ABSTRACT

ELECTRIC FIELD INDUCED STABILITY MODIFICATIONS
IN PRE-MIXED HYDROCARBON FLAMES

By: David L. Wisman II

This work focuses upon the effects of electric fields on the stability of pre-mixed hydrocarbon flames fueled by methane, propane and butane. For all fuels studied, over varying flow rates and equivalence ratios, electric-field-induced modification of the flame front stability over wide ranges has been observed and documented. Specifically, the electric pressure effect has been used to force pre-mixed hydrocarbon/air flame fronts to transition from flow-induced hydrodynamic-instability-dominated behavior, to field-stabilized laminar flow, and finally to field-induced thermal-diffusive-instability-dominated turbulent flame fronts. The effects are best described from a combustion dynamics viewpoint as a continuously variable electric-field-induced diminution of the flame Lewis number from typical values (≥ 1), past the flame critical Lewis number (0.6-0.9), to thermal-diffusive-instability dominated values below the critical Lewis number at higher applied potentials. A possible field-driven ion chemistry process that could produce those effects is the dissociative recombination of the flame ion (H₃O⁺) at the burner surface.
ELECTRIC FIELD INDUCED STABILITY MODIFICATIONS
IN PRE-MIXED HYDROCARBON FLAMES

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1. INTRODUCTION

1.1 Flame History and General Flame Structure

The discovery and subsequent development of the ability to control fire are among the greatest accomplishments in the history of humankind. Archeological findings estimate that the first controlled use of fire was between 500,000 and 100,000 years before present time. Fire became such an important part of the lives of people that around 500 BC the Greek philosopher Empedocles considered it to be one of the four elements of the terrestrial world.

As science evolved, so has our understanding of fire. Through the development of a variety of different probing techniques scientists were able to begin to understand the structure of a flame and develop new ways to manipulate it. Francis Bacon was the first person known to have conducted detailed observations of flame structures by actively probing various flames. Figure 1.1.1 shows some of the different early types of probing technologies used on flames.

Today we use two broad classifications for flame types: diffusion and pre-mixed. Diffusion flames are those where the fuel and air are initially separate and mix comparatively slowly as the combustion takes place. Examples of diffusion flames would be candles, campfires, forest fires, cooking flames, etc. Diffusion flames are the easiest to construct, and thus are more common outside of the laboratory or industrial setting. In fact, until the creation of the Bunsen burner, all flames were of the diffusion type.

With the invention of the Bunsen burner in 1855 pre-mixed flames became readily available for use in the laboratory or in industry. Pre-mixed flames are those where the fuel and air are mixed in controllable ratios prior to combustion. As suggested by Figure 1.1.2, pre-mixed flames typically contain two distinct reaction zones. The primary reaction zone can be further divided into two separate zones itself: a pre-heat zone, and the flame front. In the pre-heat zone the input reactants are heated, and are progressively dissociated and reconfigured prior to entering the flame front. The
Figure 1.1.1 Evolution of flame probing techniques.

1. Primary reaction zone
2. Secondary reaction zone

Burner Head

Figure 1.1.2 Components of a pre-mixed flame.
flame front is where most of the chemical energy is released, and thus is where most of the interesting chemistry in a flame takes place. The flame front is typically very thin, on the order of a millimeter when working at atmospheric pressure. Figure 1.1.3 shows how the reactants and temperature in a flame vary as a function of location relative to the flame front.

The secondary reaction zone consists largely of combustion end products, and is where any excess fuel can mix with any entrained air and combust further. This zone is similar in structure to that of a diffusion flame.

1.2 Combustion

1.2.1 Definition

Combustion is best defined as “a rapid oxidation generating heat, or both heat and light”. This definition illustrates the important fact that combustion converts energy stored in chemical bonds and releases it as heat. Further, the details of the emitted light provide a very important source of information about the flame that can be gathered passively. The heat released is of course more important since it can be used in variety of ways, depending on the application. Combustion is a very important source of energy in the United States, accounting for approximately 85% of the total energy used in this country at this time.

1.2.2 Reactions

Under ideal conditions, the reactions for combustion are determined by chemical stoichiometry. The reactions are a function of the particular hydrocarbon used in a given combustion process. For the work described here, three different hydrocarbon fuels were used: propane (C\textsubscript{3}H\textsubscript{8}), methane (CH\textsubscript{4}), and butane (C\textsubscript{4}H\textsubscript{10}). For propane, the reaction would be:

\[ C_{3}H_{8} + 5O_{2} \Rightarrow 3CO_{2} + 4H_{2}O \]  \hspace{1cm} (1.2.1)

For the lighter methane, the ideal combustion reaction is:
\[ CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \]  

(1.2.2)

In the case of the heavier butane, the reaction would be:

\[ 2C_4H_{10} + 13O_2 \rightarrow 8CO_2 + 10H_2O \]  

(1.2.3)

Clearly, for a given fuel flow rate, requirements of stoichiometry indicates increasing heat release as a function of the molecular weight of the raw fuel. Interestingly, the temperatures\(^8\) of stoichiometric mixtures of propane, methane and butane are very similar (2267 K, 2226 K, and 2270 K, respectively). The three fuels thus offer an opportunity to explore the behavior of electric-field-stressed hydrocarbon flames as a function of their overall molecular weight (see also Lewis number below).

The global stoichiometric chemistry just described dramatically masks the true nature of “simple” pre-mixed hydrocarbon flames. Typically there must be some energy added to the fuel and air mixture to accomplish the initial dissociation of the reactants and the formation of a wide variety of intermediate combustion species and initiate the strongly exothermic final reactions, \(N_2\) must be considered as a not-entirely-inert diluent, and other by-products are formed which may include harmful gases such as \(NO_x\) and \(CO\) as well as unburned hydrocarbons.

A simplified set of only 86 of the most important reactions for methane combustion is presented in Table 1.2.1\(^9,10\). Those reactions only focus on the most important products and intermediate steps, neglecting any reactions with nitrogen. That set of reactions includes a set near the end (reactions 74 – 86) that represent the essence of ion production (chemi-ionization) and destruction in light hydrocarbon flames. Reactions involving ions are generally ignored by those that model combustion chemistry since they minimally contribute to the overall enthalpy of the flame. They are of central importance to this work since it is the electric force impressed upon the positive ions in the flame by the external field that initiates the observed stability modifications. More crudely one can also characterize the reaction process for combustion by making a flow diagram to map out the combustion routes.
Figure 1.1.3 Plot of reactants and temperature as a function of location in the flame.
Figure 1.2.1\textsuperscript{11} shows the combustion routes taken by some small hydrocarbons during combustion with oxygen. As suggested in that figure, light hydrocarbons are dissociated and re-arranged in the preheat zone of a pre-mixed flame into a similar mix of combustion intermediary species regardless of the parent fuel molecule. As noted above, for the three fuels studied here the stoichiometric combustion temperatures are also very similar. As a result, the abbreviated reaction set shown in Table 1.2.1 is generally applicable for the three fuels of interest here. In particular, reactions involving ions would be expected to vary little from methane to propane to butane.

1.2.3 Stoichiometry

Stoichiometry is a condition where you have the precise amount of oxidant needed to completely burn all of the fuel present in a flame.\textsuperscript{12} For hydrocarbons the stoichiometric relation can be expressed by equation 1.2.4,

\[ C_xH_y + a(O_2 + 3.76N_2) \rightarrow xCO_2 + \left(\frac{Y}{2}\right)H_2O + 3.76aN_2, \quad (1.2.4) \]

where \( a = x + \frac{y}{4} \). It is also useful to determine the air-fuel ratio for the stoichiometric condition. This can be done by using the following relation,

\[ \left(\frac{A}{F}\right)_{\text{stoic}} = \frac{4.76aMW_{\text{air}}}{MW_{\text{fuel}}}, \quad (1.2.5) \]

where \( MW_{\text{air}} \) and \( MW_{\text{fuel}} \) are the molecular weights of air and fuel respectively.

1.2.4 Equivalence Ratio

It is clearly important to know the relative concentrations of air and fuel in a given flame. This can most easily be accomplished by determining the equivalence ratio of the flame. The equivalence ratio, \( \Phi \), is a dimensionless number which is used to
Table 1.2.1 Reaction mechanisms for methane flames. Reaction rates are in cm$^3$-mole-sec-cal units, and
\[
k = AT^B e^{-\frac{E_a}{RT}}
\]

