempts at measuring the electron temperature of a hydrocarbon flame when a DC electric field is present. Given that prior attempts at measuring the electron temperature do so without the presence of an electric field, any comparison with the existing literature is based entirely on temperature measurements of the unmodified flame. A comparison of results presented here with prior research using double Langmuir probes is described in Table 5.2. Electron temperatures for the present work is most in agreement with the results of Wild et al., who used a $CH_4$-$O_2$ flame at atmospheric pressure. Wild et al. record slightly higher flame temperatures, but this is likely due to $O_2$ being the oxidizer instead of air.
Table 5.2: Electron temperature of selected flames with no electric field present

<table>
<thead>
<tr>
<th>Author</th>
<th>Fuel</th>
<th>Oxidizer</th>
<th>Electron temperature (K)</th>
<th>Pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present work</td>
<td>$CH_4$</td>
<td>Air</td>
<td>2195 - 3538</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Silla and Dougherty [50]</td>
<td>$CO$</td>
<td>$O_2$</td>
<td>1150 - 2200</td>
<td>6 kPa</td>
</tr>
<tr>
<td>Porter [52]</td>
<td>$CH_4$</td>
<td>$O_2$</td>
<td>1430 - 2000</td>
<td>2 kPa</td>
</tr>
<tr>
<td>Wild et al. [22]</td>
<td>$CH_4$</td>
<td>$O_2$</td>
<td>2400 - 4000</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Clements and Smy [24]</td>
<td>$C_3H_8$</td>
<td>Air</td>
<td>1500 - 2400</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Engel and Cozens [56]</td>
<td>Coal</td>
<td>Air</td>
<td>8700</td>
<td>101 kPa</td>
</tr>
<tr>
<td>Bradley and Matthews [51]</td>
<td>$CO$</td>
<td>$O_2$</td>
<td>30000</td>
<td>10 kPa</td>
</tr>
</tbody>
</table>

Pressure is a controlling influence on flame temperature, with the two being directly proportional; higher pressure generally correlate with a higher the flame temperature. The same is generally true of the electron temperatures seen in Table 5.2. The exception is the results reported by Bradley and Matthews.

Bradley and Matthews’ CO-$O_2$ flame stands out as having the highest electron temperature for an unmodified flame, followed by Engel and Cozens’ coal-air flame. In both cases, the adiabatic flame temperature of CO and coal are higher than $CH_4$ or $C_3H_8$, which should lead to a higher electron temperature to begin with. Bradley and Matthews further state that high electron temperatures are the result of chemi-ionization, rather than collisional ionization, and that high electron temperatures can be observed in the CO-$O_2$ flame due to CO-$e^-$ collisions. The CO molecule, which is both vibrationally and electronically excited in their flame, collides with electrons such that electron velocity is increased, producing the elevated electron temperatures seen in the CO-$O_2$ flame.
5.2 Assessment of Mechanisms Affecting the Flame

Ion density is a key parameter to understanding what is occurring in flames. When ion densities change in response to an electric field, this can imply a change in ionization rate, ion-electron recombination rates, cross-field ion diffusion, or ionic wind effects. Cross-field diffusion is a nontrivial aspect of plasma physics, but is only important for plasmas in a magnetic field, which was absent from this experiment. This leaves both the ionic wind and dissociative recombination as effective mechanisms associated with changes to the ion population.

5.2.1 Ionic Wind Influence

Ion densities near the burner show a tremendous increase as the electric field strength increases. This is accompanied by a significant decline in the ion densities further downstream of the flame, particularly for the stronger electric fields observed. Diffusion acts against the formation of density gradients such as those observed in the flame, and the fact that ion populations downstream of the premixed flame become significantly depleted as the electric field becomes stronger strongly indicates a body force is acting upon the ions. This body force is the ionic wind, as has been identified by previous authors.

The strength of the ionic wind can be quantified by calculating the force density of the ionic wind. This is represented with the equation

\[ f = E n_i \] (5.1)
Figure 5.1: Ionic wind force density as a function of electrode voltage within the cathode sheath. The dashed black lines represent overall trends in growth of the force density.