<table>
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<th>Reaction</th>
<th>A</th>
<th>B</th>
<th>$E_a$</th>
</tr>
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<tbody>
<tr>
<td>1</td>
<td>OH + H$_2$ → H$_2$O + H</td>
<td>1.17E+09</td>
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<td>2</td>
<td>H + O$_2$ → OH + O</td>
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<td>15</td>
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<td>H + OH + M' → H$_2$O + M'</td>
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<tr>
<td></td>
<td>Reaction</td>
<td>ΔG (kJ/mol)</td>
<td>ΔH (kJ/mol)</td>
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<td>CO + HO$_2$ → CO$_2$ + OH</td>
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<tr>
<td>46</td>
<td>C$_2$H$_4$ + O → CH$_3$ + CH$_2$O</td>
<td>2.53E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>47</td>
<td>C$_2$H$_4$ + OH → CH$_2$O + CH$_3$</td>
<td>5.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>48</td>
<td>C$_2$H$_4$ + 0 → C$_2$H$_3$ + OH</td>
<td>2.53E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>49</td>
<td>C$_2$H$_4$ + O$_2$ → C$_2$H$_3$ + HO$_2$</td>
<td>1.33E+15</td>
<td>0.00</td>
</tr>
<tr>
<td>50</td>
<td>C$_2$H$_4$ + H → C$_2$H$_3$ + H$_2$</td>
<td>2.00E+15</td>
<td>0.00</td>
</tr>
<tr>
<td>51</td>
<td>C$_2$H$_4$ + OH → C$_2$H$_3$ + H$_2$O</td>
<td>4.40E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>52</td>
<td>C$_2$H$_3$ + M → C$_2$H$_2$ + H + M</td>
<td>3.01E+16</td>
<td>0.00</td>
</tr>
<tr>
<td>53</td>
<td>C$_2$H$_3$ + O$_2$ → C$_2$H$_2$ + HO$_2$</td>
<td>1.57E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>54</td>
<td>C$_2$H$_3$ + H → C$_2$H$_2$ + H$_2$</td>
<td>7.53E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>55</td>
<td>C$_2$H$_3$ + OH → C$_2$H$_2$ + H$_2$O</td>
<td>1.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>56</td>
<td>C$_2$H$_2$ + OH → CH$_3$ + CO</td>
<td>5.48E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>57</td>
<td>CH$_3$ + H → CH$_2$ + H$_2$</td>
<td>2.00E+11</td>
<td>0.70</td>
</tr>
<tr>
<td>58</td>
<td>CH$_3$ + OH → CH$_2$ + H$_2$O</td>
<td>6.00E+10</td>
<td>0.70</td>
</tr>
<tr>
<td>59</td>
<td>CH$_2$ + O$_2$ → CHO + OH</td>
<td>1.00E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>60</td>
<td>CH$_2$ + O$_2$ → CH$_2$O + O</td>
<td>1.00E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>61</td>
<td>CH$_2$ + O$_2$ → CO$_2$ + OH$_2$</td>
<td>1.00E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>62</td>
<td>CH$_2$ + H → CH + H$_2$</td>
<td>4.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>63</td>
<td>CH + O → CO + H</td>
<td>4.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>64</td>
<td>CH + O$_2$ → CO + OH</td>
<td>2.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>65</td>
<td>C$_2$H + O → CO + CH</td>
<td>1.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>66</td>
<td>CH* + M → CH + M</td>
<td>4.00E+10</td>
<td>0.50</td>
</tr>
<tr>
<td>67</td>
<td>CH* + O$_2$ → CH + O$_2$</td>
<td>2.40E+12</td>
<td>0.50</td>
</tr>
<tr>
<td>68</td>
<td>CH* → CH + photon</td>
<td>1.90E+06</td>
<td>0.00</td>
</tr>
<tr>
<td>69</td>
<td>C$_2$H + O$_2$ → CH* + CO$_2$</td>
<td>4.50E+15</td>
<td>0.00</td>
</tr>
<tr>
<td>70</td>
<td>C$_2$H + O → CH* + CO</td>
<td>7.10E+11</td>
<td>0.00</td>
</tr>
<tr>
<td>71</td>
<td>C$_2$H$_2$ + H → C$_2$H + H$_2$</td>
<td>6.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>No.</td>
<td>Reaction</td>
<td>Rate Coefficient</td>
<td>Reaction Temperature</td>
</tr>
<tr>
<td>-----</td>
<td>-----------------------------------------------</td>
<td>------------------</td>
<td>----------------------</td>
</tr>
<tr>
<td>72</td>
<td>( \text{C}_2\text{H}_2 + \text{OH} \rightarrow \text{C}_2\text{H} + \text{H}_2\text{O} )</td>
<td>1.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>73</td>
<td>( \text{C}_2\text{H} + \text{O}_2 \rightarrow \text{CO} + \text{CHO} )</td>
<td>5.00E+13</td>
<td>0.00</td>
</tr>
<tr>
<td>74</td>
<td>( \text{CH} + \text{O} \rightarrow \text{CHO}^+ + \text{e}^- )</td>
<td>2.52E+11</td>
<td>0.00</td>
</tr>
<tr>
<td>75</td>
<td>( \text{CH}^+ + \text{O} \rightarrow \text{CHO}^+ + \text{e}^- )</td>
<td>5.01E+11</td>
<td>0.00</td>
</tr>
<tr>
<td>76</td>
<td>( \text{CHO}^+ + \text{H}_2\text{O} \rightarrow \text{H}_2\text{O}^+ + \text{CO} )</td>
<td>1.00E+16</td>
<td>-0.09</td>
</tr>
<tr>
<td>77</td>
<td>( \text{H}_2\text{O}^+ + \text{C}_2\text{H}_2 \rightarrow \text{C}_2\text{H}_3\text{O}^+ + \text{H}_2 )</td>
<td>8.39E+15</td>
<td>0.00</td>
</tr>
<tr>
<td>78</td>
<td>( \text{CHO}^+ + \text{CH}_2 \rightarrow \text{CH}_3^+ + \text{CO} )</td>
<td>5.62E+14</td>
<td>-0.01</td>
</tr>
<tr>
<td>79</td>
<td>( \text{H}_3\text{O}^+ + \text{CH}_2 \rightarrow \text{CH}_3^+ + \text{H}_2\text{O} )</td>
<td>6.17E+14</td>
<td>-0.01</td>
</tr>
<tr>
<td>80</td>
<td>( \text{CH}_3^+ + \text{C}_2\text{H}_2 \rightarrow \text{C}_3\text{H}_5^+ + \text{H}_2 )</td>
<td>7.24E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>81</td>
<td>( \text{C}_3\text{H}_5^+ + \text{H}_2\text{O} \rightarrow \text{C}_2\text{H}_3\text{O}^+ + \text{CH}_2 )</td>
<td>7.24E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>82</td>
<td>( \text{CH}_3^+ + \text{CO}_2 \rightarrow \text{C}_2\text{H}_3\text{O}^+ + \text{O} )</td>
<td>7.24E+14</td>
<td>0.00</td>
</tr>
<tr>
<td>83</td>
<td>( \text{H}_3\text{O}^+ + \text{e}^- \rightarrow \text{H}_2\text{O} + \text{H} )</td>
<td>2.29E+18</td>
<td>-0.50</td>
</tr>
<tr>
<td>84</td>
<td>( \text{C}_3\text{H}_5^+ + \text{e}^- \rightarrow \text{products} )</td>
<td>1.50E+19</td>
<td>-0.50</td>
</tr>
<tr>
<td>85</td>
<td>( \text{CH}_3^+ + \text{e}^- \rightarrow \text{CH}_2 + \text{H} )</td>
<td>2.29E+18</td>
<td>-0.50</td>
</tr>
<tr>
<td>86</td>
<td>( \text{C}_2\text{H}_3\text{O}^+ + \text{e}^- \rightarrow \text{M} + \text{H} )</td>
<td>2.29E+18</td>
<td>-0.50</td>
</tr>
</tbody>
</table>
characterize if a flame is fuel rich, lean or stoichiometric. This can be found by dividing any given air-fuel ratio by the stoichiometric air-fuel ratio. Thus, if $\Phi > 1$, the flame is said to be fuel rich, if $\Phi < 1$ the flame is fuel lean, and if $\Phi = 1$ the air/fuel ratio is that which provides perfect stoichiometric conditions.

The overall visible color of the flame is also associated with the equivalence ratio. Flames that are fuel rich ($\Phi > 1$) tend to have a greenish tint while flames that are fuel lean ($\Phi < 1$) are more light blue. This is due to the overall concentrations of the electronically excited radicals produced by chemi-excitation processes, with fuel rich flames having more excited carbon ($C_2^*$) radicals, and fuel lean flames having a relatively higher concentration of excited CH radicals ($CH^*$). The reactions (termed chemi-luminescence) leading to the most prominent visible emissions in a hydrocarbon-air flame are shown by equations 1.2.6 and 1.2.7.

$$CH^* \rightarrow CH + photon \quad (431 \text{ nm, blue}) \quad (1.2.6)$$

$$C_2^* \rightarrow C_2 + photon \quad (516 \text{ nm, most prominent Swan Band, green}) \quad (1.2.7)$$

Other contributors to the visible emissions from such flames are from less populated vibrational levels of the first electronically excited level of the carbon dimmer ($C_2(A^3\Pi_g, v)$) radiating to the ground electronic level ($C_2(X^3\Pi_u, v')$). Specifically, Swan Bands with $(v, v')$ equal to (0,0) produce the 516-nm chemi-luminescent band of reaction 1.2.7, whereas the (0,1) transition occurs at 564 nm, (1,0) at 474 nm, and the (2,0) transition at 438 nm. Other prominent molecular bands that appear in the near UV are associated with excited CH and OH radicals. The 431-nm band of reaction 1.2.6 is due to transitions between the first-excited and ground electronic levels of CH (i.e. $CH(A^2\Delta, v = 0) \rightarrow CH(X^2\Pi, v' = 0) + photon$). Significant chemi-excitation of the second excited state of the CH radical ($CH(B^2\Sigma^+)$) also occurs in hydrocarbon flames, producing a bright band in the near UV at 390 nm. The remaining bright molecular emission from chemi-excited
radical species occurs further into the UV and is due to transitions between the first-excited \((A^2\Sigma^+, v = 0)\) and ground \((X^2\Pi, v' = 0)\) electronic states of the hydroxyl radical,

\[
OH^* \Rightarrow OH + photon \ (308 \text{ nm}). \tag{1.2.8}
\]

Figures 1.2.2 and 1.2.3 show the spectra for fuel lean and fuel rich flames, respectively. These figures clearly show that the overall flame color due to the combined chemi-excitation of CH* and C_2* is a strong function of the equivalence ratio. This further suggests that the details of the overall combustion kinetics, including the process of chemi-ionization (discussed in detail in chapter 3), should also be functions of the equivalence ratio. The overall level of ion production, and thus flame conductivity, are of central importance to the modifications of flame stability by applied electric fields that are the focus of this work.

### 1.2.5 Lewis Number

The Lewis number \((Le)\) is a dimensionless quantity used to represent how the heat and mass flow in a flame. It is most simply expressed by the relation,

\[
Le = \frac{\alpha}{D}, \tag{1.2.9}
\]

where \(\alpha\) is the thermal diffusivity and \(D\) is the mass diffusivity. This ratio is basically the rate of energy transport divided by the rate of mass transport.\(^{14}\) Less massive particles in a flame diffuse more rapidly, and thus tend to have lower \(Le\) than the heavier particles in a flame. Table 1.2.2\(^{15}\) gives a list of several important species in a flame and their associated \(Le\).

Typical laboratory flames that have not been perturbed have \(Le\) that are near unity.\(^{16}\) In this situation there is no net change in enthalpy, and the temperature across the flame front can be considered adiabatic. Changing the \(Le\) to something less than unity will cause there to be variations in the temperature along the flame front, if there are concomitant variations in the density of reactive flame species of low Lewis number\(^{17,18}\) i.e. light reactant species.
Table 1.2.2 List of species Lewis numbers.

<table>
<thead>
<tr>
<th>Species</th>
<th>Lewis Number</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>0.18</td>
</tr>
<tr>
<td>H₂</td>
<td>0.30</td>
</tr>
<tr>
<td>O</td>
<td>0.70</td>
</tr>
<tr>
<td>OH</td>
<td>0.73</td>
</tr>
<tr>
<td>H₂O</td>
<td>0.83</td>
</tr>
<tr>
<td>CH₄</td>
<td>0.97</td>
</tr>
<tr>
<td>CH₃</td>
<td>1.00</td>
</tr>
<tr>
<td>HO₂</td>
<td>1.10</td>
</tr>
<tr>
<td>CO</td>
<td>1.10</td>
</tr>
<tr>
<td>O₂</td>
<td>1.11</td>
</tr>
<tr>
<td>H₂O₂</td>
<td>1.12</td>
</tr>
<tr>
<td>HCO</td>
<td>1.27</td>
</tr>
<tr>
<td>CH₃O</td>
<td>1.28</td>
</tr>
<tr>
<td>CH₃O</td>
<td>1.28</td>
</tr>
<tr>
<td>CH₃OH</td>
<td>1.30</td>
</tr>
<tr>
<td>CH₃OH</td>
<td>1.30</td>
</tr>
<tr>
<td>CO₂</td>
<td>1.39</td>
</tr>
</tbody>
</table>
Figure 1.2.1 Combustion pathways for lightweight hydrocarbons.
Figure 1.2.2 Spectra of propane flame with $\Phi = 0.8$.

Figure 1.2.3 Spectra of propane flame with $\Phi = 1.4$. 
1.3 Flame Instabilities

1.3.1 Hydrodynamic Instability

In pre-mixed flames there are typically three distinct instabilities that are present and working in competition with each other to either stabilize or destabilize the flame. One of the most studied, and thus most well known of these instabilities, is the hydrodynamic instability. This instability is caused by the expansion of gases at the high temperature flame front, which lead to an increase in the gas velocity. The theoretical description of the hydrodynamic instability was developed independently by Darrieus and Landau at approximately the same time, and is thus often referred to as the Darrieus-Landau instability. It wasn’t until 1998 that there was an experimental study done to investigate this instability.

In 1938 Darrieus first noted that gas expansion in a pre-mixed flame will tend to deviate flow lines across the flame front. Landau also was able to independently verify the hydrodynamic instability in 1944 by calculating the instability growth rate due to gas expansion. Figure 1.3.1 shows how the deviation of the flow lines can lead to the formation of the hydrodynamic instability. As the flow lines are deviated they lead to local velocity gradients in the flame which cause it to have an overall wrinkled shape. This wrinkling of the flame front is the main characteristic of the onset of the hydrodynamic instability.

Since all pre-mixed flames have gases that expand thermally, this instability can be found in all pre-mixed flames, but is more noticeable the higher the flow velocities. This condition is typically quantified by means of the Reynolds number. The Reynolds number is a dimensionless quantity used to give a measure of the amount of hydrodynamic turbulence present in a flow, with a higher number indicating a higher degree of turbulence. This mechanism tends to have a destabilizing influence on the overall stability of the flame, and can be dominant especially at higher flow velocities. A simple expression for the Reynolds number is \( Re = \frac{\rho u R}{\mu} \), where \( u \) is the flow velocity, \( R \) is the flow size, \( \mu \) is the viscosity, and \( \rho \) is the flow density.
1.3.2 Thermal-Diffusive Instability

Though much more subtle, and thus much less commonly encountered, another widely studied instability in a flame is the thermal-diffusive instability. A thermal-diffusive instability is caused by a preferential diffusion of mass versus heat in the immediate vicinity of the flame front depending on the \( Le \) of the deficient light reactant in the flame.\(^{22} \) This type of instability can be characterized as a lowering of the effective \( Le \) of the entire flame.

The thermal-diffusive instability can have both a stabilizing and destabilizing influence on the flame. Typically, if you lower the \( Le \) of the flame to values below unity, the flame will tend to become more unstable. Raising the \( Le \) of the flame to numbers greater than unity tends to have a stabilizing effect.\(^{23} \) Figure 1.3.2 shows a sketch of how the thermal-diffusive instability works. In Figure 1.3.2, the left side is for an unstable condition, while the right is for a stable condition. In this situation, the red (or lightly shaded) arrows represent the mass diffusion and the blue (or darkly shaded) arrows represent the thermal diffusion.

Investigating the unstable situation will help us understand how the thermal-diffusive instability can destabilize a flame. First, it is important to note that there are two separate conditions in this regime. When the flame front is concave to the burnt gases, and when it is convex to the burnt gases. During the situation when it is concave to the burnt gases, the light reactants are entering a confined volume with higher temperatures at a faster rated than the heat can diffuse into the colder gases. This causes the reactants to burn faster due to injection of highly reactive species, and thus increases the local flame speed. In the situation where the flame front is convex to the burnt gases, the reactants are diffusing into a larger volume, causing them to cool and thus combust at a slower rate, which in turn reduces the local flame speed. This overall increase and decrease of the flame speed leads to a situation where the flame front becomes progressively more wrinkled, and more unstable.\(^{24} \)
Figure 1.3.1 Illustration of hydrodynamic instability.
1.3.3 Body-Force Instabilities

For purposes of completeness, it is necessary to mention a third distinct source of instability in pre-mixed flames: the body-force instability. This instability usually arises from some kind of external forces or pressures being applied to the flame, gravity being a prominent example of a body-force. The gravitational effect comes from the fact that there is a density gradient throughout the flame, and anytime you have fluids or gases of different densities, there is an associated body-force effect. (For example a lighter fluid beneath a heavy fluid is an example of a body-force effect that gives rise to the classical Rayleigh-Taylor instability.) Depending on the propagation of the flame, the body-force can have both a stabilizing and destabilizing effect on the flame. For the work presented in this thesis, the flame has a downward propagation (i.e., the burner is upright), and thus the body-force instability has a stabilizing effect.

One of the most famous body-force effects on a flame is the so called “Singing Flame”. For a singing flame to form one must first enclose the flame in some kind of chimney. This can lead to a resonance forming between the flame on the burner and the chimney surrounding the flame. Figure 1.3.3 shows an example of the effects that are associated with a singing flame.

The primary objective of this work is to investigate electrical control over the thermal-diffusive instability, and how its manipulation affects the overall flame conditions. The following chapters of this thesis will serve to develop the experimental conditions (Chapter 2) and theoretical background (Chapter 3) necessary to understand the origins of the observed experimental results (Chapter 4) and the explanation of why they occur (Chapter 5). Two appendices are also included for completeness. Appendix 1 reproduces the abstract of paper that has been accepted for presentation at the 45th American Institute of Aeronautics and Astronautics Meeting in January of 2007. It represents a highly condensed summary of the major results of this work. Appendix 2 is the result of new work accomplished largely after this thesis was completed. It is a note added in proof that offers conclusive experimental evidence that the traditional explanation of the mechanism of flame/electric field interactions known as the ionic wind is not responsible for the flame stability modifications that are of central interest in this work.
### Figure 1.3.2 Sketch of flow of gases leading to the thermal-diffusive instability.

<table>
<thead>
<tr>
<th>Unstable regime ($L_e &lt; 1$)</th>
<th>Stable regime ($L_e &gt; 1$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s_1 &gt; s_1^0$</td>
<td>$s_1 &lt; s_1^0$</td>
</tr>
<tr>
<td>Cold fresh gases</td>
<td>Cold fresh gases</td>
</tr>
<tr>
<td>$s_1 &lt; s_1^0$</td>
<td>$s_1 &gt; s_1^0$</td>
</tr>
<tr>
<td>Hot burnt gases</td>
<td>Hot burnt gases</td>
</tr>
</tbody>
</table>

### Figure 1.3.3 The “Singing Flame” as described by Gaydon.
2. Experimental Setup

This section will describe in depth the experimental setup that was used to create the pre-mixed flames of interest to this work, electrically stress those flames, and measure their electrical, spectroscopic, and dynamic response to the imposed electrical stress.

2.1 Flame Diagnostic Setup

The main setup for this experiment is presented in Figure 2.1.1, and the subsequent sections will address the individual data collection methods that were used during this experiment.

The oxidant supply for this work was regular building air. After the compressor, the air passes through an air dyer to reduce the water vapor content of the air before it is sent up to the lab. Once the air reaches the lab room, we pass it through a regulator into a secondary holding tank. The air then travels from the tank through another regulator and finally to the flow meter input to the burner. This was done to eliminate any fluctuations in the building air supply which could lead to hydrodynamic changes in the flame structure.

The fuel supplies came from tanks that were plumbed through a regulator and to the flow meter. This was done so that we could switch fuels simply by changing the tank to which the fuel-feed tubing was connected.

The flow meters used in this experiment are simple Cole-Palmer ball-float flow meters (rotameters). These flow meters are capable of a flow rates up to 66 standard liters per minute (slm), which would correspond to a Reynolds number of 5000 for air, and approximately 2.0 - 3.0 slm for the different fuels used. Figures 2.1.2 and 2.1.3 show the calibration curves for the air and fuel flow meters, respectively.

It is also possible to calibrate the flow meters so that we can calculate the flow speed in meters per second. Figure 2.1.4 shows a plot of the flow speed versus the rotameters setting. Having this number allows us to calculate different properties of the flame, and is also a good way of describing the self-induced turbulence that the flame has from the flow speed (i.e. what the Reynolds number of the flame is).
Figure 2.1.1 Flame diagnostic experimental setup.
Figure 2.1.2 Air flow calibration curve.
Figure 2.1.3 Fuel flow calibration curves.
From the flow meters, the air and fuel go to the burner mixing chamber where they are mixed prior to entering the burner head. The burner is a commercial atomic absorption burner (Perkin-Elmer) with a modified stainless steel burner head. The burner head is a cylindrical design packed with stainless steel wire mesh to enhance the mixing of the fuel and oxidant. There is a brass plug with forty-four 0.8-mm-diameter holes drilled in concentric circular patterns, which is where the mixture flows out onto an unconfined combustion zone. Figure 2.1.5 shows an illustration of the burner head assembly. On top of the brass plug is a layer of stainless steel wire mesh (typically 100 mesh) that is used to help straighten the flow and assist in electrically coupling the flame with the burner head.

2.2 Electrical Setup

Figure 2.2.1 shows an electrical schematic of the flame diagnostic setup (i.e. a subset of Figure 2.1.1). The high voltage (HV) is supplied by a Glassman series WX HV power supply. The positive bias is applied to a disk electrode which is made out of #304 stainless steel, and has 44 – 3.2-mm diameter holes drilled in a regular pattern in the base of it. The electrode is located at a fixed distance of 40-mm from the top of the burner head, and remains parallel to the burner head surface. Before the bias reaches the electrode, for safety considerations it passes through four 2-MΩ current limiting resistors wired in parallel (0.5 MΩ equivalent resistance). The current limiting resistance was chosen to be small compared to the overall flame impedance, but sufficient to limit the over-capacity primary power supply from delivering dangerously large currents in the case of a mechanical short failure.

The HV probe is a Tektronix P6015 x1000 probe. This is used to measure the voltage that is being applied across the flame and the current limiting resistor. The probe thus measures a potential that is slightly less than the value read on the front panel of the Glassman power supply since there is a voltage drop across the current limiting resistors.