Here, the force density is a linear function of electric field and ion density. As seen in Figure 5.1, this results in an increasing ionic wind strength in the premixed flame until the electrode voltage extends beyond 6 kV. At this point, growth in the force density slows significantly.

5.2.1.1 Cathode Sheath Influence

Since the ionic wind increases in strength with electric field, it would be anticipated that the ion densities would continuously increase. This is not observed; ion densities cease increasing after peaking at an electrode voltage of 6 kV, and in some locations within the flame, the ion densities decrease. The ionic wind can therefore
not be the only significant influence on the chemiionized flame.

An explanation for the ions not increasing is through the influence of the cathode sheath. As stated previously in Section 3.2, the sheath is a voltage boundary condition between the plasma and a surface. In this case, the surface is the burner, which also functions as the cathode towards which positive charges flow. As with most sheaths, the cathode sheath here is electron-repelling, so it tends to trap ions which enter it. In conjunction with the ionic wind, the cathode sheath explains why the ion densities tend to concentrate so strongly, even though diffusion should prevent this from occurring.

The size of the cathode sheath in a flame plasmas is described by Xu, [53] and more generally for collisional plasmas by Sheridan and Goree. [57] For a collisional plasma, the collision parameter \( \alpha_c \) must be > 1, where \( \alpha_c \) is defined as the ratio of the Debye length to the ion-neutral mean free path, and the ion-neutral mean free path is defined as the inverse product of the ion-neutral momentum transfer cross-section and the bulk ionization density. These take the form of the equations

\[
\alpha_c = \frac{\lambda_D}{\lambda_{MFP}} \quad (5.2)
\]

\[
\lambda_{MFP} = \frac{1}{\sigma_s n} \quad (5.3)
\]

Momentum transfer cross-sections for \( \text{H}_3\text{O}^+ \) collisions are not available, so the \( \text{N}_2-\text{N}^+ \) cross-section will be used instead. Assuming an electron temperature equal to the flame temperature, the momentum transfer cross-section is \( 1.41 \times 10^{-18} \text{ m}^2 \). [58]
Figure 5.2: Cathode sheath thickness within the flame. The red line indicates the height of the flame above the burner.

The approach taken by Xu describes the cathode sheath thickness for a collisional plasma as

\[
D = \lambda_D \left[ \frac{500 \chi_s^3}{243 \alpha_e u_0^2} \right]^{1/5}
\]  

(5.4)

Here, the dimensionless potential of the sheath \( \chi_s \) is based on the sheath potential drop, the nondimensionalized ion speed at sheath entry \( u_0 \) is 1, and the collision parameter for the flame is 154. For these conditions, the sheath thickness is plotted as a function of electrode supply voltage in Figure 5.2. The cathode sheath thickness starts to encompass the premixed flame after a 4 kV potential has been applied. After 6 kV has been applied, the cathode sheath encloses both the premixed flame and a small segment downstream of it.
While the cathode sheath expands through the premixed flame, it can trap more ions. However, a majority of ion production occurs within the premixed flame, and ion densities fall off rapidly downstream of the premixed flame. Therefore, once the cathode sheath expands beyond the bounds of the premixed flame, the number of ions it can trap falls off sharply. This causes the rate of growth in ion density in the premixed flame to halt, even though the ionic wind continues to provide a stronger influence on the ions as the electric field strength rises. In essence, the sheath swallows the ion source.

5.2.2 Dissociative Recombination Influence

Past a certain electric field strength, the ion density near the burner begins to decline. Ion density near the burner is still elevated compared to the unmodified flame, but decline is still nontrivial. The decline can either be attributed to a loss of ions to the grounded burner or dissociative recombination. If losses to ground are the cause, we would expect that the current to ground per unit area coming from the burner would experience a constant increase. The point at which ions start disappearing from the flame, however, is associated with a constant current to ground, shown in Figure 5.3. Losses to ground therefore do not cause a decline in ion density, leaving dissociative recombination as the most probable explanation.
Figure 5.3: Ion current reaching ground for increasing electric fields. The current ceases increasing past an electrode voltage of 6 kV.