For estimates of the DC current level through the flame, we have a slow-response analog ammeter placed in the circuit to measure the current. More precise time-varying measurements of current were made with a digital oscilloscope. The oscilloscope measures, in real time, the potential dropped across the flame and the voltage across a current sensing resistor (100 kΩ) which is placed in series with the ammeter.
Figure 2.1.4 Calibration curve for rotameters.
Figure 2.1.5 Burner head setup.
Ohm’s law allows simple conversion to the current through the flame. The digital oscilloscope we use to make these measurements is a LeCroy, 4 channel, 500 MHz, Wave Runner Series (LT344) digital oscilloscope. The data files are exported directly from the oscilloscope to a personal computer via a RS-232 serial connection using LeCroy’s *Scope Explorer* software.

From the data collected on the oscilloscope, we are able to construct current vs. voltage plots to check for trends in the flame, saturation limits, and to make overall characterizations of the electrical condition and how well coupled the electrodes are to the flame. We also have the ability to take fast Fourier transforms (FFT) of the current signal to check for oscillations in the current throughout the flame (i.e., oscillations in the flame conductivity, or overall ion density).

### 2.3 Optical Setup

There are several optical measurements that we perform on the flame. First, we use a 15-cm focal length, 5-cm diameter bi-convex lens positioned two focal lengths from the flame and the end of a 0.5 mm diameter 1-m fiber optic cable coupled in turn to a photomultiplier (PMT). Using this optical system we are able to make position-specific optical signal measurements of flame emission using either the broadband flame emission signal, or, using band-pass filters (10 nm, FWHM), select any of the flame chemiluminescence signals described earlier. Typically we have a bias of 1150V on the PMT (RCA 7265, S-20 response) which is supplied by a Hewlet-Packard 6516A HV power supply. Finally, we measure broadband spectroscopic data using an optical multi-channel analyzer which is also fed by a fiber optic cable (not shown in Figure 2.1.1). Spectroscopic data is taken with an Ocean Optics HR2000 multi-channel spectrometer.

Imaging of the flame is done with a Princeton Instruments PIMAX (7361-0001) intensified CCD camera. This camera is configured to view side-on to the flame at a fixed distance of 1 m. Using the imaging data, global intensity changes can be measured as well as documenting structural changes in the flame. The ICCD camera is controlled by an imaging software package called Winspec which allows us to record images, adjust gain and exposure times, and perform basic statistics on the image. Spectral imaging of
Figure 2.2.1 Electrical circuit for flame diagnostic setup.
the flame in the various chemi-luminescence band of interest is accomplished by using
the same optical band-pass described above between the flame and the camera lens.

2.4 Computer Control

In addition to controlling the oscilloscope with a computer, a LabView program
has been written which allows us to use the calibration data from the flow meters to
calculate the equivalence ratio of the flame. By doing a third order polynomial fit to the
calibration curves, I am able to derive an expression relating the flow rate in slm to the
rotameter reading. Equations 2.4.1 through 2.4.4 have the relations for air, propane,
butane and methane, respectively. In these equations, y is representing the flow rate in
slm and x is representing the rotameters reading.

\[ y_{\text{air}} = 5 \times 10^{-5} x^3 - 0.0062 x^2 + 0.6325 x - 3.6966 \]  
(2.4.1)

\[ y_{\text{propane}} = 1 \times 10^{-7} x^3 - 5 \times 10^{-5} x^2 + 0.0186 x + 0.0511 \]  
(2.4.2)

\[ y_{\text{butane}} = 6 \times 10^{-8} - 3 \times 10^{-5} x^2 + 0.0157 x + 0.0753 \]  
(2.4.3)

\[ y_{\text{methane}} = 1 \times 10^{-7} x^3 - 6 \times 10^{-5} x^2 + 0.0274 x - 0.0025 \]  
(2.4.4)

Once the flow rates in slm are known, it is possible to calculate the corresponding
equivalence ratio in an alternate way using equation 2.4.5 as a modified way of
calculating the equivalence ratio of the flame. Letting \( a = x + \frac{y}{4} \), where x and y are
defined by the hydrocarbon (C\(_x\)H\(_y\)) and fuel and air represent the flow rates of each in
slm, the equivalence ratio can be written very simply as,

\[ \Phi = \frac{4.76a \cdot (\text{fuel})}{(\text{air})}. \]  
(2.4.5)
Figure 2.4.1 shows the front panel of the program written to do these calculations. You are required to enter the fuel and air rotameter readings, and then you flip a switch for the particular fuel that you are working with. The program will give you the equivalence ratio, flow rate of air and fuel in slm, and the flow velocity in m/s as outputs.

The following chapter develops the theoretical background necessary to understand the origins of the electric field/flame interactions that give rise to the observed stability modifications. The experimental results discovered during this work are presented in Chapter 4, and the major conclusions that can be drawn from our observations are described in the final chapter.
Figure 2.4.1 LabView front panel of equivalence ration program.
3. Theoretical Development

3.1 Ion Formation

Applied electric fields will only modify the flame characteristics if there are charged particles within the flame for them to act upon. Therefore, understanding ion production in simple hydrocarbon flames is a critical component of the flame for the work presented here. The origin of ions in a flame is something that has been studied at great lengths by several different methods, with Calcote\textsuperscript{26} being one of the early pioneers attempting to discover the method for the production of ions in a flame. He theorized that the two possible sources of ionization were either cumulative excitation, or chemi-ionization, since the flame temperatures are woefully insufficient to produce observed ion densities by thermal means. Subsequent work\textsuperscript{27} has shown that the process known as chemi-ionization is responsible for the formation of ions in a flame. In this process, one of the reactants from a chemical reaction is left in an ionized state. The following relation is useful for understanding how the process of chemi-ionization works:

\begin{equation}
A + B \rightarrow C + D^+ + e^- \tag{3.1.1}
\end{equation}

where A, B, C and D can be any chemical elements or molecules, and e\textsuperscript{-} is a free electron for conservation of charge. This is basically stating that as a chemical reaction moves over a surface of potential energy, it actually crosses over to an excited potential energy surface for the products.\textsuperscript{26}

In our experimental setup, we supply the electrode over the flame with a positive bias; therefore we are interested only in the positive ions of the flame. Basic electromagnetic theory tells us that when you have two like charges, they will repel each other. In the case of our experiment the positive ions are thus being re-located toward the base of the flame, a fact that will be investigated much more thoroughly in the discussion section of this thesis.

Looking back at Table 1.2.1, it is found that the chain branching reaction for ion production can be expressed by the following relation (reaction 74 of Table 1.2.1):
\[ \text{CHO}^+ \text{ does not survive in the flame for a long time, as it is consumed by the following reaction which rapidly produces the hydronium ion, H}_3\text{O}^+. \]

\[ \text{CHO}^+ + \text{H}_2\text{O} \rightarrow \text{H}_3\text{O}^+ + \text{CO} \]  \hspace{1cm} (3.1.3)

Measurements made by Goodings\textsuperscript{28}, et. al., using a mass spectrometer have shown that the production of positive ions predominates and the hydronium ion is the most prevalent ion in a flame. Figure 3.1.1 taken from Goodings’ work shows a plot of the concentrations of the most abundant positive ions in a flame as a function of the distance from the base of a laminar flame. Thus, in our experimental setup, we supply the electrode over the flame with a positive bias and ground the burner head, providing an electric field that is generally directed counter to the flow. Electromagnetic theory then dictates that flame positive ions will be directed toward the burner head by the externally applied field. In the case of our experiment the positive ions are thus being re-located toward the base of the flame, a fact that will be investigated much more thoroughly in the discussion section of this thesis.

As would be expected, the ion concentrations are strongly dependant on the fuel used, overall flow rates and the equivalence ratio used. Figure 3.1.1 shows a plot of how the current through a flame, which can be directly related to total ion concentration, varies as a function of fuel and equivalence ratio.

3.2 Ionic Wind Theory

In typical combustion applications the presence of ions is of little concern. Ions typically only make up a small fraction of the overall composition of a flame (typically 1 in \(10^6\) - \(10^7\) particles is an ion). Nevertheless, soon after the discovery of the chemi-ionization mechanism work was begun to investigate how externally applied electric fields can impact a flame. Some of the first work to be done on this subject was in the 1960’s by J. Lawton, H. Payne and F. Weinberg.\textsuperscript{29} Their theory on how the electric field
perturbs a flame, known as the ionic wind theory, was (and to a large extent still is) accepted as the primary explanation for how flames change while under the effect of an applied electric field. The ionic wind theory states that there is a net body-force that is experienced by a flame due to a drag associated with the movement of the positive ions in the flame.\(^{30}\) This body force creates a pressure change across the flame, which results in the geometry changes seen when the field is applied. Using the equation 3.2.1\(^{10}\) they predict that there is a maximum pressure change in the flame, due to the electric field, of only 0.0004 atmospheres.

\[
\Delta p = jx \left( \frac{1}{\mu_+} - \frac{1}{\mu_-} \right),
\]

where, \(j\) is the electric current flux, \(x\) is a spatial variable, and \(\mu_\pm\) are the positive and negative ion mobility rates.

Their theory also predicts a maximum current density, \(j_s\), of 250 \(\mu\text{A/cm}^2\) using an applied electric field of 30 kV/cm.\(^{31}\) The ionic wind theory is not sufficient to explain the dramatic reaction zone geometry modifications, flame speed and combustion intensity enhancements, and radical changes in the stability of flames that are the subject of this work. The problems associated with this theory, and explanations for the ability to exceed their maximum values will be developed more thoroughly in the discussion section of this thesis.

### 3.3 Electric Pressure

Marcum and Ganguly\(^{32}\) proposed the electric pressure effect as a partial explanation for the ability of an applied electric field to drive a pre-mixed propane/air flame from laminar to turbulent flow. Electric pressure is defined as “an effective field-induced dynamic pressure on the flame front positive ions”. Electric pressure has a similar effect on a flame as that of the singing flame which was discussed in Chapter 1. The effects of electric fields on flames reported by Marcum and Ganguly were large
Figure 3.1.1 Plot of current as a function of fuel and equivalence ratio.
compared to those that could be produced by the ionic wind to the extent that an explanation other than that offered by the ionic wind had to be considered. They observed that under otherwise quiescent laminar flow conditions where Fick’s law applies and strong temperature gradients give rise to significant thermal diffusion, imposition of a closely coupled external electric field on the flame gives rise to an additional electric pressure induced ionic molecular diffusion effect. The electric field acts directly on the highly localized, chemi-ionization-derived positive ions in the flame reaction zone, significantly increasing their mass diffusivities and thereby lowering the Lewis numbers of the ionic species. The observed result is a diffusive-thermal instability, typical of flames with global Lewis numbers less than one. The increased differential diffusional velocity of the reaction zone positive ions strongly affects the bulk flame speed, and the instability results in the flame front collapsing toward the burner head and taking on a wrinkled laminar geometry. Strikingly similar responses of flames to variations in ambient pressure well above the maximum predicted for the ionic wind, suggested the term electric pressure would be an appropriate label for the observed effects. The strong wrinkling of the flame front and other indications of the onset of turbulence they reported suggest that electrically stressed hydrocarbon flames can be quite sensitive to the electric pressure effect. (See also Appendix II: ionic wind versus Electric Pressure, which is the result of new work accomplished largely after this thesis was completed. It is a note added in proof that offers conclusive experimental evidence that the traditional explanation of the mechanism of flame/electric field interactions known as the ionic wind is not responsible for the flame stability modifications that are of central interest in this work.)

The work reported here uses higher overall flow rates where the un-perturbed flame has been driven to a state of weak hydrodynamic instability (Re ~ 2500). As will be shown in the following chapters, the electric pressure effect can be made sufficiently large so as to stabilize a hydrodynamically unstable flame (i.e. force the flame to laminar burning conditions). Further increases in the electric pressure achieved by application of higher flame bias voltages and better electrical coupling to the flame causes the electric-pressure-initiated thermal-diffusive instability to dominate the flame fluidics, resulting in the collapse of the flame front toward the burner head and the onset of highly turbulent
burning conditions. This work also reports the efficacy of the electric pressure effect as a function of increasing fuel weight.

### 3.4 Instability Growth Rates

A mature mathematical model of coupled hydrodynamic and diffusive-thermal instabilities in flames has developed over time that very accurately describes the stability modifications that are central to this work. Such models have as their central feature an instability growth rate, $\sigma$, a parameter that is used to quantify the rate at which a given hydrodynamic or thermal-diffusive instability grows with time. The following relations were developed by Landau\textsuperscript{20} in 1944 to express how the hydrodynamic instability grows with time.

\begin{equation}
\sigma = \Omega_o U_b k \tag{3.4.1}
\end{equation}

\begin{equation}
\Omega_o = \frac{\sqrt{\varepsilon + \varepsilon^2 - \varepsilon^3} - \varepsilon}{1 + \varepsilon}, \tag{3.4.2}
\end{equation}

where $U_b$ is the flame speed, $k$ is the wave number of the disturbance, and $\varepsilon = \rho_b / \rho_o$ with $\rho_{b,o}$ being the densities of the burned and unburned gases, respectively. Landau treated the flame as a density discontinuity in an ideal fluid traveling at a constant velocity relative to the gases. As was mentioned in section 1.3, this type of instability is always present in flames with exothermic reactions. Figure 3.4.1\textsuperscript{20} shows a plot of the growth rates, due only to the hydrodynamic instability, as a function of flame speed. Typically, if the flame is deficient in the heavy component (lean flames) then the flame will experience only the hydrodynamic instability\textsuperscript{20}, and the above relations are sufficient to calculate the overall growth rate.

When the mass diffusivity of the limiting reactant exceeds the thermal diffusivity, then there is the onset of the thermal-diffusive instability.\textsuperscript{37} If one assumes that the gas density is constant, then any thermal disturbances cannot be transformed into hydrodynamic disturbances, and it is possible to investigate the thermal-diffusive
instability growth rate independent of the hydrodynamic instability. Sivashinsky\textsuperscript{38} has shown that under these conditions one can express the growth rate by the following relations,

$$\sigma = D_{th} [5\beta (1 - Le) - 1] k^2$$  \hspace{1cm} (3.4.3)$$

where $D_{th}$ is the thermal diffusivity, $\beta = \frac{E(T_b - T_u)}{RT_b^2}$ (also known as the Zeldovich number), $R$ is the universal gas constant, and $T_{b,u}$ is the burned/unburned gas temperature.

For the purpose of this thesis, we wish to investigate how one can manipulate the thermal-diffusive instability in a flame by electrical control. One method of doing this is to change the overall $Le$ of the flame. Using the above discussion on the separate issues of the hydrodynamic and thermal-diffusive instabilities, an expression has been derived\textsuperscript{38} to show how the instability growth rate depends on both the hydrodynamic and thermal-diffusive instabilities. In the following relation, the first term is that associated with the hydrodynamic instability and the second term is associated with the thermal-diffusive instability\textsuperscript{17},

$$\sigma = \Omega_0 U_b k - \Omega_1 D_{th} k^2,$$  \hspace{1cm} (3.4.4)$$

where $\Omega_1$ is expressed by the following relation:

$$\Omega_1 = \frac{\varepsilon (1 - \varepsilon)^2 - \varepsilon \ln \varepsilon (2\Omega_0 + 1 + \varepsilon)}{2(1 - \varepsilon)^2 [\varepsilon + (1 + \varepsilon)\Omega_0]} - \frac{\varepsilon (1 + \Omega_0) [\varepsilon + \Omega_0] \beta (1 - Le)}{2(1 - \varepsilon)^2 [\varepsilon + (1 + \varepsilon)\Omega_0]} \int_{\varepsilon - 1}^{\varepsilon - 1} \frac{\ln(1 + \xi)}{\xi} d\xi$$  \hspace{1cm} (3.4.5)$$

As is clearly visible, the Lewis number effect only comes in through the second term in equation 3.4.4. If you take equation 3.4.5 and set it equal to 0, then the value of $Le$ that you get is known as the critical Lewis number, or $Le_c$. If you have a situation where equation 3.4.5 has an overall positive sign, then the thermal-diffusive effect acts to stabilize the overall flame. This is due to the negative sign in equation 3.4.4, a positive $\Omega_1$ would cause the thermal-diffusive term to lower the instability growth rate, thus you
Figure 3.4.1 Plot of instability growth rates vs. flame speed.
get the afore mentioned stabilizing effect. A negative sign with equation 3.4.5 would have the opposite effect to the instability growth rate, causing it to increase. When there is an overall negative sign on equation 3.4.5 that is an indication that you have dropped below the value of the critical Lewis number of the flame, which is taken to be between 0.7 and 0.9 for hydrocarbons.\textsuperscript{38,17} Figure 3.4.2 shows a plot of critical Lewis number as a function of $\varepsilon$ for two different values of $\beta$.

Even though the ions represent a small fraction of the flame species, they are able to greatly influence its structure through non-linear means. Work by Sivashinsky\textsuperscript{38} has shown that the effects of the instability growth rate can be related to the flame changes by means of a forcing function. The forcing function incorporates the non-linearity into the theory by the following expression,

$$F \sim e^{(\sigma t + \mathbf{k} \cdot \mathbf{x})}.$$  (3.4.6)

Possible explanations for how one can manipulate the Lewis number, and thus the thermal-diffusive instability, will be discussed in detail in the discussion section of this thesis. The equations presented here for the instability growth rates are taken from referenced papers whose authors are experts in the fields of fluid mechanics, computational physics, and combustion modeling. The starting point for these equations is the compressible Navier-Stokes equations, which are non-linear differential equations that can only be approximately solved by the use of computer modeling. We are therefore comparing our experimental data to the best theories that are available to us at this time. The remaining chapters of this thesis present the experimental results (Chapter 4) and the conclusions (Chapter 5) that can be derived from our experimental observations.
Figure 3.4.2 Plot of critical Lewis number as a function of the density ratio.
4. Experimental Results

4.1 General Results

4.1.1 Introduction and Background

As was mentioned in Chapter 2 of this thesis, there are several methods that were employed to gather our data. This chapter of the thesis is divided into three additional sections, one for each of the fuels that we studied in the course of this experiment.

The format for presenting the results on the electric-field-induced flame instabilities will be presented in the next section of this chapter, with all subsequent sections repeating the same format. This current section is intended to present some general flame experimental results to the reader.

4.1.2 Direct Imaging versus Spectral Imaging

For the purpose of this thesis, direct imaging will refer to conditions where there is no filtering of the flame light emission and spectral imaging will refer to conditions with a band-pass filter in place in front of the camera lens which selects a given chemi-luminescence signal of interest. The following sets of figures are all for propane, at a flow velocity of 1.69 m/s, taken at equivalence ratios of 0.8, 1.2, and 1.4. For each equivalence ratio, applied voltages of 3.8 kV, and 5.0 kV as well as a zero bias case are imaged both directly and spectrally. The filters used are all 10-nm (full-width at half maximum, FWHM) band-pass filters centered at wavelengths appropriate for OH*, CH*, and C$_2$* chemi-luminescence emissions mentioned in Chapter 1.

Figure 4.1.1 is a set of images taken at an equivalence ratio of 0.8 using both spectral and direct imaging. For the direct imaging (4.1.1a) case, the exposure time of the camera was at 1 ms and as for all images presented in this thesis the gain on the intensifier was set its maximum level. The exposure times used by the ICCD camera for the direct imaging, CH*, C$_2$*, and OH* spectral imaging are 20 ms, 60 ms, and 40 ms, respectively. Figure 4.1.2 is a similar set of images taken of propane with an equivalence ratio of 1.2. The exposure times used by the ICCD camera for the direct imaging, CH*, C$_2$*, and OH* spectral imaging are 500 µs, 10 ms, 20 ms, and 20 ms, respectively for that case. Figure 4.1.3 is a set of images taken with the same conditions as Figure 4.1.2, but at an equivalence ratio of 1.4. This set of figures
clearly demonstrates that the DC-electric-field-induced modifications of the reaction zone are essentially identical for all of the flame chemi-luminescence emissions. It is not surprising that all emission signals show the same field-induced changes since we are not changing the overall flame chemistry with the small electrical input powers used. With that fact established, all future images shown will be those derived from direct imaging which allows the use of the shortest exposure time.

4.1.3 Broadband Spectroscopic Flame Monitoring

Spectroscopic data taken with an Ocean Optics High Resolution mini spectrometer is useful for reiterating how the flame emissions change with equivalence ratio. Figure 4.1.4 shows how the spectra for propane/air flames vary with fuel/air mixture for the same three equivalence ratios used for the previous direct and spectral images (i.e. \( \phi = 0.8, 1.2 \) and 1.4). The three fuels used for this work all have similar stoichiometric temperatures, which is a rough indication that the overall combustion chemistry is similar for the three fuels. It is clear from Figure 4.1.4 that as the equivalence ratio is increased, the ratio of the C\(_2\)* emission intensity to that of CH* increases dramatically.

4.1.4 Self-excited Flame Oscillation Measurements

At certain combinations of fuel/air mixture, flow rate and applied bias voltage, the electrically stressed flame fronts are observed to undergo either global or local self-excited oscillation. Such behavior is the first observation that indicated that the local chemistry of the flame changes. Measurement of the current and optical (PMT) waveforms and conversion to a power spectrum via fast Fourier transform (FFT) processing is the most revealing way to document such chemistry changes. Figure 4.1.5 shows an example of a current and optical waveform signal FFTs for propane at an equivalence ratio of 1.2 and a flow speed of 1.69 m/s with an applied voltage of 5.2 kV.