The general expression for the dissociative recombination reaction with respect to time is

$$\frac{dn_i}{dt} = n_in_e\alpha_{DC}$$  \hspace{1cm} (5.5)

The dissociative recombination rate coefficient is species specific and is a function of electron temperature, generally decreasing exponentially. An example of this is seen in Figure 5.4, which is adapted from Heppner et al. [59] Since electron temperature generally tends to increase as the electric field becomes stronger, the dissociative recombination cross section coefficient will decrease as well. If ion and electron densities were held constant, this would lead to less dissociative recombination taking place. Ion and electron densities are not constant, however, so it is necessary to know these quantities before drawing conclusions about the role of dissociative recombination in the flame.
**Figure 5.4**: $\text{H}_3\text{O}^+\text{e}^-$ recombination rate coefficient across a variety of electron temperatures. Plot is adapted from Heppner et al. [59]

Ion densities are already known from the results in Chapter 4. For the unmodified flame, where the sheath is thin to nonexistent, the assumption of quasi-neutrality applies, so the electron densities can be treated as the same as the ion densities. When an electric field is applied however, quasi-neutrality is no longer applicable due to the cathode sheath growth, which is electron repelling. Within the sheath, Boltzmann’s equation can be used to model the electron density

$$n_{e,s} = 0.61n_e \exp (-eV/kT_e)$$  \hspace{1cm} (5.6)

Here, electron density falls off very quickly as the sheath potential drop increases. Taking the electron temperature to be equal to the flame temperature, the cathode
sheath repels 99.5% of electrons present at the sheath boundary with a potential drop of 1 V. The range of conditions in the present work result in a potential drop far in excess of 1 V, so the electron density within the sheath is very low, if not effectively 0.

Not all electrons are completely repelled by the sheath; the nonzero electron temperature measurements within the sheath show that some electrons either manage to break into the sheath due to very high kinetic energies or are generated within the sheath and are present long enough to be detected by the Langmuir probe. Given that the sheath grows with electric field, the electron temperatures increasing with respect to electric field occurs because only the higher energy electrons can penetrate into the sheath. These electrons are few and far between compared to the electrons in the bulk plasma, however, so they do not contribute to dissociative recombination in a meaningful way. An illustration of this can be seen by solving the Boltzmann’s equation for electron temperature, given a known electrode voltage and ion density. For an electrode voltage of 10 kV, the ion density is $1 \times 10^{17}$ m$^{-3}$. Under these conditions, the electron temperature required to produce a steady electron population of more than 10 m$^{-3}$ is 3,200,000 K, an energy of approximately 275 eV. This does not mean that any electrons detected in the sheath must have this energy. However, this does mean that any electrons detected in the sheath likely started out at a higher energy state and shed most of their energy by the time they are detected due to collisions and the repelling influence of the sheath.

Dissociative recombination outside of the sheath does show some changes. Figure 5.5 shows the ratio of dissociative recombination rates at an electrode voltage of
2 kV to the dissociative recombination rates of the unmodified flame. At 2 kV, the locations represented in Figure 5.5 are outside the sheath, and thus do not see the sudden drop in electron density. For the locations 3 mm and 4 mm from the burner, dissociative recombination sees an increase compared to the rates seen in the unmodified flame. Electrons are repelled by the sheath and encounter the ions in the bulk plasma, resulting in increased opportunities for dissociative recombination to occur. The increase is only temporary, however, as the sheath grows quickly with electrode voltage, completely suppressing dissociative recombination. Dissociative recombination is therefore not a significant influence on flame behavior.