When looking at the FFT, the spikes that are seen are the harmonics from the modulation of the current and optical signals. This relates directly to a changing of the local chemistry of the flame, when the field is applied. Figure 4.1.6 is an optical FFT with a CH filter at the same flame conditions as above, but with no voltage applied. As
Figure 4.1.1 Imaging of propane taken with the ICCD camera using: a) direct imaging, b) CH spectral imaging, c) C₂ spectral imaging, and d) OH spectral imaging, at an equivalence ratio of 0.8.
Figure 4.1.2 Imaging of propane taken with the ICCD camera using: a) direct imaging, b) CH spectral imaging, c) C\textsubscript{2} spectral imaging, and d) OH spectral imaging, at an equivalence ratio of 1.2.
Figure 4.1.3 Imaging of propane taking with the ICCD camera using: a) direct imaging, b) CH spectral imaging, c) C\textsubscript{2} spectral imaging, and d) OH spectral imaging, at an equivalence ratio of 1.4.
can be seen in that figure, with no applied field there are no visible harmonics or spikes in
the FFT, indicative of a quiescent laminar flame.

The following sections of this chapter will be devoted to the presentation of
instability data used to reach conclusions presented in the final chapter. For each fuel,
there is a set of images showing the transformation from hydrodynamic-instability
dominated behavior to a stable, laminar flame, to the onset of thermal-diffusive-
instability dominated flame, and, when possible, the fully chaotic flame front structure
typical of highly turbulent combustion. Data sets that document that series of stability
modifications are presented for each fuel, and over a range of equivalence ratios spanning
a range from fuel lean to fuel rich. Following each set of images, corresponding flame
current/voltage characteristics are presented to illustrate the electrical properties of the
flame at a given condition. One final piece of information that included with each image
is a profile of the flame intensity as a function of location in the flame. That information
can in turn be used to estimate the increase in combustion intensity (heat release per unit
volume) caused by the field-induced stability modifications.

4.2 Propane

Propane (C₃H₈) is one of the most widely studied hydrocarbon fuels, and thus it is
the fuel we studied most intensively. Table 4.2.1 lists some important properties of
propane that are useful and necessary for any thorough study of the fuel.

<table>
<thead>
<tr>
<th>Propane</th>
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<tr>
<td>Formula</td>
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<td>MW</td>
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<td>Boiling Pt.</td>
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Table 4.2.1 Propane fuel properties.
Figure 4.1.4 Emission spectra for propane/air flames at three different equivalence ratios.
Figure 4.1.5 Optical (CH) and current FFT signals of propane at 5.2 kV applied voltage.
Figure 4.1.6 FFT of CH emission with no applied bias.
The set of conditions for propane that are reported here include the equivalence ratios 0.9, 1.2 and 1.4. The combined fuel/air flow speed was 1.94 m/s and the applied bias voltage ranged from 0 kV to 6.0 kV. With no applied bias, Figure 4.2.1 clearly shows that the flame is hydro-dynamically unstable, or at a condition of high Reynolds number (roughly 2500). When the flame has a small voltage bias applied, it is forced into smooth, laminar flame geometry as shown in Figure 4.2.2. At this condition the thermo-diffusive instability is great enough to suppress the hydrodynamic instability, i.e. the competing instabilities simply cancel each other, producing a highly stable laminar flame front. With a slightly higher voltage, the thermal-diffusive instability begins to dominate, and the flame becomes unstable once again. Figures 4.2.1 - .3 show the abovementioned transitions for an equivalence ratio of 0.9, along with the localized change in combustion intensity of the flame at applied voltages of 0 kV, 4.0 kV, and 6.0 kV.

Figure 4.2.4 shows a current/voltage (IV) characteristic for the condition presented in the images. The location on the IV plot where each image was taken are clearly marked on the IV graph as A for no voltage, B for 4.0 kV, and C for 6.0 kV. Note that the IV graph accounts for the voltage drop over our 100-kΩ current sensing resistor, which is why the voltages are slightly less than the above quoted values (i.e. numbers quoted are those that are read from the HV power supply before the drop across the current sensing resistor).

For a wide range of equivalence ratios from the lean limit to about 1.2, as the ratio is increased the flame conductivity also increases. This effect can be seen by comparing the IV characteristics Figure 4.2.5 ($\phi = 1.2$, see below) with Figure 4.2.4 ($\phi = 0.9$). This results in a greater affect on the flame by the applied bias potential, as is seen by looking at figures 4.2.6-4.2.9. Also, as is clearly visible by comparing figures 4.2.6 and 4.2.9, as the flame is “crushed” downward and confined to a smaller area, the combustion intensity increases.

Increasing the equivalence ratio to 1.4 causes a large concentration of carbon clusters (soot) to be produced from unburned hydrocarbons. Soot formation, along with lower temperature combustion causes the flame conductivity to fall. This can be seen by comparing the IV characteristics of Figure 4.2.10 ($\phi = 1.4$) to those of Figures 4.2.4 and 4.2.5. This overall reduction of the flame conductivity leads to a reduced affect
Figure 4.2.1 Image of propane at $\Phi = 0.9$, and no applied voltage including an intensity plot as a function of distance from flame central axis.

Figure 4.2.2 Image of propane at $\Phi = 0.9$, and an applied voltage of 4.0 kV.
Figure 4.2.3 Image of propane at $\Phi = 0.9$, and an applied voltage of 6.0 kV, along with an intensity plot as a function of flame central axis.

Figure 4.2.4 IV curve for propane at an equivalence ratio of 0.9.
on the flame by the applied bias voltage, which can be seen by looking at the images and combustion intensity profiles of Figures 4.2.11 – 4.2.13.

This represents all of the instability data presented here for propane. By investigating how the flame structure changes as functions of applied voltage and equivalence ratio, it is possible to bracket the range over which flame instabilities can be significantly modified, or, the range of operating conditions over which significant control over the flame instability growth rate can be achieved by applied electric fields.

It is clear that as the voltage applied to the flame is increased, the flame ions are forced to diffuse (or drift) counter to the flow and downward toward the burner head at increasing rates. The current/voltage characteristics presented in this section, in concert with the clear changes in flame stability shown by the accompanying images, make it clear why the ability to control the flame stability is a strong function of equivalence ratio. As the ion concentrations of the flame increase, so does the ability to change the instability growth rate by an applied bias – it is fundamentally a current driven process.

The following two sections will demonstrate the ability to control the stability of methane and butane-fueled pre-mixed flames. All analysis of the data will be similar to that presented here for propane, so it is not necessary to include a detailed discussion of the results in those sections.

Detailed discussions of the importance of these results, their relevance to the issue of the instabilities present in a flame and our ability to control them, will be presented in the final chapter of this thesis.

4.3 Methane

Methane is a lighter fuel than propane, which requires a larger flow rate of fuel to reach the same equivalence ratio when compared to propane. The discussion from Chapter 1 on stoichiometry explains why this is a necessary condition. These conditions with methane lead to a smaller ion density, which will become obvious when looking at the IV graphs for methane. Table 4.3.1 lists for methane the same properties as Table 4.2.1 showed for propane.
Figure 4.2.5 IV of propane at an equivalence ratio of 1.2.

Figure 4.2.6 Propane imaging at an equivalence ratio of 1.2 with no applied voltage.
Figure 4.2.7 Propane imaging at an equivalence ratio of 1.2 with an applied voltage of 3.2 kV.

Figure 4.2.8 Propane imaging at an equivalence ratio of 1.2 with an applied voltage of 5.0 kV.
Figure 4.2.9 Propane imaging at an equivalence ratio of 1.2 with an applied voltage of 6.0 kV.

Figure 4.2.10 IV of propane at an equivalence ratio of 1.4.
Figure 4.2.11 Propane imaging at an equivalence ratio of 1.4 with no applied voltage.

Figure 4.2.12 Propane imaging at an equivalence ratio of 1.4 with a 3.2 kV applied voltage.
Figure 4.2.13 Propane imaging at an equivalence ratio of 1.4, with an applied voltage of 6.0 kV.
Table 4.3.1 Methane fuel properties.

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<td>Boiling Pt.</td>
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Figures 4.3.1-4.3.4 are images and an IV for methane at a flow speed of 1.69 m/s at an equivalence ratio of 0.9. This represents the lowest equivalence ratio in methane that it was possible to overcome the hydrodynamic instabilities caused by the high flow rate.

Figures 4.3.5-4.3.8 show the case of methane at an equivalence ratio of 1.0. At this condition, it was still impossible to drive the flame into a fully turbulent state, but it was possible to force the flame from a hydrodynamic-instability-dominated state, to a stable laminar flame front, and then to the onset of the thermal-diffusive-instability dominated regime. As is clearly visible from the IV graph, the flame at this condition is much less conductive than a similar case in propane, which as mentioned above, is why it is not as responsive to the applied field.

The highest equivalence ratio where we were able to affect the flame with the field in methane was 1.1. After this condition, the flame tip was contacting the top electrode and it was no longer possible to make measurements on the flame. Figures 4.3.9 – 4.3.12 show the abovementioned case with methane, as well as an IV curve for that condition.

4.4 Butane

Butane is an interesting fuel to study because it is the only one of our fuels that has two different isomers. In fact, it is the lightest hydrocarbon for which isomers exist. The isomer of butane that was used for this work was n-butane, which is just a straight chain of carbon bonds (C-C-C-C). Iso-butane has a different branching structure of
Figure 4.3.1 Image of methane at an equivalence ratio of 0.9 with no applied voltage.

Figure 4.3.2 Image of methane at an equivalence ratio of 0.9 with an applied voltage of 4.6 kV.
Figure 4.3.3 Image of methane at an equivalence ratio of 0.9 with an applied voltage of 7.0 kV.

Figure 4.3.4 IV of methane for an equivalence ratio of 0.9 at a flow rate of 1.94 m/s.
Figure 4.3.5 Image of methane at an equivalence ratio of 1.0 with no applied voltage.

Figure 4.3.6 Image of methane at an equivalence ratio of 1.0 with an applied voltage of 3.8 kV.
Figure 4.3.7 Image of methane at an equivalence ratio of 1.0 with an applied voltage of 7.0 kV.

Figure 4.3.8 IV of methane for an equivalence ratio of 1.0 at a flow rate of 1.94 m/s.
Figure 4.3.9 Image of methane at an equivalence ratio of 1.1, with no applied voltage.

Figure 4.3.10 Image of methane at an equivalence ratio of 1.1 with an applied voltage of 4.0 kV.
Figure 4.3.11 Image of methane at an equivalence ratio of 1.1 with an applied voltage of 5.8 kV.