5.2.2.1 Electron Impact Dissociation

Another possible influence on the flame similar to dissociative recombination is electron impact dissociation, where an electron collision with a neutral molecule produces two or more neutral molecules while leaving the electron free to collide with other molecules. This process has been documented with a number of different neutral species, including CH\textsubscript{4} [60] and N\textsubscript{2}. [61] This process requires electrons with a high incident energy; electrons with a minimum energy of 12 eV are required to obtain a meaningful dissociation cross-section. Boom et al. [16] suggest this as a possible occurrence in premixed flames. Downstream of the flame, this is a strong possibility since the electrons are being energized by the electric field, but it is not certain if this occurs in the premixed flame. Even if electrons with sufficiently high energy exist
Figure 5.5: Ratio of dissociative recombination rates at an electrode voltage of 2 kV to dissociative recombination rates in the unmodified flame. Ratios are calculated at 3 mm above the burner or greater.

to cause impact dissociation, they may not exist in sufficient numbers to affect the flame.

5.3 CH* and OH* Consumption

The reactions described in Chapter 3 are viable explanation for what consumes CH* and OH*. However, only the ions and electrons produced by these reactions are directly subject to the effects of the electric field, leaving neutrals untouched. The electric field can, however, influence neutrals indirectly and make it possible for the electric field to drive the reactions between neutrals. As the electric field strength
increases, the ionic wind pushes ions towards the burner. Ions are significantly out-numbered by neutrals, but because they are subject to the electric field, they can push neutrals in the same direction as the electric field through collisions, transferring downward momentum to the neutrals.

5.3.1 Energy From the Electric Field

The effects of ion-neutral collisions are particularly strong within the sheath because the electric field is much stronger than it is in the bulk plasma. Even when the electrode voltage is only 2 kV, the electric field inside the sheath is substantial 563 kV/m. The influence of the electric field on ion-neutral collisions can be seen by first looking at the ion drift velocity, the flow velocity at which an ion travels in a direction due to the influence of the electric field. This is calculated by

\[ v_d = \mu_i E \tag{5.7} \]

This extra velocity adds translational kinetic energy to the ions on top of the kinetic energy already present due to the ion’s thermal velocity. For any generic particle, the thermal velocity is calculated by

\[ \bar{v} = \sqrt{\frac{8kT_f}{\pi m}} \tag{5.8} \]

Upon collision with a neutral, ions transfer a certain portion of their kinetic energy. The amount of energy transferred scales with the mass of the ion and the neutral,
and can be represented as

\[ E^* = \frac{2m_im_n}{(m_i + m_n)^2} \]  

(5.9)

### 5.3.2 Effects on Ion-Neutral Collisions

The first effect which the energized ions have on the flame can be seen through the bimolecular collision rate

\[ Z = \pi n_i n_n d_{i,n}^2 \left( \sqrt{\frac{8kT_f}{\pi m_{i,n}}} + v_d \right) \]  

(5.10)

where the ion-neutral reduced mass is defined as

\[ m_{i,n} = \frac{m_i m_n}{m_i + m_n} \]  

(5.11)

For the purposes of calculating the ratio Equation 5.10, the neutral density and the intermolecular distance can be assumed constant since the pressure inside the flame is assumed constant. To calculate the ion-neutral thermal velocity, all neutrals are assumed to be \( \text{N}_2 \) and all ions are assumed to be \( \text{H}_3\text{O}^+ \). The results of these calculations, plotted below in Figure 5.6, show the ion-neutral collision rate to increase by one order of magnitude compared to the unmodified flame.

In this situation, both the rate of ion-neutral collisions and the energy transferred per collision are increased compared to the case where no electric field is present. The influence of collisions on these reactions must therefore be looked at more closely.
5.3.3 Effects on Radical Consumption due to Added Energy

To illustrate the influence that ion-neutral collisions have on radical consumption, consider the following reactions

\[ CH + O \rightleftharpoons CHO \]  \hspace{1cm} (5.12)

\[ CH + O \rightleftharpoons CO + H \]  \hspace{1cm} (5.13)

For the purposes of demonstrating how collisions influence these reactions, it is assumed that CH is impacted by an H₃O⁺ ion. The energy added to CH via collision is then distributed among four different energy modes, represented by four partitioning.

Figure 5.6: Ratio of ion-neutral bimolecular collision rate to the bimolecular collision rate of an unmodified flame as a function of electrode voltage.
functions. The first mode is the translational mode, which has the partition function

\[ Q_T = \left( \frac{2\pi mkT}{\hbar^2} \right)^{3/2} V_o \]  \hspace{1cm} (5.14)

Continuing, the rotational mode partition is represented as

\[ Q_R = \frac{T}{\sigma_y \theta_r} \]  \hspace{1cm} (5.15)

The vibrational mode partition function is represented as

\[ Q_V = \left( 1 - \exp\left( -\theta_v / T \right) \right)^{-1} \]  \hspace{1cm} (5.16)

And the electronic mode partition function is represented as

\[ Q_E = g_0 + \sum_{i=1}^{\infty} g_i e^{-\theta_e,i / T} \]  \hspace{1cm} (5.17)

CH has a characteristic rotational temperature of 25 K [62] and a maximum characteristic vibrational temperature of 4316 K [63], while the electronic partition function for CH is unavailable and so is assumed to be identical to that of CO. Additionally, CHO has vibrational temperatures of 3158 K and 4278 K, [64] and its rotational temperature is taken to be 15 K [65], while CO has a vibrational temperature of 3120 K and a rotational temperature of 2.8 K. Finally, for the purposes of analysis, any products associated with CH are also treated as energized by the H$_3$O$^+$ collision.
Enough information is now present to determine how ion-neutral collisions influence the two CH reactions by looking at what happens to reaction equilibrium. This is determined through a set of nonlinear equations, which include the reaction constant equations

\[ k_{p,1} = \frac{X_{CHO}}{X_{CH}X_O} \]  
\[ k_{p,2} = \frac{X_{CO}X_H}{X_{CH}X_O} \]  

which can also be expressed as

\[ k_p = \left( \frac{kT}{V_o} \right)^{\Sigma \omega_i} e^{-\Delta H_0^i / kT} \prod_{i=1}^{\infty} Q_i^{\omega_i} \]  

Two atom balance equations are also needed to determine the equilibrium state of the two reactions

\[ \frac{C}{O} = 1 = \frac{2X_{CH} + X_{CHO} + X_{CO}}{2X_O + X_{CHO} + X_{CO}} \]  
\[ \frac{C}{H} = 1 = \frac{2X_{CH} + X_{CHO} + X_{CO}}{2X_{CH} + X_{H} + X_{CHO}} \]  

And finally, one equation summing up the mole fractions is also necessary to solve for the equilibrium state

\[ X_{CH} + X_O + X_{CHO} + X_{CO} + X_H = 1 \]
Figure 5.7: Ratio of CH mole fraction at equilibrium based on a two reaction case. The ratio is calculated against an unmodified flame as a function of electrode voltage.

The results of solving these equations are shown in Figure 5.7. Energizing CH through ion collisions has the effect of driving the formation of more products, particularly as the electric field strength increases. In this case, it is H and CO production that is favored instead of CHO formation, which sees a decline in its mole fraction compared to the unmodified flame. The gain in H and CO does nonetheless suggest that certain changes to the chemistry do occur, albeit through a means not associated with dissociative recombination, since this has already ruled out due to the cathode sheath. Increased radical production affects other elementary reactions by supplying more reactants. In turn, this leads to more complete combustion, thus a DC electric field has an indirect but nontrivial influence on the flame chemistry through ion-neutral collisions.