Figure 4.3.12 IV of methane for an equivalence ratio of 1.1 at a flow rate of 1.94 m/s.
carbon, and thus a different chemistry. Table 4.4.1 has some of the important properties of n-butane that are needed.

<table>
<thead>
<tr>
<th>Butane</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Formula</td>
<td>$C_4H_{10}$</td>
</tr>
<tr>
<td>MW</td>
<td>58.123 kg/mol</td>
</tr>
<tr>
<td>Boiling Pt.</td>
<td>-5 °C</td>
</tr>
<tr>
<td>T</td>
<td>2270 °K</td>
</tr>
</tbody>
</table>

Table 4.4.1 Butane fuel properties.

Figures 4.4.1 – 4.4.4 show the condition of butane at an equivalence ratio of 0.9, along with the associated IV characteristic. In this condition the onset of a thermal-diffusive-instability dominated flame is particularly easy to see. When moving to the higher equivalence ratio of 1.2, the ion density is higher so that a much better effect on the flame by the applied potential is observed. Figures 4.4.5 – 4.4.9 show the case of $\phi = 1.2$, and includes the IV curve for that case.

The final set of data presented here is for an equivalence ratio of 1.4 with butane. As with previous cases there are the sets of images to show the transformation to thermal-diffusive-instability-dominated flames, as well as an IV curve to characterize the electrical properties of the flame. Figures 4.4.10 – 4.4.13 illustrate this transformation for the above conditions.

The final chapter of this thesis presents a discussion of the above experimental results, where some previous theories fail, and some conclusions and future directions for research that can be derived form this work.
Figure 4.4.1 Image and intensity profile for butane with an equivalence ratio of 0.9 and no applied voltage.

Figure 4.4.2 Image of butane at an equivalence ratio of 0.9 with an applied voltage of 4.0 kV.
Figure 4.4.3 Image and intensity profile for butane with an equivalence ratio of 0.9 and an applied voltage of 6.0 kV.

Figure 4.4.4 IV curve for butane at an equivalence ratio of 0.9.
Figure 4.4.5 IV curve for butane at an equivalence ratio of 1.2.

Figure 4.4.6 Image and intensity profile for butane with an equivalence ratio of 1.2 and no applied voltage.
Figure 4.4.7 Image of butane at an equivalence ratio of 1.2 with an applied voltage of 2.6 kV.

Figure 4.4.8 Image of butane at an equivalence ratio of 1.2 with an applied voltage of 3.8 kV.
Figure 4.4.9 Image and intensity profile for butane with an equivalence ratio of 1.2 and an applied voltage of 5.2 kV.

Figure 4.4.10 Image and intensity profile for butane with an equivalence ratio of 1.4 and no applied voltage.
Figure 4.4.11 Image of butane at an equivalence ratio of 1.4 with an applied voltage of 3.2 kV.

Figure 4.4.12 Image and intensity profile for butane with an equivalence ratio of 1.4 and an applied voltage of 5.2 kV.
Figure 4.4.13 IV curve for butane at an equivalence ratio of 1.4.
5. Discussion and Conclusions

In the first three chapters of this thesis the background information on the combustion phenomena of interest, and the most widely accepted theoretical work to date that describes both electric-field/flame interactions\textsuperscript{10,30-31} and flame instabilities\textsuperscript{17,37-38}, have been outlined in detail. In Chapter 4, our experimental investigations into electric-field/flame interactions have been detailed for methane-, propane-, and butane-fueled pre-mixed flames. This chapter has two major objectives, the first being to show that the ionic wind theory is insufficient to account for our experimental observations, and second to extend a much more plausible explanation\textsuperscript{32} in terms of electric-field-induced flame chemistry changes and resultant flame instability control. While this work essentially eliminates the ionic wind\textsuperscript{10,30-31} as candidate for accounting for the field/flame interactions we observe, it serves to validate the theory of coupled hydrodynamic and diffusional-thermal instabilities\textsuperscript{17,32,37-38} in flame propagation that has been quantified most fully by the work of Sivashinsky.

As was mentioned earlier, the ionic wind theory states\textsuperscript{10,30-31} that there is a maximum current density of 250 $\mu$A/cm$^2$ that can be achieved in a pre-mixed light hydrocarbon flame using an applied electric field of 30 kV/cm. In addition to the impractical level of voltage needed to achieve such a field using reasonable anode-cathode gaps (for example, for the 4-cm gaps used for this work would require an applied potential of 120 kV!), the maximum pressure difference that is predicted by the ionic wind theory, 0.0004 atm, is much too small to account for the flame front modifications seen in our results.

On the face of it, there were two major and closely associated problems in the development of the ionic wind theory. The first and most impacting problem is the positioning of the high voltage electrode. The top electrode in the setup\textsuperscript{10,30-31} was placed a sufficient distance away from the flame so as not to disturb the flow field. That resulted in the bulk of the applied voltage drop in “cold, dead” air. That had the practical effect of requiring impractically large voltages to be used to electrically manipulate a flame. The second problem with their theory was the assumption of a uniform electric field between the electrodes. While that assumption is true when there is no flame
present since you have essentially a simple parallel plate capacitor arrangement, once a conducting flame is established between the electrodes that assumption so grossly oversimplifies the model that it cannot possibly describe reality.

The electric pressure effect\textsuperscript{32} mentioned in Chapter 3 is therefore offered as a partial explanation for the effects reported here. Our data shows we are able to reach a similar current density as predicted by the ionic wind theory with only an average applied field of less than 1.5 kV/cm. With the placement of the top electrode in intimate contact with the flame and using a disk-geometry electrode instead of a ring, we are able to much more effectively couple the applied voltage to the flame. This is the key to achieving the results presented in this thesis.

Appendix II offers data that was acquired very recently (and after the completion of the original draft of this thesis) as evidence that the ionic wind theory cannot be responsible for the flame stability modifications reported here. At best, the ionic wind theory is simply something that initiates the process, and then other process must dominate to cause the effects seen.

For a complete understanding of a possible alternate theory it is important to revisit the list of ionic reactions from Chapter 1, Table 1.2.1. It has been established by direct mass spectroscopic measurements that positive ions very strongly dominate the ion population\textsuperscript{28} in light hydrocarbon flames. Hence, we apply a positive bias to the top electrode. As previously mentioned, basic electromagnetic theory tells us that if we apply a positive bias to the top electrode, then the positive flame ions will be repelled away from this electrode toward the grounded burner head. As shown by Goodings, et al.\textsuperscript{28}, the positive ions are concentrated just at the surface of the reaction zone and diminish with distance into the area of the burned gases. By repelling them from the upper electrode we are causing them to drift toward the burner head, and closer to the pre-heat zone. Additionally, as shown in the direct and spectral images of the previous chapter, the area of the flame that is in close contact with the grounded burner head is substantially increased by the application of potentials that are well coupled to the flame.

It is important to recall that the majority flame ion, again as shown by Goodings, et al., is the hydronium ion, H\textsubscript{3}O\textsuperscript{+}. As shown in Table 1.2.1, the primary ion destruction mechanism for the hydronium ion in light hydrocarbon flames is by dissociative
recombination. Of the possible branches for dissociative recombination of H$_3$O$^+$, the three primary branches$^{39}$ yield OH + 2H (48 %), H$_2$O + H (33 %), or OH + H$_2$ as products. H, H$_2$ and OH reactants have Lewis numbers of 0.18, 0.3 and 0.73, respectively (see Table 1.2.2). All of these Le are lower than the typical hydrocarbon critical Le that was reported earlier in this thesis.

This implies that relocating the hydronium ion to the burner head can cause infusion of a continuous supply of light reactants at the burner head. According to the prevailing theory of coupled hydrodynamic and diffusional-thermal instabilities in flames propagation at subunity Lewis numbers, that will work to lower the overall Lewis number of the flame. Lowering the flame Lewis number below the critical value the will cause the instability growth rate (Equation 3.4.5) to turn positive (see the discussion in Chapter 3 concerning the instability growth rate). Since at the high flow speeds used in this work the flame is initially dominated by the hydrodynamic instability, the first change in flame propagation that is seen as the current through the flame rises is the transition to a true laminar case. This occurs because the Le term is being lowered by injecting the hydronium ion dissociative recombination products at the base of the flame. The rate of injection of those light reactants at the base of the flame increases as the current drawn through the flame increases. This Lewis number term is related to the thermal-diffusive instability, so as current is increased, so does the growth rate of the thermal-diffusive instability. This eventually reaches a level were it cancels out completely the effects of the hydrodynamic instability, which is what gives us the true laminar case. If the current is increased further, the thermal-diffusive instability will continue to grow and eventually dominate the flame leading to an overall unstable condition again. In some cases, it is possible to increase the effect to the point of producing very highly turbulent flame propagation.

This is a truly chemistry-driven process, which can be controlled by simply adjusting the voltage applied to the flame. The level of effects seen is a function of the fuel used, the overall flow velocity, and the equivalence ratio the flame. The explanation of this can most easily be understood by looking the graph of current versus the equivalence ratio of the different fuels shown in Figure 3.1.2. Obviously, since we are affecting the flame with an electric field, the most significant level of response will be
seen when the flame is most conductive. This occurs at slightly different equivalence ratios for the three different fuels, all near or slightly less than $\Phi = 1.2$.

Considerable work remains to be done to isolate the exact chemical changes that are occurring, as well as determining the level of current that is needed before the affects are seen. Future work includes setting up a method for local temperature measurements, CH absorption measurements and possibly LIF measurements of the OH concentration near the burner head.
Reference List:

Appendix I

The Effects of Electric Fields on the Thermodiffusive Instability in Hydrocarbon/Air Flames

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Abstract

We report experimental investigations of the effects of DC electric fields on pre-mixed hydrocarbon/air flames fueled by methane, propane and butane. The current study complements previous work[1,2] and extends recent results[3] which showed that dramatic modifications of the reaction zone of propane/air flames could be achieved with voltages up to 3 kV applied appropriately across the flame. Potential practical applications in a wide variety of combustors include providing for controlled flame holding and enhancement of reactant mixing, the latter possibly offering reductions in emissions of unwanted combustion products such as CO and carbon clusters (soot). More specialized applications to combustion in microgravity might also be possible.