Results of solving the nonlinear equations don’t entirely match the experi-
mental results. However, this is due to limitations of the algorithm in use. Here, it is assumed that only two reactions are occurring at most, that the reactions are occurring in a controlled volume which is not a reacting flow, and that every single CH molecule is simultaneously energized by collision with H$_3$O$^+$ molecules that all have the same energy while none of the O atoms are subject to ion collisions. In reality, CH is involved in many reactions occurring simultaneously, often involving reactants or products which have partition functions that are difficult to calculate, not every CH molecule is energized by a collision, O is subject to collisions with ions, and CH reactions in a flame occur in a reacting flow instead of controlled volume. Figure 5.7 does show, however, that the trend of decreasing CH concentrations as predicted by the partition function mirrors experimental results; including additional CH consuming reactions in the algorithm would likely produce results which follow experimental results even more closely.

It is possible to conclude, then, that an electric field can drive the consumption of radicals, albeit indirectly via collisions with ions that are subject to the influence of an electric field. This may help explain observations by Marcum and Ganguly. Although the present work has ruled out dissociative recombination as a means of influencing the flame, the reactions which consume both CH and OH produce radicals which are often low mass and may be reactive, such as the hydrogen atom. A slight increase in the production of light radicals may provide chemistry-driven explanation for a hydrocarbon flame’s response to the electric field. However, additional information, such as those coming from a mass spectrometer, is needed to conclusively demonstrate this.
5.4 Summary

Given the results of data presented and the discussion points above, a comprehensive picture of the flame behavior and how the flame interacts with the electric field can be assembled.

The unmodified flame is mostly dominated by dissociative recombination. Ionic wind effects are not present since no electric field has been applied. The cathode sheath, while present, is very thin due to the low plasma potential. Once the electric field is applied, the sheath remains thin, but begins growing rapidly as the electric field increases. Changes to ion density at this point is dominated by the ionic wind, which is generated by the electric field within the bulk plasma and depletes ions from the upper regions of the premixed and moves them towards the burner. With increasing electric field strength, the sheath expands, eventually encompassing the entire premixed flame. The ionic wind continues to increase with electric field, pushing more ions towards the burner. However, once the electrode voltage reaches 6 kV, the cathode sheath has expanded so much that it encompasses both the premixed flame and the regions slightly downstream of it. At this point, the cathode sheath has captured the majority of ions in the flame, leaving very little to be influenced by the ionic wind. Further increases to electrode voltage will not lead to any increases in ion density, since the sheath has already captured all of the ions and they cannot escape.

One final point to consider is the inverse ion gradient inside the sheath. In common low-pressure, uniform, volumetric plasmas the ion density decreases closer to
the surface or electrode. This occurs due to mass conversation requiring the flux, \( n_i v_i \), to the sheath edge be constant. Thus inside the sheath as ions are accelerated and \( v_i \) increases, \( n_i \) must decrease. The densities measured here indicate the opposite, with ion densities being largest closer to the burner surface. This can be attributed to the flame being a non-uniform and highly collisional plasma. Ion production in a flame is fixed at the flame front, which in the present work is 5 mm above the burner and does not move. The produced ions will diffuse both upstream and downstream of the flame front, though flow convection will greatly increase the ion density downstream. Thus as the cathode sheath grows toward the flame front, the ion density and thus flux at the sheath edge in not uniform and actually increases, assuming ions always enter the sheath at the Bohm velocity \( u_0 \). This will result in a continual increase in the flux, which increases ion density in the sheath. Secondly, the highly collisional nature of the flame plasma means the ion velocity is much lower than the ion velocity in a low pressure plasma. The ion-neutral mean free path is on the order of 0.3 \( \mu \text{m} \), much smaller than the sheath thickness even when the electrode potential is \(< 4 \text{ kV}\). As ions move upstream from the hot flame to the colder reactant flow, the lower temperature and slightly higher pressure will further increase the neutral density, which in turn increases the collision rate. This all combines to significantly reduce the ion velocity in the sheath as they move closer to the burner. Thus just by conservation of mass the density must increase to compensate, in addition to the ionic wind pushing more ions toward the burner.
CHAPTER 6

CONCLUSIONS

*Engineers believe that if it ain't broke, it doesn't have enough features yet.*

— Scott Adams

A premixed methane-air flame was produced on a custom-built flat flame burner at an equivalence ratio of 1.0. Methane volumetric flow rate was fixed at 0.72 SLM, while the air volumetric flow rate was fixed at 6.81 SLM. A flat sheet of perforated steel suspended 7.62 cm above the burner was connected to a high voltage power supply capable of producing up to 10 kV DC. The resulting electric field was 131 kV/m. In this configuration, the metal plate was the anode, while the burner, connected to the true earth ground, was the cathode.

Ion densities in the premixed flame were measured using a single Langmuir probe, while electron temperatures were measured using an asymmetric double probe. Both probes were connected to a sourcemeter. When measuring ion densities, the probe was biased to -5 V, while electron temperature was determined from the results of a sweep from -10 V to +10 V. Both probes were mounted to a Velmex 2-axis linear stage, which moved the probes to predetermined locations. After data collection, the probes were retracted from the flame and allowed to cool to reduce the risk.
of erroneous measurements.

Measurements indicated that ion densities greatly increased with the electrode supply voltage near the burner at the expense of ion densities downstream of the flame. This growth was attributed to the increasing ionic wind strength, which moved ions towards the burner and the growth of the burner plasma sheath. Ion density growth continued until the electrode supply voltage reached 6 kV, at which point ion densities ceased growing and started to decline in some locations. This was attributed to burner sheath encompassing the entire premixed flame and a significant proportion of the ion-containing space downstream of the premixed flame once the electrode supply voltage exceeded 6 kV. Comparatively few ions persist further downstream, thus ion density growth ceases due to fewer ions being trapped by the expanding cathode sheath. The results provide further support for the ionic wind effect as a dominate mechanism in electric field modified flames, and additionally shows the impact of the plasma sheath on the limit of the effect. The ionic wind can also be used to explain the decreasing total brightness of chemiluminescence images of the flame. Ion drift velocity towards the burner is very fast due to the strong electric field within the sheath, thus ion-neutral collisions push the neutrals closer together. This increases the likelihood of neutrals undergoing a reaction which consumes CH* and OH*, giving the chemiluminescence images less brightness as more voltage is applied to the electrode.

Electron temperatures were generally found to increase with electrode voltage, reaching in excess of 14,000 K at the highest. These results were used to help assess the validity of the dissociative recombination hypothesis. The \( H_3O^+ - e^- \) dissoci-
tive recombination cross-section coefficient decreases in response to the higher electron temperatures. However, dissociative recombination in some locations increases despite the increased electron temperature due to the increasing ion and electron density. This was only true when the electrode voltage was 2 kV or less, however. Sheath expansion stops dissociative recombination entirely due to the cathode sheath repelling electrons, thus dissociative recombination’s role in the plasma is limited at best.

6.1 Future Work

6.1.1 Combined Electric/Magnetic Fields

Of all research which covers flame modification with externally applied fields, the majority has only worked with electric fields. Research which covers the use of magnetic fields is significantly more difficult to find, and to the author’s knowledge, research which includes both electric and magnetic fields acting on the same flame is either nonexistent or is extremely rare. Research which includes combined electric and magnetic field modification would fill in gaps in knowledge of this field. For example, magnetic field may enhance or suppress the effects of an electric field, or the impact of a magnetic field may be something else entirely.
6.1.2 Real Combustors

With a few exceptions, [29] [28] [30] research in this field has been limited to small laminar flames. Speculation has been given to the benefits that Plasma Assisted Combustion can provide to real devices, but thus far, attempts at modifying a combustor used for an industrial process or in transportation has not been attempted. Applications to terrestrial power systems, jet engines, rocket engines, furnaces, and many other combustors are all possible avenues of research. Given how much research has already been done with laminar flames, attempting to apply the knowledge obtained from years of work to a combustor designed with an industrial process in mind is the next logical step.
REFERENCES


[27] Jacob Schmidt and Biswa Ganguly. Effect of pulsed, sub-breakdown applied electric field on propane/air flame through simultaneous OH/acetone PLIF. *Combustion and Flame*, 160(12):2820–2826, December 2013.