We have previously shown that modest applied voltages[3] are capable of driving the flame from laminar (stable, conical flame geometry) to a wrinkled laminar geometry (cellular; multiple laminar flamelets), with some evidence of the approach to turbulence regimes. For all fuels studied here, over varying ranges of flow rates and equivalence ratios between 0.7 – 1.5, still larger electric-field-induced modifications of the combusting flame front stability have been observed and documented. Specifically, the electric pressure effect[3] has been used to force pre-mixed light hydrocarbon/air flame fronts to transition from flow-induced hydrodynamic-instability-dominated behavior, to field-stabilized laminar flow, and finally to field-induced thermal-diffusive-instability-dominated oscillatory and turbulent flame fronts. Applied potentials up to 6 kV over a 40-mm gap encompassing the flame front have been used to force that transition sequence, and the flame response has been quantified by electrical and spectroscopic measurements, and by 2-d direct and spectroscopic imaging techniques. The effects are best described from a combustion dynamics viewpoint as a continuously variable electric-field-induced diminution of the flame Lewis number[3] from typical values (> 1), past the critical flame Lewis number[4,5] (0.7-0.9), to thermal-diffusive-instability dominated values below the critical Lewis number at the higher applied potentials.

The electric field acts directly on the highly localized, chemionization-derived $H_3O^+$, $HCO^+$ and other positive ions in the flame reaction zone, significantly increasing their mass diffusivities and thereby modifying the Lewis numbers of the ionic species.
The observed result is a diffusive-thermal instability, typical of flames with global Lewis numbers less than one; it is most likely caused by the production of H atom and OH radicals by the dissociative recombination of $\text{H}_2\text{O}^+ + e \rightarrow \text{H}_2\text{O} + \text{H}$ or $\text{H}_2 + \text{OH}$, and $\text{HCO}^+ + e \rightarrow \text{H} + \text{CO}$ near the burner head, which is the cathode surface for the polarity of the applied voltage. The increased differential diffusional velocity of the reaction zone positive ions strongly affects the bulk flame speed, and the thermo-diffusive instability results in the flame front collapsing toward the burner head and taking on a wrinkled laminar geometry. High-speed two-dimensional imaging of the flame response to a pulsed voltage have shown that the characteristic time required to complete the field-induced flame stretch is on the order of 5 ms, somewhat more rapid than that found in previous work, but still indicative of the fluid mechanical nature of the flame response to the external field. Such images also show electric-field-induced unburned pocket formation which is a characteristic of turbulent combustion.

At the higher continuous bias voltages used in this study (up to 6 kV), the flames are observed to make a further transition from cellular to highly turbulent flame fronts over a wide range of flow/bias voltage conditions. In much of the previous work, the effects of DC electric fields on flames was described in terms of the body force known as the ionic wind, which has been shown to be capable of providing a maximum pressure difference across the flame of only 0.0004 atm. This applied voltage-current induced pressure change is not sufficient to cause the observed change in either the flame speed or the flame heat release rate fluctuations through direct pressure effects, thus the introduction of the notion of electric pressure. The DC-field-induced transition from laminar to highly turbulent combusting flows is quantified by direct high-speed imaging of the flame heat release rate fluctuations by measuring the chemiluminescence intensity using 200 µsec gated ICCD wide band width, and narrow band spectral measurements of flame radical species $\text{CH}^*$, $\text{C}_2^*$ and $\text{OH}^*$ emission intensities. Typical flame images are shown in Figure 1. Fast Fourier Transforms (FFT) of the oscillating flame front emission signals were used to document the large differences between zero field (laminar flow) and DC-field driven turbulence conditions. Additionally, under DC applied voltage, the collapsed reaction zone is observed to undergo a transition from a low amplitude and low frequency oscillation typical of a laminar flame, to a large amplitude high frequency oscillation, similar to a thermo-acoustically perturbed flame. Efforts to quantify these newly observed oscillatory modes of the flame front are briefly discussed below, and preliminary results are shown for propane/air at an equivalence ratio of 1.2 and a flow speed of 1.5 m/s in Figure 2.

The burner used in this experiment produces a stable, laminar flow resulting conical reaction zones with a base diameter of 17 mm and height that varies with equivalence ratio and overall flow rate. Total flow rates varied between 12 – 30 slm, which provides flow velocities on the axis of the burner from roughly twice propane’s stoichiometric laminar flame speed (0.44 m/s) to well past the burner’s blow off flow rate at zero applied voltage. At the higher flow rates, flow Reynolds numbers of roughly 2500 (incipient turbulent flow) are achieved. DC fields are applied to the flames by connecting a 0-6 kV power supply across the grounded burner head, and positively biasing an electrode centered typically 40 mm above the grounded burner.

The magnitude of the current drawn, for a given applied voltage, with our anode configuration is nearly an order of magnitude higher than previously reported.
measurements for electrically biased pre-mixed hydrocarbon flames. Since the current is mobility limited, the larger current drawn at lower applied voltage will result in a significant increase of the positive ion flux in our experimental conditions, as compared to the previous measurements. If the dissociative recombination driven production of limiting light reactants is the source for the lowering of the Lewis number below a critical Lewis number, this could be one of the most important reasons why it is possible for us achieve a level of thermo-diffusive instability that was not possible in previous measurements.

Further improvements in the coupling of the electric field to the flame, and the use of a higher voltage DC power supply that permit larger electrical energy input to the flame are found to drive the flames toward increasingly turbulent conditions (i.e. increasingly smaller scale turbulence features) associated with the distributed reactions regime. At particular higher electric field/flow combinations (3-6 kV applied voltage), the electric pressure effect causes the flame front to undergo large amplitude, high frequency oscillations over large portions of the flame front surface. These conclusions are supported by high speed, two-dimensional images of the flame front which includes CH* emissions at 431 nm, OH* emissions at 308 nm, and C$_2$ emissions at 516 nm. All recorded images show that OH and CH radicals are redistributed by the applied electric field in essentially the same way as the overall broadband flame emissions. However, the total integrated OH and CH emission intensities are independent of the applied field. Given that the electric field depresses the flame front toward the burner head, this behavior suggests that the effect of the thermo-diffusive instability is also to increase the combustion intensity. Additionally, Figure 2 shows a series of fast Fourier transforms (FFTs) of broadband and spectrally filtered flame front emissions, as well as the FFT of the oscillating current drawn from the external bias supply. Again, the experimental conditions are the same as those used for the imaging data shown in Figure1 (21 slm overall flow and equivalence ratio of 1.2) except that the bias used was only 4 kV.

The top left FFT of Figure 2 shows the very low amplitude, featureless power spectrum of the flame’s broadband emissions under zero bias. It indicates stable laminar flow. With a 4-kV bias the flame front is driven into a large amplitude resonate-like oscillation with a fundamental frequency near 300 Hz and harmonics ranging up to 1.8 kHz. Essentially the same responses are seen again from broadband emissions and those corresponding to chemi-excited CH, C$_2$ and OH. Furthermore, the current passing through the flame also exhibits a power spectrum that is essentially identical to that of the flame front emissions. This is indicative that the voltage-current behavior is also dependent on the heat release rate modulation similar to the chemiluminescence intensity.

References
Figure 1: Direct high-speed (200 μs) images of a propane/air flame at a Reynolds number near 2500 as a function of bias voltage. The progression shows field-induced transition from a hydrodynamic instability dominated flame front (a), to a laminar flow (b), to an oscillatory mode (c), and finally to a highly turbulent flame front in image (d).
Figure 2: FFTs of broadband emission from unperturbed flame along with DC electric field induced perturbed flame, filtered CH, C₂ and OH chemiluminescence, and current fluctuations are shown.
Appendix II

Ionic Wind versus Electric Pressure – note added in proof

In Section 3.2 the limitations\textsuperscript{1-4} of the ionic wind theory were briefly discussed. The electric pressure effect\textsuperscript{5} has been invoked in place of the ionic wind theory to account for the observed dramatic effects of DC electric fields on pre-mixed propane/air flames when the field is well coupled to the flame. The electric pressure effect was proposed\textsuperscript{5} because the currents that can be drawn through closely coupled flames are larger than the theoretical maxima\textsuperscript{1-4} predicted by the ionic wind theory, and require better than an order of magnitude smaller applied field than that needed to achieve the maximum ionic wind currents. The electric pressure effect was so named\textsuperscript{5} because of the striking similarity of the observed close-coupled-electric field effects to those of the well-known singing flame\textsuperscript{6}, which is due large pressure variations due to an acoustic interaction between a flame and an enclosing combustion chamber. This thesis has documented flame stability modifications due to the electric pressure effect over a wide range of equivalence ratios for methane, propane and butane fuels.

Since the production of the foregoing thesis, further work using pulsed-DC electric fields on pre-mixed light hydrocarbon flames has been performed that definitively eliminate the ionic wind as a possible mechanism for the stability modifications described here. Substituting a pulsed-DC power supply for the DC supply shown in Figure 2.1.1, and imaging the entire flame reaction zone through a 10-nm bandpass filter centered at 308 nm onto a photomultiplier tube allowed the simultaneous measurement of current through the flame, applied pulsed voltage, and the resultant chemi-luminescence emission from excited OH* radicals. Using propane as the fuel, a flow velocity of 1.3 m/s, and an equivalence ratio of 1.1, those three signals are shown in Figure II.1. As shown, the excitation pulse is approximately 4.2 ms in duration and has an amplitude of only 2.6 kV. The average current during the pulse is only 260 μA. The OH* chemi-luminescence is modulated by better than 10%, suggesting a global flame chemistry modification that has heretofore not been observed in the continuous-DC applied field work that is the subject of this thesis. Please note that the modulation of the
Figure II.1. Current, bias voltage and total OH* emission intensity for a 4.2 ms pulsed bias of a pre-mixed propane/air flame at an equivalence ratio of 1.1. Note that the OH* chemi-luminescence signal continues to increase after the bias pulse is terminated.
OH* emission signal reaches its peak more than a millisecond after the bias pulse is terminated.

Figure II.2. shows a series of high-speed direct images of the flame front response during the pulsed-bias excitation just discussed in relation to current, voltage and OH* emission profiles of Figure II.1. Those images are 100-μs exposures spaced 1 ms apart (read left-to-right, top-to-bottom), with the top left image coinciding with the voltage rise of Figure II.1. The right-most image in the second row, and all later images in that Figure show the response of the flame after the bias pulse was terminated. Since the ionic wind is “a drag force exercised by ions which travel at velocities orders of magnitude greater than those of the gas but transfer the additional momentum they acquire from the effect of the electric field during each mean free path in the collision which terminates it,” it is clear that the ionic wind should terminate within a few collision times after the bias pulse ends. At atmospheric pressure and roughly 2000 K, a few collision times amounts to less than a microsecond. Thus, since the de-stabilization of the flame documented in Figures II.1 and II.2 persists for several milliseconds after the bias pulse ends, the ionic wind cannot possibly be the operative physical mechanism.
Figure II. 2. Direct high-speed (100 μs) images spaced 1 ms apart of a propane/air flame subjected to a 4.2-ms long, 2.6 kV bias pulse (read l-r, top to bottom). Note that the last seven frames show the flame response after the bias pulse is terminated.
References: